

2011 – Fuel Cells

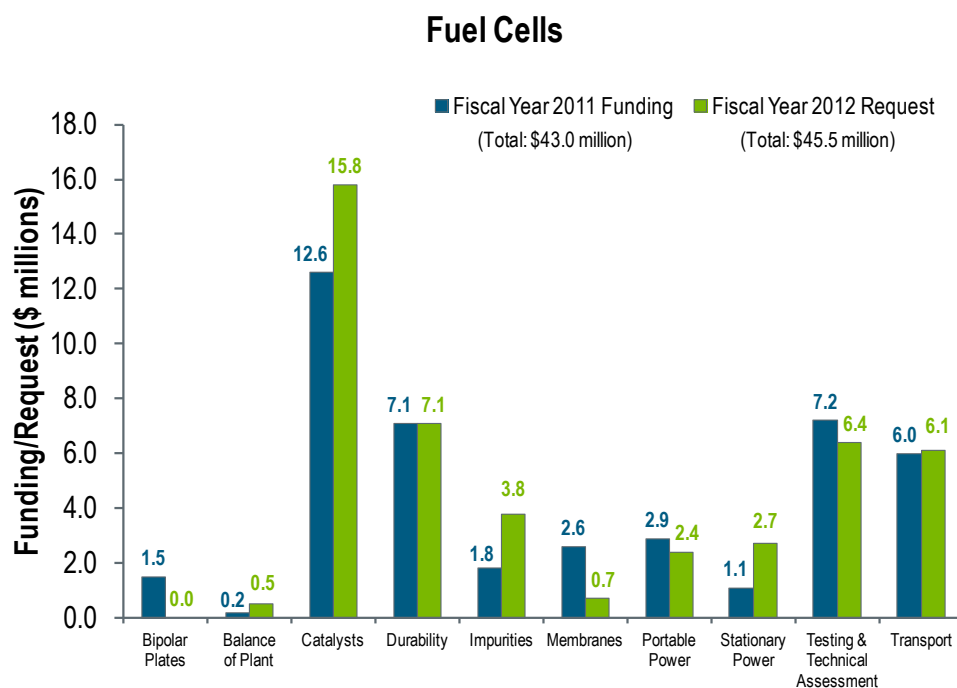
Summary of Annual Merit Review of the Fuel Cells Sub-Program

Summary of Reviewer Comments on the Fuel Cells Sub-Program:

The reviewers considered the Fuel Cells sub-program to be well managed with an appropriate portfolio of projects that address the critical technological issues. The sub-program was praised for the quality of projects and research scientists working in strong, well-managed teams across industry, laboratory, and university settings. The sub-program was also praised for effectively adjusting to shifting priorities and reduced budgets. However, the reviewers felt that some unsuccessful projects were still being funded and recommended that a more rigorous go/no-go decision making process be developed.

Fuel Cells Funding by Technology:

The Fuel Cells sub-program received \$43 million in fiscal year (FY) 2011 and approximately \$45.5 million is requested for FY 2012. The sub-program continues to focus on reducing costs and improving durability with an emphasis on fuel cell stack components. The funding profiles for FY 2011 and the FY 2012 request are very similar, with some projects in membranes and bipolar plates ending in FY 2011.



Majority of Reviewer Comments and Recommendations:

At this year's review, 73 projects funded by the Fuel Cells sub-program were presented and 67 were reviewed. Projects were reviewed by between five and eight reviewers with an average of six experts reviewing each project. Reviewer scores for these projects ranged from 2.0 to 3.7, with an average score of 2.9. This year's highest score of 3.7 was better than last year's highest of 3.6. Both the average score of 2.9 and the lowest score of 2.0 for 2011 were similar to 2010's average score of 3.0 and lowest score of 1.9, respectively.

Analysis/Characterization: Eight projects were reviewed and received an average score of 3.3 with two of these projects ranked in the top five of all projects in the sub-program. Reviewers commented that one project continues to add significant value to the DOE Hydrogen and Fuel Cells Program by providing a solid basis for decision-making

for research and development and evaluating progress toward the critical cost target, while another project provides a realistic model of fuel cell systems, exposing technological advances and shortcomings of fuel cell technology and providing a basis for cost analysis. In addition, while reviewers commended a project involving neutron imaging for providing critical analytic capabilities, they expressed some concern that features of advanced membrane electrode assemblies (MEAs) may be smaller than the neutron imaging resolution.

Water Transport: One water transport project was reviewed and received a score of 2.3. Reviewers commended the project for its overall approach, noting that the project combines modeling with experimental validation and involves effective collaborations. They observed, however, that a complete understanding of water transport issues has not been achieved and they expressed concern regarding discrepancies between the model and experimental data.

Impurities: This year two impurities projects were reviewed and received an average score of 2.7. Both projects were commended for having strong teams. Reviewers remarked that the impurities studies on both the fuel and air sides of the fuel cell are very important to the success of the Program. However, they noted that the study on the air side is making slow progress, perhaps due to the systematic approach to impurity selection. They commended one project for completing a down-select from 187 airborne contaminants, 68 indoor pollutants, and 12 roadside species that may have potential adverse effect to the fuel cell performance. The reviewers recommended that fuel cells be cycled repeatedly to failure and that the principal investigators (PIs) should carry out post-mortem diagnostics of the MEAs.

Membranes: Seven membrane projects were presented and reviewed with an average score of 2.8. The reviewers noted that progress was made toward meeting DOE targets, particularly in conductivity; however, durability remains an issue. While some projects made progress in decreasing linear swelling, improving chemical stability, and improving durability, other membrane projects needed to show improvement in mechanical durability and decreased swelling. Most of the membrane projects are ending; however, one new innovative project on corrugated membrane structures was initiated in FY 2011.

Catalysts: The average score for the 13 catalyst projects was equal to the sub-program average of 3.0. The reviewers commended projects for making advances in cathode catalysts and supports, as well as for the progress that has been made in thin film electrolyte technology. In addition, they observed that the required total platinum group metal (PGM) content continues to fall as a result of sub-program research, and higher-risk non-precious metal catalyst development efforts show progress toward mass-activity targets. However, some reviewers were concerned that the best anode and cathode compositions and structures would not match when combined in a cell/stack. Reviewers recommended less work on developing new catalytic materials and more on characterizing and diagnosing existing catalyst formulations. They also suggested using modeling to narrow the scope of materials being evaluated experimentally.

Transport Studies: Six transport studies projects were evaluated and received an average score of 3.0. The highest-rated project focused on the investigation of micro- and macro-scale transport phenomena for improved fuel cell performance considering both baseline and next-generation material sets. The reviewers praised this project in particular for relevance, approach, and progress achieved. Overall, most reviewers noted that progress has been achieved for projects in this area. However, some reviewers raised a concern regarding the lack of quantitative agreement between modeling and experimental validation for some of these projects. Also, in general, some of the reviewers felt that it was unclear how the various models relate to each other.

Degradation Studies: The average score for the five projects in this area was 3.3. The reviewers observed that much progress was made in degradation studies. They emphasized that durability improvements are critical to the Program and that these projects are effectively addressing durability issues through investigation of the degradation mechanisms of membranes, bipolar plates, catalysts, and electrodes using both modeling and experimental methods. They also recognized the value of the accelerated stress-testing methods being developed by two projects. The reviewers expressed some concern that more operating conditions, materials, and design information need to be shared, and that some of the studies were too specific to one fuel cell design or one particular manufacturer.

Hardware: Two hardware projects focusing on bipolar plates were evaluated and both received scores of 2.7. Overall, reviewers felt that developing low-cost and durable bipolar plates is critical to achieving sub-program

targets. Reviewers were impressed with some of the plated technologies evaluated in one project, but were concerned that the project lacks focus for the available amount of time and resources. Reviewers praised the management and progress in another project, but were concerned about the chemical stability of the materials being tested.

Balance of Plant: One balance of plant project was evaluated and received a score of 3.5. The reviewers felt that the project and its work in materials development were on track and showed promise toward meeting technical objectives. The reviewers recommended that the project prove long-term material stability and show applicability to stationary fuel cell systems.

Distributed Generation: Two distributed generation projects, dealing with solid oxide and polymer electrolyte fuel cells, were reviewed and received an average score of 3.0. Reviewers praised the significant progress that was made in developing and demonstrating a tubular solid oxide fuel cell system for stationary applications, in terms of performance, cost, and durability. They observed that advancements were achieved at the cell, stack, and system level. However, some reviewers thought that the cost and lifetime of the system is not currently competitive.

Portable Power: Four portable power projects were presented this year and received an average score of 2.9 with scores ranging from 2.7 to 3.5. The highest-rated project focused on improving the catalytic activity and durability of platinum ruthenium for direct methanol fuel cells. Reviewers specifically praised this project for featuring an excellent team and a sound approach to materials development and evaluation. The remaining three projects focused on MEA materials development, including membrane and anode catalysts. Reviewers commended these projects for offering a rational pathway toward component development and for assembling teams with the appropriate expertise. However, it was also noted that more MEA testing and development is required for component integration.

Innovative Concepts: Three projects presented this year fall under the category of innovative concepts and received an average score of 3.2 with scores ranging from 3.0 to 3.5. The projects involve novel approaches and include strategies for energy storage, reduced catalyst loading, and improved cell durability. Reviewers found the projects in this category to be relevant and noted that they all address critical DOE targets. The reviewers noted that one project—involving advanced materials for reversible solid-oxide fuel cells—has exceeded its targets for performance, degradation, current density, and operation duration. However, some reviewers suggested that greater interaction with utility companies and academic institutions would improve the project. Another project, involving anion-exchange polymer electrolytes, was praised for being a well thought-out, carefully planned, and systematic study of potentially useful exploratory technology; however, it was recommended that the project focus on improving membrane durability and the team should develop collaborations with groups outside the national labs. The project involving ceramic supports for polymer electrolyte fuel cells was found to be well-focused on a good range of materials, but reviewers felt that the researchers need to improve the quality of their electrochemical characterization techniques.

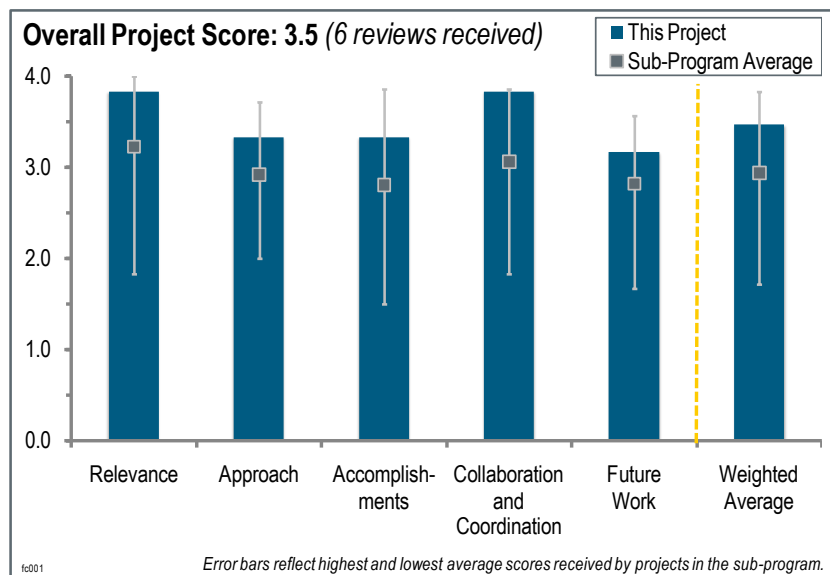
Project # FC-001: Advanced Cathode Catalysts and Supports for PEM Fuel Cells

Mark Debe; 3M

Brief Summary of Project:

The overall project objective is to develop a durable, low-cost, high-performance cathode electrode (catalyst and support), that is fully integrated into a fuel cell membrane electrode assembly (MEA) with gas diffusion media, fabricated by high-volume capable processes, and is able to meet or exceed the 2015 U.S. Department of Energy (DOE) targets. Focus topics for the past year included: (1) improving water management for cool/wet transient operation through materials, electrode structure, and boundary condition optimization and understanding; (2) continuing to develop multiple

strategies for increasing nanostructured thin film (NSTF) catalyst activity, surface area, and durability, with total loadings of less than 0.25 mg Pt/cm² (milligrams of platinum per centimeter squared) per MEA; (3) focusing on key NSTF alloy catalyst compositions and process improvements discovered and developed in 2009 and 2010; (4) continuing accelerated stability tests to benchmark durability of new NSTF MEA configurations; (5) down-selecting components for new 2010 “best-of-class” MEA for final stack testing in 2011; and (6) continuing fundamental studies of the NSTF catalyst activity for oxygen reduction reaction, and methods for achieving the entitlement activity for NSTF catalysts.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.8** for its relevance to DOE objectives.

- The catalyst work in this project is directly relevant to DOE's goal of reducing platinum group metal (PGM) content (and cost) while improving performance and durability.
- The project's relevance is related to its focus on cost (both PGM and manufacturing) and durability.
- This project has been highly relevant to DOE Hydrogen and Fuel Cells Program objectives from its inception and continues to be relevant in its last year.
- This project from 3M has always had good productivity and progress.
- There is no more relevant topic in automotive polymer electrolyte membrane (PEM) fuel cells than the development of a more active and durable cathode catalyst that is robust in all operating conditions. The project does not use technology of questionable relevance toward automotive application. Instead, it moves in the direction of higher coordinated Pt atoms that enable higher specific activity and lower surface energy. The latter hopefully allows for lower dissolution.
- The project was adequately covered and summarized. The project involved water management of the electrode and increasing the catalyst activity of low-loaded electrodes (< 0.25 mg Pt/cm² total).

Question 2: Approach to performing the work

This project was rated **3.3** for its approach.

- The approach appears to be sound and is generating good data.

- The approach in the last year of the project is very good—as a problem is identified or pointed out by reviewers, 3M formulates a path forward to resolve the issue. One example is the modification of the anode gas diffusion layer (GDL) to facilitate water removal from the anode during low-temperature operation without having to resort to sub-ambient operation to prevent flooding. 3M wisely chose to build a rainbow stack with a variety of component choices in order to down-select those with the best chance of success in testing of the final deliverable stack to DOE. The components chosen may not hold the most long-term promise, but they have shown acceptable performance and life to qualify for the final build in this project. 3M has emphasized cost effectiveness and manufacturability throughout the development process.
- This reviewer believes that direct metal deposition is the only technology that can simultaneously meet cost (low PGM) and manufacturing targets. That said, the project did not show or mention data on the uniformity of the metal deposit over the web or the stability of the allowed ratio(s) over the web. The author also does indicate that for Pt-Ni systems, the X:Y ratio can severely impact performance over a tight range, so this analytical uniformity result could be important.
- The part of the approach carried over from prior work is fundamentally solid, and includes high specific activity Pt or Pt alloys combined with durable supports to allow low-PGM-loaded catalysts for PEM fuel cells. 3M recognizes the problems in the past with water management and its approach continues to work on this issue. The approach accounts for manufacturability by investigating more efficient fabrication processes. Perhaps one weakness in the approach might concern the low open circuit voltage (OCV) that NSTF-containing MEAs often—but not always—demonstrate. Some crossover and shorting data within the presentation hint that more should be done to address lower OCV.
- The project team has addressed all of the issues, although the solution to the water management problems might be an issue on a systems level with regard to changing how water is rejected from the anode. Implementing MEAs in practical systems seems difficult, as they have not yet been accepted by industry.
- More information on the processing parameters and steps for the catalysts would be instructive. The test matrix on slide 26 bears a risk of choosing the best anode and cathode compositions and structures then finding that they do not match when combined in a cell or stack.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- The progress is as expected. Water management appears to be a major issue, especially at low-temperature, low-pressure operation. The correct hydrophobicity level of the catalyst and membrane is required to ensure adequate ion migration while not generating a gas barrier. “Whiskers,” generated in the formation and operation of a Pt-Ni catalyst, appear to be an anomaly, but do not appear to get worse with operation.
- The project achieved major accomplishments over the past year and demonstrated significant progress. The project team identified a viable path to resolving the low-temperature flooding issue that involved modifications to the anode GDL. Processing improvements yielded smoother catalyst surfaces with significant activity gains for at least some alloy compositions. The project met the DOE mass activity target in some cases, depending on the testing protocol and loading measurement. A greater understanding of the Pt-Ni system was developed through collaboration with other organizations. The amount of work involved in selecting the components for the 2010 best-of-class MEAs for final stack testing is astounding.
- Perhaps the primary accomplishment of the past year was the fundamental understanding of how to manipulate the anode GDL to achieve low-temperature performance at near-ambient conditions. While the GDL itself is not worthy of stack testing due to electrical resistance, this achievement still represents an advance that can lead to the right GDL. Accomplishments also include passing the OCV and support corrosion accelerated stress tests (ASTs), developing more cost-efficient catalyst deposition processes (P1), and discovering higher mass activity with annealing for Pt₃Ni₇. High current performance and stability of the Pt₃Ni₇ alloy remains a problem. The failure to meet the voltage cycling AST with the P1-PtCoMn material still needs to be addressed.
- Slide 32 summarizes 3M’s status against DOE technical targets and shows that most targets have been met. It is recognized that all targets may not have been met by the same formulation.
- The support stability and performance with respect to total platinum loading density (g/kW) is unquestionable. Alloy stability meets some, but not all, of the DOE targets.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.8** for its collaboration and coordination.

- There is substantial collaboration with industry partners.
- This project features a broad array of partners from basics and alloy discovery to full-scale stack testing under different flow field designs and operating conditions. There has been outstanding coordination among the partners and delineation of responsibilities.
- The collaboration includes experts in catalysis, ex-situ characterization, systems modeling, fuel cell manufacturing and systems integration, and automotive original equipment manufacturers (OEMs).
- Collaborations are strong from both the fundamental understanding viewpoint and the OEM developer side.
- 3M does a great job of teaming up with academia and others where it needs help. The project now includes teaming with the automotive industry, but the results from that collaboration were not clear.
- 3M has consistently worked with other parties to improve activity by developing new alloys. This collaboration includes the combinatorial studies at Dalhousie. A catalyst project should have collaborators on microscopy, and 3M has overachieved with its collaborations with the California Institute of Technology and the National Aeronautics and Space Administration's Jet Propulsion Laboratory. 3M has been generous in providing information or confirmations toward system and cost analysis projects that generate assumptions centered on NSTF. The collaboration with General Motors (GM) has provided perspective on the needs of a stack OEM or integrator.

Question 5: Proposed future work

This project was rated **3.2** for its proposed future work.

- The future work plan for the remaining six or seven months of the project is appropriate for a successful final stack test. Other component choices may hold more long-term promise, but they are less likely to yield as much success.
- 3M will build a "rainbow" stack using six cell/stack material sets for long-term durability (3,300 hours) testing at GM until the project finishes at the end of 2011.
- The work plan is appropriate, but it is uncertain whether there is sufficient time given the scale-up validation to address anode GDL optimization for start-up and wet conditions. Similarly, it is ambitious to achieve all of the catalyst durability criteria in the remaining time.
- The project is in the validation phase and is nearing its end.
- Given some of the remaining project issues, it may be a missed opportunity to spend the final months of the project on a stack down-selection and assembly task. Instead of stack testing, it would be more interesting to see if there is a way to address the high current performance of Pt_3Ni_7 , or investigate methods of stabilizing the alloy. It would also be interesting to see if PtCoMn could be further stabilized with processing, and why the P1 process—which created larger domains—did not produce a statistically significant advance in stabilization (as measured by the voltage cycling AST).

Project strengths:

- Hardly any company has shown greater enthusiasm for reporting all of the critical experimental details and context. While some projects may leave a reviewer wishing for clarification on a parameter, 3M can be counted on to report conditions. In terms of experimental throughput, 3M has demonstrated the ability to process through hundreds of samples and experiments on the way to an important advance. The principal investigator has consistently driven off the concept of using a high specific activity, bulk-like phase of Pt or Pt alloy, along with corrosion-resistance support. Even at a concept level, this project meets targets that other catalyst projects have to work hard to achieve. In terms of responsiveness to assigned targets, 3M moves aggressively to meet the targets and report results when given a target or a test to perform.
- The technical support, management approach, and collaboration are areas of strength for this project. It appears to be a professional job, as expected from a Fortune 500 company.
- This reviewer considers the modified deposition process with Surface Energy Treatment a breakthrough in being able to control deposit size independently of load range.

- This project has the right mix of fundamental understanding and end-user, commercial awareness. There is an emphasis on finding pathways to meet all DOE targets simultaneously, but in cost effective ways suitable for high volume processing. The project features an outstanding record of accomplishments and attention to reviewer suggestions and comments.
- 3M continues to be productive with its NSTF MEAs. This year it reported a high-performance alloy that seems to be durable and acceptable in applications.

Project weaknesses:

- The reviewer felt that there were no weaknesses.
- If both a 25-micron membrane (needed for flow of the product water back to the anode) and these super-thin metal films are routinely manufactured, it is unclear if the tolerances of the materials and stack elements (e.g., GDL, bipolar plates, and gaskets) will be sufficient to avoid shorting, pinhole formation, hot spots, etc. While a roll-to-roll metal coating method is worthy of praise, the metrics for manufacture (i.e., variations) and whether the method is sufficient.
- The talks about these catalyst/MEA systems seem so great, but it is unclear why the MEAs are not widely embraced by industry. Presumably, this is due to the flooding problem, which has plagued this project for years.
- The project has produced a high mass activity alloy, but the issues that have come with it at high current density—as well as the instability—prevent the high mass activity from being exploited. In terms of lower performance at low current, many of the polarizations shown seem to indicate that OCV is low. There is some data that suggests that crossover or shorting resistance could be improved. The project may leave some questions unanswered, such as whether PtCoMn could be further stabilized with processing, or whether an anode GDL that both allows exit and provides for high electrical conductivity could be fabricated.

Recommendations for additions/deletions to project scope:

- The reviewer had no recommendations.
- The project is over—this reviewer hopes that it transitions successfully to a fuel cell product.
- The project team should investigate manufacturing metrics, including variation.
- There is not much time or room for additional work. It would be nice to develop means to increase limiting current in the Pt_3Ni_7 catalyst.
- The project has limited time remaining, but it would be preferred to have some of the remaining project resources directed toward the following:
 - The high current performance of Pt_3Ni_7 ,
 - The stability of Pt_3Ni_7 ,
 - Understanding why PtCoMn did not meet voltage cycling targets with larger grains,
 - Anode GDLs that provide water management without an ohmic penalty, and
 - Raising OCV or low current performance.

Project # FC-002: Highly Dispersed Alloy Catalyst for Durability

Vivek Murthi; UTC Power

Brief Summary of Project:

The overall project objective is to develop a compositionally advanced cathode catalyst on a support that will meet U.S. Department of Energy (DOE) activity, durability, and platinum group metal loading targets in a structurally optimized membrane electrode assembly capable of performing at a high current density. Tasks include: (1) dispersed alloy catalyst development; (2) core-shell catalyst development; and (3) carbon support investigation.

Question 1: Relevance to overall U.S. Department of Energy objectives

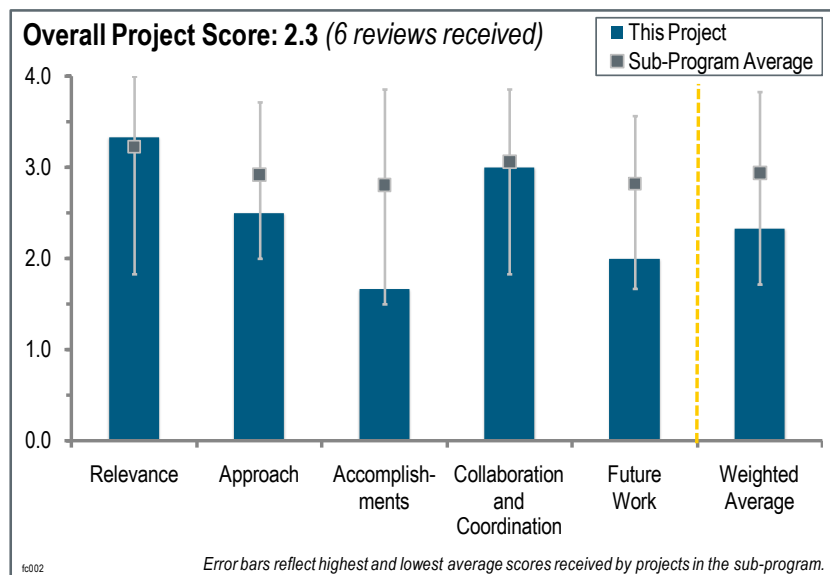
This project was rated **3.3** for its relevance to DOE objectives.

- The project is relevant to DOE Hydrogen and Fuel Cells Program objectives related to reducing polymer electrolyte membrane (PEM) fuel cell costs by reducing the precious metal loading and increasing fuel cell durability.
- High activity, robust catalysts are critical for enabling fuel cell system commercialization.
- The project addresses fuel cell cost and durability.
- The motivation of the project is relevant to DOE objectives, but the approach taken may be questioned.
- The project objectives are broadly aligned with DOE objectives, but the objectives have not been met.

Question 2: Approach to performing the work

This project was rated **2.5** for its approach.

- In general, the approach—combining high activity alloys, core-shell catalysts, and robust supports—makes sense. However, the investigators did not share any specifics about the approach for core-shell catalyst or stable carbon supports. Also, the focus and impact of the modeling work was not included in the presentation, so it is difficult to assess.
- Although the experimental approach is well designed, the project has a significant weakness, as both the performance and durability of the Pt₂IrCr catalyst do not exceed those of the Johnson Matthey (JM) carbon supported platinum (Pt/C) catalyst.
- The use of iridium (Ir) in polymer electrolyte membrane fuel cell (PEMFC) catalysts is always a concern, given the rarity of the element. There also appears to be performance shortfalls not attributed to the catalyst, such as activity, magnitude of current density in air, and stability.
- Weaknesses of the approach include (1) the fact that there appear to be separate activities in core-shell work (now stopped) and ternary alloys that are unrelated and uncoordinated, and (2) the lack of contingency plans in the event that nothing meets the objectives. The project continues to its conclusion regardless.
- Using Ir in the catalyst may be risky. The current cost benefit is low compared to Pt, and the Ir supply is very limited, so cost is likely to increase if a substantial market for it develops.



Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **1.7** for its accomplishments and progress.

- The initial results indicated improvements in activity versus Pt/C. However, benefits have not translated to MEA performance. New results (slides 9 and 10) suggest that the Pt-Ir-Cr catalyst shows little to no performance or durability benefit over platinum in MEA testing. Performance at high current density and durability both appeared worse than JM Pt/C. Data on the distribution of metals in the catalyst particles is lacking and the project team did not illustrate any plans to obtain this data. Data regarding Ir or Cr dissolution was not presented.
- The team has achieved a significant amount of work, but it does not look like it has enough time to overcome the barriers on durability, as the project ends in 2011.
- There seems to have been limited progress in addressing the concerns of last year's reviewers.
- The membrane electrode assembly (MEA) performance struggles to reach the baseline. There is no clear value. A great deal of time has been spent on core-shell materials. It is not clear if MEA optimization will reach the targets. This reviewer sees no input from the modeling activities.
- After four years, the mass activities are far from DOE targets. The fuel cell performance at high current densities is well below standard Pt electrodes at the same loadings. Even the low current density performance only seems to match platinum.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.0** for its collaboration and coordination.

- This project features well-coordinated collaboration with partners from academia and industry.
- The partners include material suppliers (JM), national laboratories (Brookhaven National Laboratory [BNL]), an end user United Technologies Corporation (UTC), and academia. It is a shame that there is so little sign of the academic contribution.
- The investigators are collaborating with JM, BNL, and Texas A&M University. It does not appear that catalyst technology is transferring well from BNL to scaled-up production at JM and UTC.
- It was unclear from the presentation and slides available how the subcontractors have contributed to the project. All of the data presented seemed to be from UTC.

Question 5: Proposed future work

This project was rated **2.0** for its proposed future work.

- The future work should be less focused on optimization of the Pt₂IrCr catalyst because of problems with its stability.
- The work is closing in on the four-year mark with 90% of the project complete. Some of the barriers to implementation have not been surpassed, and it remains to be seen how useful this work will be in attaining DOE PEM fuel cell performance and durability goals.
- The proposed work appears to aim at simply completing the tasks and building a stack. It is not expected to meet the targets or overcome any of the identified barriers. For example, there are no details regarding the plan to solve the optimization of the catalyst layers to improve mass transport at high currents.
- Short stack testing is not justified (task one) until the high current density performance is improved (task three), which will involve MEA optimization through ink and processing optimization. The tasks for membrane and gas diffusion layer selection are a distraction. The value of the continued core-shell work (task two) is unclear.
- It is not clear how the proposed future MEA optimization will help improve the activity or durability of the PtIrCr alloy catalyst.

Project strengths:

- The strengths of this project include its well coordinated collaboration between industry and academia, fast progress in fuel cell optimization, and fast progress in down-selecting durable supports.
- This project brought together key industrial partners to develop novel catalytic systems and bring them to an end product demonstration. Involving academia in the fundamental modeling studies brings in defined skills that are not available in industry to further understanding and guide research.
- This project's strengths include its ability to provide catalysts at quantities suitable for fuel cell testing, and generate fuel cell performance and durability data at relevant conditions.
- This project's biggest strength is its team of a fuel cell provider, catalyst provider, and catalyst developer.
- The project has been performed by an excellent team with good collaboration and guidance from industry leaders.

Project weaknesses:

- The project is still focused on Pt₂IrCr, which performs poorly in the fuel cell operating region. The investigation of the structure-property relationship requires more attention.
- The project, particularly the demonstrated catalyst stability, appears to be lagging behind DOE targets. Catalyst costs have not been addressed.
- The initial plan was uncoordinated, and there seems to be no link between the core-shell tasks and the Ir alloy tasks. There appears to be no clear plan to address the technical barriers that were identified during the project, or a contingency or backup plan.
- This project has not been successful at meeting its goals. While there was a solid overall approach, it is hard to see the reasons it was not successful because no details were provided about the core-shell catalyst, stable carbon supports, or modeling work.
- The project's inability to get a scaled-up version of core shell catalysts with comparable improvement in activity over Pt in an MEA was observed in rotating disk electrode experiments.
- Unfortunately, it appears that the choice of catalyst (Pt-Cr-Ir) is perhaps not very good in terms of stability, and has marginal, at best, advantages over Pt in terms of activity. While this project was well directed technically, the resulting outcome is more related to eliminating a system of oxygen reduction reaction catalysts rather than identifying a new and outstanding class of catalyst.

Recommendations for additions/deletions to project scope:

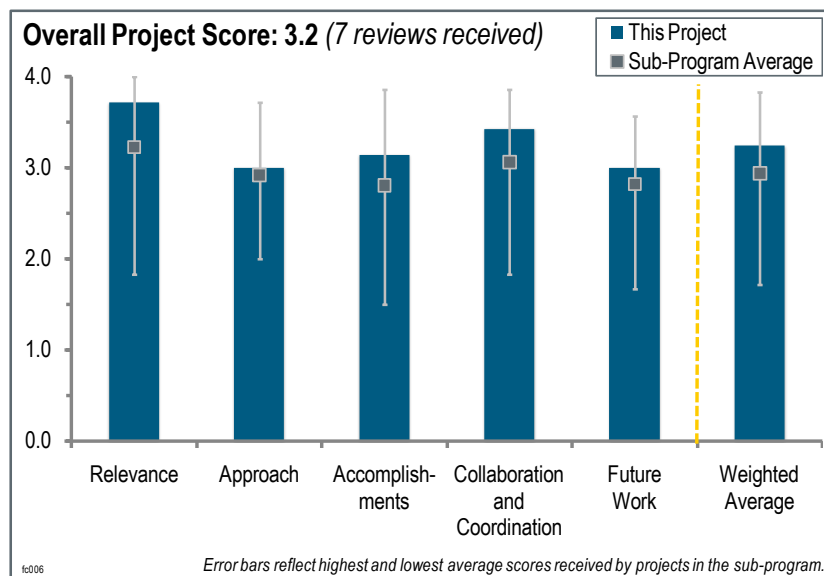
- This project is close to completion and has a no-cost extension to complete the delayed stack testing. This reviewer recommends that the stack testing be completed and the project be allowed to finish. The stack testing should include start-up/shut-down accelerated testing.
- The reviewer has no recommendations as this project is in final stages.
- The investigators should not conduct stack testing. The focus should be on improving the activity of the base catalyst, which is still less than one-half of the DOE target, and improving the high current density performance of the MEA with C4 support.

Project # FC-006: Durable Catalysts for Fuel Cell Protection During Transient Conditions

Radoslav Atanasoski; 3M

Brief Summary of Project:

The overall project objective is to develop catalysts that will enable proton exchange membrane fuel cell systems to weather the damaging conditions in the fuel cell at voltages beyond the thermodynamic stability of water (greater than 1.2 V [volts]) during the transient periods of start-up/shut-down and fuel starvation. The catalysts will prevent damage by favoring the oxidation of water over the dissolution of platinum and carbon. Such catalysts are required for fuel stacks to satisfy the 2015 U.S. Department of Energy (DOE) targets for performance and durability.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.7** for its relevance to DOE objectives.

- Durability issues resulting from automotive start/stop cycling can be severe. This project uses a passive approach to control cell and electrode potentials to mitigate support corrosion. A passive approach could replace system-based strategies and reduce system cost.
- 3M electrode research and development has taken a new direction. The intent is to add oxygen evolution catalysts to the fuel cell cathode electrodes to attempt to enhance durability. The science is certainly acceptable. However, the addition of more precious metal to a system that already costs too much seems like a move in the wrong direction. It is unclear how this approach will cut costs, and why the oxygen evolution reaction (OER) catalyst will not mix into the oxygen reduction reaction (ORR) catalyst in ways that are detrimental to the primary fuel cell task of making electrons.
- There may be some controversy about testing protocols that realistically address start-up/shut-down and reversal tolerance, as well as what the associated targets should be. However, automotive fuel cell commercialization does face the durability barrier, and facilitating the oxidation of water at the cathode and the anode for different operating modes can significantly decrease degradation. The project is focused on OER catalysts on the nanostructured thin film (NSTF) catalyst. Because this catalyst is relevant to future automotive fuel cells, the project relevance remains intact.
- Unexpectedly high potential for cathodes and cell reversion for anodes are imperative challenges for durability, particularly for low Pt loading electrodes. Looking at OER catalysts is a corrective action for the fundamental part of these problems, and this project is expected to achieve great accomplishments.
- This project is very relevant to achieving the fuel cell durability requirements for the automotive application.
- The project is extremely relevant. Increasing catalyst durability during transient conditions is very important.
- This project is relevant to DOE objectives.

Question 2: Approach to performing the work

This project was rated **3.0** for its approach.

- Simultaneously addressing both electrodes by modifying electrocatalytic properties, without significantly impacting the electrocatalysis of the primary electrode reactions, is an ideal approach to improve durability during start-up/shut-down and fuel starvation.
- Using a second catalyst material to mitigate potential excursions and protect the Pt and support from corrosion during start/stop cycles is interesting, and has shown some benefit with NSTF. Ex-situ characterizations effectively support the project. The test cycle needs to be widely vetted.
- This reviewer questions why durability studies are being conducted on shorted membrane electrode assemblies (MEAs), as was pointed out in the presentation. Also, focusing on catalysts utilizing iridium seems to limit the practicality of the project, given iridium's low natural abundance.
- It is recommended to pursue materials screenings for the OER catalyst, including Ir and Ru oxide and alloy, which expect more effectiveness. It is unclear why those materials were not in the scope of work. It is also unclear about how to identify "real" materials for low ORR anode catalysts.
- The project proposes two approaches to prevent damaging high potentials on the cathode during start-up and shut-down—(1) modifying the anode catalyst to reduce ORR activity while maintaining high hydrogen oxidation reaction (HOR) activity, and (2) modifying the cathode catalyst to enhance OER kinetics. The former approach appears to be more feasible, as the HOR is a fast reaction compared to ORR and can tolerate changes of the catalyst with minimal impact on the overall cell voltage losses. Also, the conditions on the cathode are much more corrosive than on the anode, so any material added to the cathode catalyst must be extremely resistant to corrosion. Based on the number of slides devoted to each approach, the project is devoting much of the effort to cathode catalyst modification. The rationale for modifying the cathode catalyst with an OER catalyst is to prevent high potentials, which can corrode the carbon support and platinum. However, the NSTF support used in this project is already corrosion resistant, and the application of the OER catalyst to more traditional Pt/C does not appear to prevent electrochemically-active surface area loss.
- While some hydrogen/air testing would be useful to see if down-selected concepts perform in a similar trend versus voltammetric experiments, the voltammetric experiments are acceptable for screening. Some tweaking needs to be done in order to ensure that platinum oxide formed at higher potentials has an opportunity to electro-reduce at lower potential. The materials are mainly limited to Ir and Ru. It would be interesting to see if the project could expand beyond platinum group metals (PGMs). Investigators should vary the ramp rate and anodic potential limit to see if more degradation is produced at conditions that could be realistic under automotive operation.
- This new activity represents another investment of almost \$6 million, which seems like lots of money. Perhaps it would have been better to do a few experiments now that explore the feasibility, and then consider the next steps after those results were in. There are other ways to address the start-up/shut-down concerns. It must also be remembered that a fuel cell is just a heterogeneous reactor, and chemical scientists know how to control reactors. For example, there is no reason that "hydrogen starvation" should occur. There is a precise tool to measure how much H₂ is consumed (current), and how much H₂ has been injected (flow meter). It is unclear why the fuel cell system should ever experience a H₂ shortage. Likewise, everyone knows that O₂ must be kept from the anode compartment. Hydrogen has been there, and all surfaces are H₂ contaminated (reduced). The introduction of O₂ generates a fire that is perhaps small. There are many rather simple approaches to control the anode so that O₂ is always excluded. It makes more sense to focus on reactor engineering than to keep making the catalyst system even more complex and expensive.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.1** for its accomplishments and progress.

- The project has made advances in demonstrated trends, such as OER protection of Pt, OER activity being independent of Pt loading, accelerating the voltage increase after 1.7 V, and the advantage of platinum NSTF with OER over Pt/C with OER. Some shorting is evident in the Pt/C cyclic voltammograms. For a given current pulse, this shorting may decrease the measured voltage, and should be corrected. Much of the project thus far has focused on developing the experimental context—protocols, observed phenomena with baseline OER on Pt

NSTF, and comparisons between Pt/C with OER and OER-less platinum NSTF. However, the project does need to demonstrate a plan for how it will develop OER materials, including Ru/Ir ratios, OER particle sizes, and OER loading. The performance impact of OER addition should be reported more extensively, both with respect to normal polarization conditions and operational sensitivity (temperature, relative humidity, stoichiometry, pressure, H₂ concentration). The results for both the anode and cathode formulations were positive. Ex-situ characterizations have been used to support formulation and understanding of cell behavior.

- This project is showing good results and effectiveness of OER for high cathodic potentials and cell reversal (anode). The project team has identified a detailed structure of the electrode, including OER materials, the loading amount, and how it is loaded and dispersed.
- The development of an oxygen-tolerant anode catalyst through collaboration with Argonne National Laboratory (ANL) is significant progress toward achieving a materials solution to the catalyst degradation caused by start-up and shut-down. The effort involving addition of OER catalysts to the cathode catalyst has made progress and shows promising durability, but degradation is evident.
- The project has surpassed what it set out to do. The results are impressive, and the newly developed catalysts are promising for incorporation in stacks.
- The excellent 3M work persists. However, this reviewer notes that progress seems to have stalled, and perhaps the project has reached the optimum plateau. Efficiency is the primary attribute of a fuel cell, so perhaps 3M could consider increasing efficiency from a fuel cell system point of view. It would be good to have some really good people focusing on just efficiency.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.4** for its collaboration and coordination.

- The project collaborators account for the roles that would be expected—an OEM (Automotive Fuel Cell Cooperation [AFCC]), a materials synthesis partner (Dalhousie), and characterization specialists (ANL and Oak Ridge National Laboratory [ORNL]). AFCC input on performance and durability targets would be useful to report. Dalhousie appears to have contributed more in the opening stages of the project and not as much in the past year. It may be useful to show the work tasks and explain whether Dalhousie's task has ended or declined. Both ANL and ORNL were well used in showing data that related the different interaction of Ru to the whiskers in contrast to either Pt or Ir.
- The collaborators and the assembled team appear capable of addressing project objectives.
- It is good to collaborate with industry partners to implement proper test protocols that mimic real-world conditions.
- This project features outstanding use of external collaborators in providing needed insight into morphology and interactions in OER catalysts, as well as into the development of oxygen-tolerant anodes.
- This project features good collaboration with Dalhousie. The collaboration with partners is appropriate.
- 3M has built a quality, well-organized technology team. However, 3M's biggest resource is the talent that resides within its organization. It is apparent that the principal investigator (PI) gets considerable support from 3M, which is a center of polymer excellence.
- A stack integrator is among the collaborators. Automotive original equipment manufacturer (OEM) involvement would enhance the project.

Question 5: Proposed future work

This project was rated **3.0** for its proposed future work.

- The proposed future work addresses perceived weaknesses in the project and will provide guidance for further understanding. The cycle protocols will be vetted through the Fuel Cell Technical Team and the DOE Durability Working Group.
- The project planning and future objectives seem appropriate for meeting the stated project goals.
- The future work looks reasonable, but it is hard to judge. The project team should include a timeline showing tasks and decision points.
- Investigators should place more focus on understanding the source of the apparent 1.6 V onset of degradation, as well as on developing and demonstrating the O₂ tolerant anode material.

- It seems like the detour into oxygen evolution catalysts has certain risk and not much reward. There is a long set of data that suggests that the catalyst design for O₂ reduction and the catalyst design for O₂ evolution from water cannot be identical. Oxygen reduction requires access for a gaseous reactant, while O₂ evolution requires access for liquid water. It is unclear how a catalyst can be both hydrophobic and hydrophilic. The project team should consider addressing reactor engineering—operating the stack in ways that the highly damaging operating conditions are prevented by using system designs that keep the stack in sensible reacting environments. DOE might redirect some of the 3M work in a more sensible direction.
- The PI is correct in identifying the reduction of PGM loading as a priority in the future work. Furthermore, more attention should be spent in the future on high HOR/low ORR selective catalysts for the anode. Changing the test protocols with outside input is also a reasonable path for the project, as pointed out by the principal investigator. The targets identified are based on low PGM loading and provide for a sufficient amount of electrochemical stress events. Other 3M catalyst projects (those on NSTF) maintained a fairly active degree of alloy exploration throughout their durations. This project identified Pt, Ru, Ir, and Ti as metals of interest in 2010, but has not explored outside of this scope. This year may represent the final opportunity to engage Dalhousie on studies to explore other combinations, particularly those beyond PGMs.

Project strengths:

- Studying methods to protect catalysts from deterioration caused by transient conditions is important, and requires projects like this one.
- The capability of material fabrication, including working MEA with targeted OER catalyst materials, is a strong part of the project. Implementing proper test protocols for unexpected high cathodic potentials and cell reversals is also a strength of the project.
- The biggest strength of the project is the use of NSTF, with its inherent stability against corrosion due to the non-carbon support and larger Pt crystallite size. Using multiple approaches to solve the problem of high cathode potentials is also a strength of the project.
- This project features an excellent technical approach and well qualified participants.
- The team is excellent and has a demonstrated ability to produce quality work and successfully address DOE targets. The team has a global reputation of highest quality. The main strength is the people.
- This project's strength is its proactive approach to test protocols. Despite investigating a topic without assigned protocols or targets, the project has moved successfully toward establishing both. The established protocols will need to be modified, but the project has been able to use them to screen materials for activity and durability. This project also has good experimental curiosity. The project has explored many of the aspects associated with both activity and durability. Investigators have observed comparisons with Pt/C and platinum NSTF, as well as the effects of Pt loading, nanowhisker interactions, and PGM loss during cycling. The investigators have a willingness to engage the industry. The project has reached out to stack OEMs to understand what the needs are, which is crucial for a topic that is exploring matters that can be system-dependent.

Project weaknesses:

- It is difficult to find anything very wrong with this project (i.e., something that is serious enough to call it a “weakness”).
- The project team is performing extensive investigations on MEA samples that are shorted before or after break-in. Catalyst cost is not addressed by focusing on the incorporation of Ir and, to a lesser extent, Ru.
- Sharing details of the electrode/catalyst structure with OER catalyst would have been expected.
- The majority of the effort is on the most difficult approach, the cathode catalyst modification, rather than on the anode modification approach.
- The project has confined itself to precious metals, which forces the need for lower loading. There is limited reporting of future material developments. The project has identified a desire to change the architecture of the Ir/Pt/Ru system, but the plans for doing so were absent from the presentation. For the reviewers' sake, the project should describe what is presently known about the material development intentions in order to gain some line-of-sight toward whether loading and durability targets could be met.
- The reviewer felt that there were no weaknesses.

Recommendations for additions/deletions to project scope:

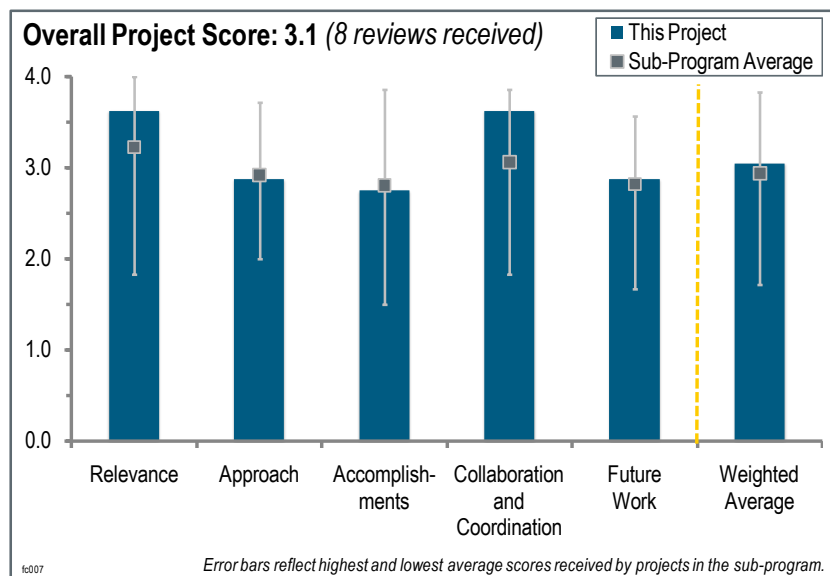
- The project should include molecular scale modeling to optimize configuration and materials screening to form an electrocatalyst or electrode with an OER catalyst.
- It seems more valuable to add intelligent controls to the fuel cell reactor in order to eliminate experimental conditions that accelerate corrosion reactions. It is well understood in catalysis that a catalyst must be activated to achieve high performance. For example, CuO/ZnO must be carefully reduced to Cu/ZnO, the useful catalyst. However, that active material is pyrophoric in air, so the reactor is built to exclude air. That is the way all heterogeneous catalytic reactors are designed and operated, and it is unclear why a fuel cell heterogeneous reactor should be operated casually.
- The investigators have observed that Pt remains more oxidized in the presence of an OER catalyst. It would be interesting to observe whether this causes tradeoffs among failure modes, depending on OER loading. In other words, higher OER loading might suppress start-up potentials, but lead to increased Pt dissolution at low current or idle operation. Explorations of more non-PGM materials would be welcome. Cell testing should be checked to ensure that shorting resistance is not too low. Additional x-ray photoelectron spectroscopy could be used to clarify the oxidation states of Ru and Ir. The project has already reported that these materials can oxidize with cycling, but—for purposes of understanding the loss of OER activity—it would be useful to know how, and at what potentials, these oxidation states change.

Project # FC-007: Extended, Continuous Platinum Nanostructures in Thick, Dispersed Electrodes

Bryan Pivovar; National Renewable Energy Laboratory

Brief Summary of Project:

The overall project objective is to produce novel catalysts based on extended platinum surfaces with increased activity and durability. Demonstrated improvements by 3M and others in specific activity and durability using similar materials have shown significant promise. This project focuses on limitations in terms of mass activity and water management. In 2010–2011, this project seeks to: (1) produce novel extended thin film electrocatalyst structures (ETFECS) with increased activity and durability, moving toward simultaneously meeting all 2015 U.S. Department of Energy (DOE) catalyst targets; and (2) begin studies of electrode incorporation of ETFECS with the highest potential to address membrane electrode assembly targets.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.6** for its relevance to DOE objectives.

- This project has a very high relevance due to the focus on reducing the Pt loading and potentially providing electrode design capability for novel structures in dispersed electrodes. The current focus of novel catalysts does not generally extend to the practical electrode design, so this project may have wide applicability to various catalyst structures.
- The basic idea of incorporating an extended film catalyst into a more traditional dispersed electrode is good. If this project can obtain uniform deposition of the catalyst and good mass activity, and employ the improved transport and water management of the dispersed electrode, it could reach all the DOE 2015 objectives. However, meeting the new platinum group metal (PGM) total loading for 2015 will be challenging.
- This project, if carried out along the lines originally proposed, has the potential to make major step improvements in both catalyst cost and catalyst durability. The original concept—utilizing the specific activity advantages of continuous-layer catalysts while also building thicker electrodes to give more water storage volume to improve low-temperature performance—is excellent. The only concern is that the people in the project are becoming frustrated with the difficulty of growing thin continuous layers, and are getting sidetracked into semi-continuous layers. These semi-continuous layers can provide only marginal improvements in activity, and would likely be worse than standard catalysts in durability due to the lack of spatial separation of the individual Pt particles making up the porous structures they are now developing. The investigators should keep the faith of continuous-layer catalysts and go for the “home run,” instead of settling for a “single.”
- This project is focused on multiple approaches to generate many varieties of extended thin film catalyst structures, from synthesis of the supports and catalyst structures to coatings on the supports by multiple processes. By the time the project is complete, it may provide insight on the feasibility of some approaches versus others in terms of generating improved electrocatalyst activity or durability (by rotating disk electrode [RDE]). The project is only 30% complete and incorporation into membrane electrode assemblies (MEAs) has just barely begun, so it is too early to judge whether it will have any impact on the three barriers for MEA—cost, durability, and performance. The project is too broadly based and may not have time to conduct in-depth

study of any approach, and thus may not be able to draw solid conclusions. This is the only reason that it is rated “fair” versus “good” on this criterion.

- This project appears to be an attempt to develop a novel fuel cell catalyst support. It has the possibility to meet the DOE objects at three levels—cost, durability, and performance.
- Reducing the Pt loading amount with required durability is one of the most important factors for making automotive fuel cells commercially viable. Looking at bulk properties rather than nanoparticle properties to improve specific/mass activity is expected to achieve the goal.
- This project is focused on catalysts and electrodes.
- High-activity, robust catalysts are critical for enabling fuel cell system commercialization.

Question 2: Approach to performing the work

This project was rated **2.9** for its approach.

- The original approaches, with a wide range of supports and several distinct approaches to platinum deposition were excellent. It is probably too early in the project to be down-selecting to a single Pt deposition technique. The project appears to be going in the direction of excessive concentration on spontaneous galvanic displacement as the only Pt deposition technique to be used. This process is producing porous layers with only part of the originally anticipated specific activity advantage, but with higher-than-originally-planned specific surface areas. This leads to mass activities that, so far, appear to be only about double those of classical carbon supported platinum (Pt/C)—this project should be aiming higher. The surface area of the Pt deposits, with the concomitant large concentration of low-coordination surface Pt atoms, will likely lead to lower durability than could have been expected from this project. On the bright side, the higher surface area of these deposits would lower the local current density and perhaps improve high current density performance in air, if the apparent local transport limitations seen under those conditions could not be mitigated by other means.
- The “screening” phase of catalyst-making should be reduced and the focus on electrode making should be emphasized. The overall approach is a good “portfolio” design that maximizes success and minimizes the risk of a single solution or approach failing.
- The approach is well integrated. This reviewer understands the need to pick the best method of fabrication with continuous film formation, but now would be a good time to focus and optimize a particular method.
- This project appears by design to be very broadly based on different approaches to generating a plethora of extended surface area catalyst structures. At this initial stage, the project is not expected to impact a key barrier. Also, because the ultimate manufacturability of the various approaches is not a consideration, the project will not really address the questions of cost. The approaches for depositing the catalysts and the types of structures generated do not seem to have any unique qualities compared to other state-of-the-art extended surface area catalysts, such as 3M’s nanostructured thin film (NSTF), so it is not clear what will be achieved. With so many different support and catalyst deposition processes being considered, only superficial studies are possible in the relatively short time of the project.
- Extended thin films have proven to be stable, highly active oxygen reduction reaction catalysts. This reviewer is concerned about the leaching of Ag and Cu from the metal nanowire and nanoplate coated materials. Atomic layer deposition (ALD) on Ti dioxide and other non-metal nanowires seems more promising. The presentation was unclear about the impact of the modeling work and how it contributes to materials development.
- The different structures studied are promising. The approach to leverage other work on catalyst structures where appropriate is efficient. The vertically aligned carbon nanotubes (VACNTs), analogous to 3M’s NSTF whiskers, will provide an important comparison. The number of structures should be further streamlined, or a plan to incorporate understanding of the parameters associated with the large number currently under investigation should be clearly laid out. Incorporating carbom and understanding durability effects are important. The next phase of electrode work and modeling will be the more valuable aspects of the work.
- The proposed technical approaches are too widely spread. The approaches should be systematically reorganized for thin film synthesis and substrate selection. The project should focus on the mass activity target—it should be conscious about the thickness of the thin film and the number of the atomic layer, which are the most important metrics to investigate for the mass activity target. There are too many options of substrates—such as metal wires, tubes, etc.—and carbon nanotubes. Critical characteristics of substrate configurations and materials should be identified and these substrate options should be theoretically screened before fabrication. Achieving the mass activity target should occur before electrode design consideration and MEA testing.

- The approach appears to be applying technology from other projects in the generation of various catalyst supports, catalyzing the supports, and then evaluating the results. This is a tried and true analytical approach.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.8** for its accomplishments and progress.

- The project has worked with a well-chosen collection of support materials, wisely choosing to use materials with preparations that have already been well developed and described. The principal investigator and coworkers have realized the problems inherent in the use of “vertically-aligned” carbon nanotubes. On the scale relevant to this project, the tubes are not sufficiently vertical or sufficiently aligned as the 3M NSTF perylene red whiskers are. These whiskers should perhaps be substituted, per Mark Debe's explicit invitation, for the VACNT in future work (though probably using a Pt deposition method other than the sputtering that 3M uses). The project has employed a well-chosen variety of Pt deposition methods, but has had limited success in achieving the smooth, continuous-Pt layers less than 2 nanometers thick that would be needed to achieve the original vision of the project. Such deposition is very difficult to achieve, but should not be impossible. The project team would do well to continue its efforts in this regard, and to keep searching for ideas on how to achieve the desired thin continuous layers. The fuzzy Pt layers achieved to date by spontaneous galvanic displacement have given specific activities below those anticipated for continuous layers, but above those for standard Pt/C nanoparticles. This is a modest achievement, but even with the larger-than-originally-anticipated specific surface areas, the mass activities—which should have been reported instead of the specific activities—appear to be only about twice those of Pt/C, rather than the targeted four-fold increase. Additionally, the fuzzy layers are unlikely to be as durable as continuous layers would be. The original concept of this project has so much promise on both activity and durability that it would be a shame to now concentrate only on the fuzzy spontaneous galvanic deposits, which may give easier, but much smaller, gains. For \$9 million, the taxpayers should expect this excellent team to continue dedicated efforts toward the full anticipated promise of the original approach, not a retreat to incremental improvements.
- There seems to be some confusion about seeing bulk property or nanoparticle property. The project team should identify the “ideal” configuration of the catalyst to meet mass activity targets such as specific activity, thickness of thin film, number of atomic layers, and surface area per area of electrode (roughness factor). This reviewer wants to know if Pt on a metal nanowire is considered a nanoparticle. If so, the project should get back to the original approach of focusing on thin film and its bulk property. The atomic layer target is unclear, regarding the ALD. This reviewer wonders if the target range is in the hundreds. Performance data without the thin film thickness or the number of the atomic layer is not meaningful.
- This reviewer agrees that if sputtering is used, a shorter, more spread out array would be better to coat for good uniformity. Because of line-of-sight, more material is deposited on the top of the whisker or tube. The whisker end with the larger amount ends up being immersed in the electrolyte in the 3M process, but in nanotubes for mass activity, this smaller, stubby tube would probably work better.
- The mass activities measured are generally well shy of DOE targets, even on RDE tests. The MEA tests show low, high-current density performance. The transmission electron microscopy images shown suggest that there are not always true extended thin films created, so the ALD and spontaneous galvanic displacement (SGD) methods require more optimization.
- The team has shown a catalyst preparation achieving 40 square meters per gram, and several preparations hitting DOE analytical catalyst targets.
- To date, supports based on nanotube, nanowire, and nameplate technology have been generated and catalyzed. Their performance is equivalent to commercially available electrodes circa 2001. There is no indication that water management is being addressed at this point. The progress is more than adequate and the accomplishments are interesting.
- Overall, the investigators have made good progress on identifying catalyst structures.
- Taking the definition of the ratings for this category literally, it would appear that a rating of “one” (poor) is required, as the investigators, to this point, have just made and tested their first five MEAs, so there is little chance of showing any progress toward meeting the MEA targets. However, the catalysts the investigators have fabricated do not appear to have any extraordinary properties that would suggest overcoming the barriers at a more fundamental level.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.6** for its collaboration and coordination.

- The National Renewable Energy Laboratory has put together a strong collaborative team consisting of some of the leading experts in the field, and each partner has clearly defined roles. The team appears to work well together.
- This project has a large, collaborative team.
- This project features excellent collaboration.
- The collaboration is impressive.
- The various groups participating in this project appear to be working together very well, and the principal investigator seems to be providing excellent coordination.
- This project features many partners and good coordination among such a large team. This reviewer did not see any modeling results.
- This project has many solid and experienced collaborators, but their contributions to date were unclear.

Question 5: Proposed future work

This project was rated **2.9** for its proposed future work.

- The proposed work appears to be a rational progression based on the progress to date.
- It would be appropriate to continue the modeling work. It would be good to see some effort related to the scale-up of whichever fabrication method is chosen with MEA fabricating and testing.
- The proposed future work is well thought out, but is perhaps a bit too conservative—retreating to the easier modest gains of the fuzzy galvanic displacement deposits rather than continuing to pursue the difficult growth of thin continuous layers. The fuzzy deposits might turn out to be useful for high current density performance in air if local transport issues cannot be rectified.
- It is good to continue focusing on metal oxide cores. The project team should not abandon the whiskers. The primary near-term focus needs to be on developing SGD and ALD processes to create the continuous thin films, rather than discreet particle dispersions. Investigators should make sure the modeling work drives materials development, such as the effect of adding carbon and ionomer to ETFECS electrodes.
- The team should start to focus on a few of the potential methods, and shift focus to electrode and gas diffusion layer (GDL) efforts. The unique hydrophilicity of these catalyst structures poses numerous challenges in being able to realize the potential shown in the analytical results.
- An increased effort to “incorporate these structures into highly performing MEAs over wide operating conditions...will be a primary focus of the rest of the project...”
- The investigators should focus on the characteristics of thin film and ALD catalysts to meet the mass activity. It is too early to address the electrode design and MEA testing. Achieving the mass activity target should occur before electrode design consideration and MEA testing.
- Electrode studies and models will provide valuable information and should move the technology forward significantly compared to catalyst-synthesis-only focused projects. However, the probability of success is not clear. The investigators did not present a clear plan regarding a structured study in terms of models or design of experiment approaches.

Project strengths:

- The project has a number of strengths based on the different supports being evaluated.
- The investigators have diverse, complementary backgrounds. The team can draw from outstanding analytical resources. The portfolio approach is also a strength.
- This project features a very strong team that has many concepts to consider. The team possesses strong materials, and analytical and modeling expertise. There is much room for improvement on reasonably good progress.
- Looking at the bulk property of Pt catalysts is a good approach to meeting the mass activity target. The capability of the material fabrication is a strong part of this project.

- Strengths of this project include incorporating novel catalyst structures into dispersed electrodes, and the large team for collaboration.
- This project features great integration of the work. The investigators designed a good concept by essentially taking two types of catalysts and merging them together to overcome the weaknesses of each.
- This project has an excellent initial concept, a good choice of substrates, good range of Pt deposition methods, and thoughtful analysis of the results.

Project weaknesses:

- It is unclear whether these various carbon supports would be more cost effective than the current carbon/Teflon supports. There is no information indicating the cost or durability of cells using these various supports.
- The challenge of electrode making and GDL matching may be underestimated. Using carbon as an additive to offset hydrophilicity may be undermined by carbon corrosion, unless high-graphitic carbon is used.
- There is no clear plan of how to down-select the most promising concepts. There is no clear connection between modeling and experimental work, and no clear feel of how thin the ETFECS layers need to be in order to meet the mass activity targets.
- The theoretical modeling to identify the targeted configuration of the bulk property of the Pt catalyst is weak. It is recommended the investigators add a collaborative partner to work on this area.
- Using transition metal nanowires may result in MEA contamination. The project may have more catalyst structures than optimal for moving the work forward as effectively as possible.
- This reviewer is still not convinced about adding carbon, mainly because of long-term stability and possible problems during start-up and shut-down, as well as reverse conditions where carbon corrosion is a serious issue. Cycling from 0.6 to 1.0 V (volts) is not good enough to determine long-term durability. This reviewer is not surprised about silver, because it migrates easily. This reviewer questions whether silver nanotubes are needed. The investigators should specify how the PGM loading affects durability and fuel cell testing of ETFECS. The project team should also include humidification, temperature, and pressure conditions.
- The investigators were too quick to abandon their focus on thin, smooth continuous layers, and too quick to concentrate on spontaneous galvanic displacement. They placed too much emphasis on specific activity in the presentation, though demonstration of specific activity is a good first step in proof of concept. Also, the investigators should show progress against the economically critical metric of mass activity.
- The theoretical part of this project seems to be weak in identifying targeted configurations of thin film or ALD catalysts to meet mass activity targets. Involving molecular scale modeling would be good.

Recommendations for additions/deletions to project scope:

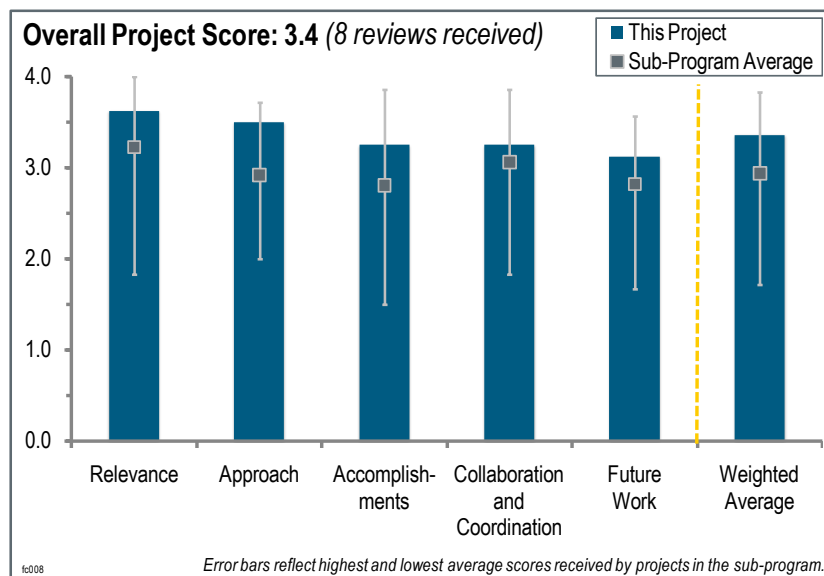
- This project is expected to make breakthroughs and meet the cost and durability target of the electrocatalyst. It is important to identify the targeted configuration of the bulk property of the Pt catalyst to meet the mass activity. The entire project should focus on this task. This reviewer recommends deleting the electrode design and its modeling. It is too early in the project for that work.
- The investigators should add cost and durability estimates and something to indicate the end game.
- Although there is a vast array of approaches, this reviewer wonders if there is a unifying target (not an end goal such as DOE analytical results) that will allow the team to eliminate non-viable approaches. For example, ink processability could perhaps become a screen for viable methods.
- The project team should limit work on Cu- and Ag-coated nanowires, and stick to the primary objectives of making very thin ETFECS layers. Electrode development studies should follow.
- This reviewer is not sure that ALD will ever be a viable technique without much work on processing conditions, and believes this work could be removed from the project. The team should put more focus on sputtering or SGD.
- The project team should improve the clarity of the plan forward.
- The investigators should stay true to the original concept of the project.

Project # FC-008: Nanosegregated Cathode Catalysts with Ultra-Low Platinum Loading

Nenad Markovic; Argonne National Laboratory

Brief Summary of Project:

This project focuses on developing a fundamental understanding of the oxygen reduction reaction (ORR) on PtM bimetallic and PtM₁M₂ ternary systems that would lead to the development of highly-efficient and durable real-world nanosegregated Pt-skin catalysts with low Pt content. Argonne National Laboratory's (ANL's) materials-by-design approach will be used to design, characterize, understand, synthesize/fabricate, and test nanosegregated multi-metallic nanoparticles and nanostructured thin metal films.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.6** for its relevance to U.S. Department of Energy (DOE) objectives.

- This project is focused on catalysts and the impact on cost and durability.
- This project's main objective is to examine the feasibility of using nanosegregated Pt catalysts as candidates for the ORR. This activity was related to meeting and exceeding the DOE cost targets for platinum group metal (PGM) catalysts as well as durability in accordance with DOE mandated protocols. These tasks are in line with the DOE requirements and objectives.
- This project addresses the most critical fuel cell research and development material issues—those of catalyst performance, cost, and durability.
- This project is highly relevant. The ANL team has taken years of fundamental and applied research and models, and is finally proving them out in viable membrane electrode assemblies (MEAs).
- This project is very relevant, especially considering the new, much lower PGM loading requirement for 2015.
- This project is seeking to build off of prior discoveries to generate higher activity and more durable catalysts for the ORR. This topic is most relevant to the commercialization of automotive fuel cell vehicles. The project is seeking to address the same targets identified by DOE.
- The need for low-loading, high-performance catalysts is central to DOE's goals of reducing cost.

Question 2: Approach to performing the work

This project was rated **3.5** for its approach.

- This project has a strong approach. ANL does things the right way, with strong scientific support of the work. The project also could have a big impact because of its applicability to several different support approaches (e.g., 3M nanostructured thin film, carbon supports). Careful work by an outstanding researcher eliminates guesswork with respect to findings and directions. This is a high watermark for the DOE Hydrogen and Fuel Cells Program.

- This project features a very comprehensive approach including modeling, highly controlled synthesis, processing, and analytical testing. This is probably the best designed project to truly understand structure-function relationships.
- This project addresses barriers very well and with sound logic.
- The science is excellent.
- This year's approach primarily focuses on ternary alloys of Pt with two of the following three metals: Co, Fe, and Ni. It also includes the study of Pt monolayer coatings on alloys and Au as a core. These three efforts are interesting, although not necessarily synergistic. The “materials-by-design” approach that was quite logical in using theory to guide the binary alloy effort in 2010 is not as clear this year. The milestones given are vague and lack dates and metrics.
- This year, the approach is excellent. In the past, ANL has been criticized for working on highly idealized models. In their presentation this year the investigators showed that they have “vertically integrated” their whole research approach with actual results in an applied system.
- The team seeks to generate atomic segregations within a nanoparticle, which can be done with combinations of acid and heat treatments. These processes can be made manufacturable without adding significant cost. The project seeks to use high ORR activity for a Pt-Ni catalyst (Pt₃Ni) that has already been demonstrated with bulk materials, and transfer such activity to a nanoparticle, which is entirely appropriate to investigate. There is some risk involved with developing nanoparticle catalysts that may allow base metal access to the particle surface. Base metal dissolution has foiled many Pt-alloy developments in the past. However, investigating possible stabilization is worthwhile.
- The approach is based on a model nanosegregated profile that would allow for further enhancement of activity and durability. There was no clear idea regarding how the small-scale, careful, laboratory bench synthetic approach would translate into actual scale-up and ultimately to an application.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- The team identified a leading candidate and a framework to understand why it performs well. The transition out of the laboratory to larger preparations and MEAs was not clearly addressed.
- The performance and durability of the novel materials demonstrated continue to be impressive and lead the field. The results obtained are of interest to the community, but the lack of a traditional catalyst manufacturer, the demonstration of significant quantities, and evaluation of the catalysts in fuel cell systems under operating conditions continue to be weaknesses.
- The project team has performed a good volume of quality work. The focus on a scientific and rational approach is appreciated.
- The project features great results in a practical MEA. ANL should run durability tests with a range of conditions. The test they are running is just for Pt dissolution, but they should also look at the impact of the catalysts on carbon corrosion. A change of the relative humidity (RH) might have an impact as well. Companies running higher RH seemed to have more problems with alloy dissolution.
- ANL has made steady progress toward goals. Conducting more fuel cell testing would be outstanding.
- ANL has delivered a PtNi/C catalyst that exhibited only a 12% activity loss over 20,000 0.6–0.925 V (volt) cycles. The DOE test still needs to be conducted (higher RH, triangle wave up to 1.0 V, 30,000 cycles), but this appears to be an improvement over other Pt alloys. Higher activity catalysts were demonstrated by rotating disk electrode (RDE), including PtNi/C and Pt ternaries. These activities also need to be demonstrated *in situ*, as has been done with PtNi/C (three-times the mass activity of Pt/C. The project features a good demonstration of a skin structure with PtNi/C. The project would benefit from being able to show a skin structure (if possible) with the Pt -ternary catalysts. A skeleton structure for Pt ternaries was not explicitly shown in the microscopy images. Au/PtFe/C mass activity should be normalized by precious metal loading (Pt+Au).
- The accomplishments are good and in line with the targets in the proposed effort. More specifically, the following points need some attention:
 - No mention was made regarding the efforts on chemical synthesis, which is the focal point of efforts at Brown, including what synthetic routes were used and how they would translate on a scale-up effort.
 - There were no specifics of the theoretical efforts at Indiana University - Purdue.

- In the ternary alloy synthesis, it is unclear how the synthetic approach pairs with the nanostructured model presented in slide 4.
- This reviewer is wondering why three monolayers are needed for protection of the Ni, and if this is the same with other materials, such as Co. It is unclear why the PtNi nanoparticles with multilayered skin are so much more active. The reviewer wants to know if this is an electronic effect or if it is structural on the surface from the distribution of the underlying layers. It is unclear what is used to leach the excess material and at what temperature this is done.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.3** for its collaboration and coordination.

- This is a close collaborative effort with close ties between electrochemical measurements and testing efforts at 3M. The theoretical modeling aspect was missing and needs to be increased.
- The inclusion of a traditional platinum catalyst on carbon supplier would greatly strengthen this aspect of the project. The group presents and publishes frequently.
- This project features a good team make-up.
- This is a properly organized project.
- The collaboration with Oak Ridge National Laboratory is substantial, as evidenced by the transmission electron microscopy images. The collaboration with General Motors (GM) has benefited the project by providing MEA data that shows the mass activity and durability enhancements. The recent project information has not clearly reported the roles of Brown; Indiana University, Purdue; and the Jet Propulsion Laboratory (JPL). The experimental efforts appear to overwhelm the modeling efforts. It is difficult to see the impact of modeling on the project's direction.
- This reviewer would like to rank this project "high" on collaboration, but it is impossible to tell from the presentation what JPL; Brown; Indiana University, Purdue; and 3M contributed. From the overall progress, it seems that there has been progress among the collaborators. The investigators claim to be transitioning to GM, but the details of that transition are not clear.
- It is not clear if a catalyst maker or an MEA team is or was part of this effort.

Question 5: Proposed future work

This project was rated **3.1** for its proposed future work.

- This project is an excellent example of using fundamental work to drive applied goals. The plan is right on target.
- There is good delineation of the future efforts, though researchers should have further described the approach toward durability validation.
- ANL has a good plan for future work, but the team should pursue larger batch sizes on the most promising materials sooner.
- The author acknowledges the need for an MEA maker to see if the analytical results can be realized in a working MEA.
- It is unclear if further enhancements can be achieved by the approach proposed. It would be more advantageous to focus on applying the advances to date to more fuel cell testing.
- The team should really be focusing on MEA work. The investigators plan to take this approach for the next year.
- More details are needed on the thin film work. This reviewer wants to know what the support will be, and how the deposition will be carried out. The high current performance of the PtNi/C catalyst in an MEA was not reported, and the reviewer is wondering if it suffers in a similar fashion to the PtNi catalysts in the 3M project. It would be good to see a plan for further synthesis of the Pt ternaries. The reviewer wants to know which base metals will be used and how the processing will lead to a skin structure (as opposed to the skeleton structures shown thus far). ANL should state whether the stability of the Pt ternaries will be measured in an RDE or in an operating fuel cell.

Project strengths:

- This project features an outstanding example of how to proceed from theory to practice, including efforts to understand the impact of external perturbations (such as acid etching and annealing), and obtaining a final “active” structure.
- The investigators on this project are perhaps the world’s best in terms of understanding how to standardize electrochemical experimentation, purify reagents and samples to avoid artifacts, and extract meaning from measurements. The collaborators on this project for material characterization are well known for capturing atomic resolution, which is required for determining whether atomic segregation was achieved. This project has a strong understanding of atomic segregation; the investigators entered the project knowing that the surface segregation of Pt from base metals can enhance oxygen reduction activity.
- This project features an outstanding researcher, plan, and approach, as well as excellent execution. The whole field benefits from these findings.
- This project has been funded for years, and it looks like the team pulled it all together this year. The team members have been able to pull together a lot of basic research into an MEA that apparently works.
- The project is well formulated and timely. More needs to be done in regard to examining such systems.
- Strengths of this project include its great performance, great durability, and great fundamental understanding that serve to educate the community.
- The team is excellent and ably led. Other strengths include a focus on science, exploration of innovative approaches, and outstanding results reported in terms of activity.
- The research team has made excellent progress in meeting objectives with a process that appears very scalable. This reviewer wonders if large batches of materials have been prepared—for example, enough for 50 cm² MEAs. It might be worthwhile to try this with promising candidates sooner rather than later to determine what problems arise.

Project weaknesses:

- Analyses of the cost of making the catalyst (surfactant approach), as well as the feasibility to scale-up this approach, are missing.
- The findings of this project have been slow to find their way into the commercial materials that are available for fuel cells. Increasing the focus on making the scientific advance materials relevant would be beneficial. Several template “required” slides did not appear in the supplemental or reviewer-only slides, including the response to last year’s reviewer comments.
- Scalability is a concern. Despite results, the concern remains that Ni will leach out with catastrophic consequences. This would be especially concerning if the catalyst was mass-manufactured, which would inevitably lead to at least a small fraction of particles improperly coated, among other worries. In general, this criticism applies to all core-shell systems employing less noble metals.
- A lot of work is still being done in RDEs. There should be more focus on MEAs.
- For the ternaries, a reviewer wondered if the Pt₃M₁M₂ structure is the final stable form after a leaching process. This reviewer wants to know if the team has any ideas as to why Pt₃CoNi/C is so much better than FeCo and FeNi. MEAs were made with PtNi nanoparticles, and the reviewer wonders if polarization curves could be shown. The reviewer also wants to know why the PIs are cycling from 0.6–0.925 V, while others cycle from 0.6–1.0 V. It is unclear why this voltage was chosen. The reviewer wonders why the ternary Au/Fe/Pt₃/C was chosen and not another ternary, such as Au/Ni/Pt₃/C.
- The use of base metals in the best concepts introduces risk. There are many base metal-containing Pt alloys in the literature that have suffered from dissolution. Pt base metal alloys also suffer from poor performance at higher current density, and some base metals can produce negative effects on the membrane. ANL needs to place greater emphasis on MEA measurements. MEA measurements are the surest method by which to measure durability, thanks to the reproduction of the fuel cell environment. While some catalyst projects might correctly focus on RDE for activity, this project contains many samples with assuredly high activity, but questionable durability.
- The project team could practically implement its findings a little faster, in case “surprises” emerge from fuel cell work.

Recommendations for additions/deletions to project scope:

- The scope of the project is good, and an automotive partner is needed at the final stage for validation of the scale-up and durability.
- The project is within its scope.
- ANL should partner with a catalyst company to assess surfactant method cost and viability. Similarly, as the author has acknowledged, the project should transition to MEA-making to assess whether analytical results can be transferred to working MEAs.
- The project has too much of “more of the same” increased systems investigation and too few efforts to implement the materials improvements that have been found into fuel cell systems and at a larger scale.
- This reviewer would like to see an attempt (perhaps with an industry partner taking the lead, with added funds if needed) to prepare the catalyst on a large scale, mimicking best manufacturing practices. The reviewer wants to know if such a catalyst will demonstrate similar activity and stability.
- The durability work needs to be clearer—others have shown that alloys are stable in RDE and MEAs, but their catalysts ultimately failed in practical conditions. The team should make sure that there are no big surprises in the durability. They need to run multiple ranges for their durability tests (e.g., ranges of RH and potential).
- While the Au/PtFe stability that has been demonstrated is intriguing, the concept should advance to another PtM shell to avoid the possibility of Fe causing Fenton degradation.
- The Pt ternary alloy work should consider other metals beyond the three-dimensional base metals that may present either dissolution or poor high current performance. That said, the existing ternary work should continue in case there is a possibility of stabilizing the materials. The investigators involved in this project are exceptional and if stabilization is possible, they should be able to achieve it.

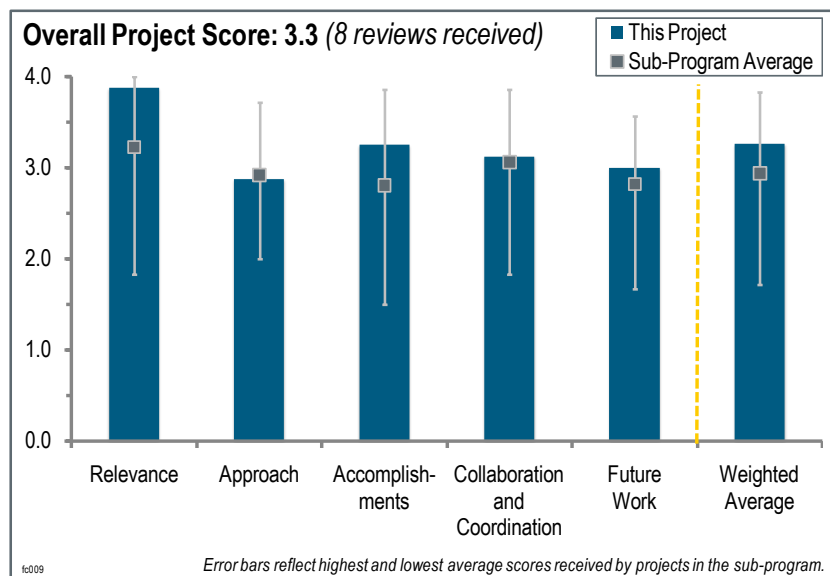
Project # FC-009: Contiguous Platinum Monolayer Oxygen Reduction Electrocatalysts on High-Stability, Low-Cost Supports

Radoslav Adzic; Brookhaven National Laboratory

Brief Summary of Project:

The overall project objective is to develop high-performance fuel cell electrocatalysts for the oxygen reduction reaction (ORR) comprising a contiguous Pt monolayer on a stable, inexpensive metal or alloy, including nanoparticles, nanorods, nanowires, hollow nanoparticles, carbon nanotubes, scale-up syntheses of selected catalysts, membrane electrode assembly (MEA), and stack testing. An additional supporting objective is to increase the stability of cores and supports.

Question 1: Relevance to overall U.S. Department of Energy objectives



This project was rated **3.9** for its relevance to U.S. Department of Energy (DOE) objectives.

- The development of low-cost, highly durable, and high-activity platinum group metal (PGM) containing catalysts is one of the key activities for the successful commercialization of automotive fuel cell electric vehicles.
- This work is of significant value to the future of fuel cells. It supports the main objectives of the fuel cell multi-year plan.
- The project is very relevant to the focus of lowering Pt loading and increasing catalyst durability. The work provides an approach to utilize Pt better by using a core shell or other support with monolayer Pt coverage. The issue of stability of the structure appears to be addressed with durability testing and investigation into the Pt structure. However, the use of Pd in the core is a concern regarding the cost objectives. The future work on lower-cost core materials is important. Again, the use of Ir should be approached with caution with respect to Ir prices and Ir contents in the catalyst. The potential for metals contamination of the MEA is also a concern.
- The project is very relevant and is focused on decreasing the cost and increasing the durability of fuel cell catalysts.
- The project is very relevant in keeping with the scarcity of Pt and its overall global availability.
- This project addresses the most critical fuel cell research and development material issues: catalyst performance, cost, and durability.
- Brookhaven National Laboratory (BNL) continues on the quest toward a higher-performance ORR catalyst system. This task is clearly tied to DOE Hydrogen and Fuel Cells Program targets, specifically getting cost down and durability up.
- The principal investigator (PI) uses very fundamental approaches to explore new catalyst material systems to address all three of the critical barriers. However, the PGM loading target stated on slide 2 is not correct and should be 0.125 g PGM/cm².

Question 2: Approach to performing the work

This project was rated **2.9** for its approach.

- This project incorporates several key fundamental properties recognized as necessary for optimizing the ORR on Pt surfaces into novel nanoparticles with controlled shapes and compositions. The approach features a good balance of fundamental characterization and modeling with performance characterization.
- The approach is sound. The team has the right set of expertise and the facility supports such work. The team should focus more on materials science and solid state chemistry.
- The project features an excellent, rigorous approach that includes modeling. The use of Au and Pd, which are both expensive, is a negative aspect of the work. The overall approach to replace these with Ir and Ni is good; however, areas of concern for these include stability, interactions with other MEA performance and degradation mechanisms, and cost sensitivities of Ir to volumes. The study into the stability mechanism of the core shell catalysts is important. There appears to good Pd stability, but there is a loss of Pd. It is also recommended that further *in situ* durability testing is completed to understand interactions with operating conditions and other degradation mechanisms. The first MEA results are an important step, and it is not expected that the performance on air will be good. The results were only shown for oxygen. Performance on air should be assessed as well. The work has been done with a monolayer of Pt. It may be beneficial to do an overall stability/cost trade-off with more than one monolayer, particularly with other core materials.
- The approach has the potential to meet DOE targets. The nanoparticle and hollow nanoparticle work is very good and has shown high activity. Some calculations of how thin the Pd nanowire needs to be to meet loading targets and how thin BNL can make them would be useful. The PI needs to progress to more MEA testing, especially for demonstrating the durability of the catalysts.
- The formation of contiguous Pt monolayers on inexpensive metals and alloys has been the holy grail of several patent applications in the last three decades, including a lot of efforts for the phosphoric acid fuel cells. The biggest challenges have been the ability to reproduce such systems, and the stability of such underlying structures in the aggressive pH and voltage conditions. The use of Pd nanorods seems difficult to justify from the perspective of nanostructure cost, which at the moment is several times higher than Pt and the issue of scalability around making large quantities of such catalysts. Putting Pt on multiple-wall carbon nanotubes is even more problematic from both cost and scalability standpoints.
- The approach is quite varied, focusing on Pt or Pd nanowires, hollow nanoparticles, coated carbon nanotubes, and metallic core shell materials. The premise of using monolayer (thin) coatings is a common theme that makes sense for utilization issues, but often relies on a precious metal core. Most techniques for particle synthesis have significant challenges for cost or scale-up.
- The BNL accomplishments are well known and impressive, and many workers in chemical science are watching, admiring, and copying. There has been considerable progress during the last decade. However, previous success in a task increases the difficulty of the next steps. It is possible that BNL has come to the end of the core-shell catalyst road. The BNL report has an element of disorder, and many synthesis approaches are being tried without the usual planning for the experimental method. Fuel cell catalyst performance involves a number of parameters, including a number of unfortunately closely coupled parameters. Experiments that focus on just the ORR catalyst must factor into a range of design elements, including mass and energy transport to the reacting fuel cell catalyst sites.
- The concept of core-shell catalysts is a very effective way of reducing the loading of Pt.
- The PI has replaced Pt with other very expensive commodity metals such as Pd and Au. While this reviewer understands that the cost is now shared among different metals, commodity pricing and traders will not.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- Very good progress has been made in meeting the stated DOE 2015 targets for mass activity and Pt cycling durability. Credible test results are being obtained from outside laboratories, including original equipment manufacturers (OEMs). One concern is that the activities measured in MEAs are substantially less than the values measured by rotating disk electrode (RDE) experiments. Lower activities are primarily due to the different protocols used, but suggest that the MEA values should be compared against the DOE targets. The

processes for generating the novel structures appear somewhat complex, so the fabrication costs to get high yields at higher volumes should be seriously considered. It may help to down-select from the multiple approaches that the PIs have demonstrated.

- The results are excellent.
- The results for both performance with very low Pt loadings and stability are excellent. There is a delay in some of the other approaches by collaborators and it is not clear how successful this work will be due to the stage of the activities.
- The progress toward nanoparticle targets is excellent. The PIs have developed Pt/PdAu and Pt/IrNi nanoparticles that meet the DOE mass activity targets.
- The overall progress is very good. The data presented does not address the true nature of the Pt deposited or the cause of the observed higher mass activity. There is no explanation of the reported durability of these nanoparticles, especially on the “cathodic protection effect.”
- The project continues to show good mass and specific activities for novel catalysts. At least part of the precious metal core or multiple shell materials has shown good durability and performance. Obtaining materials that can be produced cheaply at scale while demonstrating both performance and durability remains a challenge.
- The core shell concept has been actively pursued for years. The concept of a thin precious metal layer covering a nanoparticle containing transition metal elements is interesting. However, alloy electrocatalysts have been explored for 50 years or more. The question remains if these designs can be durable for extended periods, long enough for the fuel cell hardware to prove useful. Moreover, transition metal cations, certainly Fe, Cr, and Ni—are known to degrade polymer electrolyte membrane fuel cell performance. The “durability” experiments exclude a wide range of experimental conditions that could impact system durability in contrast to electrode durability. The accomplishments would have been stronger if those system durability issues were addressed.
- Catalysts are obviously incredibly active when investigated in RDE experiments. The vital hurdle, however, is transplanting that level of activity to an actual single cell and having the testing show significant real performance.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.1** for its collaboration and coordination.

- The partnership with Johnson-Matthey Fuel Cells, Inc. (JMFC) is an excellent collaboration that will enable future commercialization of this technology if it proves to be viable. This reviewer is looking forward to seeing Toyota's fuel cell results when real MEA testing (beginning of life performance, start-stop cycling, load-cycling) takes place.
- This project has all the bases covered from fundamental modeling, synthesis, and characterization to OEM testing, with excellent collaborators involved in the critical work.
- The collaboration is excellent. This reviewer wants to know if there is any chance BNL will work with U.S. catalyst/MEA companies.
- The project includes a number of collaborations and it appears to be well aligned. MEA tests have occurred at both 3M and Toyota. However, it is difficult to tell the amount of catalyst collaboration that is occurring.
- BNL collaborates with several other catalyst projects.
- This is a good collaborative effort. It is assumed that JMFC is the catalyst scale-up partner and its role in this effort in terms of scale is not yet reported. The collaboration with 3M is also not yet reported. The modeling effort by the University of Wisconsin is not clearly spelled out.
- The institutions involved are excellent choices. The project is very broad in terms of scope, and it is not clear that all institutions are being used as effectively as possible.
- There is much collaboration that could make sense, some of which is in place. However, there needs to be more emphasis on determining the implications of dissolution of the core constituents, as many researchers—more recently Argonne National Laboratory—have shown that not all core shell catalyst designs are stable. It is unclear to this reviewer what is exactly known about that. The reviewer wants to know how long the high catalytic activity persists once the core elements are gone and how the core corrosion can be minimized.

Question 5: Proposed future work

This project was rated **3.0** for its proposed future work.

- Future work plans are solid and well defined. It would be nice if other non-PGM materials are examined besides Pd and Au.
- The work proposed is clearly leading to scale-up of the more promising approaches for serious MEA characterization. In fiscal year 2012, there is still a wide diversity of material approaches being explored that might be able to be down-selected to fewer, more promising candidates to develop by the 2015 target date.
- The proposed work is appropriate.
- The proposed future work is logical. While not appropriate at this time, by the end of next year the project would benefit from a down-select and more focus on one or two options.
- The strong collaborative effort is ongoing.
- The project team should switch to lower-cost core materials as soon as possible. The team should also conduct MEA tests for both performance gaps (including on air) and durability effects, considering possible interactions with existing degradation mechanisms. Finally, the PI should increase the alignment of catalyst approaches with collaborators and outline a clear path for how the approaches will mesh.
- Most of the work proposed for next year focuses on Pd, which only offers marginal benefits when addressing precious metal concerns. Scaling-up catalyst and MEA fabrication at JMFC are highly anticipated components of the project.
- There is a history of continuation on the core-shell path. The proposed future work focuses on new approaches for making such active nanoparticles for ORR. There needs to be a pause, and some thoughtful experiments that document durability. This reviewer wonders if the activity enhancement lasts for 500 hours, for example, if it makes any sense to pay for that extra performance with a stack that needs to last for 20,000 hours. This issue should not be ignored.

Project strengths:

- This project features a world-class electrochemist and strong collaborations with industry (e.g., JMFC and Toyota).
- The PI is clearly the visionary for these very successful approaches. The high productivity of this group is also a strength. The other strength is the breadth of the collaborators' expertise.
- The team is solid. Other strengths include the facilities, the national laboratory, and the partners involved who bring a broad spectrum of talent.
- This project's strengths include its catalyst nanoparticle synthesis and the stability of the nanoparticles due to the Pd interlayer.
- This project's strengths are the team of excellent scientists and the approach toward preparing monolayers of Pt on stable constructs such as WC, TiC, and oxides.
- This project has a strong science thrust with very interesting results using diverse techniques.
- The BNL team has many excellent members and their skill sets impress.

Project weaknesses:

- As reported and covered in the presentation, the PIs seem too focused on some scientific areas with lower priorities. The project needs more materials science.
- Replacing one very expensive metal with another expensive metal may end up being fruitless from a cost-perspective unless some other very unique catalytic features present themselves.
- The project's weakness is that these ideas and concepts for monolayers of Pt may be difficult to translate to large-scale production.
- The project is very broad for the project funding level, and there is no real discussion about weighing different approaches and down-selecting or prioritizing research direction. The team is strong, but the actual team roles and interactions were not communicated effectively.
- The durability issue must be addressed because these nanocatalyst advances may prove to be not useful.

Recommendations for additions/deletions to project scope:

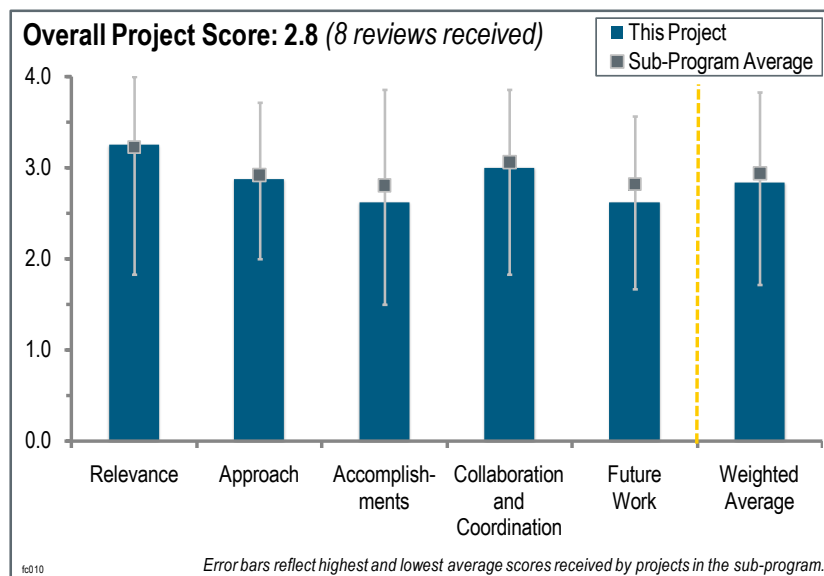
- The project team should investigate other non-PGMs and conduct extensive fuel cell testing.
- Investigators should add more materials science and solid state chemistry expertise (or at least report on it). The work on the carbide supports is good, but most such systems exhibit non-stoichiometric ratios (e.g., WC[1-x]), which is common, depending on the synthesis of such materials. The value of “x” will then induce different characteristics of the support itself (even semi-conducting). This reviewer suggests that this be factored into the effort.
- The work should concentrate on the more promising nanoparticles that have demonstrated they can meet the targets, unless there is some direct evidence the Pd nanowires can be made thin enough to meet DOE’s overall PGM loading targets.
- The project team should focus on materials scale-up and the processes that could allow for cost-effective catalyst production.
- Investigators should focus on durability and the implications of core-shell corrosion, a process that has been well documented.

Project # FC-010: The Science and Engineering of Durable Ultralow Platinum Group Metal Catalysts

Fernando Garzon; Los Alamos National Laboratory

Brief Summary of Project:

The objectives of this project are to: (1) develop durable, high-mass activity platinum group metal (PGM) cathode catalysts that enable lower cost fuel cells; (2) elucidate the fundamental relationships between PGM catalyst shape, particle size, and activity to help design better catalysts; (3) optimize the cathode electrode layer to maximize the performance of PGM catalysts and thereby improve fuel cell performance and lower cost; (4) understand the performance degradation mechanisms of high-mass activity cathode catalysts to provide insights to better catalyst design; and (5) develop and test fuel cells using ultra-low loading and high-activity PGM catalysts to validate advanced concepts. This project will help lower the cost and the precious metal loading of polymer electrolyte membrane (PEM) fuel cells as well as improve catalyst durability.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.3** for its relevance to U.S. Department of Energy (DOE) objectives.

- The project addresses the DOE goal of reducing the cost of fuel cells and increasing their durability.
- This project's stated goals are well aligned to the need for cost reduction in fuel cells.
- The effort addresses catalysts and use in electrode structures.
- The overall relevance to the DOE mission is very good and timely, considering the cost and availability of Pt.
- The project attempts to address several issues for catalysis in PEM fuel cell cathodes.
- The project is attempting to develop new catalysts for oxygen reduction, which is an entirely relevant pursuit in the development of automotive fuel cells. The project is attempting to go beyond Edisonian approaches and understand the relationships between activity, shape, and size. The most advanced catalyst projects in the DOE Hydrogen and Fuel Cells Program have generally attempted to do this.
- This project addresses optimizing PGM catalyst activity to reduce the amount of PGMs required and reduce fuel cell cost.

Question 2: Approach to performing the work

This project was rated **2.9** for its approach.

- Los Alamos National Laboratory (LANL) uses a very strong combination of theory and experiments to develop deeper understanding. The approach comprises a good combination of conventional and novel morphology catalyst assessment, as well as novel approaches to develop platelet catalysts to optimize performance. The project will provide critical information regarding the viability of reducing catalyst size and loading. It incorporates important consideration of electrode structures and performance beyond simple model catalyst studies to correlate purely catalyst materials properties with fuel cell performance. The tasking is well defined, and the task participants are appropriate based on individual team member expertise areas.

- The project is well designed with respect to combining experimental and modeling approaches.
- The approach of this project is somewhat diffuse due to its broad plan. It seems to be a very exploratory type of project and its objectives are pretty far removed from real systems. Exploring the geometry of catalysts seems like a novel idea.
- This is a truly outstanding example of incorporating the relevant modeling into each aspect of the project to complement the development of catalysts and electrode structures.
- The overall approach of the project is very good, with the correct combination of theoretical and experimental aspects. Some of the approaches, however, are not completely in line with the DOE objectives. For example, the idea of putting Pt on nanowires is difficult to reconcile with the specific activity requirements of DOE. In most of these approaches, scale-up would be a considerable challenge.
- The project seems to repeat others' work. Vienna ab initio simulation package (VASP) calculations have been published on Pt₃Ni. There has been much work on tubular Pt structures and carbon stability. It is not clear why LANL is pursuing these paths. LANL should stop doing rotating disk electrode (RDE) in sulfuric acid. The data look poor and cannot be compared to the up-to-date body of literature. The data also look like the slope is too great, which suggests impurities. CeO₂ (ceric oxide) is unstable in acid, or so say the Pourbaix diagrams. Any doping to give CeO₂ electronic conduction abilities will probably not work because electronic conduction in CeO₂ relies on a M³⁺/Ce⁴⁺ hopping mechanism, which is an activated process and unlikely to work at low temperatures.
- The approach begins by using theoretical modeling to understand catalysts, catalyst layers, and catalyst-support interactions. However, the modeling can only be as good as the data inputs, and inputs for many of these topics are immature. The models used here do not encourage increasing particle size, which is in opposition to data that suggests larger particles provide higher specific activity. The intrinsic activity of Pt has not played a role in the models. Pt/Pd nanoplates are conceptually interesting, but a lower cost core is preferred. The likelihood of CeO₂ being soluble in acidic media is fairly high. The Pt/polypyrrole concept is interesting, but the Pt thickness must decrease.
- This reviewer is not entirely convinced that this project has a central theme or focus. From the presentation, it seems that there are a lot of independent thrusts, and it was difficult to see how they linked together to yield an overall picture. The baseline data presented (mass activities) appear to be exceedingly low per the standard in existence today. It was unclear how the work differed from approaches proposed by other researchers (e.g., Markovic, Adzic, etc.) other than the much lower activities obtained in this study. The team is fine, the objectives are worthy, and it is also acceptable to study similar systems as proposed by others. However, it will be much better if the principal investigators (PIs) identified 1–2 key areas as a “go”, discarded the rest, and focused on exemplary work (which they are certainly capable of performing). The reviewer believes that the project is currently handicapped by the different thrusts and not by the personnel, who are excellent.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.7** for its accomplishments and progress.

- The project team achieved key milestones within each task area during the prior year. The findings regarding Pt particle dispersion on carbon illuminate a key challenge going forward: increasing catalyst nucleation site densities on stable carbon supports. The initial results of the nanoplatelet and Pt/ceria/carbon catalyst are promising. The initial modeling results are intriguing; it will be interesting to see how they influence future experimental work.
- LANL has made significant progress with respect to understanding through modeling, as well as progress with respect to the synthesis of materials with promising electrochemical properties toward the oxygen reduction reaction.
- LANL has made good progress. It was very gratifying to see a model predict how one should approach the solution (as opposed to explain what happened). Two examples include the simulation of the inherent instability of a 10-nm (nanometer) diameter metal nanotube, and, more importantly, showing that a support surface with tailored holes can actually promote catalyst stability by inhibiting ripening.
- The accomplishments have been very good to date. All efforts are in line with the theoretical efforts.
- The progress has been fair—some of the data do not really seem useful, such as the RDE results. The modeling team needs to be careful about how it considers the electrochemical potential in VASP. This reviewer's experience is that VASP only works for electrochemical systems with validated electrochemical results.

- RDE results have been compromised thus far by the use of sulfuric acid, compared to standards. Even with sulfuric acid, the baseline data are lower than expected. For example, 20% Pt/C is shown to provide only 11 A/g (amps/gram) of Pt. Due to expected fluctuations in precious metal price with automotive demand, Pt/Pd nanoplate mass activity data should be normalized based on total PGM loading. If this normalization is done, the activity is likely very low for these materials. For Pt/polypyrrole, it would be interesting to know the character of the Pt surface and whether the Pt film could be described as conformal. The connection between the modeling results and the driver for the three experimental families is not clear.
- The PI presented a large amount of work. Because of the approach and a somewhat disorganized presentation, it was difficult to assess how close the team is to its objectives.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.0** for its collaboration and coordination.

- This project features very strong collaboration and coordination with other institutions. Institutions participating in the project provide significant expertise in catalyst development and characterization, as well as electrode and cell development and testing. Team members provide complementary expertise that covers all aspects of the project.
- The project features well-coordinated collaboration between LANL and universities.
- The partners seem to bring broad experience to the team, and the scope of work is generally well suited to their expertise.
- There is well-delineated and coordinated effort between partners, including a stack partner ready for validation.
- This project seems to be a well-coordinated effort.
- The original equipment manufacturer (OEM) partner (Ballard) is fairly unpublished on RDE/rotating ring disc electrode (RRDE) testing, which is important in the opening stages of this project. Other partners are unlikely to assist. The microstructural model contributed by Ballard appears to confirm what has been generally known about desired ionomer loading. Nanoparticle growth and nucleation models from the University of New Mexico have not had a major impact on the material development aspects of the project. The presentation slides could better point to collaboration efforts where they exist.
- The LANL team would benefit from looking at literature about the state-of-the-art in many of their chosen research areas more carefully.

Question 5: Proposed future work

This project was rated **2.6** for its proposed future work.

- The outline of planned future work for each task area includes next logical steps, including work toward a key go/no-go decision point on the polypyrrole work.
- The plans address overcoming barriers.
- The future plans seem pretty well aligned to the objectives. This reviewer would like to see more effort on focusing activities and simplifying the presentation of progress. The reviewer would also like to see some assessment of the practicality of using Pr in the catalyst, and wonders how rare this element is relative to Pt. Additional rigor regarding the definition of “optimizing” the catalyst and what that implies would be helpful.
- The PI notes the need for membrane electrode assembly (MEA) results, and this reviewer agrees.
- The proposed future work is in line with the milestones.
- Pt/ceria catalysts need to move toward *in situ* durability testing immediately. Before improving dispersion, Pt/polypyrrole catalysts need to be fabricated with a lower thickness Pt film. Pd platelet thicknesses will likely need to decrease from 15 nm, or a low-cost replacement for Pd needs to be found. The future work slide shows that modeling needs to progress toward addressing the material development concepts.
- LANL is pursuing some avenues that do not seem productive. LANL needs to straighten out its RDE problems, and make sure that its VASP code is validated.

Project strengths:

- This project's strengths include using insights from modeling for catalyst design and employing novel synthetic approaches.
- The project shows a balance of modeling and experimentation. Good collaboration is also shown.
- This project represents an outstanding demonstration of the power of modeling to lead developmental efforts, and is run by a good team.
- Project strengths lie in the overall goals of lowering Pt loading while increasing its durability. The approach of trying the coating on various surfaces, including conducting polymers, is novel and merits such an effort.
- This project has a good team.
- The strength of this project is how it examines a broad range of topics.
- Images of all catalyst concepts are excellent and provide a clear identification of sample morphology.
- The general concept of Pt layered conformally onto a nanoplate is in agreement with increasing activity through the use of higher coordinated surfaces of Pt.
- Although Pt was layered too thick on the polypyrrole, the concept of a conformal layer of Pt on a polymer could possibly yield activity and stability benefits.

Project weaknesses:

- The project would benefit from a clearer discussion of how and when theoretical methods will be validated. For example, which aspects of the theory will be validated first, and why.
- The predictions of the density functional theory modeling with respect to the stability of Pt nanotubes have not been validated.
- Careful RDE characterization is necessary for this project. To make a valid comparison, Pt/C standards should be characterized in the same electrolyte as oxide-supported catalysts. Pt loadings of $50 \mu\text{g}/\text{cm}^2$ (micrograms/centimeter squared) seem too high for thin-film measurements. If RDE measurements are not performed in a thin-film limit, no reliable activities can be extracted from polarization curves. To be consistent with data in the literature, it would be better to use 0.1 M HClO_4 as an electrolyte.
- Core-shell structures for Pt/Pd nanoplates have not been confirmed. High specific activities for low-loaded, oxide-supported catalysts do not guarantee the same for high-loaded catalysts.
- The modeling is not connecting with material developments. The modeling shown in the presentation involves nucleation sites on carbon, as well as ionomer loadings and restraining particle sizes. However, the materials developed generally do not involve Pt nanoparticles on carbon, but instead involve Pt monolayers or other conformal layers, or Pt on oxide supports.
- The material development needs to address possible barriers. The Pt/ceria work needs to move aggressively toward stability measurements. In the Pt/Pd nanoplate work, the PI should seek to decrease Pd significantly or choose a non-PGM. In the Pt/polypyrrole work, the PI should seek methods for decreasing Pt layer thickness.
- The principal weaknesses of this effort are the disconnect with the need to enhance specific activity on a real surface area basis and some of the approaches, which are exotic at best and do not contain any effort showing how they could translate to scale-up.
- The team is dabbling in many areas and is doing cutting-edge problem solving. The electrochemical data look poor.
- Weaknesses of this project include the project focus and connection implications for practical fuel cell stacks and systems.
- The electrode and MEA fabrication is an area of weakness.

Recommendations for additions/deletions to project scope:

- Modeling performance as a function of nanoplates packing would be helpful for future design of the catalyst layer. With respect to oxide-supported catalysts, the project should focus on the synthesis of catalysts with higher Pt loading.
- If carbon is used as an additive to increase porosity on platelet type supports, the type and impact of the carbon should be investigated as it may introduce a source of corrosion into the system.

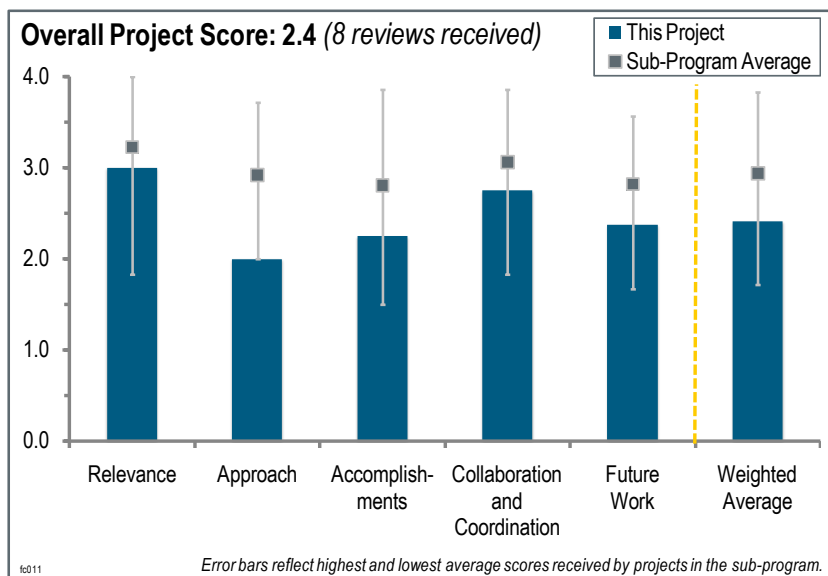
- The project team should clean up the RDE experiments and move to an HClO_4 electrolyte and meet state-of-the-art performance for Pt/C standards. It should also stop work on Pt/CeO₂ because it has little chance of being stable, or having any likely catalytic or electronic benefit. The PIs should validate the VASP results.
- It would be good to report crystallographic orientations of the Pt surfaces in the nanoplate and polypyrrole work. The nanoplate work should expand beyond Pd. The deposition of Pt onto polypyrrole/starch nanowires could be done by a variety of methods—atomic layer deposition, various types of galvanic deposition, etc. These could be explored to decrease the Pt film thickness.

Project # FC-011: Molecular-Scale, Three-Dimensional Non-Platinum Group Metal Electrodes for Catalysis of Fuel Cell Reactions

John Kerr; Lawrence Berkeley National Laboratory

Brief Summary of Project:

The project's objectives are to: (1) demonstrate that non-platinum group metal (non-PGM) catalysts can be used for oxygen reduction in polymer-coated electrode structures based on polyelectrolyte membranes; (2) incorporate catalysts into polymer binders of composite electrodes for the construction of membrane electrode assemblies to demonstrate that the matrix is effective for testing new catalysts; (3) demonstrate that the three-dimensional structure of polymer-coated electrocatalyst layers can offset the slower kinetics of the catalyst centers when compared with two-dimensional Pt or non-Pt catalysts; (4) demonstrate the possibility of significant matrix stability; and (5) demonstrate the design, synthesis, and scale-up of new catalysts capable of performance that is superior to platinum group metals.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.0** for its relevance to U.S. Department of Energy (DOE) objectives.

- Creating better catalysts for oxygen reduction is the most relevant subject of study toward commercializing automotive fuel cells. While non-PGM catalysts are not necessarily beneficial if they cannot provide adequate power density, they could provide cost savings and decouple fuel cell stack economics from the volatilities of precious metal markets.
- Total replacement of Pt on polymer electrolyte membrane (PEM) fuel cell cathode catalysts would tremendously advance the DOE Hydrogen and Fuel Cells Program toward a sustainable energy future. The potential benefit of this “Holy Grail” is so great that even projects with high risk in this area should be supported if they are properly grounded in knowledge of past efforts and are thought through in a logical manner. This project probably satisfies the latter criterion with highly creative, if perhaps surprising, logic, but may be lacking in the former.
- Development of non-PGM catalysts is obviously a major goal for the successful commercialization of fuel cell electric vehicles when considering the overall cost of the system.
- This project is relevant to the Program goal of lowering fuel cell costs by using a non-precious-metal catalyst.
- This project addresses DOE barriers A, B, C, and E.
- This project is very relevant, especially with the drive to substantially lower PGM loadings.
- Relevance is a generic problem for non-PGM catalyst projects. To achieve the DOE cost target, even though using non-PGM catalysts, good fuel cell performance at very high current density regions is needed because the material costs related to the fuel cell area (e.g., membrane, gas diffusion layers, bipolar plates) will need to be reduced. The PIs should evaluate whether the current non-PGM catalyst target is relevant to the ultimate goal of automotive fuel cells.
- This project is organized so that it has little or no chance of meeting any of the DOE goals for catalyst performance. It would be better as a DOE Office of Basic Energy Sciences project, since it does not seem responsive to the Office of Energy Efficiency and Renewable Energy (EERE) metrics.

Question 2: Approach to performing the work

This project was rated **2.0** for its approach.

- The project is coherent and comprehensive in that catalysts were/are being developed first, and that they will be incorporated into electrode layers in later stages. The project is well focused on the DOE targets. In the third and fourth years, researchers will investigate the layers and work on stability.
- The principal investigator (PI) claims an order-of-magnitude intrinsic advantage for homogeneous catalysts over heterogeneous catalysts. This reviewer wonders if tethered, and thus partially immobilized, catalysts would still be expected to have the full pre-exponential-factor advantage that the researchers claimed. The approach seems to have properly abandoned the pH-sensitive enzymes of last year's proposed work to concentrate on more stable macrocycles with a history of some oxygen reduction activity. However, such macrocycles—admittedly without the formal tethering attempted here—have been studied as oxygen reduction reaction (ORR) catalysts since the 1960s and, though more stable than some of the systems proposed last year, have (in molecular form) shown very poor stability in the acid electrolytes proposed to be used here. Extensive pyrolysis of such molecules has been necessary to provide any semblance of stability in acid. The use of the ferrocene redox couples and electroactive polymers to conduct electrons between the current collector and the tethered molecular catalysts is an improvement.
- The background described in the approach appears to indicate that a demanding turnover frequency will be required to allow the concept to work. A list of materials is shown, but it is unclear whether any particular material has already been shown, via proof-of-principle, to demonstrate the turnover frequency needed. While models may show that decent polarization curves may be achieved, this reviewer questions how much is known about the modeling inputs—such as transport parameters, kinetic parameters, and factors—that contribute toward the open-circuit voltage (OCV).
- The approach is a bit scattered and does not really focus on any one path. It comprises a bunch of feeble attempts in a variety of directions. The PI is relying too much on his past work on imidazole-based membranes.
- The approach does not seem feasible, as an increased amount of ionomer in the catalyst layer leads to decreased electronic conductivity of the layer due to the non-electronic-conducting ionomer blocking the carbon support.
- The PI seems to be taking a theoretical model approach to identify catalyst materials and the optimized electrode structure. However, it is unclear what part of the model developed in this project is dedicated for non-PGM catalysts and electrodes. The material experiments seem to be ad-hoc.
- To achieve a good approach, the project team should explain which group is responsible for what part of the project on slide 3 in the relevance section.
- This project is unlikely to contribute to overcoming the barriers. Other institutions have tried to make homogeneous catalysts for polymer electrolyte membrane fuel cells (PEMFC) and failed. The approach does not seem viable.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.3** for its accomplishments and progress.

- The technical accomplishments are very good on all levels, although the membrane electrode assembly (MEA) tests show that it is very difficult to substitute Pt (see chart 19).
- Each test provides informative data. However, this is very generic and it is questionable how the project team will achieve proper fuel cell performance with such a low OCV performance (slide 19). The OCV target should be identified for material screenings.
- Achieving any ORR activity from these tethered systems for any length of time is an accomplishment, though the molecular catalysts in untethered form have generally shown some temporary activity in the past. The potential at which substantial activity has been achieved have been well below those needed for a practical fuel cell.
- It appears that 600 mV (millivolts) is the highest potential at which any electrochemical activity is seen in these systems, and most of the halfwave potentials were around 200 mV. The Lawrence Berkeley National Laboratory (LBNL) calculation that their approach has the potential to match Pt is interesting and deserves more discussion than could be given in a short presentation. The observation from calculations that outer

substituents on porphyrins can modify ORR activity is certainly not news. The use of free or polymer-bound redox couples to provide some conductivity between the molecular catalysts and the current collector or electrolyte is an improvement over the state of this project last year.

- Tetrakis (N-methyl-4-pyridyl) porphyrin (TMPyP) catalysts do not show oxygen reduction onset until they reach potentials lower than 0.6 V (volts). Adding ferrocene increases onsets by only 200 mV, which is still not high enough. Dipyrromethane (DiPM) and 5- (4-aminophenyl) dipyrromethane (APDPM) catalysts do not show high onsets. Despite attempts to represent the catalysts through modeling, the project does not contain a morphological study that investigates whether there is something similar to a porous polymer layer on a support. Given the relative success of similar metal-N-C catalysts, it would be worth knowing why onsets continue to be low. This reviewer wants to know if the site density can be quantified, and if metal-N bonds are preserved. MEA test results show extremely low performance.
- This reviewer wonders if modeling has predicted the optimum thickness that can be used before oxygen transport limitations occur. Researchers mentioned that a turnover frequency of 10^5 is needed for 1 A/cm^2 (amp/centimeter squared), but this requirement does not appear to be discussed any further.
- The project has made some progress toward Program objectives, but has not made any progress toward EERE barriers.
- Rotating disk electrode (RDE) experiments thus far have not indicated anything of real interest. The fuel cell performance curves are very off target from the state-of-the-art, even for non-PGM catalysts.
- The rate of progress is slow. The project is far from reaching the DOE goals.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.8** for its collaboration and coordination.

- This project features well coordinated collaboration between academia, national laboratories, and industry.
- For this work, LBNL took strong partners with an outstanding track record in fuel cell research and development, such as Los Alamos National Laboratory (LANL), 3M, and the University of California, Berkeley.
- There seems to be a lot of interaction between the groups.
- The project features good collaboration with other institutions and companies.
- The main collaboration appears to be with LANL, which provides both catalytic moieties and MEA integration. However, LANL has guided successful non-PGM catalyst projects in the past. This reviewer wants to know if LANL is contributing more than materials and testing, and if collaboration could be extended so that LANL could suggest techniques (e.g., thermal and acid treatments) that would help to increase metal-N site density. The perspective of a stack original equipment manufacturer or integrator is missing. The project should be guided toward addressing concerns with activity before fabricating MEAs. The role of 3M is not entirely clear.
- This project includes some people who must be familiar with the study of some of the molecules used as catalysts in the project. One would hope that some of the accumulated knowledge of the past behavior of non-Pt catalysts would be transferred between the groups, along with samples of the materials.
- Collaboration is pretty evident between the two national laboratories, but the overall level of work is pretty poor.
- The number of collaborative partners in the project is appropriate; however, the project management needed to orchestrate each task and material experiments toward the project goal is unclear.

Question 5: Proposed future work

This project was rated **2.4** for its proposed future work.

- The basic research approaches and the down-to-earth testing of the catalysts to be developed are well-balanced.
- This project has a good, very complete plan for future work.
- The proposed future work is based on overcoming barriers.
- The PIs should measure actual parameters to compare to the “reasonable” parameter values used in the calculation that is purported to show that this approach should be able to match Pt. They should also develop a way to incorporate non-Pt catalyst centers with a better history of stability (e.g., Fe-C-N catalysts or pyrolyzed macrocycles). While incorporation of the redox wires in the catalyzed polymer layer makes this approach seem

more plausible, further clarification is needed regarding how electronic and ionic conductivity pathways can make this concept work. It also might be good to demonstrate Pt tethered in the polymer as a catalyst to demonstrate proof of the structural concept before going with a non-Pt system.

- The criteria for materials screening is unclear.
- This reviewer is not sure if the plan for future work will allow the PI to match the current state-of-the-art for non-PGM catalysts, as the PI is currently so far away.
- The goal of showing catalysts in MEA with a stability target of 10 hours is irrelevant.
- Preparing MEAs with the present materials is entirely the wrong direction. If no materials can demonstrate OCVs greater than 0.9 V, then no MEAs should be made. The team needs to examine carefully why OCVs are low, and why ORR onset is at low potentials. Any existing catalyst materials cannot proceed further until extensive modifications are made toward improving performance. Investigators need to understand what ORR mechanisms are taking place and why performance is not what it was expected to be.

Project strengths:

- This project features two leading national laboratories.
- The experimental design is based on insights from modeling. The PI has a strong background in fundamental electrochemistry that helps him understand reaction mechanisms.
- The project features strong partners, great catalytic approaches, and realistic testing.
- This project has a highly motivated team that is looking at homogeneous catalysts.
- It appears that the project is on track to complete its goals and reach DOE targets.
- Areas of strength for this project include the high level of creative logic in the development of the concept, and the attempt to test the concept with an electrode model. Another strength of the project is the change from the earlier biomimetic choice of catalytic molecular centers, as these would be stable only near neutral pH, where electrolyte conductivity is inadequate for achieving practical energy densities.
- One area of strength for this project was the investigators' willingness to consider a new idea, such as homogeneous catalysis. Except for this project, all of the catalyst projects funded in the Program derive their concepts from heterogeneous catalysis. This project team has been willing to see if a homogeneous concept could work. Another strength is the PI's ability to generate new organic chemistry. Although a catalyst with sufficient performance has not been shown, the project has shown the ability to at least deliver an impressive amount of different organic structures in a short period of time.

Project weaknesses:

- The current level of catalyst activity is well below where it needs to be, even for non-PGM based materials.
- The project is not focused on achieving DOE targets. It seems like it belongs in the Basic Energy Sciences portfolio.
- The project planning and management is a weakness in this project. Criteria should be relevant to the ultimate fuel cell performance goal for material screenings and should be clarified.
- This is a very long-term project, which can eventually be a project strength.
- This project has poor electrochemistry and needs to follow the methods developed for RDE of Pt-based catalysts, such as using 0.1 M HClO₄ and report loadings. Methods are described in the literature. The reported catalyst performance is poor. The selected materials are unlikely to be stable in a PEMFC cathode.
- Turnover frequency and a catalyst density sufficient to meet DOE goals are not scheduled until the third year—maybe they should be evaluated earlier. Evaluation using RDE and MEAs should come earlier, in parallel as much as possible with the catalysis synthesis work.
- The PIs do not give adequate attention to the historical development of non-Pt ORR catalysts for use in acidic electrolytes, particularly with regard to durability. Another weakness is the treatment of ORR currents around 200 mV RHE (reference hydrogen electrode) as if they portend eventual practical success in a useful fuel cell.
- The PI has simply not delivered a tenable catalyst. All catalysts have shown low onsets, and researchers must investigate why this is so.
- Vehicle efficiency will not be achieved unless the polarization curves begin from a fairly high OCV, at least above 0.9 V. Lower OCVs are not acceptable.

- MEA testing is for active catalysts that need to demonstrate some measure of durability. In this case, the catalysts are simply not active, and none of them are worth the time and effort of an MEA test.

Recommendations for additions/deletions to project scope:

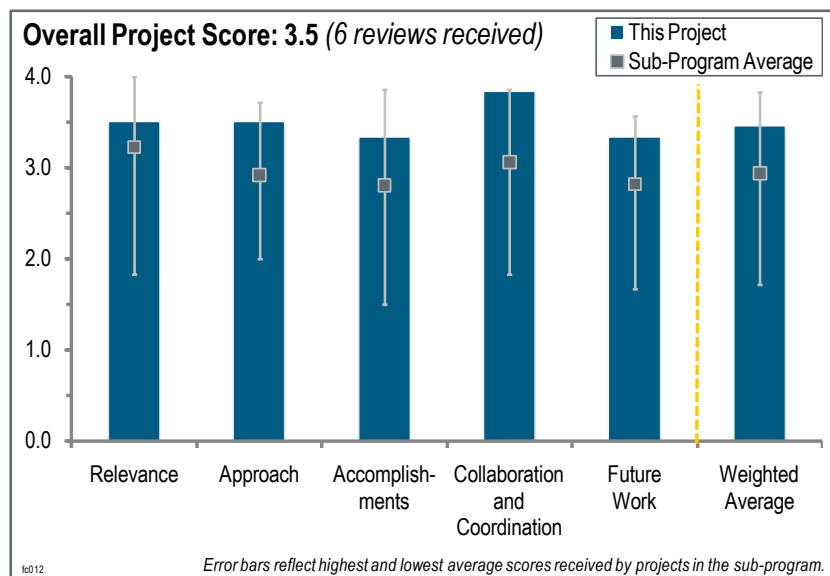
- The project team should focus on two or three key approaches and quickly try to reach acceptable activities.
- Criteria that are relevant to the ultimate fuel cell performance goal should be clarified for material screenings. An OCV target should be defined, at the very least.
- Someone from a credible fuel cell organization needs to explain to the team how ORR catalysts work, the standard methods for analysis, and prior methodology taken for modeling. Even with the high potential payoff of a successful non-Pt ORR catalyst, and with due respect for the need for trying unconventional approaches, the investigators should reconsider whether this project is plausible on the grounds of durability and activity.
- The project team should remove MEA testing and examine why the catalysts are not performing. This reviewer wants to know if there are critical differences between LANL non-PGM catalysts and those shown in the LBNL project, what the sites are in the catalyst, and if there is characterization that might reveal that expected sites are not being produced.

Project # FC-012: Polymer Electrolyte Fuel Cell Lifetime Limitations: The Role of Electrocatalyst Degradation

Deborah Myers; Argonne National Laboratory

Brief Summary of Project:

The project objectives are to: (1) understand the role of cathode electrocatalyst degradation in the long-term loss of polymer electrolyte membrane fuel cell performance; (2) establish dominant catalyst and electrode degradation mechanisms; (3) identify key properties of catalysts and catalyst supports that influence and determine their degradation rates; (4) quantify the effect of cell operating conditions, load profiles, and the type of electrocatalyst on performance degradation; and (5) determine operating conditions and catalyst types or structures that will mitigate performance loss and allow polymer electrolyte fuel cell systems to achieve the U.S. Department of Energy (DOE) lifetime targets.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.5** for its relevance to DOE objectives.

- This project is extremely relevant. Degradation is the most critical unresolved issue. This project addresses the key limitation of the entire fuel cell industry—membrane electrode assembly (MEA) degradation—and is attempting to provide scientific explanations.
- Understanding lifetime issues in polymer electrolyte fuel cells is of paramount importance, and presently constitutes a major barrier to commercialization.
- This project features a very quantitative approach to resolving durability issues for Pt and Pt alloy catalysts.
- This project is very relevant because degradation mechanisms for catalyst electrodes and polymer electrolyte membranes must be determined for long-term success.
- To meet the DOE Hydrogen and Fuel Cells Program objectives on cell durability, it makes sense to first identify and quantify the various mechanisms that would adversely impact durability.

Question 2: Approach to performing the work

This project was rated **3.5** for its approach.

- This project features by far the most comprehensive work on degradation mechanisms. The team goes far beyond the prior superficial analyses by looking at the fundamental science and physics of the issues. This solid state and materials science approach is what has been missing in past efforts. This group's approach is to study the degradation at a much more comprehensive level. This project is long overdue.
- The approach is a very good mix of experiments, spectroscopy, and theoretical modeling.
- This project features an excellent and balanced approach for experimentation and modeling.
- This project appears to have a sound approach.
- Argonne National Laboratory (ANL) has elected to define a series of cell degradation tests to quantify predictably the amount of degradation to performance, as well as generate a number of characterization methods

to quantify the structural damage. These data are then used to generate a model to predict the damage and performance degradation as a function of operating history.

- Much of the data reported demonstrates well-known trends. While an avalanche of data have been gathered, it was not immediately evident how these data would be analyzed to provide a grass-roots understanding of catalyst durability.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- The project team had made excellent progress on the elements reported at the Annual Merit Review. Most major degradation mechanisms are being addressed, and the progress has been significant.
- The accomplishments to date are good. A lot of data have been collected and analyzed. With this much data, some novel approaches are needed to extract the critical information. Statistical methods are needed.
- This project features lots of good, quantitative data.
- The progress to date appears to be appropriate for the time elapsed on this activity.
- It appears that for Pt₃Co, the 14.3 nm (nanometer) size is the most stable (i.e., there is less degradation at 30,000 cycles). This reviewer wants to know if the principal investigators (PIs) expected this and if the results would be even better if the particle size was larger than 14.3 nm for Pt₃Co, or if that is the limit for particle size that can be made. The reviewer also wants to know if the PIs have a chart for Pt₃Co similar to the one on slide 19 for MEA cycling, and what size MEAs are being tested.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.8** for its collaboration and coordination.

- The collaboration is excellent. The team is contributing on many fronts and includes the right expertise and technical capabilities. The team is also looking at other sources of information and data, in particular the Durability Working Group. This non-competitiveness is refreshing. New collaborations were mentioned at the Annual Merit Review, allowing the team to evaluate different MEAs that are fabricated by different MEA suppliers.
- The team seems very well coordinated and very productive. ANL did a great job pulling together an outstanding team, and seems to be managing the project well.
- This project features excellent coordination of the work. The Kinetic Monte Carlo modeling work is excellent, and this reviewer is very interested in the future work concerning optimal concentrations and sizes for alloy particles.
- The collaborators involved appear to have adequate breadth to address this task.
- Outstanding collaborations have been established.

Question 5: Proposed future work

This project was rated **3.3** for its proposed future work.

- The future work is clear and the approach is very good.
- The team should keep up the good work.
- The proposed work appears to be a rational approach to reach a satisfactory completion of this project.
- The continuation of the existing effort is significant enough. The results of the current tests and analyses will guide the path forward. The testing of non-carbon-supported catalysts is an obvious next step that this team already has plans to do. This reviewer would like to see non-ionomer-based electrodes included in the work plan. The reviewer would also like to see the impact of fluoride ions on the MEA, as they are released at significant levels in the first 100 hours of operation. Most electrodes are not washed after deposition. The reviewer recommends understanding the impact of the chemistry of such impurities on the electrode performance.
- For modeling, the proposed future work is excellent. This reviewer has tried Pt₃Sc and was not impressed with the results. The reviewer hopes the project team has better results.

- More emphasis should be placed on data analysis and experiment design, as opposed to routine experimental measurements.

Project strengths:

- The project strength appears to be the participants' skill sets.
- The project features a phenomenal team, which is producing results as expected from a national laboratory. The team is solid enough, asking the right questions, and focusing on the entire picture. The collaborative effort is appropriate.
- The project is well laid out in terms of approach and choice of partners, tools, and methods.
- Areas of strength include the project's good team and excellent collaborations.
- The project has a very well-organized, focused, and productive team focused on addressing key Office of Energy Efficiency and Renewable Energy questions. The team is able to carry out difficult experiments to achieve useful results.
- The project has achieved very interesting and important results so far on particle size effects. This reviewer is interested in Pt-Co catalysts, and thus enjoyed this report and looks forward to what can be achieved in the next year and a half.

Project weaknesses:

- The reviewer would like to see testing of the U.S.-based MEAs. The only issue is that there are too many topics to address.
- This reviewer wants to know if the project team has identified all of the degradation modes, and what happens if it misses one or two.
- Others have had some excellent results with Pt-Ni, and this reviewer wants to know if ANL has thought about trying this material.
- More statistical tools are needed to cull the critical information from all of the complex data obtained to date.

Recommendations for additions/deletions to project scope:

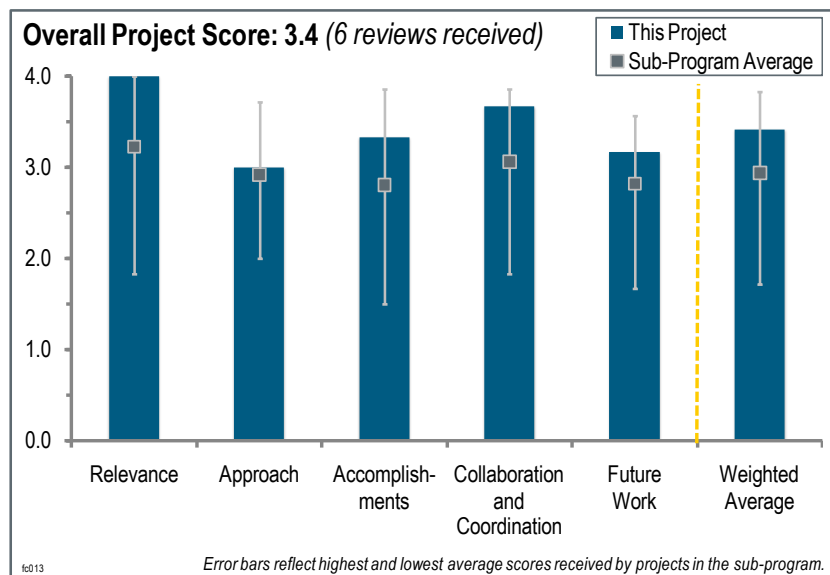
- When it makes sense, the PIs should include the impact of electrode architecture and the microlayer composition and chemistry, as well as determine if the gas diffusion layer (GDL) has an impact. (It was not uncommon to bake out the sulfur at 400°C from the as-received GDL prior to preparation for the fuel cell application.) The impact of air quality on cathode chemistry and materials stability will eventually have to be addressed in actual road conditions.
- Alloys are tricky to work with because they are so variable. The team might consider looking at different variations (i.e., vendors) of Pt₃Co. ANL might want to look at catalyst durability in sulfuric acid to match the MEA data.
- The team should possibly remove Sc and try other alloys.

Project # FC-013: Durability Improvements through Degradation Mechanism Studies

Rod Borup; Los Alamos National Laboratory

Brief Summary of Project:

The objectives of this project are to: (1) identify and quantify degradation mechanisms, including (a) degradation measurements of components and component interfaces, (b) elucidation of component interactions, interfaces, and operating conditions leading to degradation, (c) development of advanced *in situ* and *ex situ* characterization techniques, (d) quantification of the influence of an inter-relational operating environment between different components, and (e) identification and delineation of individual component degradation mechanisms; (2) understand electrode structure impact; (3) develop models relating components and operation to fuel cell durability; and (4) develop methods to mitigate degradation of components through new components, properties, designs, and operating conditions.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **4.0** for its relevance to U.S. Department of Energy (DOE) objectives.

- Durability is one of the critical challenges to commercializing fuel cells. This project addresses the durability of several individual components (i.e., electrodes, membranes, gas diffusion layers [GDL], bipolar plates), and includes modeling efforts to understand the impact of combined degradation on performance.
- This project is in direct alignment with DOE goals.
- Identifying and quantifying a degradation mechanism is a fundamental part of fuel cell research that could lead to commercialization-enabling technology development. It is important to take a systematic approach in order to identify the critical factors among the many degradation factors.
- This project is providing excellent mechanistic and parametric understanding of degradation in polymer electrolyte membrane fuel cell systems, specifically lifetime hours and cyclic durability, without sacrificing cost. The characterization of membrane degradation, catalyst/electrochemically active surface area loss, and mass-transport effects is addressed in a holistic, largely well-integrated approach.
- This project explores many failure mechanisms with different cell components, which is clearly relevant to DOE goals.
- This project seeks to increase fuel cell durability, but not at the expense of component cost. Identifying the factors affecting degradation (or lack of degradation) and monitoring the effects of these factors will help guide what to use and what not to use as an efficient, long-lived, and practical fuel cell power source.

Question 2: Approach to performing the work

This project was rated **3.0** for its approach.

- This project includes a wide range of component durability investigations. The impact of interactions between layers will be important to understand, and models may need more empirical data to accurately reflect component interactions.
- This project features an extensive approach with many variables to manage.
- This project includes direct measurements and metrics for finding and characterizing degradation mechanisms, as well as correlating to fuel cell durability over the gamut of components, including catalysts, electrolytes, bipolar plates, sealing gaskets, etc.
- Durability measurement would be varied with materials. The project should make sure to capture promising technology to meet cost and performance targets with the durability metrics. For the catalyst/membrane electrode assembly (MEA) area, low-Pt-loading technology that could potentially meet cost and performance targets should be covered. An example of this technology includes the Pt alloy catalyst bulk property concept, including 3M's nanostructured thin film (NSTF).
- Analysis to define individual component contributions to loss in performance is important, and if the investigators successfully integrate all of these modes of performance degradation, they will have delivered something very important. Quantifying changes in surface species and morphological changes are critical to developing the necessary key insights. Coupling the membrane and electrocatalyst seems very important. It is not clear how the transport-related phenomena addressed in the bipolar plate are tied in with the other key modes.
- This project explores known failure modes and current accelerated stress tests. The scope and the group are a bit large; a more focused effort would have been preferable.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- This project has made excellent progress. This reviewer did not see a quantification of the percentage completed or a project start date.
- This project met a number of milestones using a parallel effort by multiple partners. The investigators studied the major components of fuel cells and their integration, including catalyst activity (and effects of water), support stability, ion conducting membrane performance and stability, and bipolar plate and sealing gasket durability with modeling performed to organize results.
- This team has made very good progress and has produced good insights. It was interesting to see the MEA variability, as well as the results from the performance degradation model.
- The project team has done excellent work on the carbon corrosion and the change in effect on water retention. The investigators have done similar good work on polymer aging and the effect on polymer structure. These types of fundamental work can help investigators on many platforms.
- Lots of backup slides substantiate claims that significant progress was made on all project milestones.
- The test data is interesting, but tested materials should include promising technology that can meet cost and performance targets with the durability metrics. Test data without this consideration may not be meaningful. For example, if researchers pursued durability testing for various ionomers (e.g., short-side chain versus long-side chain, and carbon/ionomer ratio) with a conventional Pt/C catalyst electrode, the testing would not be applicable for low-Pt loading technology.
- The membrane crystallinity change was a good finding. It was necessary to identify that this change would lead to failure, and determine what kind of failure mode and its mechanism.
- Seal (gasket) degradation would be highly dependent on design configuration. More detailed design information should be shared. Material robustness, such as the compression set for various temperature profiles, should be covered. Both high- and low-temperature (sub-zero) regions should be included.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.7** for its collaboration and coordination.

- This project features extensive collaboration with relevant partners.
- The project team has done excellent work coordinating so many organizations.
- The work of the various team members appears complementary and well coordinated.
- This project features a very good division of labor among team members who are well-qualified for their roles.
- The project appears to have a well-integrated team, with good communication and results. The bipolar plate work seems a little bit removed from the rest of the project, though it might not require the same degree of integration.
- More detailed information of the material property and design should be shared along with the testing data.

Question 5: Proposed future work

This project was rated **3.2** for its proposed future work.

- The future work is in alignment with the objectives.
- The planned work on degradation mechanisms, electrode structures, component interactions, and modeling seems well thought out. The future results should bear this out.
- Looking at various materials and parameters is the next logical step.
- The principal investigator (PI) should make sure to capture promising technology to meet cost and performance targets with the durability metrics. For the catalyst/MEA area, low Pt loading technology that could potentially meet cost and performance targets should be covered. An example of this technology includes the Pt alloy catalyst bulk property concept, including 3M's NSTF. For seal and bipolar plates and short stack testing, it is necessary to share detailed design information along with the test data.
- The project team needs to clarify how the bipolar plate studies are integrated with the rest of the efforts to characterize overall degradation. Most other areas are one-dimensional or localized in nature; bipolar plate studies are highly platform specific and might be best handled in terms of corrosion or impurities.

Project strengths:

- This project's strengths are the excellent collaboration and world class team.
- A comprehensive examination of the major factors affecting fuel cell durability is being carried out by some of the best for each factor.
- The testing capability of the MEA durability testing is an area of strength for this project.
- This project features an excellent team and a good approach to understanding the fundamentals.
- This project has a very strong team and division of labor. Examining different materials and operating conditions adds relevance to a large number of investigators.

Project weaknesses:

- The team is geographically spread out. This has to have some impact, but the PI appears to be handling it well.
- It is unclear how the transport/GDL effort is integrated with or differentiated from other transport-related efforts, such as Giner's.
- The project is a large patchwork of collaborators and topics. Many of these topics have no overlap at all. It would likely be much better to have this project be a number of smaller projects with a PI for each topic.

Recommendations for additions/deletions to project scope:

- This is one of the better projects this reviewer heard about during the Annual Merit Review. The reviewer's only suggestion is that the conductivity of ion-conducting membranes only appears to have been studied indirectly, as was shown in iR-free fuel cell plots. The study of the direct correlation of conductivity to failure modes seems like a good idea for future work.

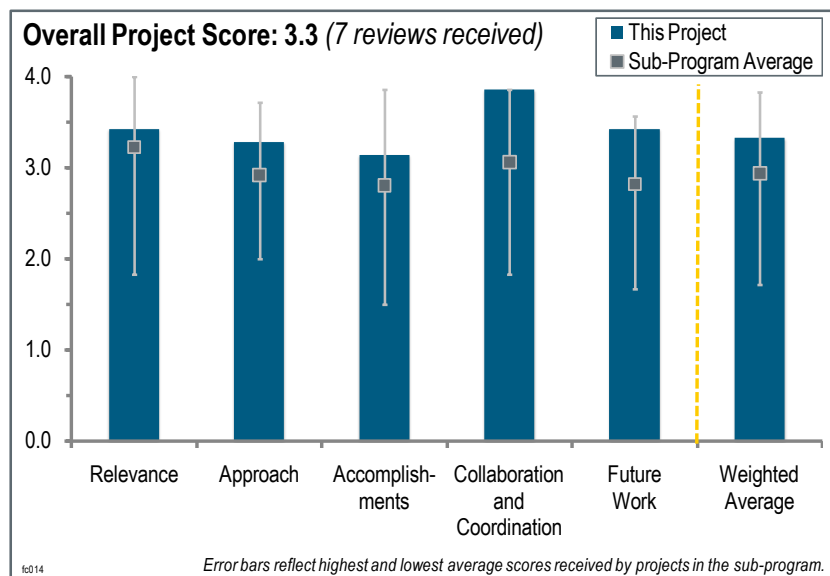
- Investigating the contamination from plate and seal materials may be a separate study.
- Materials selection is important for this project. Materials with promising technology that could potentially meet the cost and performance targets should be included with the durability metrics. For seal and bipolar plate durability, it is necessary to share detailed design information along with the test data.
- This reviewer recommends expending less effort on bipolar plates and more effort on understanding coupled effects and feedback loops.
- Few original equipment manufacturers are pursuing carbon composite bipolar plates; this effort could be dropped.

Project # FC-014: Durability of Low Platinum Fuel Cells Operating at High Power Density

Olga Polevaya; Nuvera Fuel Cells

Brief Summary of Project:

The objective of the Sustained Power Intensity with Reduced Electrocatalyst (SPIRE) program is to study decay mechanisms and identify strategies to ensure the durability of fuel cells capable of achieving the U.S. Department of Energy's (DOE's) 2015 cost target. The most significant enablers for achieving stack cost goals are increased power density and reduced Pt loading. The technical approach of the SPIRE program is to elucidate the critical durability mechanisms for a stack operating at a power density and Pt loading that can achieve DOE's 2015 cost target. The key deliverable of this program is a durability model that has been experimentally validated over a range of stack technologies operating at high power.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.4** for its relevance to DOE objectives.

- This project, which seeks to maintain higher power density operation with reduced catalyst loading over operating time, is relevant to DOE Hydrogen and Fuel Cells Program goals and durability objectives.
- Durability and cost are very relevant to DOE objectives.
- It is imperative to achieve both the targets of ultra-low-Pt loading and ultra-high-current density (power density) to meet the cost target. The durability metric at high power density is also important. The definition of this power density should be fixed (slide three). This Nuvera project leveraged a lower voltage target at rated output power (0.6 V [volts]) than DOE targets. Currently, DOE defines 0.67 V per cell at rated power from the thermal management standpoint, and this number should be used in this project.
- The project's targets of reducing fuel cell cost and increasing fuel cell durability are highly relevant.
- This project fully supports two critical DOE objectives—durability and cost. A durability model associated with low-Pt loading has the potential to define the conditions that would meet the DOE 2015 cost targets.
- Understanding the performance and durability of stacks at lower catalyst loadings is very important to the overall industry. One major overall criticism of this project is how it is very specific to Nuvera technology, as it is based on Nuvera's Single Cell Open Flowfield (SCOF) design.

Question 2: Approach to performing the work

This project was rated **3.3** for its approach.

- Verification of the modeling using a specialized single cell experimental arrangement is excellent.
- The technical approach used in this project is adequate, systematic, and well planned.
- The approach used in this project is fairly standard, and is directed toward increasing incremental understanding of degradation mechanisms affecting durability using modeling tools complemented with experimental validation.

- Using some cell designs that are public was a good choice that allows the results to be duplicated and shared.
- The overall approach seems good. The open flow field/“Orion” stacks are specific for the new durability metric of high-power density region. This design information should be shared. The platinum group metal (PGM) loading level for a series of testing is unclear (this reviewer wants to know if it is 0.5 mg PGM/cm² or 0.2 mg/cm²). Low PGM loading should be covered in this new durability metric and model validation. The catalyst technology for 0.2 mg PGM/cm² is unclear. This information should be shared.
- The approach is viable and has a good balance of experimental work and complementary modeling work. The experimental design work seems to be well thought out and should provide success. One major weakness is how the approach only really benefits Nuvera’s SCOF cell architecture

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.1** for its accomplishments and progress.

- The experimental design, experimental data, and modeling work are all proceeding very well, especially considering the relatively early stage of the project. The fundamental investigation of low loadings of cathodes is very interesting and insightful. The durability studies and post-mortem analysis are excellent.
- This project is fairly new and is only 1.25 years into an extended four-year schedule. The progress to date seems reasonable. The results on the open flow field architecture are encouraging.
- This project has made excellent progress, considering it is only 25% complete.
- Very good progress has been made. The project team demonstrated a good agreement between the performance decay model and experimental results for a single cell.
- Some comparisons of the specialized single cell experimental results to other single cell fuel cell tests have been done.
- It is not appropriate to use iR-corrected data for high power density (high current density regions) durability metrics. For catalyst degradation, durability should be evaluated by potential change at low current density (iR-free) during and after high current density durability protocols. The potential change at high current density should not be iR-corrected. Also, it is unclear if the partial oxygen pressure measurement (slide 10) of 1.2 W/cm² (watts per centimeter squared) at 0.2 mg PGM/cm² is iR-corrected. The outcomes of the membrane chemical stability evaluation (slide 13) are also unclear. It is recommended that the researchers measure the leached fluorine ion during and after durability cycling.
- The objective and key deliverable have changed since last year, but it is not clear how the presented project milestones will be used to evaluate progress toward the project objective. Specific validation criteria was not explicitly stated anywhere in the presentation. The results are not compared to DOE goals.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.9** for its collaboration and coordination.

- The work with national laboratories, while small in nature, is well coordinated and progressing as planned. Los Alamos National Laboratory's post-mortem durability membrane electrode assembly (MEA) analysis is very valuable to the project.
- This project features good collaborations with national laboratories for testing and characterization, as well as collaborations with an industrial MEA developer. The interaction with and participation in the DOE Durability Working Group are important.
- This project has an excellent team and mix of industry and laboratories.
- This project features very good integration of modeling and experimentation, as well as excellent team integration.
- The project activities are well coordinated among the team members.

Question 5: Proposed future work

This project was rated **3.4** for its proposed future work

- The plans for future work are reasonable and consistent with the project's scope.
- There is a good plan for future work among collaborators.
- This reviewer wants to know when comparisons between the model predictions and specialized cell results and experimental results from actual operational fuel cells will begin.
- The proposed future work is dependent on the project's progress and integrated in a logical manner. The incorporated go/no-go decision is appropriately positioned.
- The proposed future plan seems good, but detailed information of the cell design (open flow field, etc.) and low Pt loading catalyst technology should be shared. This information is necessary to clarify membrane chemical stability metrics.
- This project is well managed with an excellent design of experiments and programs. The major criticism is that this work is only really applicable to Nuvera's cell design.

Project strengths:

- This project is well managed and has an excellent balance of experiments and modeling. Progress thus far has been very high. The project features a strong, experienced, and capable team.
- This project's areas of strength include its strong team and good progress.
- The project's testing capability is an area of strength.
- This project targets the key issues for fuel cells, and employs an excellent combination of modeling and experimentation to verify model predictions.
- The project has a very good team. Each team member brings relevant expertise to the project.

Project weaknesses:

- The project is only really applicable to Nuvera's SCOF design.
- The performance and durability results should be compared with DOE targets. The model validation criteria should be explicitly stated. The milestones should relate technical progress to the project objective.
- The design and material information sharing is an area of weakness for this project.
- It will be quite important to verify that the single cell modeling and experimental results are applicable to actual fuel cell stacks.
- It is not clear whether sufficient data will be collected in time to make the go/no-go decision in the first quarter of 2012.

Recommendations for additions/deletions to project scope:

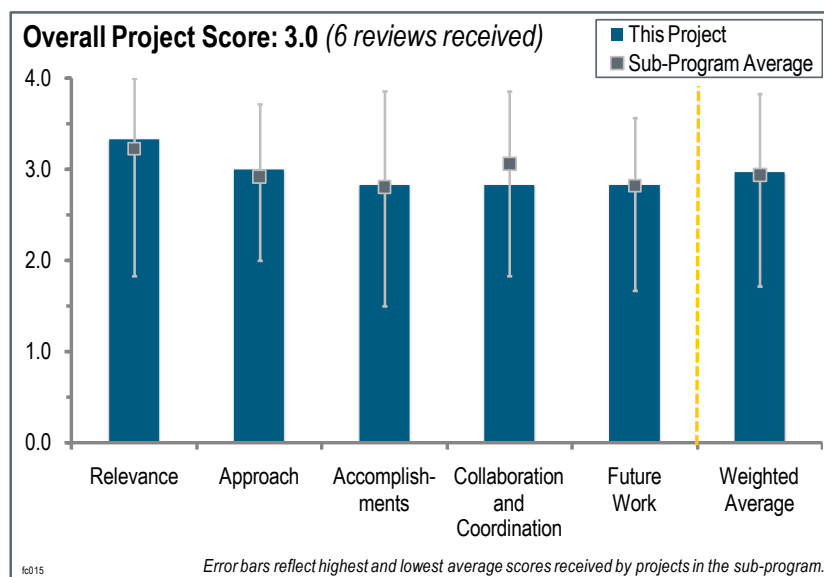
- The project should include mitigation strategies based on project findings and identify degradation mechanisms that are affecting durability. Understanding factors that contribute to reduced durability is important, but finding solutions that improve durability without increasing stack cost is even more important.
- This project leverages unique flow field and specific low-Pt-loading catalyst technology. Information sharing is important for these cases. The membrane chemical degradation evaluation and investigation of the mechanism with proposed test protocols (new stress tests) should be clarified.
- To exclude factors such as flow and heat distribution, the performance decay and durability tests should include a single cell stack with the same active area as the eight-cell stack.
- One reviewer had no recommendations and felt the investigators should continue the excellent work as is.

Project # FC-015: Improved Accelerated Stress Tests Based on FCV Data

Timothy Patterson; UTC Power

Brief Summary of Project:

The objectives of this program are to: (1) compare conditions and materials in bus field operation with U.S. Department of Energy (DOE) accelerated stress tests (ASTs), (2) develop acceleration factors for DOE AST mechanisms and recommend modifications, and (3) identify life-limiting mechanisms not addressed by DOE ASTs and recommend new ASTs. Tasks are to: (1) analyze performance data and characterize degraded materials from 2,850 hour stacks in bus service; (2) analyze data and degraded materials run in DOE ASTs (same as in bus stacks); and (3) correlate results for all current DOE ASTs including platinum group metal decay, carbon corrosion, and membrane mechanical and chemical degradation.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.3** for its relevance to DOE objectives.

- Correlating data from DOE ASTs with real-world operation and then validating the data is very relevant to the DOE Hydrogen and Fuel Cells Program. Understanding the degradation mechanisms and suggesting modifications based on findings are also good aspects of the project.
- Connecting real-world data to laboratory ASTs is important for next-generation development.
- This project compares actual performance and materials characterization from a bus stack operated for 2,850 hours to similar results from ASTs. The results will be correlated for catalyst decay, carbon corrosion, and membrane degradation. ASTs may also be developed for other stack components, such as the gas diffusion layer (GDL).
- The project offers a good statement about the issue for the current DOE ASTs and their gap to real-world usage profiles. However, filling the gap may be challenging. Materials and technologies that are available for real-world usage are not promising technologies that can possibly meet cost, performance, and durability targets. For example, only high-Pt-loading technology is available for current real-world usage, but acceleration factors developed from these data would not be applicable for ultra-low-Pt-loading technologies.
- The project may help UTC Power with its own system durability; however, it is the only company to use its system with porous plates, leading to very different system stresses. Great work will have to be done in order to make this work relevant to other investigators, and this has not been done appreciably.
- While it is important to see this data, it is largely diagnostic and post-mortem. Industry should be paying to develop these standards.

Question 2: Approach to performing the work

This project was rated **3.0** for its approach.

- The project starts with the real-world operation, performance decay, and teardown/postmortem characterization of materials that have been used in bus service. The project conducts ASTs in the laboratory for direct

comparison, sets up a laboratory breadboard system for accelerated life test comparisons, and develops models to reflect the performance and decay data obtained in the real-world and laboratory settings.

- The approach is good. It would be nice to have a third party such as the National Renewable Energy Laboratory Technology Validation Group analyze the data from the real-world operation of UTC Power's stack compared to other companies' stacks and the AST data. This third party could see if similar correlation can be made between the AST data and other companies' stack data. This suggestion is complicated by the fact that different materials and operations have been used in other companies' stacks, but even a general correlation would be a good general validation of the ASTs. It was unclear whether the ASTs were carried out on single cells or stacks. Tests on stacks would be more relevant. It is also unclear whether the modeling approach will be able to predict membrane durability. This reviewer wants to know if modeling will be used to predict the life of other components as well.
- A degradation model that incorporates ASTs to predict stack lifetime, or a more in-depth assessment of how ASTs can be used to predict stack lifetime, would be helpful to translate the work of others to this body of work. In other words, data from ASTs for the top failure modes used to predict rather than correlate to lifetime in hours or degradation rate would be useful.
- Sharing materials and design information would make the reported data more meaningful. The materials and technology should be promising enough to meet an end-game goal, such as low Pt loading, etc. The operating conditions of the ASTs and the real-world usage should be clarified and considered to analyze acceleration factors. The relative humidity cycle should be included in the ASTs. Adequate statistical data collection should be planned, particularly for the latest data collection activity (2010 bus data).
- It feels like an opportunity was lost, as the investigators had three stacks with three different failure mechanisms. It would have been great to look at the different conditions and material builds, and show how the ASTs could have predicted these failures. The investigators have tried some of the standard ASTs, but have not suggested changes or new tests when the correlation was weak or non-existent.
- These diagnostics are nice, but do little to advance the field. Perhaps DOE should shift focus away from this type of project and more intensely emphasize solutions to problems rather than diagnoses.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.8** for its accomplishments and progress.

- Good and relevant progress has been made.
- The project has completed several aspects of the planned work. While detailed results were given in various slides, a useful summary of the results from real-world and accelerated testing was given in slide 18. Several causes of performance decay were summarized: Pt dissolution and sintering, carbon support corrosion, and the mechanical and chemical degradation of the electrolyte membrane.
- The data presented are informative. Information about materials, design, and operating condition should be shared to make the reported data more meaningful. In particular, relative humidity data are missing. More detailed failure mode analysis for real-world data, such as membrane mechanical damage (slide 13), should be investigated whether for material degradation, for understanding how to correlate the data with mechanical failure during AST, or for other causes. The membrane hydration state should also be investigated.
- Other than the loss of catalyst surface area, very poor correlations have been made with the other ex-situ testing, and there is little reason to believe that more will be made in the remaining time. The investigators see degradation at the stack inlet, but do not have much insight into how this varies from the rest of the stack. It is unlikely that they will find an ex-situ test to help them to address this failure mode when they do not understand why this degradation is different.
- The project should define the hypothesis and mechanism for variation in failure across the cell (inlet to outlet) in terms of temperature, relative humidity, compression and mechanical stress, and potential.
- While this project ostensibly addresses durability goals and includes plenty of well executed experiments, it is hard to understand how this project substantially increases our understanding.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.8** for its collaboration and coordination.

- The project team includes a major fuel cell developer, an industrial research organization, and two national laboratories. The research relates to a specific fuel cell technology, however.
- The established relationships are good. A further improvement would be to ask other industry experts (e.g., Ford and General Motors) to peer review the ASTs and proposed degradation mechanisms.
- It is very hard to take this experience with a bus drive cycle at low temperatures with porous plates and help the fuel cell community. The work being done by the collaborators on post-mortem is a project strength.
- The collaborations are adequate. However, this project is one more example of the same group of people doing the same sets of tests with a new rotating principal investigator. This project is highly duplicative in effort with other projects. This project exemplifies a growing negative trend in the DOE Program toward more routine, duplicative work and away from true innovation.

Question 5: Proposed future work

This project was rated **2.8** for its proposed future work.

- The proposed future work seems appropriate.
- The project has a good future plan. It seems to need more analysis items that do not show stronger correlation between real-world usage and ASTs before new or modified ASTs are developed.
- Tasks 1–3 have essentially been completed. Tasks four (develop and validate additional AST protocols) and five (further develop membrane hydration strain model) will be undertaken in the remaining seven months of the project.
- The investigators have very little time left, but are proposing new AST methods. Unfortunately, these ASTs are not based on recreating failures that UTC Power has seen.
- This reviewer agrees with the recommendations except for the GDL AST, unless new GDL concepts are being prototyped with drastically different materials than the industry standards. In the reviewer's opinion, the GDL will likely be the last component of the membrane electrode assembly to fail, and if it does fail, it will more likely be due to a fuel starvation or unit cell issue than a weak GDL. It would be valuable to compare the degradation propagation of buses to other fuel cell applications (e.g., stationary, auto, etc.).
- The work should be outsourced to a standards organization for a fraction of the money.

Project strengths:

- Having real-world data and comparing it with ASTs is a strength. The partners for the AST testing and development and post-mortem analyses are also good.
- The project is trying to take systematic approach.
- Strengths of this project include having access to, and making good use of, real-world performance, performance decay, and failure mode data from bus fuel cell systems. Another strength is the project's direct comparison of real-world and laboratory test data obtained under comparable conditions.
- Having strong partners for catalyst degradation and post-mortem characterization is an area of strength for this project.
- This project features great use of field data.
- This project features solid work and high quality methodologies.

Project weaknesses:

- The results may mostly be relevant and beneficial to UTC Power because the project evaluates UTC Power materials, stack design, and operating conditions.
- The materials and design are unique and outcomes from this project would not be directly applicable for other materials or fuel cell designs.

- The results of this project are specific to the fuel cell technology of UTC Power. This reviewer wants to know what the implications are for fuel cell degradation for other polymer electrolyte fuel cell technologies, and how these results can be extended in a more general manner.
- One area of weakness is the project's poor relevance to most fuel cell systems being pursued. The attrition of principal investigators has hurt this program significantly.
- GDL ASTs have low value unless new GDLs are being developed.
- This project is routine and barely advances the understanding of state-of-the-art. It certainly does not advance the technology in a meaningful way. This work should be done by industry without DOE support.

Recommendations for additions/deletions to project scope:

- Investigators should bring in a third party to analyze the data. Investigators should also consider testing other materials.
- This reviewer recommends using the latest materials and technologies that can potentially meet (or nearly meet) the end-game targets. Materials, design, and operating condition information should be shared as much as possible to make reported data meaningful.
- The investigators should put less emphasis on GDL corrosion AST development—UTC Power's latest test results show no GDL corrosion. Unless there is data that indicates that the GDL corrodes before the catalyst, the likelihood of GDL corrosion being the life-limiting failure is low. The project team should engage more experts (e.g., industry and national laboratories) to peer review the proposed degradation mechanisms. To ensure UTC Power proprietary information is kept confidential, investigators should consider working with other fuel cell developers that are not in direct competition, such as in the automotive industry. Sharing the half cell potentials with a small group may offer further insight into the mechanisms.
- DOE should cut this project.

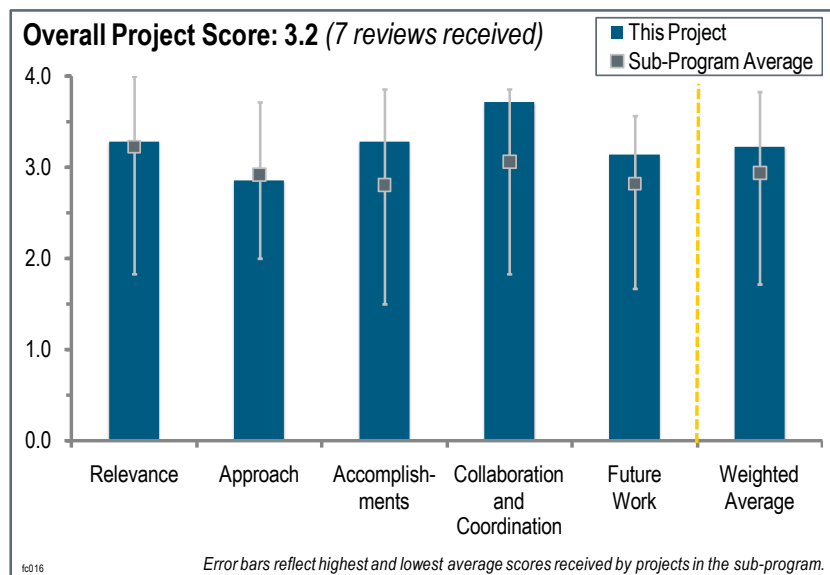
Project # FC-016: Accelerated Testing Validation

Rangachary Mukundan; Los Alamos National Laboratory

Brief Summary of Project:

The accelerated stress test (AST) allows faster evaluation of new materials and provides a standardized test to benchmark existing materials. The objectives of this project are to: (1) correlate the component lifetimes measured in an AST to real-world behavior of that component; (2) validate existing ASTs for catalyst layers and membranes; and (3) develop new ASTs for gas diffusion layers, bipolar plates, and interfaces.

Question 1: Relevance to overall U.S. Department of Energy objectives



This project was rated **3.3** for its relevance to U.S. Department of Energy (DOE) objectives.

- Developing, understanding, and correlating ASTs is very important for the industry.
- This project supports fuel cell durability as one of the critical DOE Hydrogen and Fuel Cells Program objectives. The project objectives—which include correlation and validation of existing and development of new ASTs for the membrane electrode assembly (MEA) and fuel cell components—will enable fuel cells to reach durability targets for automotive and stationary applications.
- The project has a high relevance to DOE objectives. The development of ASTs is a necessary factor for predicting the life and performance of fuel cell components and reduces the requirements for many thousands of hours of extended testing. There is a strong need to correlate the data to real-world data, and this project attacks that issue.
- The objectives are very relevant to Program goals.
- A great deal of this project is reliant on the partnership with Ballard and its fleet of buses. Failure in bus stacks is of fairly limited interest to the overall community because they have unique operating conditions and drive cycles.
- The principal investigators (PIs) gave no insight into Ballard's build of materials, which greatly limits what can be gained by other investigators.
- Studies of catalyst degradation are important to meeting DOE's fuel cell goals for all applications. Buses are an important early application for fuel cells because they are inherently a fleet operation refueled from a limited number of points; therefore, it is relatively easy to surmount the fueling infrastructure problems for H₂ fuel cells in the bus application.
- Buses are a heavy-duty application in which the power plant runs very near its rated power most of the time that it is not idling. Buses do not take advantage of fuel cells as much as automobiles, in which the power plant runs at the lower power levels at which fuel cells have maximum fuel efficiency advantages.

Question 2: Approach to performing the work

This project was rated **2.9** for its approach.

- The overall approach is very strong and comprises trying to collect and correlate laboratory AST results with ones from the industry.

- Completed milestones are not clearly stated and do not even represent what is in the presentation, despite what the PI says. This reviewer wants to know if the PI will be able to correlate the ASTs to the various material sets the PI has listed.
- The project has a well-thought-out plan and approach.
- The technical approach used in this project is adequate and well defined.
- The use of materials by Gore and Ion Power is relevant to all investigators and is a strength of the project. There is far too much overlap with other investigators; for example, another group is doing voltage cycling and open-circuit voltage hold.
- It is not clear if the dataset from the actual buses is of high enough quality to warrant all of this laboratory work to try to match it with accelerated tests. Investigators did not run any controlled polarization curves, so the only available data are the highly dynamic results that had to be averaged to provide any form of comparison with the accelerated laboratory tests. Averaging does not give a clear picture, as most of the damage would likely come from the extreme points.
- The voltage distributions shown for the buses show no values high enough to approach air-air open circuit. It seems doubtful that air-air open circuit could be entirely avoided, and the lack of quantification of this potentially damaging condition draws into question the value of the vehicular dataset (though during the talk a comment was made that the highest degradation rate correlated with the highest number of air-air starts).
- Los Alamos National Laboratory (LANL) ran such high Pt loadings in the bus stacks that the loadings provide a poor comparison with the low-loaded stacks that will be needed for mass-production vehicles.
- The stack with the more durable materials was not run in vehicles, but rather on a test bench. More complete data were available from this stack, including periodic controlled polarization curves, but this is not real vehicular data.
- Bus vehicular data should not be taken as a good model of automotive applications, so tuning laboratory accelerated tests to match bus data does not give much confidence that the accelerated tests will be meaningful for automotive applications. The presentation did not clearly identify plans for all of the MEAs being prepared by Ion Power. It is not clear why the project team was pursuing all of the different loadings of Pt on carbon.
- The approach is consistent with the development of testing procedures and correlation of the testing procedures with commercial fuel cell components. The approach incorporates the fleet data from Ballard. Correlation of the commercial demonstration and laboratory data is an important contribution to the development of fuel cell components. Not having the correlation of the ASTs and laboratory data with automotive fleet data available is a weakness in the approach.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- LANL has made very good progress and generated a significant amount of high quality results. There is good agreement between the performance loss results presented and those obtained in other projects.
- The results are on track with the overall project status and goals.
- The project demonstrates good correlation of the AST data with the field data. The data is a strong indication of the successful implementation of the AST approach.
- LANL has made a lot of progress in correlating the work in the laboratory and the field, especially with Ballard's material.
- This reviewer is shocked by the very high Pt loadings of Ballard's MEAs (1 mg/cm^2) and wonders how applicable this will be to the automotive industry, which is rapidly approaching a tenth of this amount.
- The results from the Ballard MEA are poor; the electrochemically active surface area and durability are both quite low. This is not the fault of the LANL PI, but the applicability of the results to the industry is unclear.
- The performance of the baseline materials and Ballard stacks has been completed and will serve as a baseline to future tests.
- Some numerical acceleration factors were mentioned in the presentation, but the level of confidence in these factors cannot be high, even for the bus application. It is not clear if the vehicular data, from a limited number of buses, has provided significant insight toward evaluating the validity of the ASTs, and the laboratory vehicle-like profiles have not been tuned to emulate bus operation. Apparently investigators did not observe any membrane thinning in the buses, so no comparisons can be drawn with the chemical degradation AST.

Investigators have generated and summarized some interesting results from the ASTs, but it is not clear if they are making progress in generating real correlations to vehicular behavior.

- The use of dynamic mechanical analysis tests to run the system to failure after vehicular or AST runs to give a remaining life number is somewhat ingenious, but is of dubious validity.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.7** for its collaboration and coordination.

- This project features an excellent group and the team effort is evident.
- The project activities are well-coordinated among the team members.
- LANL and Ballard seem to be working together rather well, though this project would have been really valuable if it was initiated before the buses were run so that proper data gathering could have been planned.
- Incorporating materials from various companies and collaborators is vital for this project to succeed, and it appears this is happening quite well. Access to real-world data from Ballard buses is open, which is great and a real asset to the project.
- The comparison with a different generation of Gore materials will only be helpful if it is clear what changes are made. This reviewer offers the same criticism for Ballard. If LANL reports differences between two materials, they must also describe what the materials did and did not do to highlight the difference between these materials, otherwise the comparison is of no benefit to other investigators.
- The team is obviously well qualified to take on the work.
- The explanation of the coordination of the LANL efforts was unclear and little confusing. The interaction with Ballard, Lawrence Berkeley National Laboratory, and Ion Power should all be beneficial.

Question 5: Proposed future work

This project was rated **3.1** for its proposed future work.

- The future work is well-planned.
- The proposed future work is adequate and laid in accordance with the project objectives.
- The proposed future work is consistent with the requirements of the Program.
- Testing of other cutting-edge materials is planned, which is good. This reviewer recommends clearer future milestones and plans from the PI.
- LANL should concentrate on public materials so there is a benefit to other investigators.
- Efforts should be taken to avoid overlap with other investigators.
- Not enough vehicles were tested.
- The vehicles were not the correct type to be correlated with ASTs designed for automotive applications. LANL does not seem to have plans to get around this gap, and it therefore appears to be just a laboratory project from this point forward.

Project strengths:

- Strengths of this project include the world-class team and collaborators. The project team has an excellent opportunity to correlate laboratory testing with real-world buses.
- This project's many accomplishments and great collaboration are areas of strength.
- Examining Ballard materials at different stages in their lifetimes is valuable.
- This project features an excellent team that has generated results.
- One strength of this project is the investigators' attempt to correlate vehicular data with laboratory tests, both accelerated and nominally non-accelerated. Such comparisons are needed.
- The project's strength is its strong team.

Project weaknesses:

- The AST effort should be applied to automobile applications.
- The low technology of the Ballard MEA, which is being used as a major baseline material, could weaken the applicability to other materials.
- The Ballard MEA's total loading is approximately 1 mg Pt/cm². This could be a totally different regime of catalyst degradation than typical current commercial MEAs (~0.4 mg Pt/cm²) and the DOE target (< 0.2 mg Pt/cm²). Also, any system-level mitigation of the bus stack should be considered in evaluating the relevance of real-world degradation data to light-duty automotive systems.
- There is far too much overlap with the work being done by the Borup group (at LANL). Many of the same team members (e.g., Ballard, Ion Power, and numerous national laboratories) are involved in both projects. If there is going to be overlap on the same topic, the teams should be as different as possible.
- Not much can be learned from this project because Ballard did not disclose the build of materials. Ballard's motivation in not doing so is certainly understandable, but this work will benefit only Ballard and one should ask why DOE is funding it.
- The automotive real-world drive data is not included in the project even though ASTs for automotive fuel cells are generated based on the automotive drive cycles.
- The vehicular data are from buses, but the non-accelerated laboratory vehicular cycle test instead emulates light-duty vehicles, and ASTs were also chosen to emulate light-duty vehicles. The bus dataset appears to lack important details needed for proper comparisons with laboratory ASTs.
- The PIs are looking at too many different material sets in too many different ways to draw coherent correlations.

Recommendations for additions/deletions to project scope:

- This reviewer suggests accelerating the plan to test cutting edge materials from Gore and Ion Power when possible.
- Either this group or the Borup group should drop bipolar plate studies.
- The PIs may have to give up their original goal of correlating vehicle and laboratory data, and concentrate instead on laboratory evaluations of different protocols.
- The project team should include Daimler, Toyota, General Motors, etc. in the activities.

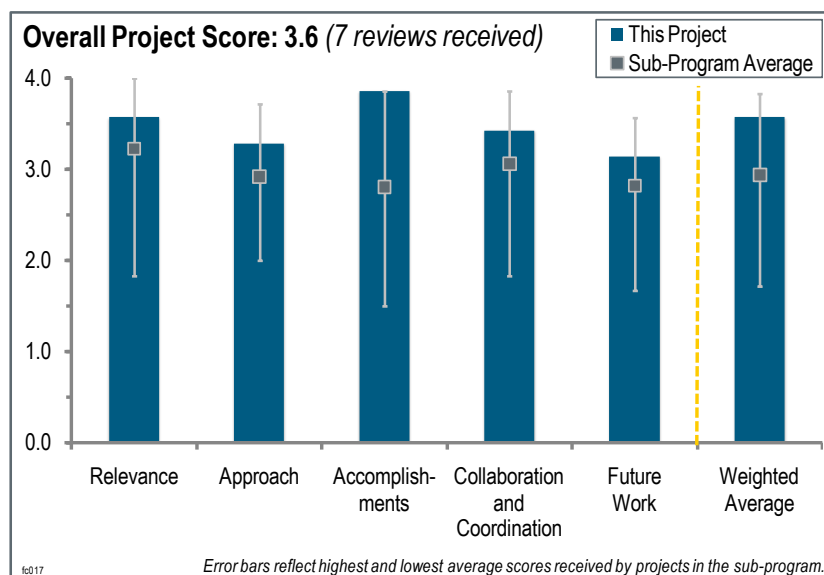
Project # FC-017: Fuel Cells Systems Analysis

Rajesh Ahluwalia; Argonne National Laboratory

Brief Summary of Project:

The overall objective of this project is to develop a validated system model and use it to assess design-point, part-load, and dynamic performance of automotive and stationary fuel cell systems.

Objectives are to: (1) support the U.S. Department of Energy (DOE) in setting technical targets and directing component development; (2) establish metrics for gauging progress of research and development projects; and (3) provide data and specifications to DOE projects on high-volume manufacturing cost estimation.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.6** for its relevance to DOE objectives.

- The modeling tool developed in the project is critical to benchmarking progress achieved in the DOE Hydrogen and Fuel Cells Program and providing input to cost analyses.
- It is important to have a model to correlate fuel cell system operating information, as technology and knowledge advances and systems are evaluated. Modeling the dynamic operations of the system is important to help understand the effects of variables in the system.
- This project's models are especially important to cost models and to estimating system progress.
- Most automotive original equipment manufacturers (OEMs) have their own systems analysis effort in place, which means that this project does not directly benefit them. However, the project is certainly a benefit to other DOE projects, particularly those involving cost analysis or those attempting to define targets related to balance-of-plant (BOP) components or system operation. The relevance of the project has improved in recent years as both high- and low-power operation have been examined in the course of component selections, and as sensitivities to operating conditions have also been examined.
- The need for a system model to define both automotive and stationary fuel cell systems is high. This project's model supports the Program.
- There is a concern that the model does not cover the industry designs; for example, the model does not include porous plates used by some industry researchers. Adzic reported the use of core-shell catalysts with more than 200,000 cycles of tests by a foreign automobile company, but these catalysts were not mentioned in this report. Companies such as Ballard deliver stacks for many applications and have very high catalyst loadings (e.g., 0.4 mg Pt/cm²) on the cathode. This reviewer did not see the Ballard catalyst discussed. Companies such as UTC Power and ClearEdge Power are selling stationary power systems in the United States that were manufactured in the United States, but there is no mention of this technology. DOE is missing an opportunity by not including the emerging business opportunities in the system study.
- This project develops and maintains technical excellence in the detailed understanding and analysis of fuel cell systems. This year the principal investigator (PI) reported on a number of projects, but the sum is greater than the parts. The overriding relevance to the Program is the expertise and competence that this project brings to the table.

- It is helpful to continue looking at system components and challenge the industry status quo. This reviewer questions this level of pressure and the addition of the turbo-compressor, but will keep an eye on these developments.

Question 2: Approach to performing the work

This project was rated **3.3** for its approach.

- This project features a sound approach based on model validation through experimental tests and interaction with a variety of industrial partners.
- This project develops and disseminates system design and analysis tools and validates them with laboratory data and external collaborations. The approach benefits from a solid engineering understanding of fuel cell technology.
- The models are good and the tools are correct. This reviewer wants to know how design choices were made.
- This project features a very thorough analysis.
- The approach has continued along the improved direction that was established in 2010. This direction assumes more realistic system components and explores system operation variations with changes in operating conditions and high power efficiencies.
- Limitations at low power, such as surge line limitations on the compressor and ejector/pump limitations, have been explored.
- The PI has begun investigating various water transport membranes with different water transport rates to determine which serve the system best throughout the range of operation.
- The project team needs to validate the model against OEMs, and compare the projected performance and performance limits with them to validate model capabilities and reliability for dependable DOE guidance and direction.
- The approach is focused only on polymer electrolyte membrane (PEM) vehicle applications and is therefore limited, but the objectives identify stationary fuel cells as part of the project. The strong influence of the U.S. DRIVE Technical Teams appears to be guiding this project. To develop a sense of independence, it might be valuable for DOE to sponsor a system modeling team separate from this team.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.9** for its accomplishments and progress.

- The analyses and collaborations are outstanding. The investigators focus almost entirely on vehicle applications.
- Developing the dynamic modeling capability is an excellent addition.
- The PI reported on a variety of technical accomplishments (slides 6–18) that increased basic knowledge about fuel cell technology.
- This project has achieved numerous accomplishments in parameter variation effects. This reviewer suggests that the purge is due to crossover, not impurity.
- Manufacturers may be able to drop pre-cooling equipment; this issue is not clear yet.
- The investigators have performed a thorough analysis of system component limitations, finding regimes for ejectors versus pumps (or both). They have also incorporated the effects of hill climb on thermal management.
- Using humidifier data from the Honeywell device (with Gore membranes) is the right direction, although examining alternatives is also helpful. It would be interesting if the project were to provide information as to whether the RH (relative humidity) is sufficient throughout the entire operation.
- Argonne National Laboratory (ANL) has exhaustively looked at the effects of lower efficiency regarding the impact on the thermal system.
- The presentation left some question as to whether the 3M data were collected using carbon or metal plates, and whether some adjustment in performance needs to be made to assume the use of metal plates.
- ANL has made very good progress, and the approach based on a turbo-compressor opens intriguing alternatives to the standard system design. This reviewer recommends further exploration of the effect of high pressure on anode-to-cathode pressure drop and the potential impact on fuel economy and control strategies. This reviewer also wants to know if the H₂ tank will deplete faster if anode pressure needs to closely match higher cathode pressure, and how a low pressure drop will be maintained during transients.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.4** for its collaboration and coordination.

- This project is highly collaborative. It interacts with leading fuel cell component providers (e.g., 3M, Gore, and Honeywell), the open standards community (e.g., the International Organization for Standardization Technical Committee 192), other DOE laboratories (e.g., Los Alamos National Laboratory), the projects studying manufacturing costs (e.g., Directed Technologies, Inc. [DTI] and TIAX, LLC), and many others.
- This project features a great list of collaborators with value flowing into the project and out to the partners.
- This project has nice collaboration partners.
- Because this project relies upon the inputs of other parties, collaboration has always played an important role, including the inputs from balance of plant (BOP) component suppliers, particularly Honeywell. 3M has been providing cell data, along with performance sensitivities for low-Pt-loaded nanostructured thin film (NSTF) membrane electrode assemblies (MEAs). The project has delivered outputs to cost analysis projects. Despite the continued use of collaborations, this project began turning the corner a few years ago when it embraced inputs from automotive OEMs, particularly with respect to how dynamics and low power operation can affect BOP component selection. The project has improved significantly since then.
- The network of partners is well developed. For air management subsystem and stack performances, this reviewer recommends increasing the number of partners providing feedback to the project.
- The collaboration list, especially with OEMs, is limited. The project needs outside validation of the model. One on one interactions, rather than Tech Team group responses, can provide valuable inputs for model validation by OEMs.
- The project team does not include a stationary fuel cell manufacturer. Nuvera's website addresses material handling equipment and other vehicle applications. This project needs to engage a stationary fuel cell manufacturer.

Question 5: Proposed future work

This project was rated **3.1** for its proposed future work.

- The plan to extend the model capabilities to simulate non-automotive applications will greatly enhance the value of the tool for the fuel cell community.
- The first bullet on the future work slide is perhaps the most important. The continuity of this project is important for DOE development efforts at system, component, and phenomenological levels in many ways—including those that may not be obvious today. Sharp and dedicated researchers will identify the most significant needs and opportunities as the technology develops, and adapt their project accordingly.
- The proposed future work is appropriate for the tasks assigned in the statement of work.
- Based on the modeling capability and information received for the inputs, the proposed future work is to generate and publish projections regarding the effects of various operating conditions to define the direction and value of improvements in any of the several key operating parameters.
- Six of the seven future work activities are on vehicle systems, which is good. The work on performance of stationary systems involves PEM fuel cells, but it is unclear if this means backup power or distributed power. For distributed power and most stationary applications, PEM fuel cells operating on natural gas are too inefficient. Other fuel cell systems such as molten carbonate fuel cell, phosphoric acid fuel cell, and emerging solid oxide fuel cell technology are more consistent with stationary applications other than backup power.
- The project must involve continued collaboration with 3M to improve the stack model. It would be interesting to see how well performance versus anode hydrogen concentration is represented in the model, especially considering that the setting for the purge has been arbitrarily set at 10% inerts (N₂ and He). The continued validation of air, water, thermal, and fuel subsystems is also important. The humidifier is particularly important.
- Durability is mentioned in the future work. While the project is at a point where this can begin to be explored, it would be interesting to see what would be defined as a stressor in the drive cycle. In other words, the reviewer wants to know what kind and how much of a voltage cycle would be expected to cause degradation. The reviewer also wants to know how much of a RH cycle or low RH event would be necessary to cause degradation of a component.

Project strengths:

- Strengths of this project include its relevance and approach.
- This project's engineering excellence is a strength.
- This project has a high value to DOE in evaluating systems, and helps cost analysis projects.
- Model development has progressed nicely and includes dynamic operations.
- One strength is the investigators' willingness to receive inputs from component suppliers and OEMs. Recently, the PI began taking OEM suggestions about low-power operation more seriously, and it has made a considerable difference. Another strength is the investigators' willingness to explore operational variations. ANL has made significant strides by recognizing that performance gains can be revealed by looking at changes in operating conditions (e.g., RH, stoichiometry, temperature, pressure, and hydrogen %) and how they affect both stack performance and BOP efficiency.
- The analysis methodology has matured and the system model is more consistent with real-world PEM fuel cells. The correlation with automotive applications is strong; however, it is not clear if the system model is consistent with automotive applications.

Project weaknesses:

- Relying on only one partner for air management (Honeywell) and two for stack (3M and Nuvera) could influence the objectivity of the assumptions. This reviewer recommends seeking further collaborations in these areas, especially with an automotive OEM.
- It is not clear that the designs are optimal, or how the PIs determined them.
- The project team should corroborate the model findings and projections with OEMs for validation to help guide DOE direction.
- The project is too focused on low-loaded NSTF. Operational sensitivities for NSTF may be different than a dispersed Pt alloy catalyst of similar loading. The project is also occasionally over-reliant on 3M. While it is worthwhile to examine freeze dynamics in the context of a microporous layer (MPL)-less anode, an MPL-less anode may pose durability concerns for the membrane. The PI should still be aware of operation with the presence of anode MPLs and request stack performance data from MEA suppliers in that context. The project's focus on low loading is also an area of weakness. Given the cost benefits that may derive from a lower active area with slightly higher platinum group metal (PGM) loading, it may be worthwhile for this project to work with DTI to explore active area and PGM loading tradeoffs.
- It is unclear why the project only addresses NSTF catalysts. Other catalysts are much further along the commercial path. Ballard is the recognized leader in stack development and sales, but Ballard does not use NSTF catalysts. The project does not address stationary fuel cell systems and appears not to have categorized the stationary applications with fuel cell type, which is probably the simplest task to undertake.

Recommendations for additions/deletions to project scope:

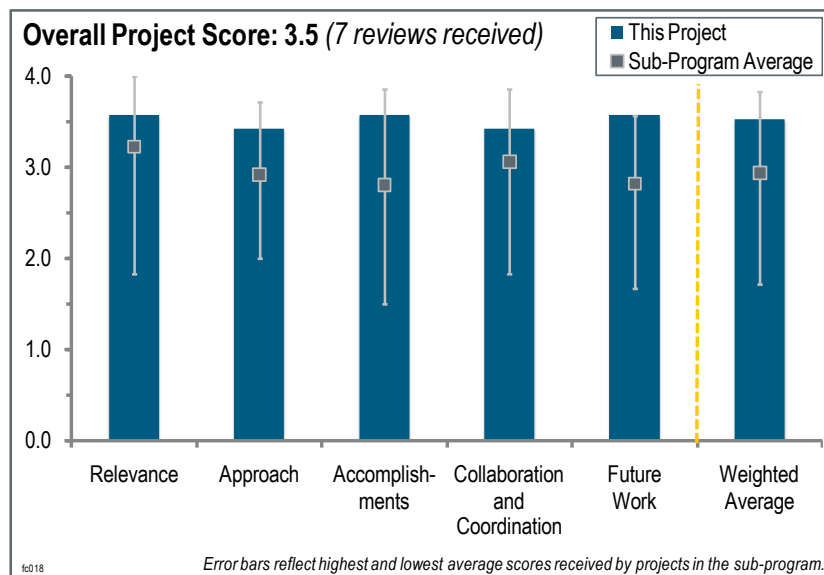
- The project team should produce better documentation on how the design choices were made and what the implications of alternate designs might be.
- The project team should consider more model validation work at controlled conditions. The data points are a little confusing.
- The project team should explain whether stoichiometric sensitivities are based on small single cell or stack module operation. With a module, sensitivity can be greater and fairly design-dependent. The PI should also examine freeze dynamics with the assumed presence of an anode MPL, and outline expected durability stressors and the extent of degradation expected from different levels of stress. Finally, the PI should provide information on stack model sensitivity to anode H₂ concentration, as well as examine active area and loading sensitivity in conjunction with the cost analysis project.
- The project is not really addressing stationary fuel cells, so this should be deleted from the project scope.

Project # FC-018: Manufacturing Cost Analysis of Fuel Cell Systems

Brian James; Directed Technologies, Inc.

Brief Summary of Project:

The overall objective of this project is to help the U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program develop fuel cell systems by assessing the cost status, identifying key cost drivers, and exploring pathways to cost reduction of automotive and stationary fuel cell systems. The specific objectives of this project are to: (1) identify the lowest cost system design and manufacturing methods for an 80 kWe (kilowatt electrical), direct-hydrogen, automotive proton exchange membrane fuel cell system based on current technology and 2015 projected technology; (2) determine costs for these technology level systems at varying production rates from 1,000 to 500,000 vehicles per year; and (3) analyze, quantify, and document the impact of system performance on cost, using cost results to guide future component development.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.6** for its relevance to DOE objectives.

- This project is a very important activity for creating a basis for future decisions.
- This project presents a very good overview of fuel cell cost estimates. It is good to see a breakdown of costs, etc.
- This project seeks to realistically assess the mass production cost of prospective automotive and stationary fuel cell systems. This project's highly detailed research is essential for informing overall policy on the realistic prospects that fuel cell systems will be an affordable alternative to other power plants (e.g., batteries and internal combustion engines). The detailed study of individual components and subsystems focuses planning and elucidates what additional research might be done to further reduce costs.
- The cost of fuel cell systems has been traditionally higher than what is tolerable in a commercial market, so work focused on analyzing and addressing the major cost drivers of systems and infrastructure is required prior to wide market adoption of the technology. To that end, this project aligns with the needs of both DOE and industry. Specifically for this presentation and work, this reviewer believes that it is important to develop a context, and thus a cost structure, for high-volume production of stack membrane electrode assemblies (MEAs). The reviewer thinks it is equally important to address other balance of plant components, specifically the air compressor, H₂ storage system, and humidifier. This recent work seems to have focused primarily on the MEA.
- The project evaluates the cost of automotive fuel cell systems and is critical for measuring the progress toward achieving a cost effective fuel cell for automotive applications. Task 4.1.3 reports on optimizing the operating pressure versus catalyst cost balance, which appears somewhat strange because pressure has little to do with catalyst utilization.
- Quality control will play a large role in fuel cell cost, so it seems that the impact of parameters such as waste, recycling, and numbers of rejects, as well as a range of sensitivity to quality control (page 12), should be included. The stated objective "Identification of lowest cost system design" seems to be overstated, as the system design used is more correctly based on the modeling information provided by Argonne National

Laboratory (ANL). Additionally, this system includes as many features as possible for such as study without proprietary information being provided from an actual commercial system.

Question 2: Approach to performing the work

This project was rated **3.4** for its approach.

- This project features good use of ANL information and experience in system modeling and equipment providers. It would be useful to provide some information as to the degree of uncertainty in Honeywell quality control (page eight). It is not clear how investigators can evaluate system cost effects by changing oxygen stoichiometry when it was held constant at 1.5 in the chart on slide 10 of the presentation. The reviewer wants to know if the \$2/kW total cost increase with listed parametric relaxation falls within the analysis sensitivity (slide 11).
- Directed Technologies, Inc.'s (DTI's) approach to the project was not clearly stated in its presentation; however, one can determine the approach from the information that was presented. Primarily by using Design for Manufacturing and Assembly® (DFMA) and engineering judgments, investigators are making projections for material and manufacturing costs of systems at high volumes (up to 500,000 units per year, based on DOE direction). This is a sound approach, but this reviewer would like to see the results of the work done on a more step-by-step basis in order to make the impact of individual contributors to the unit costs (specifically increased pressure compared to decreased membrane area) more easily recognizable. The current approach washes out the effect of several significant (and potentially conflicting) individual factors.
- The “new” high-pressure stack comes as a surprise—the benefit stated is not explained transparently.
- The principal investigators are laboriously developing a reference design in close consultation with the national laboratories (e.g., ANL) as well as leading firms that are developing fuel cells and their components. A major advance this year was the inclusion of the new ANL polarization model for the 3M membrane material. The investigators are using well established methodology (e.g., DFMA) to estimate materials and manufacturing costs.
- The presentation did not discuss the approach in detail. The statements that “DTI practices a blend of ‘Textbook’ DFMA, industry standards & practices etc.” and “Analysis includes effects of bulk purchasing, manufacturing methods, tooling amortization” did not provide sufficient information regarding how investigators used the analysis method.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.6** for its accomplishments and progress.

- This project features good progress and an established methodology for conducting the last phase of the work.
- The main accomplishment for this project is an accurate assessment of the state of the technology, regardless of the DOE goals, and the investigators seem to be doing a credible job of it. Other major accomplishments include the continuing reference values on cost trends (slide 20), re-optimizing the reference design based on ANL's new models (slides 6–9), and a detailed analysis on the cost of implementing a manufacturing quality control system (slide 12).
- The presentation featured a good report on progress. The project team has completed a comprehensive cost model for vehicular systems as well as a risk assessment and sensitivity analysis, which addresses some of the key issues in the ability to actually achieve the system cost as modeled (specifically the compressor and humidifier). The team has also addressed concerns from previous reviews. The high-volume cost estimates are on track with DOE objectives, but this reviewer would like to compare the costs as reported with the expected costs when the cost of capital and full corporate burdens (profit) are added in. The largest single step function of cost reduction in the stack is from 1,000 units to 30,000 units, and is defined as a cost down due to going from low- to high-volume manufacturing processes. With such a large number of plates and MEAs in the stack unit (approximately 369), this reviewer suggests that the fabrication of 1,000 systems already represents a relatively high-volume rate of the individual components, especially considering the current capacity of the industry. The reviewer wants to know what the basis is for this step function change in costs.
- The increase in operating pressure affects the minimum storage pressure, and therefore the quantity of usable hydrogen.

- The optimization of operating pressure versus catalyst cost result is impressive. This reviewer wants to know if this has been confirmed through experimentation by a research laboratory. The lifecycle cost benefit was surprisingly flat. The reviewer requests additional explanation regarding the lifecycle data. The capital equipment and research and development needs are very informative. The compressor/expander motor information is not consistent with some industry inputs from an independent review of DTT's previous work. The project team should address this. The membrane industry input should be used to crosscheck the results. It is unclear if the membrane facility cost was built on production of 500,000 vehicles. If it was not, the reviewer wants an explanation of why not. The cost trends at low rate production need to be harmonized with present cost and price information.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.4** for its collaboration and coordination.

- The project features excellent collaboration with government and industry partners. The level of input is significant, and represents a substantial increase from 2010.
- This project is highly collaborative. The contractor (DTI) seeks information broadly from a variety of sources, including the national laboratories (e.g., ANL and the National Renewable Energy Laboratory) and industry (e.g., Ballard, Nuvera, Ford, and Honeywell).
- The project team needs to expand original equipment manufacturer (OEM) involvement, considering those who will be supplying and providing vehicles in the United States. Also, the project team should include some of the solid oxide fuel cell developers (e.g., Bloom and FCE) as collaborators.
- The reviewer wants to know why investigators did not choose General Motors, Toyota, Hyundai, Daimler, or Honda—companies that are the leaders in automotive fuel cell development—as collaborators. The non-U.S. companies all have production facilities in the United States.

Question 5: Proposed future work

This project was rated **3.6** for its proposed future work.

- The future (ongoing) work on three different designs of stationary fuel cells should be a valuable addition to the work already accomplished on automotive systems. Perhaps the most important future work of this project would be the continuity of the project itself. Fuel cell cost is one of the greatest impediments to the widespread adoption of this technology. It is vital to the Program to have reliable, fact-based assessments to demonstrate the maturation of the technology.
- Stationary analysis is the next phase of this project. This reviewer is looking forward to an equally substantial report on the costs of stationary and home fuel cell generating units. The reviewer wants to know if a review of current results could be done in the context of determining the impact of specific cost factors. Perhaps investigators could review costs with the inclusion of capitalization costs and corporate profit burden.
- The system design should simultaneously evaluate range with a given H₂ volume in a tank and at power, i.e., from a customer's perspective, at power and range simultaneously—this will presumably favor low-pressure solutions.
- This project features good inclusion of stationary fuel cell systems. The project team needs to correlate the findings from task 4.1 with several OEMs before issuing the final report.
- This reviewer wants to know what fuel cell model will be used for high-temperature polymer electrolyte membrane (PEM) and solid oxide fuel cell systems, and what the stationary low-temperature PEM system is. The reviewer also wants to know if stationary analyses will be on H₂ systems or hydrocarbon systems.

Project strengths:

- One strength of this project is the comprehensive review of the costs. An intelligent analysis of the system being costed out showed good judgment by team leadership. Good cross-section of industry collaboration was included.
- The project successfully integrates a wealth of detail, developed through intensive investigation and interaction, into succinct reference designs with highly credible cost estimates. These are essential

benchmarks for the current state of the Program. Further, the detailed analysis highlights what areas continue to need attention to further reduce costs.

- The cost analysis knowledge combined with technical insight is an area of strength of this project.
- The project integrates modeling and experience from ANL in system design, and from equipment suppliers for tying costs into system operation information. The project features a good outline of manufacturers' considerations about quality control and other equipment for mass production. The project also follows a natural progression of using the cost basis from vehicle to stationary fuel cell systems.
- Strengths of this project include its well developed analytical experience and consistency in data development.

Project weaknesses:

- While this is not really a weakness, this reviewer desires to see more definitive effects of individual cost factors—specifically, how the suggested compressor target cost could be achieved. The reviewer suspects that the cost will be much higher than anticipated due to the context created by the size and speed required for this unit.
- The project relies on a number of assumptions, as documented, that could potentially cause a variance between the models and real-world product behavior. However, no particular deviation is expected.
- The limitation to a specific system layout is an area of weakness.
- The project team needs to have more input from OEMs and corroboration of costs with a wide range of suppliers.
- The proposed stationary activity does not appear to have sufficient external direction. For stationary activities, this reviewer wants to know if applications are chosen, if there is a system design, and who the industry leaders are. A better explanation of the approach is needed. There was no explanation why lifecycle cost was so flat compared to efficiency.

Recommendations for additions/deletions to project scope:

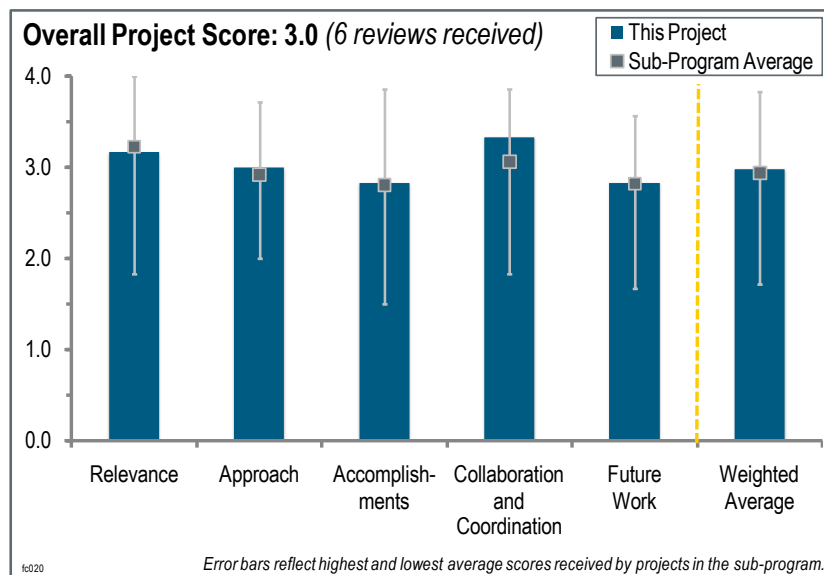
- This reviewer had no recommendations, other than the already recommended analysis of stationary systems. The investigators have done a good job, and presented a good report.
- The investigators should add to their analysis a low-pressure fuel cell system with a blower instead of the compressor-expander.

Project # FC-020: Characterization of Fuel Cell Materials

Karren More; Oak Ridge National Laboratory

Brief Summary of Project:

The objectives of this project are to: (1) identify and optimize novel, high-resolution imaging and compositional/chemical analysis techniques, as well as unique specimen preparation methodologies for the micrometer to angstrom scale (μ -Å-scale) characterization of the material constituents composing polymer electrolyte membrane (PEM) fuel cell membrane electrode assemblies (MEAs), (2) understand fundamental relationships between the material constituents within fuel cell MEAs and correlate these data with stability and performance as per the guidance of the entire fuel cell community, (3) integrate microstructural characterization with other U.S. Department of Energy (DOE) projects, (4) apply advanced analytical and imaging techniques for the evaluation of microstructural and microchemical changes to elucidate microstructure-related degradation mechanisms contributing to fuel cell performance loss, and (5) make techniques and expertise available to PEM fuel cell researchers outside of Oak Ridge National Laboratory (ORNL).



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.2** for its relevance to DOE objectives.

- Understanding the factors that cause proper functionality and durability are well-correlated with the goals of actually advancing those factors.
- The equipment, facilities, and personnel of this project offer excellent opportunities and capabilities to fuel cell material investigators.
- This project is focused on critical barriers and fully supports DOE objectives. The collaboration and support theme of the project objectives requires that the projects assisted by this project are relevant to the DOE objectives. Although there was little time to explain how ORNL's efforts fit into the projects that it supports, this information could have been conveyed in backup slides to provide further support for project relevance.
- It is important to develop the capability of high-resolution imaging and related analysis techniques to achieve the fuel cell end-game goal, particularly ultra-low Pt loading catalyst technologies such as thin film and atomic layer deposition (ALD) catalyst layers.

Question 2: Approach to performing the work

This project was rated **3.0** for its approach.

- This project is consistently developing the best techniques and methods. The tools used are very appropriate for the mission.
- While the laboratory serves an important function in materials characterization for fuel cell research, there seems to be a need for more problem and information analysis to determine ways in which the facilities can provide “the next step” toward improved material understanding and mechanisms insights. This may involve modifying the equipment and facilities to permit the availability of advanced analysis concepts.

- The investigators should determine if this project should improve analysis techniques and provide users with analysis services or pursue research such as material characterization by using its own analysis techniques. The former (analysis technique provider) is the original objective of this project. However, the project recently seems to favor the latter (own research), which would mean that the project is deviating from its original objectives, and investigators should consider the research contents in order to make it relevant to the fuel cell research goal or potential customers.
- The channel wall makes a difference. The presentation features a rib-rib structure; however, in a real stack, it is very hard to realize the design of rib-rib for MEAs.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.8** for its accomplishments and progress.

- The data of the recast ionomer in the catalyst layer are interesting and demonstrate the capability of analysis techniques including high resolution imaging and chemical bonding, as well as atomic level resolution imaging for Pt particle nucleation and its growth. Particularly later in the project, one of these must be an important and useful technique for ultra-low Pt loading catalyst technologies such as thin film and ALD.
- This project has achieved diverse accomplishments. The process of Pt nucleation and growth is found at step edges. Another accomplishment is related to changes to the ionomer. ORNL used elemental maps to link to spectroscopy to show ionomer association with the catalyst membrane structure.
- ORNL has made many outstanding contributions to several research projects; however, the next level of detailed information about catalyst activity degradation, migration, and alteration will require information on the dynamics associated with catalyst change. Working closely with the fuel cell researchers with whom ORNL is already familiar may provide this guidance. For example, cathode catalysts that have been contaminated with a particular airborne contaminant may be investigated at ORNL to determine alterations to the catalyst that may give insight to the poisoning effect.
- The mapping technique may not be suitable for fuel cells because too many factors affect the performance of the components. Catalyst growth under fuel cell operation is not new and has been well-studied.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.3** for its collaboration and coordination.

- This project includes an excellent list of collaborators.
- This project features good cooperation.
- The PI is working with many of the best projects in the portfolio, and many others. ORNL gets a lot of perspective, but this also increases the importance of the team maintaining excellent quality and continuing to innovate on technique.
- Many well respected organizations are listed as collaborators; however, the information provided about collaborators is not sufficient to evaluate the effectiveness of the collaboration. Details, such as the scope and objective of each collaboration, could be included in backup slides.

Question 5: Proposed future work

This project was rated **3.0** for its proposed future work.

- This project features worthwhile activities to work on that are essential to understanding and promoting improvements in the fuel cell subsystems.
- Further advanced work will be incumbent upon identifying how the ORNL facilities and personnel can enhance their expertise to gain new insights into catalyst stability and activity.
- The PI should continue to pursue a more systematic study of degradation.
- Different MEAs have different performance levels. Different hardware causes different levels of performance. It is not clear how to do long-term material characterization *in situ*. There are papers that address the catalyst growth problem.
- The priority of each year's future research tasks is unclear.

Project strengths:

- Scientifically, this project is of great interest.
- This project features the strong capability of high-resolution imaging and its related techniques, including specimen preparation.
- This project's strengths include the PI's willingness to back off on areas in response to guidance, fantastic technique, and innovation in methods.
- Strengths of this project include its analytical facilities and experimental personnel.

Project weaknesses:

- Investigators should be more actively seeking new methods of investigation.
- The relevance for further optimization of practical systems is questionable; there are many observations, but no clear means or suggestions on how to improve membranes, etc.
- This project has lots of data, but the functional meaning in some cases is less clear. For example, in the ionomer work it is unclear what the association of S and Pt mean.

Recommendations for additions/deletions to project scope:

- This reviewer is looking forward to the results of the element mapping versus function and aging. This could be helpful and deserves high attention.
- ORNL should determine whether this project should improve analysis techniques and provide users with analysis services or pursue research such as material characterization by using its own analysis techniques. If the latter is chosen, ORNL should consider prioritizing research contents by relevance to the fuel cell research goal and customer expectations.
- Based on the past work on catalyst investigations on carbon supports, examining the relationship between heterogeneous carbon supports and carbon on catalyst stability and ionomer reconfiguration may result in additional insight into the interactions between catalysts, supports, and ionomers. Thermal cycling *in situ* may also offer a means by which reconfigurations on the support take place.

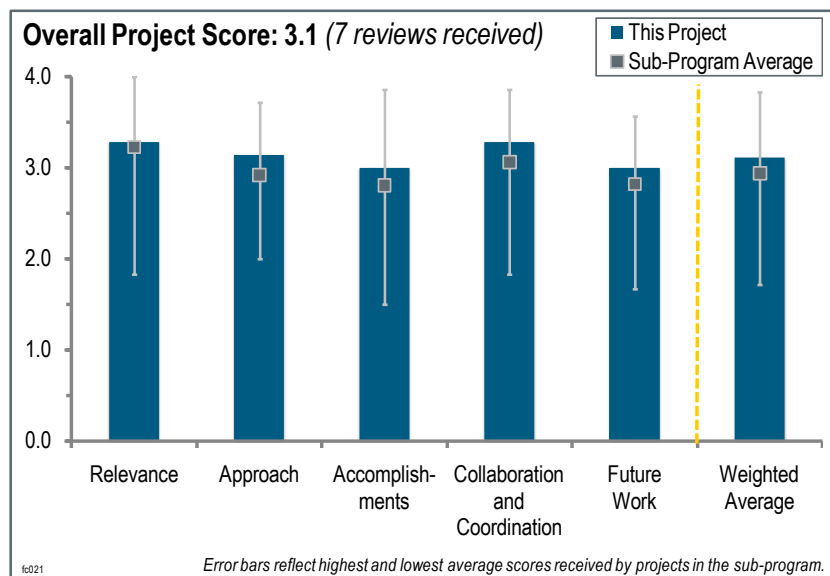
Project # FC-021: Neutron Imaging Study of the Water Transport in Operating Fuel Cells

David Jacobson; National Institute of Standards and Technology

Brief Summary of Project:

The National Institute of Standards and Technology (NIST) aims to develop and employ an effective neutron-imaging-based, non-destructive diagnostics tool to characterize water transport in polymer electrolyte membrane (PEM) fuel cells. The project's specific objectives are to: (1) form collaborations with industry, national laboratory, and academic researchers; (2) provide research and testing infrastructure to enable the fuel cell/hydrogen storage industry to design, test, and optimize prototype to commercial-grade fuel cells and hydrogen storage devices; (3) make research

data available for beneficial use by the fuel cell community; (4) provide secure facilities for proprietary research by industry; (5) transfer data interpretation and analysis algorithm techniques to industry to enable them to use research information more effectively and independently; and (6) continually develop methods and technology to accommodate rapidly changing industry/academia needs.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.3** for its relevance to U.S. Department of Energy (DOE) objectives.

- Most aspects of this project align with the Hydrogen and Fuel Cells Program's research, development, and demonstration objectives. The technique is good. Water management of fuel cell stacks is one of the most critical elements of meeting PEM fuel cell performance targets, and the neutron imaging technique provides very powerful analysis for this area of research.
- Water management is a major issue in the practical implementation of fuel cells in vehicles, and this is a highly relevant area.
- This work provides excellent support to the fuel cell projects of the collaborating partners.
- NIST has continued to provide key analytical capabilities for DOE fuel cell research in understanding the role of water in fuel cell operations.

Question 2: Approach to performing the work

This project was rated **3.1** for its approach.

- Using a relevant fuel cell *in situ* in a neutron imaging system is excellent. Featuring higher resolution is also excellent. Including industrial input in the testing is wise.
- Continual modifications and improvements in facilities to meet the challenges in understanding how water affects PEM fuel cell performance and durability are commendable.
- The principal investigator is continuing to improve testing capability, including imaging resolution and testing infrastructure (e.g., larger cell and freeze capability).

- This project is academically interesting, but it may not be practically useful. In a real full cell environment, the membrane electrode assembly (MEA) is clamped under force. Different areas have different force, which changes the local area behavior; this was surprisingly not mentioned in the presentation.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- NIST used the illustrations of monitoring water distribution and migration to understand different aspects of fuel cell performance changes, which has provided significant insights into the fuel cell operating dynamics.
- NIST is developing new methods to get better resolution, as well as developing clues about chemistry based on mass distribution and operation conditions. NIST is able to show that a hydrophilic plate can pull water out of the cell. NIST made a flat stack to look at the impact of multiple cells, and showed that cell crashes were due to gas diffusion layer water issues, not the channel.
- The investigators have demonstrated significant improvements in imaging resolution; response time (frame time) is still challenging. Response time is limited by data processing or generic issue.
- The improvement in spatial resolution is significant.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.3** for its collaboration and coordination.

- This project has many valuable partners that seem to both provide and receive value.
- This project features a good diversity of collaborations with university, industrial, and national laboratory partners.
- The collaborator list continues to remain strong and includes key organizations in PEM fuel cell development.
- This project features good cooperation and a strong team.

Question 5: Proposed future work

This project was rated **2.9** for its proposed future work.

- The plans to develop low-temperature operating capabilities will be an added benefit that will assist the projects that are investigating effects of freezing on PEM fuel cell performance.
- The future work is logical—improving the field of view and/or increasing spatial resolution would both be beneficial.
- The proposed future work will help support DOE Hydrogen and Fuel Cells Program goals.

Project strengths:

- Scientifically, this project is of high interest.
- This project's strengths are its obviously strong capability to provide novel analytical methodology and improve its capability.
- This project features rare and valuable tools, industry guidance, good partners, and a talented team, and is focused on meaningful problems.
- There is not really a better way to image water inside of a fuel cell.
- This project has excellent facilities and personnel.

Project weaknesses:

- Advanced electrodes are 0.5–10 microns (μ) thick, and advanced membranes are $\sim 10 \mu$ thick. This technology has limited value for advanced MEA development due to the resolution limits. The technical path and odds of success for the new goal of a 1μ resolution were not sufficiently explained. Information about funding, milestones, and go/no-go decisions related to the 1μ resolution subtask should have been included in the presentation.
- The transfer of the knowledge gained to the systems being improved is not clear.

Recommendations for additions/deletions to project scope:

- NIST should use low-temperature capability investigation to determine where the onset of ice formation takes place and identify possible mitigating actions that could be implemented. Including the facility's dynamic operations will be valuable in following the freezing phenomenon.

Project # FC-023: Low Cost PEM Fuel Cell Metal Bipolar Plates

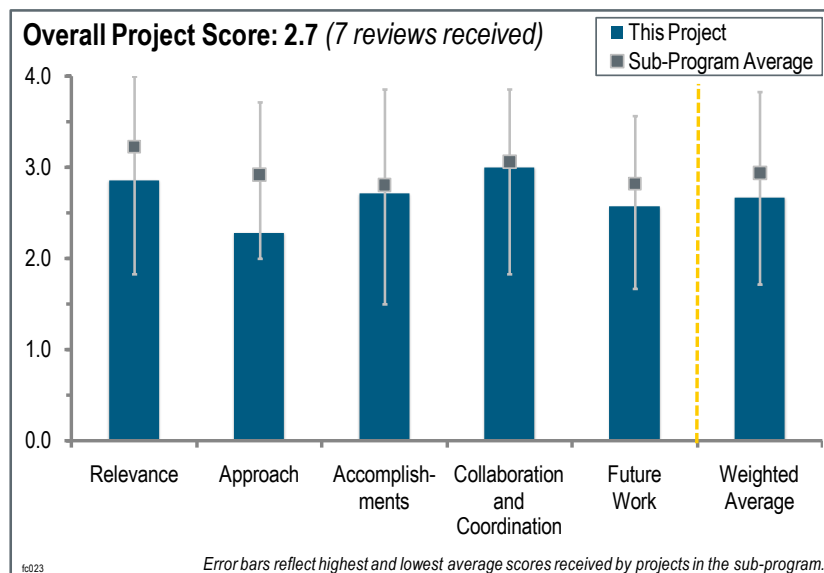
Conghua Wang; TreadStone

Brief Summary of Project:

The objective of this project is to develop low-cost metal bipolar plates to meet the U.S. Department of Energy (DOE) 2015 performance target at a cost of less than \$3/kW by: (1) developing carbon-steel; (2) reducing or eliminating the use of Pt; and (3) demonstrating TreadStone metal plate applications in portable, stationary, and automobile fuel cell stacks.

Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **2.9** for its relevance to DOE objectives.



- Developing low-cost and durable bipolar plates is critical to achieving fuel cell cost targets. The exploration of carbon-steel or aluminum plates is therefore relevant.
- This project represents an attempt to reduce plate cost, which is in line with overall objectives.
- Bipolar plates are an unheralded but potentially expensive part. Finding alternative development strategies is generically worthwhile.
- Bipolar plates represent a significant (15%–20%) component of the total fuel cell stack costs. Alternative materials, designs, and structures are needed to lower the costs to the 2015 target value of \$3/kW.
- This project seeks to develop low-cost, metal bipolar plates. Low-cost, metal bipolar plates are needed for the best manufacturing processes; however, this reviewer is not sure about the cost analysis references. Normally the catalyst dominates the cost in large volume (around 50% of cost).
- Bipolar plate research is important to lower the cost of fuel cells. This reviewer is not convinced that the TreadStone approach will meet this objective.

Question 2: Approach to performing the work

This project was rated **2.3** for its approach.

- The proposed approach is correct. Regarding previous Annual Merit Review reviewer comments, Aluminum plate development should have had earlier milestones. Intermediate cost analysis would have avoided the risk that, at the end, there is a good technical solution that is too costly to be industrialized.
- This project is investigating the use of conductive vias (dots) through an otherwise non-conducting protective layer on steel or aluminum metal bipolar plates. The conductive materials that investigators have tested or will test include Au, Pd, carbon nanotubes, and carbides, among other materials. Composition of the protective layer—through which the vias penetrate—is not clear, except for aluminum plates where Cr plating is identified. The plates are being tested in short stacks.
- Although the project features good data, it was not clear how investigators selected some of these options. This reviewer wants to know if the driving factor was a model or hypothesis. The project team should add an assessment of other cell properties that would need to change in order to enable these technologies (e.g., gas diffusion layer [GDL] conductivity and GDL compressibility). The methodical approach to materials selection

based on the hypothesis or models is not clear. The reviewer wants to know how the investigators design for cost. The process, materials, and utilization should be addressed.

- The project approach looked at three methods to improve previous Au nanodot technology: Pd dots with Au, carbon nanotubes, and CrC. The investigators' cost analysis did not show any advantage of the carbon nanotubes, so it is unclear why they did not abandon that approach immediately. For substrates, they explored carbon steel (with a no-go) and Al with a Cr coating. A no-go on the carbon steel is likely the correct decision for the near future. The investigators appropriately proposed different materials and coatings, and moved on when those approaches did not show promise.
- The TreadStone project has produced little supporting data that its bipolar plate technology does not impact the performance of proton exchange membrane fuel cells. The project needs significant validation work, fuel cell performance polarization curves, and cycling test data to validate the claims of a stable bipolar plate material.
- This seems like a very difficult approach to take. The investigators will likely face difficulties from the pinhole formation in the plate. This reviewer has serious concerns about stability under aggressive cycling conditions. This seems like it will always generate concern about electrical contact and other issues.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.7** for its accomplishments and progress.

- Given the concerns about the approach, this project features solid execution and a good focus on the technical problems. The project's steady progress seems to be continuing.
- Investigators have completed testing of several alternative conductive materials. Plate processing is at the stage of producing hundreds of samples for evaluation by others in stacks, including hot-rolled polymer gasket for continuous bipolar plate manufacturing. The 10-cell, 20-cm²-per-cell short stack shows good performance at up to 500 mA/cm² (milliamps/cm²) current densities. The through-plate voltage drop and metal ion leaching from the plates appear to be able to meet DOE targets.
- Showing all of the important metrics (e.g., material properties and cost) compared to a baseline would be valuable. The durability metrics are not clear.
- The results are globally correct but are not in line with the project timeline, and no concrete new results have been shown. Specific comments include:
 - The results presented were in cells or stacks and are considering stainless steel material with initial gold vias. Material and coating developments were delayed, so investigators should have allocated increased resources to these activities instead of testing standard solutions. The added value of these results regarding the project targets is unclear.
 - Contact resistance using carbides before and after test corrosion has been given, but without precizing the test duration.
 - Concerning the "optimal" solution proposed—Cr-plated aluminum plates with Cr carbide coating—no complete sample has been tested yet, 3.5 months before the end of the project. Moreover, aluminum plate process development is not completed. Thus, no long-term test or performance stack test with these new plates is really expected before the end of the project.
 - The cost analysis that was presented with stainless steel material was not clearly explained. The reviewer wants to know the expected value and the expected active area. Researchers have not given any indication of aluminum plates.
- The investigators have demonstrated a large active area short (10 cell) stack based on the Au-dots approach and 316 stainless steel. The activity for 2011 was unclear, as this stack was demonstrated in 2010 for 800 hours. They conducted corrosion measurements of coatings on aluminum, but have not yet shown the applicability of the coating with their conductive dots and in fuel cell mode.
- Most of the approaches examined by TreadStone have not yielded a low-cost, durable bipolar plate.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.0** for its collaboration and coordination.

- The collaboration and coordination between the partners appears fair regarding the obtained results. Resource reallocation may have been proposed to adapt to the material and coating development delays.

- The project team includes an automobile original equipment manufacturer, a university, an industrial research laboratory, and a national laboratory, along with an organization conducting manufacturing cost analyses.
- This project features good collaborations with laboratories and industry.
- The team is well-rounded, but does not comprise the best players in the field.
- The investigators have good collaboration with the Gas Technology Institute, Oak Ridge National Laboratory, and Ford, with IBIS Associates doing cost projections. It is unclear what the State University of New York is providing to the project.

Question 5: Proposed future work

This project was rated **2.6** for its proposed future work.

- This project features a solid work plan to deliver on the proposed technology.
- In the remaining months of the project, investigators will complete the plate processing development and test stacks of 200 W, 1 kW, and 5 kW rated power for up to 2,000 hours.
- The proposed work for the end of the project is correct. However, as the project ends in 3.5 months, it appears not fully feasible, in particular the 2,000 hour operation at Ford with a 20-cell stack using the optimized materials. The test should already have started.
- This project ends in August 2011, but the investigators have proposed a substantial amount of work to be completed before then. It is unlikely that they can accomplish the proposed work in the next few months, especially as it includes a 2,000 hour (approximately three months) stack test.
- This reviewer does not see how the suggested approaches will improve the durability of the project team's bipolar plates.
- This reviewer is not convinced that the production line should be scaled up. The reviewer wants to know if the "customers" of this material (e.g., Ford) are pulling this technology.

Project strengths:

- Strengths of this project include its interesting initial proposed approach and the competencies of the different partners.
- The conductive nanodot technology is showing promise. The cost projections look good for the materials that the investigators continue to pursue.
- This project uses lower cost materials compared to other approaches.
- This project's strong project team is an area of strength.
- Strengths of this project include its novel ideas and industry support.
- The cost reduction achieved by replacing dots of precious metals with carbides is an area of strength.
- It seems like there is lots of room to introduce new approaches.

Project weaknesses:

- Investigators planned to test many different options to achieve the project targets. Apparently, there were too many options. The project team did not react as fast as needed and adapt the resources to focus on the one or two best solutions in order to be in line with the project timeline. In consequence, there might be neither real stack demonstration of the new optimized solution nor an associated cost evaluation.
- An evaluation of mass production costs of carbon composite plates and this technology is needed. More data on the materials demonstrations is needed for good evaluation.
- This project does not demonstrate a good understanding of corrosion science.
- As discussed in the presentation, it is critical to avoid pinholes or micro-cracks in the coatings. Even if these are not present at the beginning of life, any imperfections may lead to the formation of such defects, which may then lead to accelerated decay in performance.
- It is hard to follow the project. A summary chart comparing all of the options to a baseline and the 2015 targets would be helpful.

- This specific project was based on a 316 stainless steel substrate. Investigators work on 304 stainless steel and aluminum substrates, which might have corrosion issues. A comparison between 304 stainless steel and 316 stainless steel substrates would be helpful.

Recommendations for additions/deletions to project scope:

- This project is ending in August 2011. It would be nice to see the Cr/Al with dots demonstrated, if possible, and particularly demonstrated in a single cell fuel cell. The 2,000 hour test is nice, but it is based on the investigators' older technology, and they simply do not have time to do this (2,000 is approximately 83 days).
- The investigators should conduct load and thermal cycling of the stacks to be tested for up to 2,000 hours to verify the integrity of the protective coatings.
- The use of accelerated stress tests would be valuable, for potential and thermal cycling. An assessment of the interface requirements of the GDL for a Au particle plate would also be valuable. This reviewer wants to know if the GDL compressibility needs to be altered, and how that would change the contact resistance.
- Investigators should conduct a cost analysis with 304 stainless steel and Cr-plated aluminum.

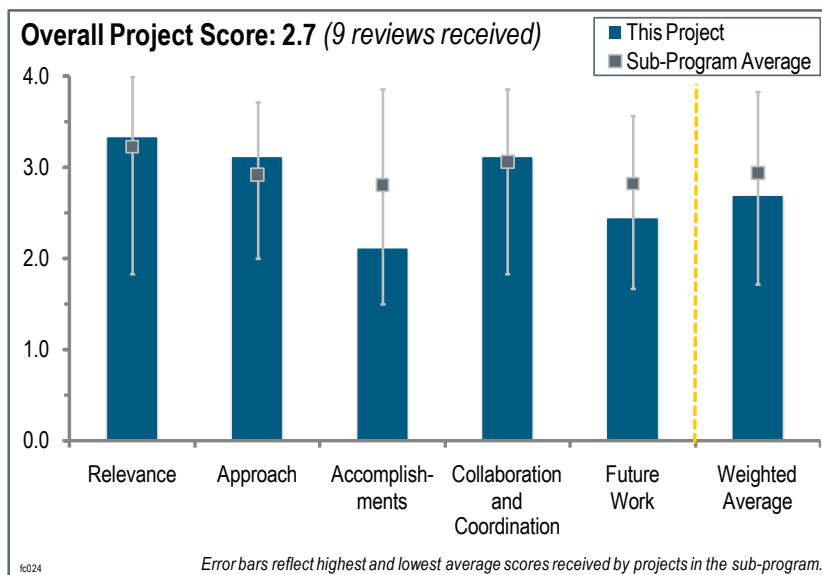
Project # FC-024: Metallic Bipolar Plates with Composite Coatings

Jennifer Mawdsley; Argonne National Laboratory

Brief Summary of Project:

The objectives of this project are to: (1) create a coated aluminum bipolar plate that meets U.S. Department of Energy (DOE) performance and durability targets for bipolar plates that are thinner and more durable than machined graphite bipolar plates and up to 65% lighter than stainless steel; and (2) develop a composite coating that is electrically conductive and corrosion resistant using a mixture of a fluoropolymer and inorganic filler.

Question 1: Relevance to overall U.S. Department of Energy objectives



This project was rated **3.3** for its relevance to DOE objectives.

- Developing low-cost, highly durable, and thin bipolar plates is one of the keys to the successful commercialization of fuel cell electric vehicles.
- Developing low-cost and durable bipolar plates is critical to achieving fuel cell cost targets.
- Developing low-cost, durable coatings for metal plates is very relevant to DOE objectives.
- The development of inexpensive and durable fuel cell bipolar plates is highly relevant.
- This project supports cost, durability, and performance, three main challenges for fuel cells called out in the DOE Hydrogen and Fuel Cells Program. The development of low-cost, metal-coated bipolar plates is critical to the Program.
- Replacement of coated stainless steel bipolar plates with coated aluminum bipolar plates would produce modest cost and mass decreases and would make fuel cell costs independent of the historically major variations in the price of nickel. However, while coatings are needed on stainless steel mainly to keep the contact resistance down (and can be highly discontinuous), aluminum is so susceptible to corrosion in polymer electrolyte membrane fuel cell conditions that coatings must be absolutely pinhole-free. Pinhole-free coatings generally must be thick so that the electronic conductivity needed for the composite is very high. Given the relatively small cost and relatively unimportant mass benefits if this concept were to work, in addition to the high risk of it not working, the relevance to the furthering of DOE goals is rather limited.
- The project has important objectives, but these approaches have been tried and reported in the literature and at Annual Merit Reviews over the last 10 years. The metal borides are a repeat of work at Los Alamos National Laboratory (LANL) approximately 10 years ago. The redo should be justified. The chemical handling industry (for pinhole-free coating justification) is not a good comparison because the electrochemical conditions seek out film weaknesses. Thick coatings using “cladding” techniques cannot be justified for the bipolar plates.

Question 2: Approach to performing the work

This project was rated **3.1** for its approach.

- Coating aluminum bipolar plates with a thin layer of conducting and protective film is a good approach to achieve this target. Developing a valid *ex situ* durability test certainly helps the Program tremendously.
- The proposed approach is correct and in accordance with the announced barriers to overcome.

- The focus on polymer coatings with conductive fillers on the aluminum surface is a good one.
- The technical approach used in this project is adequate and in accordance with the set objectives.
- It is not clear just how much experimentation should have been needed to assess this concept adequately. The coating subcontractor seems to have recognized how thick of the fluoropolymer coating was needed to give a pinhole-free film before the start of this work. Bulk conductivities of the filler materials should be known, and by scaling these by the ratio of the conductivity of carbon black-filled polymers to that of bulk graphite, one should be able to estimate the upper limit of the conductivity of the filled polymer. This estimation would also demonstrate if one is within an order of magnitude of that required to give adequate conductivity of the filled polymer through the minimum thickness required for pinhole-free behavior. But, with the exception of this apparent lack of planning, the investigators seem to have conducted this project in a reasonable manner. One additional experiment that investigators probably should have performed would have been to make a conductivity measurement of the filler powder after it had been through the acid-exposure test. For most of these materials, there is a danger of surface oxide layers forming that would electrically insulate one particle from another and from the plates. In at least one case, the X-ray diffraction (XRD) study showed evidence of such oxide formation, but one could also get unacceptable resistance from oxide layers that are too thin or too amorphous to show up in XRD analysis.
- The coatings are very thick and will not lead to thin plates even if successful.
- DOE, through LANL, has already explored metal boride. The composite coating was not well-explained in the approach.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.1** for its accomplishments and progress.

- Developing new testing protocols has significantly strengthened the project and has allowed the principal investigator (PI) to down-select a number of coatings.
- The bipolar plates are much too thick.
- Fuel cell testing at the Gas Technology Institute (GTI) is obviously the end goal; the data collected are poor and lack real value because they do not include electrochemical impedance spectroscopy studies or even high-frequency resistance results.
- The results are globally correct but are not in line with the project timeline and have the following inconsistencies or areas lacking precision:
 - It is not clear if area-specific resistance measurements examine one or both interfaces.
 - It is unclear why the coating of 50% polychlorotrifluoroethylene (PCFTE)/25% graphite/25% TiC has not been compared to the composite-coated aluminum panel.
 - This reviewer wants to know where the Orion coating comes from. It is difficult to understand the link with the previous results. It appears that it has been tested because nothing else was available.
 - With respect to single cell stack testing, there was no precision about the tested composite-coated aluminum.
 - The coating thicknesses cover a wide range (30–130 micron [μ] on each side). This wide range may impact the stack assembly and gas tightness quality, as well as the MEA compression and, therefore, MEA performance. This thickness range is not acceptable for fuel cell coatings where a μ -level range is expected. With these thicknesses, there will be no stack volume decrease.
 - This reviewer questions the mechanical strength of these coatings.
 - The wording “single cell stack” is contradictory, since a stack is usually composed of multiple cells.
- Considering that the project is 80% complete, it does not seem like it will reach its objectives. The PI should not compare new materials to uncoated aluminum because uncoated aluminum is not a popular option.
- The through-plane conductivity results with TiC and PCFTE are still five times lower than the target conductivity. Corrosion results look good.
- The major accomplishment is the preparation of a corrosion-resistant bipolar plate with a small active area. The milestone for producing and testing the bipolar plate with a large active area has slipped from the schedule.
- At the thickness needed for pinhole-free behavior, the conductance of the composite is about 10-times too low. This gap is probably too great to bridge unless it is due to some avoidable passivation of the particle surfaces. It appears that the PI did a good job of down-selecting materials and testing composites. She probably has succeeded in putting to rest this concept as impractical, which is a significant and useful, if not particularly

happy, accomplishment. But one wonders whether such an experimental project was really necessary to reach this conclusion.

- People have been working on aluminum-based bipolar plates due to their low weight. The challenge is still there, though the team made significant efforts. The fundamentals of an Al-based alloy need more understanding and breakthroughs before investigators can overcome the barriers.
- It is unclear why the investigators synthesized TiB_2 . H. C. Stark manufactures this material at reasonable quantities with controlled purity. This reviewer wants to know if the project team used graphite flake composite film for hydrophilic surface treatments. If so, it is well known that heat treatment in air will make graphite surfaces hydrophilic. The reviewer also wants to know what was new for hydrophilic treatment, and whether the sulfuric acid was analyzed for aluminum after the test of the coated plate. There were no data in charts on the corrosion rate of the composite coating. It is unclear if the composite coating was sacrificial.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.1** for its collaboration and coordination.

- The collaboration with GTI is evident and the researchers have made very strong progress. It is evident that this project is well-organized and that the technology is being transferred and tested.
- The collaboration and coordination have been fair regarding the obtained results and, in particular, the unacceptable coating thicknesses. The involved industrial partners should have pointed that out.
- The collaboration between the parties in this project appears to have been good, though perhaps the coating company could have been a bit more insistent from the start about the minimum thickness needed to give a pinhole-free coating.
- This project has strong national laboratory support.
- The collaborations are very good.
- The project activities are well-coordinated among the team members.
- The collaborations were consistent with the objectives.

Question 5: Proposed future work

This project was rated **2.4** for its proposed future work.

- The proposed future plans are well-defined and will likely be achieved. With such thick coatings and a poor understanding of fuel cell testing, it is unclear if the future goals will be achieved.
- It will be difficult to perform the announced work in the remaining three months of the project.
- If the resistance is still an order of magnitude too high at the minimum thickness for nearly-pinhole-free behavior, then the bi-layer coating and other proposals made for future work seem unlikely to close the gap. Conductivity measurements after acid-compatibility testing might reveal surface oxidation effects that could be major, and might be fixable with proper handling or with an additional coating layer on the particles.
- It is unclear why fiscal year 2010 activities are in the future work section. In the presentation, if this reviewer remembers correctly, the composite coatings were identified as too thick. If so, it is unclear why the investigators should continue the work.
- The proposed future work is adequate and laid in accordance with the project objectives.
- This project is 80% complete.

Project strengths:

- Strengths of this project include the project management, collaborations, and original concept of the proposal.
- The technical and scientific competence of the partners for addressing the project target is an area of strength. The current results provide potential solutions to be applied on aluminum bipolar plates to achieve lighter stacks.
- Coatings are very important in meeting the low cost goals for plates. Coatings for aluminum that are pinhole-free would be very significant.
- There is a high payoff in both cost and durability if aluminum bipolar plates can be made to work instead of the current graphite plates.

- The team showed a great ability to produce and test a wide variety of the bipolar plate coating compositions.
- The project features a straightforward, effective methodology. Other areas of strength include the reasonable choice of materials in this past year and the assembly of a team with relevant expertise.
- This project started with an out-of-box idea.

Project weaknesses:

- Weaknesses of this project include the coating thickness and the lack of expertise in fuel cell testing and characterization.
- No clear and fast applicable outcomes are expected from the project. Much work remains to be done to integrate the proposed coating into a stack.
- The coatings must be thinner.
- The through-plane conductivity of the coated aluminum bipolar plates may not be high enough as compared to graphite.
- The targeted plate resistance may not be achieved due to schedule delays.
- The researchers might not have done enough estimation of likely resistances at hole-free thicknesses prior to the initiation of the project. It is not clear whether enough attention was given to possible particle-to-particle and particle-to-plate resistances due to superficial oxidation of the particle surfaces.
- The thin coating is not working, while the thick coating will introduce higher resistance and cost and shorten life in thermal cycling. These effects relate to the fundamentals of the materials.
- The project team was unaware of previous DOE sponsored research and development (R&D) on boride coatings at LANL. This effort was not a very creative R&D effort for a national laboratory.

Recommendations for additions/deletions to project scope:

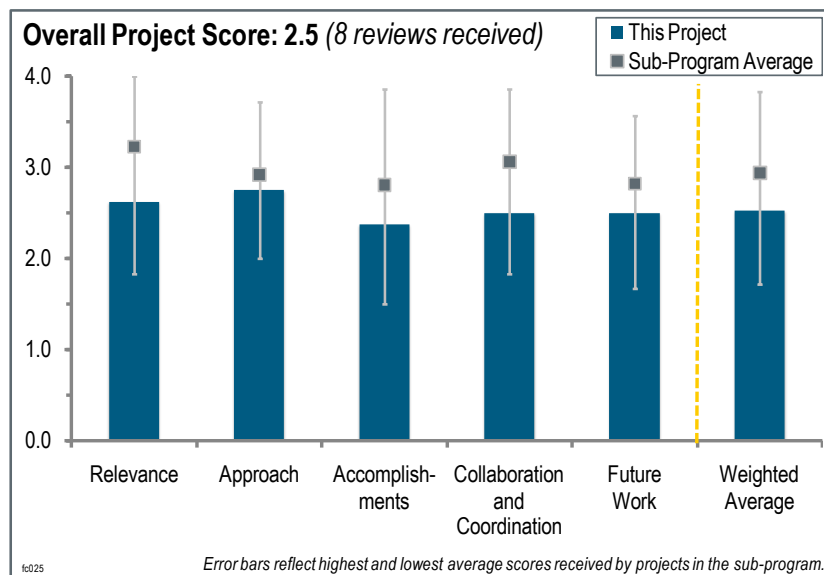
- The researchers should pursue thinner coatings and high-value fuel cell testing.
- This reviewer is not sure why in-plane conductivity was considered important.
- The project team should include thermal cycling as a critical test for coating-metal adhesion evaluation.
- The PI should do conductivity measurements of powders after acid exposure tests and then wrap-up the work and write the report so that these efforts are not repeated in the future.

Project # FC-025: Air Cooled Stack Freeze Tolerance

Dave Hancock; Plug Power, Inc.

Brief Summary of Project:

The project objectives are to: (1) evaluate and develop the stack and system together to meet durability, cost, performance, and freeze tolerance requirements; (2) develop an understanding related to integrating air-cooled stack technology into a dynamic materials handling system with frequent start-up cycles; (3) test and evaluate air-cooled stacks and components developed to increase freeze tolerance and durability; (4) evaluate failure mechanism mitigation in stack and/or system design; (5) perform life-cycle cost analyses for freeze tolerance strategies; and (6) document and publish a summary of stack freeze failure analyses.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **2.6** for its relevance to the U.S. Department of Energy's (DOE) objectives.

- The key topics focus on major issues and barriers of fuel cell technologies. They address durability, cost, and performance by increasing understanding of air-cooled stack technology with a focus on the freeze start behavior of stacks and systems. Studying freeze start effects and mitigation is an important issue that is required for moving fuel cells toward the marketplace.
- The project does address the two major barriers—cost and durability.
- This project is very focused on a specific cost and durability barrier—freeze tolerance in air-cooled stacks—that is relevant to the near-term implementation of fuel cells for materials handling. The information gained will therefore be highly important to the application, but perhaps of less general relevance and less likely to generate an overall “breakthrough” for fuel cells. However, the project fits well within the diversity of projects that DOE should support.
- The project supports the DOE goal of applied research and a portfolio of fuel cell technologies. This project's targets, and how they are related to DOE targets, are unclear.
- The simplified, air-cooled stack is a good concept, but this reviewer questions if the material handling equipment market is the correct market target because of the durability, power density, and market size of the extreme cold conditions for which this project is designed.
- The results of investigating one special stack model are perhaps not easy to generalize to a broader range of applications.
- Although the business case for air-cooled polymer electrolyte membrane (PEM) fuel cell systems is clear, the scientific impact of this work is lacking. The project needs input from fundamental degradation mechanisms. The conclusions from this project are convoluted by several additional variables associated with a low-temperature system.

Question 2: Approach to performing the work

This project was rated **2.8** for its approach.

- This project features a clearly structured approach.
- The project appears to be tailored to the development of one particular system. The development of detailed scientific understanding of freeze processes and their mitigation is lacking. The project is specific to one architecture family and one system. Investigators did not present a strategy to allow interpretation of the data across a variety of architectures. Outreach and data dissemination is missing in the approach.
- The combined modeling and experimental approach is good. There needs to be more discussion of model validation and tuning though, especially because the durability projections seem to depend on the modeling results.
- It is difficult to assess the approach based only on the Approach slide because the language used is rather opaque—for example, “Baseline Freeze Failure Analysis,” “Generate Hypothesis for Freeze Function,” and “Freeze Testing for System Input.” However, from the presentation, one can deduce the kinds of characterization and modeling used to achieve the results.
- The actual work being accomplished has been poorly communicated.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.4** for its accomplishments and progress.

- The project has shown impressive progress toward improving the durability of air-cooled stacks, as exemplified on slides 7 and 11. This reviewer assumes that the membrane electrode assembly (MEA) strategies (Ballard) and system operation strategies (Plug Power) have been or will be combined. Regarding freeze specifically, identifying the appropriate conditions to rely on system control was a major outcome—the temperature should be kept above -10°C. This reviewer is not certain, based on the presentation, if the issues with having an undesirable stack inlet temperature gradient, as shown in slide 15, would remain if the cathode air recirculation strategy described on slide nine is implemented. It seems that some work remains in developing a sufficiently freeze-tolerant stack, but perhaps this overall goal can be reached by the project’s end.
- Progress has been good, but it is not clear what has actually been done and if the improvements are actually new technology or simply implementing technology developed by others.
- The results of the materials development are promising. The modeling results are sufficient, but there needs to be more analysis of the dominant losses in the system and the inefficiencies. It is not clear whether the investigators are appropriately considering the costs of the start-up energy and hydrogen fuel in terms of mitigation, etc. The project focuses on freeze, but this does not seem to be studied, as the temperature and stack do not go below -5°C or -10°C where ice may not be present in the materials. This reviewer wants to know if the correct physics are employed in the model and degradation mechanisms and how confident the project team is in the lifetime predictions.
- The set of results was not conclusive. Stack cost projections showed that cost will be reduced when moving from liquid-cooled to air-cooled architectures. The presenter pointed out that this stack was not aimed at automotive applications, though lifetime targets of 5,000 hours were used as criteria. The presenter, when questioned, pointed out that some data plotted as air-air freeze start was actually start-up from idle states. That is obviously very different and the results are thereby questionable. The presenter did not identify the material improvements that they made during the project to improve the baseline material for testing. They also failed to identify the failure mechanism for leak development. They did identify the critical temperature for freeze start for the given architecture, and developed an engineering mitigation strategy that prevents the stack from dropping below a certain temperature. This may not be practical for many applications, specifically automotive applications. This reviewer’s impression from last year that the fuel cell community will not benefit from this project was confirmed.
- This reviewer would like to see the time period of the testing.
- The mitigation methods described for start/stop are obvious to one skilled in the art. Investigators need to focus on degradation mechanisms associated with low-temperature operation (perhaps liquid water).

Question 4: Collaboration and coordination with other institutions

This project was rated **2.5** for its collaboration and coordination.

- The project is centered on the efforts of the two partners, which may be appropriate given its near-term product focus.
- There are only two parties, and it seems that Ballard is doing more of the research and development work while Plug Power is only integrating the system. Some more collaboration with material suppliers would be beneficial.
- The commercial collaboration seems sound, but the lack of laboratory and academic partners seems to leave the research community in the dark. This is highlighted by having no publications associated with the project.
- The collaboration consists of two companies. Collaborations with academia or national laboratories to create fundamental results are missing. This is reflected in the results and in the fact that no information of general interest is shared with the public by presentations or publications.
- The only collaboration is between two businesses that already have a strong original equipment manufacturer-supplier relationship (i.e., Plug Power and Ballard). Additionally, the communicated results are so vague and non-transparent that nothing much is gained.

Question 5: Proposed future work

This project was rated **2.5** for its proposed future work.

- The future work is appropriate for the length of time remaining in the project, which is scheduled to end in November 2011.
- Future improvements may result in some additional incremental improvements, but not any major improvements.
- The project is ending. The future work is sufficient, although some model validation would be beneficial for the community.
- The proposed future work is mainly focused on air handling systems. The reviewer wondered why it is not focused on materials and designed for liquid water associated with low temperatures.
- This appears to be a product development project. This reviewer does not believe that the scope for such a project should involve the development of new filter systems. Instead, researchers should investigate the general effects with gas flow. The proposed modeling work needs to be experimentally validated, but it is questionable if that is possible in the remaining timeframe of the project.
- The proposed future work seems like quite a bit for the remaining nine months. Because of the short timeframe, the quantity and remaining timeline should have been included.

Project strengths:

- Strengths of this project include its use of real fuel cell stacks and the fact that the modeling supports the experiments.
- The partnership between two industrial partners with a vested interest in product development has resulted in significant durability advances for the specific technology of air-cooled stacks.
- This project features a systematic approach to a specific issue of an air-cooled stack.
- This project's simple, low-cost design is an area of strength.
- This project features solid input on the business case.
- An area of strength for this project is its development of complete PEM fuel cell systems for a commercially viable, near-term application. Another project strength is the focus on cost reduction and durability improvements utilizing both stack and MEA improvements, as well as system design and decay-mitigation strategies.
- This project has made good progress and has a good combination of modeling and experiments. The improvements and strategies seem to be making progress.

Project weaknesses:

- This project did not feature any sharing of information that would be of real value to the community. It also lacked a science aspect and did not present any scientifically interesting results. It is a product development oriented project with only one or two companies as beneficiaries.
- It is unclear the degree to which the lessons learned from this project can be translated to other fuel cell technologies or if the project's main value is advancing the cause of early market penetration.
- The project is very specific to one stack model.
- Weaknesses include the fan turn down capability and power density.
- The project lacks scientific information.
- There is a lack of clarity regarding what is actually being done to improve cost and durability. It is unclear how the fuel cell community benefits. For example, it is not at all clear what is meant by "reduced starts strategy" (on slide 11) when the chart shows an increased number of "air-air starts." A reviewer question seemed to reveal that investigators were employing both a reduced number of air-air starts (by utilizing "idle" strategies) and potential control during the remaining air-air starts. This information should be clearly communicated. Slide 11 makes no sense without an explanation.
- The control and prediction of durability are questionable. There is a lack of true freeze-related studies, as the concept is to keep the stack warm.

Recommendations for additions/deletions to project scope:

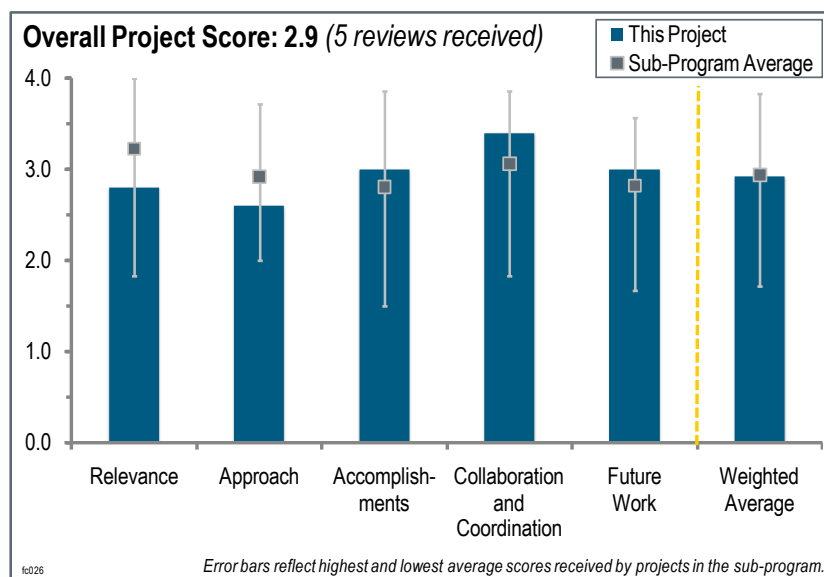
- The project is ending. Investigators should publish some findings to help the broader community.
- The investigators should add a science aspect to the project. They should also redirect the work so that it increases general understanding of freeze-start phenomena and interpret the findings with respect to a reference to increase the usefulness of the findings and share them with the fuel cell community.
- The project team should generalize the results to other air-cooled stack systems and show the relevance of air-cooled stacks in the framework of fuel cell mobility.
- Investigators should refocus on degradation mechanisms associated with low-temperature operation versus any fuel cell system. They should also isolate key durability mechanisms associated with air-cooling versus low-temperature operation, as well as propose a system strategy that will prevent the system from freezing (e.g., start-up at $< 5^{\circ}\text{C}$) and reevaluate cost and durability. Perhaps first-generation systems could afford this, as it seems that leaving a fork truck in a freezer will be a rare event.
- Investigators should provide a better explanation of the technology (e.g., explain how the air-cooled stack design looks at a high level) and the technical improvements they are implementing. Otherwise, this project has limited value to the fuel cell community.

Project # FC-026: Fuel-Cell Fundamentals at Low and Subzero Temperatures

Adam Weber; Lawrence Berkeley National Laboratory

Brief Summary of Project:

Project objectives are to: (1) provide detailed understanding of transport phenomena as well as water and thermal management at low and subzero temperatures using state-of-the-art materials by (a) examining water (liquid and ice) management with thin film catalyst layers, and (b) enabling development of optimization strategies to overcome observed operational and material bottlenecks; and (2) elucidate the associated degradation mechanisms due to subzero operation to enable development of mitigation strategies. Improved understanding will allow U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program targets to be met with regard to cold start, survivability, performance, and cost.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **2.8** for its relevance to DOE objectives.

- This is a good project with a good approach to understanding the fundamental issues of polymer electrolyte membrane (PEM) fuel cells.
- Frozen start fundamentals are needed for automotive fuel cells. Freeze failures are primarily associated with the cathode catalyst layer. The current presentation indicated an increased focus on this component as compared with last year.
- It is important to expand understanding of the material and performance limits of PEM fuel cells due to low temperatures.
- Freeze starting is a non-issue for original equipment manufacturers. Though it may be interesting to understand the fundamentals, there is almost an unlimited number of other problems with fuel cells that do not have established engineering solutions that could be looked at instead. Additionally, although studying freeze starting with nanostructured thin film (NSTF) is novel, NSTF currently has other, more cumbersome, limitations prohibiting use in commercial fuel cell systems.

Question 2: Approach to performing the work

This project was rated **2.6** for its approach.

- In general, it was a very solid and complete approach to looking at freeze starting.
- The approach is focused on critical barriers and investigating all components. This reviewer suggests considering the clamping force effect on the components under sub-zero conditions.
- Lawrence Berkley National Laboratory is adding new diagnostic techniques and modeling to develop an advanced understanding. However, because NSTF is a fundamentally different structure than dispersed electrodes, the approach of using data from both structures to make one model seems overly complicated. There are many additional elements to this project, but electrode modeling and characterization will be the most critical to predicting freeze start performance.

- Investigators did not put the approach outline into the context of the entire project milestones, which would have given a more complete picture of the plans and deliverables as well as the roles of the collaborators over the course of this work. The continuity and relevance of the early work to the later efforts and iterations of lessons learned for guiding model development were not clear.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- The project features thorough analysis of different components.
- It is not clear how capillary properties in dispensed catalyst layers correlate to NSTF.
- It would help to put error bars on the plots (e.g., on slide 17 of the presentation). The principal investigator (PI) needs to discuss the effect of gas diffusion layer (GDL) fiber composition angles and open areas in the onset of ice formation. This reviewer wants to know whether the surface of the GDL would be expected to form ice earlier than the interior due to the increase in water accumulation and change in surface energy. Also, the researchers may want to measure H₂ transport change across the membrane electrode assembly during freezing operations to determine the change in membrane and catalyst layer pores and compare this to larger molecules, such as Ar, to determine the pore size change as freezing takes place.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.4** for its collaboration and coordination.

- This project features very strong teamwork.
- The project utilizes the strengths of the individual collaborators and institutions. Considering the number of collaborators on the project and the progress to this point, the overall level of collaboration is excellent.
- It would be helpful to show which collaborator provided which data and information, such as on slides 14 and 19.

Question 5: Proposed future work

This project was rated **3.0** for its proposed future work.

- The PI needs to address temperature cycling with voltage cycling.
- It would be helpful to show which collaborator is contributing or is responsible for information in each of the listed tasks to be conducted over the next year. Also, information regarding the relationships between controls in experiments and findings should be shared with stack manufacturers to assist them and to solicit feedback on the testing protocols.

Project strengths:

- Overall it is a very well organized project that thoroughly investigates water management and freeze starting, and has a very good team.
- This project has a strong connection between modeling and experiments. The increased focus on the catalyst layer is good because most real-world fuel cell stacks start-up from a cold or frozen state with a dry GDL. Under these conditions, water or ice in the catalyst layer will shut down the stack before GDL saturation reaches a critical level, regardless of degradation.
- The project features a good team and a very appropriate topic area to investigate. The approach seems to be appropriate to gain better insight into the freezing mechanism.
- This project looks like a basic scientific activity.

Project weaknesses:

- The relevance for and transfer to system development is not clear.
- In general, freeze start-up is not a very relevant issue at this time.

- Other reviewers have suggested that the project scope should cover catalyst structures other than just NSTF. This reviewer disagrees and believes that NSTF freeze and cold start performance is more critical, as this technology is the best path to DOE targets. The PI should not dilute the project with dispersed catalyst work; understanding the fundamental limitations of NSTF is more important.
- The model complexity and relation to the inclusion of many of the parameters, such as nano-delamination, channel development in the catalyst layer, and others, is not clear.

Recommendations for additions/deletions to project scope:

- Investigators should keep focusing on NSTF.
- The project team should correlate gas transport with freezing phenomena to help determine pore size and channeling changes during the onset and subsequent freezing.

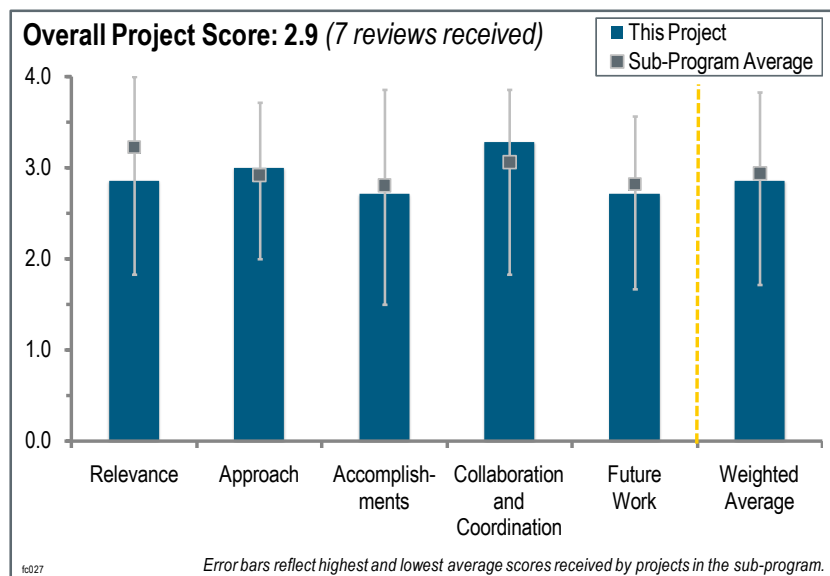
Project # FC-027: Development and Validation of a Two-Phase, Three-Dimensional Model for PEM Fuel Cells

Ken Chen; Sandia National Laboratories

Brief Summary of Project:

The project objectives are to: (1) develop and validate a two-phase, three-dimensional transport model for simulating polymer electrolyte membrane (PEM) fuel cell performance under a wide range of operating conditions; (2) apply the validated PEM fuel cell model to improve fundamental understanding of key phenomena involved, identify performance limiting phenomena, and develop recommendations for improvements to address technical barriers and support the U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program's objectives; and (3)

employ the Sandia National Laboratories (SNL) toolkit for design, optimization, and uncertainty quantification (DAKOTA) with the PEM fuel cell model's computational capability to improve and optimize PEM fuel cell design and operation. Consequently, the project helps address the performance and cost technical barriers, as improving performance will use less material (e.g., catalyst) or minimize operation cost (e.g., reduce pumping power).



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **2.9** for its relevance to DOE objectives.

- This project is highly relevant to the Program's objectives. The project seeks to develop a validated PEM fuel cell model that can be employed to improve and optimize PEM fuel cell designs and operation. It will decrease costs while increasing performance and potentially durability.
- This project addresses the barriers of performance improvement and cost reduction. It will support fundamental understanding of the effect of vapor phase and liquid water distribution in fuel cells. It also allows for predictive modeling that will reduce costs for experiments and generate insights that cannot be achieved through experiments.
- This project is a very important activity to enable industry and technology.
- Fuel cell modeling provides insight into cell and stack operation that contributes to and supports design and operation understanding.
- The model offers a thorough representation of the physical phenomena. It is not clear what is required to adapt the model to different flow-field geometries.
- The Program needs advanced modeling to guide characterization and validation work regarding the complex physics of a fuel cell stack. However, at this point, a three-dimensional model is overkill because a consensus on one-dimensional physics in the membrane and electrode still does not exist.
- The principal investigator (PI) claims that there are multiple relevant objectives. However, this reviewer does not agree with any of them. The PI asserts that the project will develop a three-dimensional, two-phase PEM fuel cell model. This reviewer does not think that anyone will be interested and wants to know what it will be used for. The PI also argues that the model will be used to improve fundamental understanding. One does not need such a complex model to identify limiting phenomena. No recommendations have been generated to date, and it is doubtful that any will result from the big, complex model. Finally, the PI states that the big model will be useful in developing advanced cell designs and operation. The reviewer doubts that this model will be

capable of making predictive recommendations that cannot be done (sooner and with fewer resources) with simpler models.

Question 2: Approach to performing the work

This project was rated **3.0** for its approach.

- The proposed approach should allow the project team to reach the project objectives.
- The combination of three-dimensional, two-phase modeling and experimental verification will provide insight into cell and stack performance and operation. The role and usefulness of DAKOTA needs to be discussed in more detail.
- The approach is effective, shown by the experimental validation of the results extracted from the model. Coupling with DAKOTA extends the model's worthiness from pure simulation to optimization of the described fuel cell architecture.
- The approach uses a number of experimental results for model validation, which is essential for model development. The use of a segmented cell system will produce insights that will be of great value for the intended model. Spatial information will be very beneficial for three-dimensional model development. The selection of a 10-by-10 segmented cell with high resolution is positive; however, segmentation of the current collector with a conductive one-piece flow-field may lead to some resolution loss that needs to be investigated.
- This reviewer likes the approach, but would like to understand how the industry can get access to and use the tools as they are developed.
- The approach slide claims that the polarization validation work is complete, but the spread in the experimental data is high. More validation data should be taken to improve the confidence interval, as a resolution exceeding 70 mV (millivolts) should result from such a high-fidelity model.
- The rationale for such a complex model is tenuous. The "sub-models" developed for use in this model may be valuable.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.7** for its accomplishments and progress.

- The preliminary validation of the model with Los Alamos National Laboratory (LANL) data is encouraging.
- This reviewer likes the data validation work. Channel flooding will be a challenge.
- The project seems to be on schedule. The investigators have successfully performed the two-phase model convergence and experiments on current distribution mapping. Although the accuracy of the model is within the uncertainty of the experimental data, this reviewer recommends that the researchers explore the larger gap between the model prediction and the experimental results at a high current density.
- The investigators have clearly presented progress toward the objectives. The model appears to be applicable to different cell geometries. The first model comparisons to the experimental data show acceptable agreement regarding the serpentine geometry used with the LANL setup. It was not the easiest geometry. Further improvements are nevertheless expected. Investigators achieved another accomplishment regarding the project schedule. The model explanations should be better described for topics such as the model assumptions and main physical equations. The model parameters sensitivity study should be better presented in order to identify the main relevant parameters to take into account for the design phase (e.g., membrane electrode assembly or bipolar plate designs and operating conditions), be able to assess the model applicability, and compare with the experimental uncertainty.
- The computed segmented cell data versus the non-segmented cell data did not reflect the actual segmented cell hardware used in this project. Experimental confirmation of the computed current collection would be valuable. The error values of the model were not so impressive when compared to the segmented cell. Deviations of up to 30% at individual segments do not yet indicate full understanding. The team should investigate if these deviations are hardware or model related.
- The investigators are only validating dry conditions. The "chevron" flow-field is not correctly modeled. The anode and cathode channels should be out-of-phase with each other.
- To date, this project has produced nothing of significant value. Obviously, this is a major undertaking, so it requires a significant investment just to develop the capabilities that the PI has in mind. However, the PI seems

to think that some new contributions have been made, which just reflects the PI's lack of knowledge regarding the state-of-the-art understanding of fuel cell technology.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.3** for its collaboration and coordination.

- The collaboration between the different partners appears to be well established and working. Industrial involvement is clearly critical to developing a designing model that is applicable to many situations. Therefore, it may be interesting that the Ballard stack designs proposed in the next steps refer to stationary applications, while Nissan will cover transportation applications.
- The collaboration comprises a healthy mixture of national laboratories, industry, and academia. The interaction between the individual partners is apparent. The exchange of data, information, and materials will require a significant exchange and will help the project to succeed.
- There is nice collaboration with Penn State, Ballard, General Motors, and LANL.
- The project features a good exchange of information with the partners to define (e.g., the work with Lawrence Berkeley National Laboratory) and validate the model (e.g., the work with Ballard and LANL).
- The project team includes stack developers and integrators, automobile original equipment manufacturers, and national laboratories, all with extensive experience and knowledge.
- The project features a good list of collaborators, but the interaction appears to consist of (1) subcontractors generating data and sub-models that are often useful and interesting by themselves and (2) subcontractors feeding these results to the PI for use in the big model, which is a "black hole" with respect to useful results. The subcontractors do not appear to get anything of value from the PI.

Question 5: Proposed future work

This project was rated **2.7** for its proposed future work.

- The proposed work is in accordance with the technical results and the project objectives. Investigators should address the points mentioned in the Accomplishments and Progress sections, as well as the relative humidity and temperature variations inside the cell.
- There is a straightforward path to complete the model, but model validation may need to be emphasized a little more.
- Additional experimental validation is planned, including open-circuit voltage and neutron imaging using LANL and Ballard data and National Institute of Standards and Technology images, respectively. The researchers should exercise caution when using partially segmented cells for validation.
- Further model validation is encouraged. The project team should combine through-plane neutron spectroscopy with water balance techniques.
- This reviewer would like to see a path to getting access to these tools.
- This project could be stopped and nothing of significance would be lost as even the useful tasks being done by the subcontractors are also being done in other projects. The continual development and validation of the big three-dimensional, two-phase model is not worthwhile.

Project strengths:

- A strength of this project is its good partnership and balance between modeling and experimentation. Another strength is the integration of "real" stack modeling in the sense that the model will consider the cell stacking and not just a single cell that will be repeated. However, this will only be effective in the following years.
- This project features a strong team with a lot of expertise. The team's capabilities are put to good use.
- The low computational time described during the Annual Merit Review enhances the effectiveness and the usability of the modeling tool.
- The state-of-the-art computational fluid-dynamics modeling for dry operating conditions in fuel cells is an area of strength.
- The project is starting to make a considerable effort toward validating the big model. However, this is a very difficult task and it is not clear if this complex model will ever be capable of matching the experimental data

from the different cell designs without adjusting the parameters (beyond the geometric parameters that should be varied to account for the different geometries). It is highly doubtful that this model will be capable of generating believable predictive results for novel cell designs.

Project weaknesses:

- The current agreement between modeling and validation has to be further improved in order to meet the announced project targets. Investigators should do a better job of pointing out sensitivity analysis in order to better assess the most relevant parameters in designing a PEM fuel cell.
- The model validation may need some improvements.
- It seems the software platform (Fluent) and model are limited in their ability to be used by a large number of players in the fuel cell community.
- The physics and validation for wet operation are insufficient (in and outside this project).
- To date, it appears that nothing new has resulted from this project. For example, the results and conclusions shown in slides six through eight are exactly as expected (i.e., there are no surprises here).

Recommendations for additions/deletions to project scope:

- It is very difficult to appreciate how different modeling related projects complement each other. From that point of view, this can be seen as a new model among many others.
- Investigators should try to improve the dry predictions before moving to wet.
- Investigators should stop working on the three-dimensional, two-phase stack model for the reasons stated above.

Project # FC-028: Transport Studies Enabling Efficiency Optimization of Cost-Competitive Fuel Cell Stacks

Robert Dross; Nuvera Fuel Cells

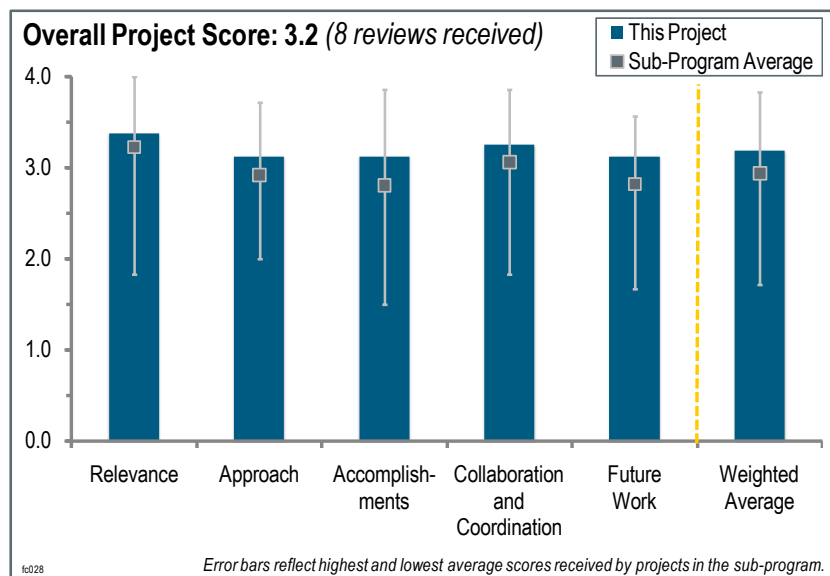
Brief Summary of Project:

The objective of this project is to investigate transport limitations at high current densities in order to optimize the efficiency of a stack technology meeting U.S.

Department of Energy (DOE) 2015 cost targets of \$15/ kWe (kilowatt electric), with stack power density of 2,000 W/L (watts/liter) and stack efficiency of 65% at 25% of rated power, and 55% at rated power.

The project is on schedule, and the 2010 go/no-go milestone has been met.

Question 1: Relevance to overall U.S. Department of Energy objectives



This project was rated **3.4** for its relevance to DOE objectives.

- Lowering cell and stacks cost while maintaining performance and efficiency is critical to the achievement of DOE Hydrogen and Fuel Cells Program goals.
- Water transport is an issue in the polymer electrolyte membrane (PEM) fuel cell.
- Transport studies to improve fuel cell performance and decrease costs are extremely relevant.
- A detailed electrochemical model for cost analysis perfectly suits the requirements of advanced cost optimization of fuel cells.
- The presentation provided a good overview on progress from the Nuvera testing project. Though the project is 50% complete, it has passed the go/no-go decision at 1.11 W/cm² on a four-cell short stack.
- The intention of the project is to produce a published model that will help stack designers improve efficiency and lower costs for either open flow field or channel and land cell designs. In principle, this objective is relevant to the Program goals. While the details of the open flow field design are likely shrouded in the model, the model will still include channel and land inputs, which makes the model relevant to a wide breadth of stack designers.
- The objective makes sense, but it is very general and does not have any details about the low-cost stack technology.
- The presenter stated that the overall objective of this project is to optimize the efficiency of a stack technology that meets DOE 2015 cost targets, yet the deliverable is a model capable of predicting high current density operation in different architectures (slides 9 and 18). This reviewer has to agree with one of last year's reviewers and restate that it is "not clear what fundamental findings will be derived from this project that can be shared and can help the wider fuel cell community." Nuvera contends that the model itself will answer many questions. The project is useful to the extent that it shows what one has to do to reach 65% efficiency at 25% part load to reach the 2015 cost goals and operate at approximately 3 A/cm² (amps/cm²) or at 7.5 W/mg of Pt. The investigators will probably achieve these goals by using special open architecture (porous media flow fields) and low equivalent weight ionomer membranes, all of which are intellectual property to the project, and which no model development will reveal to the fuel cell community as a whole.

Question 2: Approach to performing the work

This project was rated **3.1** for its approach.

- The model verification is important.
- The “two-dimensional+1” model looks like an opportunity to significantly reduce computational efforts.
- The approach of starting with a low-cost cell and stack design concept is unique and will provide valuable insights into operating envelope and efficiency limits. The concept involves an open flow-field design operating at very high current density with very low Pt loading. Nuvera understands that this approach may result in other problems such as heat rejection. Modeling will apply to both open flow-field and land-channel designs.
- The project does a good job of addressing DOE barriers B, C, and E. The methods applied are detailed and scientific; the research subject is of high technical relevance, though.
- Should the DOE and the U.S. DRIVE personnel not change the efficiency standards at full and part load on the 80 kW automotive demonstrator, then this approach is the only viable method to meet these standards and the cost goal. After the costs of the membrane and the coated metallic bipolar plates are reduced, operation at a very high W/mg of Pt level is the only option. This approach leads to operation at 3 A/cm² (amps/cm²), which is perhaps viable for Nuvera, but not for the fuel cell community as a whole. However, with this approach, this project is sharply focused on overcoming the critical barriers.
- While driving stack power density higher may lower stack cost, investigators still need to consider cell voltage to ensure that the need for heat rejection does not drive up thermal system weight, volume, and cost. The modeling approach is standard, but two-phase treatments for the Nuvera empirical model and the multi-physics model were not shown in the presentation slides. It would be interesting to understand if condensed water is treated empirically, by a pore network model, or with something else. Heliox and air contrasts begin showing differences at much lower current densities than electrochemical impedance spectroscopy (EIS) data indicate. It would be interesting to know what accounts for the difference and which set of data constitutes the preferred inputs for the modeling.
- The approach of integrating model development with materials development is fundamentally a good one. The direction of the project (and of Nuvera's approach in general), which uses relatively low operating voltages (as low as 0.5 V [volts] quoted in the presentation), is questionable, as (1) the heat removal problem becomes more difficult, and (2) decreasing system efficiency undermines one of the principal advantages of fuel cells relative to internal combustion engines.
- Nuvera has many years of experience with real fuel cell stack assembly and testing. It is unclear why Nuvera chose the fuel cell membrane electrode assembly (MEA) clamping “rib-to-rib” diagram, as there is no way a real fuel cell has the “rib-to-rib” structure. The holding force is key to the transport issue; however, no holding force was addressed. Without the clamping force for fuel cell components, the model is very hard to use to address real fuel cell issues. Therefore, the approach is not valid.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.1** for its accomplishments and progress.

- Investigators have built and validated the model, and passed the materials development go/no-go decision. The open flow-field design shows marked mass transport advantages over land-channel.
- The project is producing modeling results that are in reasonably good agreement with the experimental results, and is producing materials capable of high-performance operation (i.e., meeting the 1 W/cm² go/no-go).
- The project team has achieved 50% of its goals.
- The progress in modeling and verification is very convincing. The researchers completed an important step with the implementation of a multi-phase physics model, which was verified with an existing empirical Nuvera model. Moreover, an electrode model for Pt agglomeration was implemented in cooperation with the Lawrence Berkeley National Laboratory (LBNL). This model is now ready to be used to improve designs and materials. The materials work is sharply focused on the targets—see, for example, slide 15 in the 2011 Annual Merit Review presentation.
- Modeling inputs have been delivered from heliox and air polarizations as well as from EIS. The Nuvera empirical model appears to provide a match with single-phase and two-phase operation. As with any empirical model, the question remains as to how far the model can “walk” toward operational extremes such as low or

high temperature, and combinations of low pressure and high relative humidity (which imply low oxygen partial pressure). Low-loaded MEAs have achieved 2010 go/no-go targets. The researchers have completed the initial model verification. The slide did not say, but it can probably be assumed that this verification was for single-phase operation. While electro-osmotic drag fluxes may be low at low current density, it is unclear if the electro-osmotic drag coefficients are themselves near zero.

- While significantly more progress was made during the second half of this project, some of the key first steps were addressed in the first half of the project.
- At this point, the project personnel should have realized that meeting both the DOE cost targets and the U.S. DRIVE efficiency at full and part load is not feasible if operation has to go to approximately 3 A/cm². This reviewer listened with high attention to the presentation by the system modelers at Argonne National Laboratory and asked them what they recommend to meet both the efficiency and cost goals for 2015. They stated that to forego operations such as Nuvera's, they are going to recommend that U.S. DRIVE lower the efficiency at full and part load for 2015. If U.S. DRIVE does not do so, DOE may find itself with only one viable PEM fuel cell stack in 2015—one that operates at very high current density, which introduces its own problems. Otherwise, Nuvera has made good progress toward overall project and DOE goals.
- Although the approach is questionable, Nuvera and its partners did a good job with the modeling. Many researchers know that the fuel cell active area is very sensitive to the fuel cell performance. Any fuel cell with a small active area has a different decay rate and performance than one with a large (i.e., > 100 cm²) active area. Using 1.9 cm² active area is dubious. Researchers can use small cells for each component development for comparison; however, for the integrated cell modeling and test, small cell is not enough.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.3** for its collaboration and coordination.

- This project features good collaboration with national laboratories.
- LBNL and Pennsylvania State University are excellent partners for Nuvera in this project.
- Nuvera chose strong and appropriate partners for this project.
- This year, Nuvera demonstrated that all of the partners are fully involved.
- Johnson Matthey has sufficed as material input to the project. LBNL is delivering an agglomerate electrode model, which is slated to be completed. It is unclear whether this has been incorporated into the existing cell-level models. Many of the modeling and single cell testing slides show both Penn State and University of Tennessee logos. Penn State appears to have delivered the beginnings of the two-dimensional+1 model. The University of Tennessee's contribution is unclear, but appears to be substantial.
- Stronger collaboration with automotive original equipment manufacturers (OEMs) would be beneficial.
- The roles in the collaboration are not clear.

Question 5: Proposed future work

This project was rated **3.1** for its proposed future work.

- The proposed future work is reasonable.
- Investigators will undertake material development and broader parametric model validation.
- It would be interesting to explore how the new material inputs (e.g., ionomer equivalent weight, electrode structure, and graded loadings) will affect modeling parameters. It would also be interesting to hear whether the modeling performed to date has driven the desire to change these material parameters. Continued fine-tuning of the model is an appropriate part of the future work. The future work should describe the eventual stack verification task. Moving from single cell to stack will introduce challenges related to flow sharing, stoichiometry, and thermal gradients. This reviewer wants to know if the model products can be adjusted to account for scaling to the stack module level.
- Investigators should give more details regarding how they will achieve the goals.
- The transport studies should be extended to 95°C. Investigators should prove the relevance of the values attained at 60°C.
- Nuvera, in pursuing high amperage operation as the only way to meet both DOE cost goals and Freedom Car efficiency in 2015, is sharply focused on its future work, which at this point is developing a new membrane and

model tuning and validation. This reviewer fails to see how the model is going to help the fuel cell community as a whole if high current amperage operation is the only solution.

- The method needs to be modified; therefore, the future work in the project is questionable without changing its approach. This is a good topic, but it is unclear why Nuvera, a good fuel cell developer, did not adequately consider the difference between the real scaled-up fuel cell and the tiny cell.

Project strengths:

- Lowering cost without sacrificing efficiency is important.
- This is a very goal-oriented project that has clear objectives.
- Strengths of this project include its overall good approach and good partners.
- This project has strong partners. It also has a very scientific and detailed approach that is performed with a strong focus.
- This project's strengths include Nuvera's open architecture form involving porous flow fields, high-current density operation (if such is required), development with partners of a new membranes permitting high-current density operation, and development of new two-dimensional+1 modeling. This approach, however, helps guide Nuvera's internal effort more so than the fuel cell community as a whole.
- The project's expansive breadth of cell designs is an area of strength. Because Nuvera can study open flow fields, the project can deliver a model that, in principle, should be flexible toward many possible cell designs. Another project strength is the attempt to provide a model product. One of the weaknesses of the water transport efforts in the DOE Program is the lack of model product delivery. This project aims to produce such a model, which should benefit stack OEMs and integrators. Lastly, the materials are meeting early go/no-go targets. The project involves some fairly aggressive performance and loading targets, so there is some self-regulation to ensure that they are using relevant, high-performing, low-loaded materials. Passing the go/no-go mark implies that the project is moving in this direction.

Project weaknesses:

- This project does not have any weaknesses worth mentioning.
- This reviewer discussed the project's weaknesses in the first question.
- The impact on stack durability is an area of weakness.
- The knowledge gained from the high current density work may not be useful to the majority of developers who are targeting higher voltages.
- Many other water transport projects have recognized that water transport is not by uniform displacement, but rather by capillary fingering, which can affect the flux of condensed water from the catalyst layer to the flow channels. Furthermore, condensation and evaporation of water must be accounted. The researchers need to explain how the project is addressing water transport mechanisms on the microscale. This project has another weakness related to the verification performed at the cell level, which may hide errors in important parameters. Sometimes models can match experimental data despite the fact that errors in certain parameters offset each other. The plot of low-current density electro-osmotic drag coefficients raises this concern.

Recommendations for additions/deletions to project scope:

- Investigators should consider adding a durability aspect because transport and durability are closely linked.
- The project team should include transport studies at 95°C.
- Investigators should discuss their approach with the U.S. DRIVE Fuel Cell Technical Team to determine if this approach is the only one justified. Operation at sustained high current amperage brings new sets of problems. If Nuvera has anticipated all of the new problems and still feels that only this solution will work, then it should continue.
- It may be interesting to consider the use of nanostructured thin film as another possible material variant. Investigators should expand the reporting of two-phase modeling methodology. It would be interesting to see how the model verification holds up from the perspective of voltage loss breakdown. In other words, if the model was expected to match the experimental data in terms of kinetic, ohmic, and mass transport losses, this reviewer wants to know how close the model would be.
- It is unclear if any fundamental results will be derived from the project that can be shared with the fuel cell community.

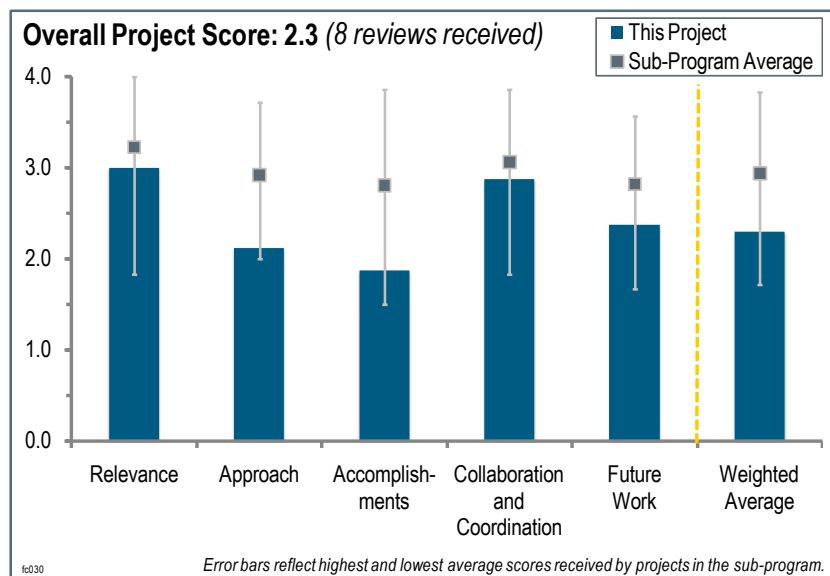
Project # FC-030: Water Transport in PEM Fuel Cells: Advanced Modeling, Material Selection, Testing, and Design Optimization

Vernon Cole; CFD Research Corporation

Brief Summary of Project:

The overall objectives of this project are to: (1) improve understanding of the effect of various cell component properties and structures on the gas and water transport in a polymer electrolyte membrane (PEM) fuel cells; (2) demonstrate improvements in water management in cells and short stacks; and (3) encapsulate the developed understanding in models and simulation tools for application to future systems.

Question 1: Relevance to overall U.S. Department of Energy objectives



This project was rated **3.0** for its relevance to U.S. Department of Energy (DOE) objectives.

- It is important to have a good understanding of two-phase flow in PEM fuel cell systems.
- Water transport is a key aspect of understanding performance, degradation, and freeze start/stop issues. Accurate models are critical to ultimately guiding research into improving the design of water transport in fuel cells.
- Water management is very important for optimum fuel cell performance.
- A better understanding and improvement of water management in stacks is critical to fuel cell commercialization. This project was ambitious and attempted to include a variety of factors in the models used to understand water management. Other stated goals were the examination of gas diffusion layer (GDL) materials and the suggestion of components and operational strategies. However, the wide variety of topics perhaps led to some dilution of effort.
- Transport is important and requires treatment of porous media, fluid mechanics in flow channels, surface energy, and membrane materials. This effort seems focused more on the porous materials and plate materials.
- The DOE Hydrogen and Fuel Cells Program needs advanced modeling to guide characterization and validation work regarding the complex physics of a fuel cell stack. However, at this point, a three-dimensional model is overkill because a consensus on one-dimensional physics in the membrane and electrode still does not exist.
- The project addresses thermal and water management. It is unclear whether the model and project will move the knowledge base forward.
- Although water transport is a very important part of the Program's research and development plan, it is unclear how this project (85% complete) has contributed to the Program's objectives. The modeling was supposed to lead to input on how to design better materials, but this was not evident from the presentation.

Question 2: Approach to performing the work

This project was rated **2.1** for its approach.

- The overall approach is good, but there is a lack of experimental data on the electrochemical performance. There is too much focus on flow in the channel. Although researchers presented models at various scales, there is a lack of multi-scaling and it seems that the approach is not truly as comprehensive as mentioned. Polarization curves indicate more severe limitations than expected with no real explanation.
- The approach of combining modeling and experimenting is a good one. The source of the disagreements between experimenting and modeling in this project is unclear. This reviewer wants to know how the pressure drop measurements were calibrated to determine membrane electrode assembly (MEA) water content. Other experimental approaches, such as neutron imaging, could have been used to determine the location of the problems.
- The measurement method of water in MEA seems inherently inaccurate and should not be used for model validation. Manifold design is the cause of the poor flow distribution in the channel/channel studies. Microporous layers (MPLs) should be included in GDL studies because it dominates two-phase transport resistance of GDLs.
- The overall goal of this project is: “Improved Water Management through Improved Component Designs and Operating Strategies.” Investigators indicated that they will use modeling, characterization, and design efforts to achieve this goal. Although this is appropriate in general terms, it seems that investigators are executing two distinct types of work (modeling and GDL testing) that have not yet been integrated. Researchers have explored a rather limited set of variables within each category.
- This project features a nice approach, in theory. Investigators use modeling that is verified by experimental data to design better materials that lead to improved performance. This reviewer is not sure how GDL aging (slide 14) ties into a water transport project. There are other durability projects looking into GDL aging in detail; it should not be a focus of this project.
- Validation has lagged throughout the entire project.
- This project is nowhere near as integrated or as accomplished as other transport projects. Investigators appear to be measuring parameters in isolation, without integrating them into the larger plan.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **1.9** for its accomplishments and progress.

- The project has certainly contributed to modeling and GDL studies. However at the 85% completion level, the outcome should not resemble a series of independent observations, but rather a nearly complete set of suggestions for flow field design, GDL selection for that flow field, etc., for certain specified conditions. Also, per slide 12, the models fail to capture the shape of the water profile or the water content. This may be common in the field, as stated by the speaker, but it remains a shortcoming. Such discrepancy could be overlooked if the project demonstrated some important trends that dictate fuel cell performance. It also seems that the commercial Toray paper continues to outperform the in-house developed materials in the latter section of the presentation. The slides do not clearly explain if the investigators have achieved any improvements for water management.
- This project should either generate or collaborate with someone to get water distribution data.
- The modeling predictions are far from reality. For example, in slide 13, even with the improved model, there is a huge discrepancy with the measured data. In fact, both models under predict the MEA water content significantly. This indicates that there is something fundamentally flawed with this model or the data. The reviewer wants to know how the model ties into Ballard's flow field designs and if they are being used to just verify the model or if the model is actually influencing flow field design. It seems like this project is looking only at the former (validation), while the latter (actual design improvements based on model results) is what has value to the Program.
- The model predictions of MEA water content are a factor of two lower than experimentally determined values. Considering the size of this project, the modeling results appear relatively sparse.
- After three years, CFD Research Corporation is still far from developing a validated channel model, and is unlikely to deliver one before project completion.

- It does not appear that the results will be useful for making predictions and optimizing designs of fuel cell stacks. The model does not even do well with more basic designs used for testing.
- This project lacks validation and quantitative agreement, considering it is almost finished. The model predictions are too far from the data and are far from state-of-the-art. It is not clear if the model has been useful in predicting designs, etc., or if it has been just trying to model what has already been experimentally tried. A lot of work needs to be done on model improvement, but the project is ending.
- To be this far along in the project and not have better agreement between the model and the experimental data is extremely disappointing.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.9** for its collaboration and coordination.

- There is strong collaboration with Ballard.
- The project features good collaborations, including with several industry partners who provide and characterize materials and provide model verification data, and one university partner who provides more fundamental characterization.
- There are a number of organizations involved in this project.
- The direct contributions of the partners is clear, but a coordinated effort to reach a single, organized goal of better fuel cell designs for water management is not as apparent.
- The project has good partners who are performing, but it is unclear how they interact with each other. The data transfer seems one-way and not collaborative. It is not clear whether the aging studies, etc. will be incorporated into the model.
- The project seems ad-hoc and not very well integrated.

Question 5: Proposed future work

This project was rated **2.4** for its proposed future work.

- The two upcoming milestones are very reasonable. Hopefully these can be achieved, especially the one on “assemble, test, and demonstrate improved self-humidified cell by October 2011.” Hopefully the model development can be validated better with experimental data.
- The project is 85% complete.
- This reviewer recommends including MPLs in both the cell and GDL ex-situ studies.
- The final milestone is a final version of the CFD code; however, it is not clear if the result will be useful. Investigators need to do more validation work using a much simpler fuel cell design before one can say that this model is finalized.
- The following upcoming milestones are ambitious, and will not be easy to achieve based on the results presented to date: (1) complete final model improvements and code package development by September 2011 and (2) assemble, test, and demonstrate an improved self-humidified cell by October 2011. The cell model and the materials and flow field selection seem to be behind schedule.
- This reviewer is skeptical that the work proposed can be completed to satisfactory levels by the completion of the project.
- The model predictions are not accurate, which is somewhat expected because the two-phase physics are still being debated. However, identifying the key relationships that cause the prediction to vary is more critical than pushing the model forward.
- The project is ending. The model dissemination needs more clarity regarding format and other issues. The model should borrow more from the work in the literature and needs better validation, including current distribution, etc.

Project strengths:

- The strong collaboration with Ballard is an area of strength.
- Combining sophisticated modeling and a strong industrial partner (Ballard) is the correct approach for understanding water transport.

- This project features good collaborations.
- The consideration of liquid water in the flow field channels is an area of strength for this project.
- A strength of this project is the good experimental water flow work at Ballard.
- This project has a comprehensive approach that was not perfectly enacted. It has made good progress, especially on the ex-situ experimental side.

Project weaknesses:

- The project has made minimal progress over the last year. Other areas of weakness include the project's inability to validate the model and the questionable experimental methods used for model validation.
- The project features poor agreement between the experiment and the model on the most important aspect—water transport.
- The project has not delivered a package of actionable suggestions for improved water management, and it is late on the timeline.
- The model predictions need to be better validated with experimental data. Maybe MEA water content is not the correct metric.
- The coordination of the various institutions and activities involved in this project seems a bit weak. The modeling activities also seem weak.
- The contact angles for GDL samples are not useful due to various surface and underlayer effects. The model is behind where it should be in terms of data validation and is not predictive. The initial sub-model work has been disregarded, especially in terms of properties and multi-scaling relationships.

Recommendations for additions/deletions to project scope:

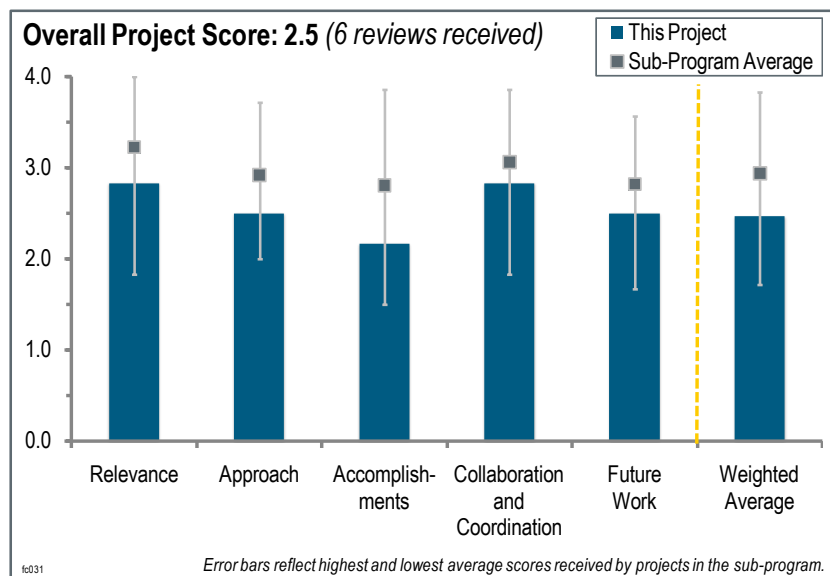
- Investigators should include MPLs in transport studies.
- The information gained to date should be transformed into concrete examples that can be understood by the fuel cell community at large.
- Researchers should consider water balance measurements and controlled variation of specific material properties to validate the model.
- The project team should not move on to transient simulations until the steady-state predictions are accurate.

Project # FC-031: Development and Demonstration of a New Generation, High Efficiency 10kW Stationary PEM Fuel Cell System

Durai Swamy; Intelligent Energy

Brief Summary of Project:

The overall objective of this project is to develop a high-efficiency 10 kW (kilowatt) polymer electrolyte membrane (PEM) fuel cell combined heat and power (CHP) system and demonstrate it in an International Partnership for the Hydrogen Economy country (United Kingdom [UK]). Project objectives for 2011 are to: (1) study the impact of operating stacks on 99% hydrogen as an approach to improving system level efficiency; (2) build and test an integrated system with multiple heat recovery streams to demonstrate greater than 70% efficiency; and (3) perform a real-world conditions field demonstration with system health monitoring to demonstrate 40,000-hours durability.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **2.8** for its relevance to U.S. Department of Energy (DOE) objectives.

- The project objectives of attempting to address efficiency, durability, cost, and start-up time of CHP systems are aligned well with DOE objectives.
- PEM CHP system design and deployment are relevant to DOE goals and objectives.
- As demonstrated in Japan, CHP systems have a large market potential that can lead to deployment of a significant volume of systems. This, in turn, promotes technology innovation and attracts volume production manufacturers. Cost reduction and improved efficiency are key components of producing a viable product.
- High-efficiency, long-durability, stationary distributed fuel cell systems are important to DOE objectives.
- This project is relevant to the DOE Hydrogen and Fuel Cell Program's stationary fuel cell objectives. It is integrating a different type of reformer to increase system efficiency.
- Although this work will result in a small PEM fuel cell-based system that can operate on reformat, it is not clear how it advances DOE goals. The primary barriers are cost and durability. A system that includes a pressure-swing adsorption (PSA) is certainly not a low-cost approach, nor does a PEM fuel cell operating on almost pure H₂ provide any advances in durability. The goal of this project is unclear. If one simply wants examples of CHP PEM fuel cell systems, then Japan's New Energy and Industrial Technology Development Organization project can provide ample data (i.e., greater than 20,000 hours operating on natural gas reformat).

Question 2: Approach to performing the work

This project was rated **2.5** for its approach.

- This project features significant industry involvement from utilities that can introduce this technology to very large markets. It also has an interesting trade-off on H₂ purity with the focus on reducing cost, good use of academia and national laboratory resources, and an effective project plan.
- This project investigates a natural gas reforming system that produces H₂ for use in a 10-kW fuel cell stationary power system. The project features a prototype followed by a demonstration unit.

- The project involves designing the system so that a drop in replacement fuel reformer can be substituted for the conventional steam reformer or PSA fuel processor. It is reasonable to use a more conventional fuel processor subsystem to fulfill the contract objectives. Once the milestone is completed, the absorption enhanced reformer subsystem will replace the steam methane reforming (SMR) or PSA system. Designing a system with flexibility will likely entail compromises that will make the design less efficient than if it was designed specifically for the absorption-enhanced reformer.
- The current approach is much simpler than the planned approach because of technical difficulties. The new approach is not novel and, thereby, provides a smaller benefit to DOE.
- It does not appear that this project has made significant progress on any of the barriers it is intending to address, particularly compared to systems that have been built before. The future work proposed may have some impact on the cost and efficiency of the reformer portion of the system, doing little to address deficiencies elsewhere.
- If investigators had chosen some path other than PSA, then more knowledge may have resulted from this project.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.2** for its accomplishments and progress.

- The project features a very impressive demonstration unit and good results in reducing package costs. The automated control system is impressive. Investigators have achieved the majority of their targets and have made a good pathway to achieving those in which they have come up slightly short.
- Intelligent Energy has reduced the size of the system by about 30% according to the presentation. The individual components of the system—the combustor, steam-methane reformer, PSA, and fuel cell—have been tested. The absorption medium has been selected and tested. The absorbent is commercially available. The project is apparently on schedule for demonstration in the UK this summer. Conformité Européenne (European Conformity) certification is in sight, although the impact of changing the reformer system is not known. There does not appear to be enough time left in the project to actually implement the absorption-enhanced reformer approach. The efficiency of the demonstration unit is not projected to meet the 40% overall electrical generation efficiency target. The path to meeting the efficiency target by means of the absorption-enhanced reformer is not clear and is not supported with data.
- This project claims to make progress on the DOE barriers of 40% lower heating value electrical efficiency, 85% total thermal efficiency, 30,000-hour durability, \$650/kW, and 45-minute start-up time. The project thus far has demonstrated 33% electrical efficiency, 61% thermal efficiency, 6,100 hours on the reformer, and 730 hours on the fuel cell stack (not without failures), start-up times exceeding one hour, and undisclosed costs. In order to boost efficiency and match the voltage requirements of the commercial off-the-shelf power conversion devices, investigators deployed two stacks, which are running at very low current density. This approach will increase capital cost. Much of the capacity of the fuel cell stacks is unutilized. Much of what was demonstrated appeared to be laboratory-scale disaggregated subsystems with very little operation as an integrated system. The project appears to be far from commercial maturity. Others have previously demonstrated CHP systems with better performance in several areas (e.g., efficiency and durability). There appears to be very little innovative, new, or groundbreaking work here.
- The field demonstration system has been developed a little late in the game, as the project is almost over. Thereby, the durability of the demonstration system will not be demonstrated to the 40,000-hour target.
- The original technical goals and objectives were not achieved, resulting in a backup plan of marginal benefit.
- Researchers are making progress, but the end result will be the same as for other fuel cell-based CHP solutions, which are namely too complex, big, and expensive.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.8** for its collaboration and coordination.

- The collaboration features a good mix of academia, national laboratories, and industry. Having a major utility partner is a major contribution to achieving the project objectives.
- The project makes use of relationships with universities and other governmental organizations outside of the United States. University collaborations appear to be crucial to project execution.

- It looks like the project features good collaboration with the International Partnership for the Hydrogen Economy.
- Collaborations cover most aspects of the project. It would be instructive to learn more about the stack design and materials.
- Collaboration with entities in the UK/Ireland market is evident. There does not appear to be any activity with the aim to expand the market to the United States. The lone U.S. collaborator is California Polytechnic State University, which is only training students who might have an interest in fuel cell technology.
- It is not clear if there is really much collaboration among the partners, especially those in the United States.

Question 5: Proposed future work

This project was rated **2.5** for its proposed future work.

- This is a well planned and executed project. The project is close to completion and there are no obvious areas that need to be changed.
- This project is 82% complete. Hopefully, a suitable field demonstration of the technology can be conducted.
- The project is scheduled to end in August 2011—just enough time to demonstrate the current system, which is not projected to meet the efficiency target. There will not be time or, presumably, resources left to complete the absorption-enhanced reformer development.
- The proposed future work focuses solely on the reformer section of the system. If realized, the claimed improvements in efficiency from the reformer will make progress toward DOE barriers. However, work is also needed on the stack, power electronics, overall system efficiency, and cost. There will be no magic bullet here; the system needs to be improved as a whole on many levels.
- A six-month system demonstration, which is planned to complete the project in August 2011, is of marginal benefit.
- The project is essentially complete. It is not obvious why this work should continue with DOE support.

Project strengths:

- This project features good collaboration with others.
- This project is well planned and executed. Strengths include the close involvement of a major utility and a good technology transfer plan.
- A complete system demonstration can showcase the true status of the technology.
- The strength of this project is the demonstration of a stationary distributed power generation PEM fuel cell system with the H₂ generated from natural gas sources.
- The project is focused on achieving the DOE target regarding efficiency for small CHP systems.

Project weaknesses:

- Without a follow-on project, it is unlikely that the 40,000-hour durability of the demonstration system can be demonstrated.
- Investigators presented limited durability data, and what was presented appears to be on individual system components and not an integrated system.
- This project lacks real commercialization focus. This system is many years from commercial viability. Without commercial viability, any technology developed in this project will not make real progress toward addressing DOE barriers. It is difficult to see where real, measurable progress has been made toward DOE objectives. The future work only addresses some of the above weaknesses, and will not be sufficient.
- This project is doing the same thing over again while expecting a different result (i.e., it is a fuel cell based CHP system with no real technology breakthroughs).

Recommendations for additions/deletions to project scope:

- The project team should focus on stack efficiency, power conversion, and system cost.
- DOE should not fund system demonstrations unless they incorporate some truly new technologies that can significantly address the key barriers in a commercially viable manner.

Project # FC-032: Development of a Low Cost 3-10kW Tubular Solid Oxide Fuel Cell Power System

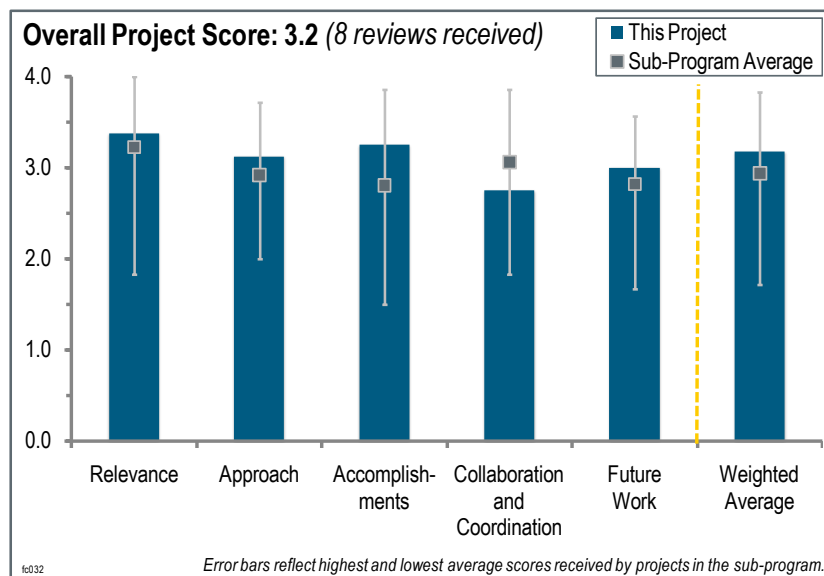
Norman Bessette; Acumentrics Corporation

Brief Summary of Project:

The objectives for this project are to: (1) improve cell power and stability; (2) reduce the cost of cell manufacturing; (3) increase stack and system efficiency; and (4) integrate the system in remote power and micro combined heat and power (micro-CHP) platforms to allow short-, medium-, and long-term market penetrations.

Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.4** for its relevance to U.S. Department of Energy (DOE) objectives.



- This project makes progress on stack power, cell power, cost, efficiency, and lifetime. Overcoming all of these barriers is important for the adoption of this technology.
- The project seems to be making more progress with a renewed focus on field demonstrations and showing progress on efficiency.
- This project fully supports the DOE Hydrogen and Fuel Cell Program's objectives.
- Acumentrics is lowering the cost of its tubular solid oxide fuel cell (SOFC) system for remote and micro-CHP applications. The project is relevant to the Program's stationary fuel cell objectives.
- The project is based on high-temperature SOFCs. It is mainly targeted for small-scale stationary applications and is showing steady progress.
- The project is advancing deployment of the technology focusing on: prototype testing to meet system efficiency and stability goals (3–10 kW [kilowatts]) and developing remote power and micro-CHP platforms to allow short- and longer-term market penetrations
- Work on SOFCs continues to be vital to the DOE objectives in transforming a critical energy conversion device into a viable market. SOFCs hold great promise because of the variety of materials that can be used to build cells and stacks, and the great abundance of these materials relative to some of the materials used in other types of fuel cells.

Question 2: Approach to performing the work

This project was rated **3.1** for its approach.

- Last year's future work plans were executed as planned and generated significant results. The future work plans continue on the roadmap toward commercial viability.
- The approach to reducing the cost of these systems appears sound. The project is concentrating on decreasing the processing cost for making the individual tubes and fabricating the recuperator. Increasing the power density of the cells can reduce the size and cost of the units, but at the expense of increased thermal management issues. Seeking automated manufacturing solutions also moves in the direction of decreased costs. This reviewer wants to know if a sponsor other than DOE has been identified to provide the needed development funds to further commercialization efforts.

- The approach is focused on the appropriate priorities (i.e., increased stability and reduced cost). The emphasis on optimizing system integration is also appropriate. Replacing welding with brazing appears to be a good step forward in terms of cost reduction. Also, it appears that investigators have made progress in terms of reducing the cost and complexity of the cathode current collectors.
- Acumentrics has made good progress in the development of its technology. Tubular SOFCs are attractive because of their resilience to thermal and mechanical shock, and because they are easy to manifold and seal. For instance, the replacement of a bad or broken cell is essential and can be done with Acumentrics' design. However, the interconnection between the tubes is a major issue that limits power and is a cost factor, so the use of a different alloy that eliminates Ag must be the greatest priority. The power density is modest, but not a problem. The stability of the cells and stacks is excellent.
- This project represents a good combination of field demonstration learning and modeling. This reviewer would like to see a little more definition of what has been learned from the field demonstrations. The researchers should also define efficiency targets to clarify why 35%–40% is adequate.
- Acumentrics' integrated research and development approach is focused on the four most important areas of research, development, and demonstration. In terms of cell technology, the project seeks to improve the power and stability of the cell building block. With cell manufacturing, the project aims to improve processing yield and productivity while decreasing material consumption. Regarding stack technology, investigators are trying to refine stack assembly and improve heat removal and integrity while reducing the cost of components. Finally, for system performance, the project seeks to develop simplified controls and balance of plant (BOP) to allow for a reliable and highly efficient unit.
- The approaches to improving cell performance and reducing manufacturing costs are logical. However, critical analyses and studies on performance losses and manufacturing cost breakdowns are needed to guide development paths and focus.
- While there has been steady progress over the years, specific cost reduction was not discussed. For example, this reviewer wants to know what the cost reduction is with the co-sintered approach. Also, it appears that the stable system operation of greater than 10,000 hours is on a 200 W (watt) system/stack (power 0.2 kW, slide 23) and not a 3–10 kW system, which is the project's objective.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- This project has made significant progress on a number of the barriers that it intends to address, including power density, efficiency, and cost. The primary remaining work is to further reduce costs through system design and manufacturing. More information on reliability issues is desired, such as what they are and how they will be resolved.
- The systems development and applications have been well considered and are moving forward very quickly. The micro-CHP appears especially well designed and compact. The small volume may even become interesting to American consumers, as it probably already is with the Europeans. The tubular design lends itself to ruggedness in design.
- Accomplishments include good progress in field demonstrations and increases in efficiency.
- The project team has made significant progress in demonstrating systems for remote power.
- The increases in power density have been very significant. The increased power density has not come at the expense of decreased durability. Commissioning a high-temperature furnace has increased the throughput by a factor of four. Investigators have reduced the time for fabrication of one tube by a factor of two by co-sintering the base tube and the spray applied electrolyte layer. The co-sintered tubes show only a 2% performance deficit compared with the current dip coating sintering process. Moving the recuperator design and fabrication in-house has resulted in significant cost savings compared with purchasing the component. In total, the accomplishments have been good.
- Working on multiple areas to lower cost and improve reliability is an area of strength. The progress on power density improvement in single cells is impressive.
- Progress has been made in the following areas: (1) in manufacturing, where progress includes a four-fold increase in furnace throughput, with a reduction in firing times and the energy requirement by cell; (2) the recuperator, where the project team lowered the operating temperature allowing for lower-cost raw materials, lowered the required effectiveness through better thermal balancing, and simplified the design and

manufacturing process; (3) partial oxidation, where a 100% increase in power per stack was demonstrated while maintaining thermal balances; (4) increased efficiency, from 30%–39% to 40%–49% (4,000–5,000 hour runs); (5) achieved 615 mW/cm² (milliwatts/centimeter squared) peak power density; (6) increased cell stability as current increased; (7) achieved co-sintering of the electrolyte and the green tube; and (8) conducted 11,000 hour durability testing and many in-field tests.

- It is good to see the improvements in efficiency and power density over the past year. It would have been nice to see more details regarding current and projected manufacturing yields. It was noted that some field units experienced downtime due to BOP issues, and it was not clear how those issues were being addressed (for example, it was unclear if more expensive components will be required to ensure reliable performance).

Question 4: Collaboration and coordination with other institutions

This project was rated **2.8** for its collaboration and coordination.

- Acumentrics works with a wide variety of collaborators, including government, military contractors, universities, and three energy companies. The investigators should continue to expand this network to include more energy providers, especially natural gas, propane, and biofuel provider companies. The rugged design of the Acumentrics SOFC system should lend itself to excellent fuel flexibility.
- The project would benefit from additional relationships with backup and remote power companies.
- Acumentrics is partnered with the Italian government program that was granted to Ariston Thermo Group and 14 other partners. Acumentrics is the first foreign company to be issued an Italian government grant for a green energy program.
- Acumentrics has not built a broad coalition of partners for this project; however, investigators have mentioned work with the Italian government and the Office of Naval Research.
- The collaboration with strategic partners was indicated, but not explicitly mentioned in the talk.
- This project does not feature any major collaborations.

Question 5: Proposed future work

This project was rated **3.0** for its proposed future work.

- The future work seems appropriately focused. This reviewer wonders whether the proposed work on liquid fuels may be a distraction that will slow down the excellent progress that has been made thus far. It might be better to continue to improve the existing product rather than broaden the fuels of the existing one, unless liquid fueled military applications are a critical first market. Lifetime and cost need to be dramatically improved to hit the micro-CHP market. This will require additional effort in design, manufacturing engineering, and building early volume.
- The proposed future work has a good focus on continued learning in field demonstrations. The investigators should focus a bit more on root cause analysis from fielded units.
- The future work is logical and effectively planned.
- The future work appears to be appropriately planned. It is good to see the emphasis on increased reliability and reduced cost, and it was helpful to be provided with some specific examples of how the investigators are pursuing cost reductions.
- The investigators have identified major barriers, and the work is focused on removing those barriers. Also, the proposed future work includes significant effort in field testing of demonstration units that should provide valuable testing experience.
- Acumentrics is doing the correct things to proceed with commercialization. The proposed future work includes: ensuring cell stability by continuing to test at the 250–400 milliamp(s) per cm² current density; further demonstrating stability over thermal cycles through cell and stack testing; continuing cost reductions on each product platform; reducing generator and BOP costs to levels allowable for remote power products; moving from field testing of first-market products to second-market products; continuing to build on the success of remote power units and accept commercial orders; and field demonstrating liquid-fueled military units in the 1–3 kW range.
- No commercialization timetable was provided for the future work. The first priority should go to the stack, and the first priority of the stack should be to develop interconnections, which was not listed in the proposed future

work. This reviewer wonders if this is a major cost factor both in materials and in manufacturing. Otherwise, the focus on systems BOP cost reduction and manufacturing automation is well emphasized.

Project strengths:

- The project delivered on its future work roadmap, and has units in the field gaining valuable reliability data.
- The project's SOFC design benefits the materials selection, general availability, and cost. DOE has invested well by supporting this technology. The project's tubular SOFCs are rugged and easy to manifold, and their tube manufacture and power density have improved to an acceptable point. The system appears to be well designed and compact. The lifetimes of the cells, stacks, and systems have shown strong performance.
- Strengths of the project include its good progress, interesting market, and focus on deployment.
- The remote power system demonstration is the project's strength.
- The project appears to be making headway in reducing the cost of the system.
- Strengths of this project include how it is results-oriented with a commercialization focus, has identified barriers and is working toward resolving the issues, has a good field-test plan, and is focused on multiple applications.

Project weaknesses:

- The cost and lifetime are weaknesses of this project.
- One weakness is the limited partnerships—the company will likely need more to be successful.
- Studies and analyses are needed to identify key performance losses to guide performance improvement and development efforts.
- Specific project objectives were not mentioned. The project objectives are generally stated as reducing costs and increasing durability. The answers to questions on how far away the current system really is from a competitive cost were evasive and uninformative. Without a list of specific quantified project objectives, it is not possible to determine the chances for success for this project. The project was scheduled finish at the end of this fiscal year, but the final deliverable to DOE was not described.
- The cost target was not mentioned, system degradation was not shown, and investigators have not mentioned factors affecting degradation.
- A stack life of only three years is projected at this time.

Recommendations for additions/deletions to project scope:

- The investigators should remove the liquid fuel scope unless it is absolutely critical to early market opportunities.
- It would be helpful to Acumentrics and other SOFC developers if a consortium is developed to further develop the BOP in the hot box.
- The investigators should focus more on the root cause analysis of field units.
- The project team should provide details on specific project objectives and a sense of the prospects for commercial success.
- The project's headway in reducing the cost of the system needs to be placed in the context of what is really needed for commercialization.
- Film coating versus wire winding for the current collection may be an important development and should be explored by investigators.
- The investigators need to continue working on the interconnect material.

Project # FC-036: Dimensionally Stable Membranes

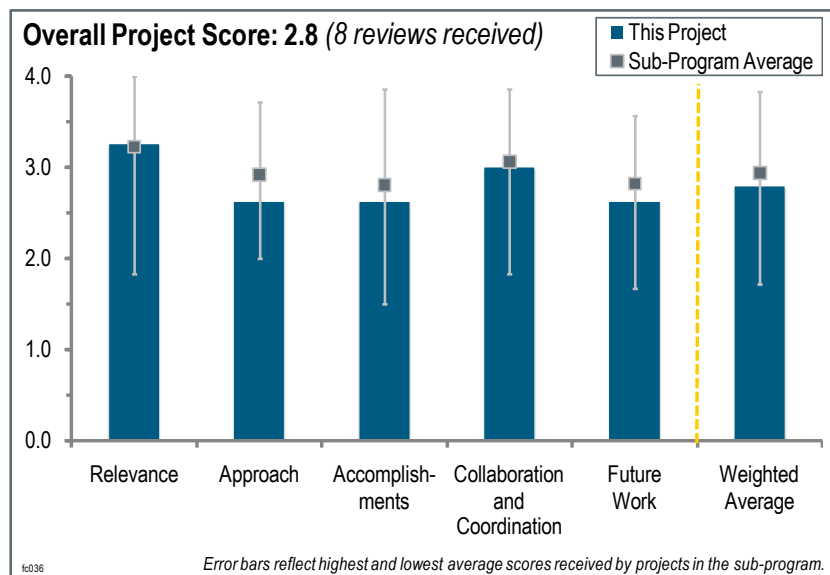
Cortney Mittelsteadt; Giner Electrochemical Systems, LLC

Brief Summary of Project:

The ultimate goal of this project is to meet U.S. Department of Energy (DOE) performance targets with a membrane film that can be generated in a roll at DOE cost targets. Project objectives are to: (1) determine the effect of pore size and substrate thickness and demonstrate polymerization of the perfluorosulfonic acid (PFSA), (2) achieve 0.07 S/cm

(siemens/centimeter) at 80% relative humidity at room temperature, (3) demonstrate membrane conductivity greater than 0.1 S/cm at 25% relative humidity at 120°C using non-Nafion® materials, (4) demonstrate

the ability to generate these materials in quantities suitable for automotive stacks, (5) build short stacks with optimized materials and demonstrate its durability, and (6) demonstrate how these materials can be produced to meet DOE cost targets.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.3** for its relevance to U.S. Department of Energy (DOE) objectives.

- The project is relevant to the objectives and research and development (R&D) targets of the DOE Hydrogen and Fuel Cells Program. The initial activities were very much aligned with the Program's goals. The development of a low relative humidity and dimensionally stable membrane is critical to the success of DOE's hydrogen research initiatives.
- Membranes with improved conductivity and durability are needed to meet DOE targets.
- The development of new membranes that have the potential to operate under hotter and especially drier fuel cell operating conditions is essential toward the goals of fuel cell commercialization.
- Producing membranes with high conductivity at low relative humidity with reduced cost is a key DOE goal, and there is a clear economic rationale for doing so.
- This project features good relevance to DOE performance and cost goals. The project has maintained focus on the goals despite some setbacks, and the principal investigator (PI) has shown versatility in adapting the project appropriately to keep the project's relevance strong.
- This project proposes to build a superior ion exchange membrane that could work in ways that would enhance fuel cell performance. It addresses the targets of cost and durability.
- High temperature membranes are still needed.
- Dimensionally stable membranes have the potential to improve fuel cell durability, particularly at elevated temperatures. The membrane may improve fuel cell catalyst layer and membrane interface, and minimize hydrogen crossover.

Question 2: Approach to performing the work

This project was rated **2.6** for its approach.

- The approach of making two- and three-dimensionally stable membranes (2-DSM and 3-DSM) is reasonable for meeting the Program objectives. Examining triflic acid and benzene sulfonic acid (BSA) conductivity data is good to understand the feasibility of achieving the DOE low relative humidity membrane target using presently available sulfonic acid-based ionomeric membranes. The data in slides 8–10 is very convincing. From this data, it is clear that it will be difficult to achieve the DOE target in a polymeric membrane, as neat BSA just meets the target. Adding any additional atoms to the BSA for preparing a polymeric form membrane will certainly result in a conductivity penalty.
- A very low equivalent weight (EW) material in the right support may meet the area-specific resistance target, but durability issues—such as ionomer solubility—should be addressed. The very low PFSA was described as an “oligomer.”
- It is not clear if this approach can meet DOE goals, but the path taken—low EW ionomers in a supportive network—needs to be investigated for feasibility.
- The approach is mechanical with essentially known materials or variations of known materials. Therefore, it is not especially innovative, but it does allow the focus on the DOE goals to be maintained. The variety of supports that could be examined is limited and the project’s continuation might consider a wider variety of materials. Mechanical measurements are lacking, which is surprising because the goal is to provide roll-to-roll manufacturing.
- The general thrust is apparent, find a lower EW poly PSFA polymer that will have higher conductivity and the ability to operate at a higher temperature. However, the identified polymers tend to be water soluble, so the approach is to add “reinforcement” to stop the dissolution process. The intent is clear, but the approaches do not seem sensible—soluble things dissolve.
- This approach, which seemed promising based on early work, seems to have hit a wall. It is not clear whether the targets can be met with even the three-dimensional approach. In general, this approach does not address the underlying difficulties of achieving high temperature conductivity. Rather, it provides a platform to stabilize someone else’s solution.
- Giner Electrochemical Systems, LLC relies on commercially available supports despite some issues, such as non-uniformity, incompatibility with ionomer solution, undesired thickness, and high conductivity penalty. Giner has not yet answered if those issues can be fixed through its approach.
- The approach of stabilizing a PFSA in a support is not new and this project does not offer anything particularly novel, but the project team has world-leading characterization abilities.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.6** for its accomplishments and progress.

- Progress has been very good. The investigators have covered a lot of ground and adopted appropriate responses to setbacks.
- The team has made good progress in the development of two- and three-dimensionally stable membranes. The team’s strategy seems to be to lower the EW of the ionomer to achieve low relative humidity and high temperature conductivity. The team is evaluating low EW ionomers from 3M (660 EW) and the State University of New York College of Environmental Science and Forestry (SUNY-ESF). The project hinges on the use of low EW ionomers in a thin reinforced membrane matrix. To ensure the stability of ionomers in such a configuration, the team should also consider evaluating the dissolution behavior of low EW ionomers during humidity cycling. Under a fully humidified condition, these low EW ionomers and homopolymers tend to dissolve and leach out of the reinforcement matrix. Therefore, the team should also test these membranes under automotive humidity cycling conditions. The 660 EW PFSA ionomer from 3M may not be stable under relative humidity cycling conditions. The team needs to think about an alternative strategy, such as incorporating inorganic particles to achieve low relative humidity conductivity while maintaining the stability of the ionomer under humidity cycling conditions.
- This is a nice, systematic study. Showing data addressing the chemical stability of the support is crucial.

- The investigators have achieved chemical stability gains per slide 22. However, the inability to sufficiently thin the membranes compared to Nafion 211 means that the materials perform similarly to Nafion 211 in terms of conductivity at 120°C, H₂ crossover, and cell performance (slide 21). At this point, the project team has indicated, but not entirely achieved, what may be possible. The project is nearing completion, so some experiments that clearly demonstrate the advantages of the new materials would be desirable. For example, if membrane electrode assembly (MEA) testing could demonstrate the most promising membranes' similar performance along with greater stability, this would be valuable information for the fuel cell community. Also, given the importance of "dimensional stability" to the overall concept, some data reinforcing that it has been actually achieved versus Nafion 211 would be welcome, at least in the supplemental slides.
- It seems that the new work reported this year took a step backwards; all the films seem to have lower conductivities under the conditions prescribed by DOE. The team is benchmarking against Nafion 211, which may be of higher EW and thicker than the current state-of-the-art. The suppression of fluoride release rate is impressive.
- This project is 90% complete and the progress to date seems marginal. Test data on the produced test articles was generated at the Florida Solar Energy Center. In the end, a polymer very much like the "Dow polymer" was perhaps the best candidate, which is the material used in the majority of commercial PEM fuel cell stacks. The PI claimed that the developed membranes will have "commercial applications in the GES [Giner Electrochemical Systems] electrolyzers," which is not exactly the right target, as this project is funded for fuel cell progress.
- This project got close to its targets, but will not likely meet them with the remaining resources.
- There are no expected reductions in H₂ crossover and related increases in fuel cell performance. The reduction in resistance does not contribute to increases in fuel cell power density. The investigators also need to do more durability evaluation.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.0** for its collaboration and coordination.

- The close interaction with multiple partners is apparent, including materials exchange and testing.
- The close interaction with SUNY-ESF seems to be an ideal collaboration, bringing strong, synthetic capabilities outside the ring of the "usual suspects."
- The team consists of good collaborators, but collaboration with national laboratories may benefit the team. The team should also try to implement some of the testing protocols developed by its industrial partner General Motors (GM) to test newly developed 2-DSM and 3-DSM membranes.
- The project features good collaboration with SUNY-ESF, but GM's role is not clear.
- The project has good collaboration with university and original equipment manufacturer (OEM) partners, but the project team really should be talking with an MEA manufacturer as well as an OEM.
- The investigators seem to be collaborating with their own team members and other suppliers such as 3M. Collaboration does not seem to be strongly emphasized.
- The collaborations, as structured, resembled live vendor interactions. Collaborators are those who are assigned important technical roles.
- One collaborator contributes new ionomers and test results. However, the intrinsic challenges listed in the approach section have not been appropriately addressed by selecting partners or collaborators.

Question 5: Proposed future work

This project was rated **2.6** for its proposed future work.

- Given the remaining time and funding, the proposed future work is appropriate.
- The extension of the project to demonstrate the cycling goal for 12–15 micron film is reasonable. However, given the challenges of making thin film, demonstrating this goal by the end of October 2011 seems challenging.
- The team will push the concept to as thin as possible with as low an EW as possible, but there is insufficient information to determine whether this approach will work. It may not be worth using project resources to demonstrate cycling if DOE conductivity or area-specific resistance (ASR) targets cannot be met.

- The proposed future work is not very likely to meet DOE targets; however, it is very likely to provide improved membranes and materials insights. The presenter provided very little information on how the thin membranes might be achieved.
- The future plans are perfunctory. This is disappointing because the project has made considerable progress and it would be good to know how this would be developed and what kinds of issues could be solved by further R&D.
- Giner wants to utilize a lower EW ionomer in thinner supports. However, there is no solid plan to fix the challenges, such as completely filling the ionomer in the support and making them uniformly.
- There were no clear descriptions of what would occur should this project be extended. As for now, it appears as if work has stopped.

Project strengths:

- The team has access to automobile test protocols and real-life drive protocols at its partner's (GM) test laboratory. The team also has individuals with solid understanding of the field and related challenges in such membrane development work.
- The systematic study of these materials provides a valuable data set for future research.
- The project is tightly focused on improving the mechanical properties of thin membranes using two-dimensional and three-dimensional scaffolds in order to address cost and durability issues simultaneously. It is clear that the investigators have explored a wide variety of low EW ionomers and scaffolds, and have overcome many technical problems with materials compatibility during this project. In the end, they have prepared well-functioning membranes with comparable properties to Nafion 211 and perhaps greater chemical stability.
- This project features excellent focus on DOE cost and performance goals. The project team has been sufficiently versatile in its response to setbacks in terms of overcoming them and making progress.
- This is an interesting approach that the investigators have taken probably as far as it can go. The project has good collaboration.
- Giner may understand what it needs to improve its fuel cell or electrolyzer performance.
- The concept is good, but the novelty is in doubt.

Project weaknesses:

- The team is exploring the avenue of low EW ionomers, which is a common strategy that most researchers are pursuing. The team should think of an alternative strategy to circumvent the traditional approach to low relative humidity membrane conductivity.
- The investigators should look at durability more broadly and include the chemical stability of the support and stability toward hot water.
- The chemistry being employed in the PFSA development is not being divulged, so nothing can be said about it.
- This reviewer desires a more thorough evaluation of the properties of the most promising materials in MEAs compared to Nafion 211, given the late point in the timeline of this project. Proof of the dimensional stability and the benefits of that stability in MEAs should be obtained. Some relative humidity cycling results were presented in 2010 showing that Nafion 211 starts to show slight instability after 4,500 relative humidity cycles, whereas one version of the three-dimensional membrane shows no degradation up to 5,000 relative humidity cycles. Without statistics and longer testing, however, this comparison is not complete.
- Innovation is not a strong point of this project. Others, such as researchers in Japan, have reported similar systems with good results. The mechanical properties should be reported.
- The project started on April 3, 2006. Very little was accomplished. For example, the project plan called for a task of building a short stack with the best new membrane. There has been no membrane or stack.
- This project should address issues at a more fundamental level.
- Giner has not showed how to overcome the challenges.

Recommendations for additions/deletions to project scope:

- Investigators should look at durability more broadly and include the chemical stability of the support and stability toward hot water.

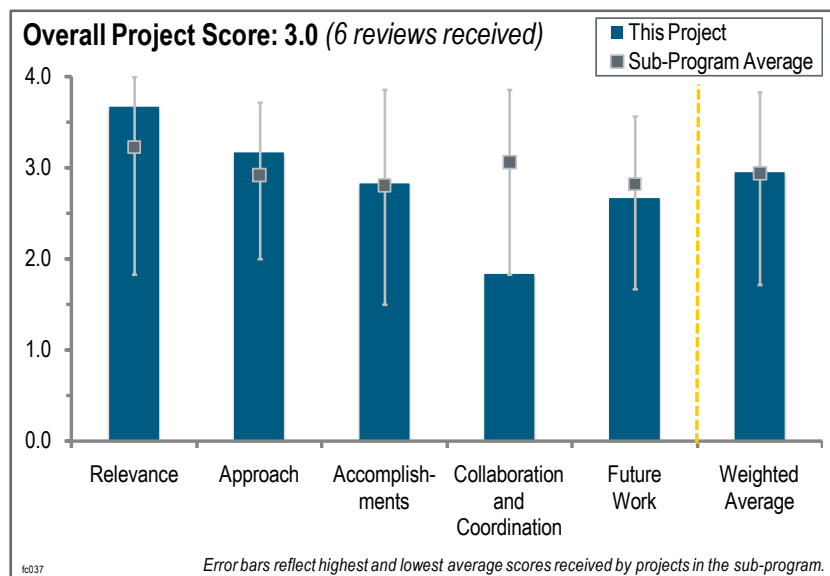
- Investigators should concentrate on achieving DOE conductivity or ASR targets.
- The project team should add MEA testing with a focus on mechanical stability so that the value of the materials produced in this project will be clear to the fuel cell community at large. This would make the adoption of such materials across the community much more likely, which would be a desirable outcome for DOE investment.
- Investigators should submit new proposals that would investigate issues such as delamination from the support, more detailed chemical and mechanical measurements, and how researchers can make MEA electrodes with this material.
- A membrane manufacturer may have a better chance to make a desired membrane if the approach is to use commercially available membrane support and get an ionomer from collaborators.

Project # FC-037: Rigid Rod Polyelectrolytes: Effect on Physical Properties; Frozen-In Free Volume: High Conductivity at Low Relative Humidity

Morton Litt; Case Western Reserve University

Brief Summary of Project:

The project's objectives are to: (1) synthesize polyelectrolytes that reach or exceed the U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program's low humidity conductivity requirements, (2) use materials and synthetic methods that could lead to cheap polymer electrolyte membranes (PEMs), (3) understand structure-property relationships in order to improve properties, and (4) develop methods to make these materials water insoluble and dimensionally stable with good mechanical properties.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.7** for its relevance to DOE objectives.

- Developing membranes with high conductivity at low relative humidity and high temperatures will enable simpler and less expensive fuel cell systems.
- Membranes with improved conductivity and durability are needed to meet DOE targets.
- This project is critical to the Program because it fully supports the fuel cell research, development, and demonstration objectives of enabling practical fuel cells through the development of PEMs that require no external humidification for operation.
- The project is very relevant to Program goals and directly addresses the performance goals. The project provides not only results, but also soundly based rationales as to why the results are achieved—thereby providing fertile ground for others to build upon. This reviewer's only complaint is that the rationale for changing some of the monomers is a bit shaky.
- Improved membranes are critical to fuel cell stack performance, life, and cost.
- The project is well aligned with the DOE objective of developing membranes that have adequate performance (especially conductivity) at low relative humidity.

Question 2: Approach to performing the work

This project was rated **3.2** for its approach.

- This ambitious, novel approach has provided excellent conductivity. The current work to improve mechanical properties is the appropriate next step. Cross-linking may prevent dissolution in water, but it might not help with brittleness and poor mechanical properties.
- This project is sharply focused on the objectives of producing a PEM that conducts under hotter and drier conditions with excellent mechanical conductivity and at a low cost. The polymer chemistry is world-leading.
- This project features an excellent approach, given the limitations of the funding.
- This is an interesting approach that holds promise to work at elevated temperatures.
- The project addresses a couple of key barriers, and the approach to the synthesis of the materials is sound.

- Professor Litt is focusing on improving the mechanical stability of his PEMs, which is good. However, his grafting and cross-linking approaches have not been successful.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.8** for its accomplishments and progress.

- The current cross-linking approach has the potential to provide stable cross-links that can help prevent dissolution in water. Researchers need to show if these materials can have suitable mechanical stability for fuel cell use.
- The progress toward a dimensionally stabilized PEM with the group's chemistry is ongoing; each year investigators have made progress, but the perfect solution appeared to be elusive until this point. It is not clear why the membrane electrode assembly (MEA) with the crack was patched and run. It would have been better to have made another and marked the corner with a permanent marker rather than a notch—that did show that the materials are still too brittle.
- The progress is outstanding, given the resource limitations. Achieving the goals is very impressive. Some more extensive characterization would be useful, which is where the project does not do so well.
- The project team achieved good conductivity at 120°C and low relative humidity. The team needs to improve MEA performance and durability at 120°C and low relative humidity, as well as membrane electrical resistance.
- The project made progress on the collection of the mechanical and MEA testing data, and helped to clarify the potential of and problems with the polyelectrolytes. The grafting chemistry has helped with the swelling and stability of the polyelectrolytes in water and high relative humidity environments. However, the materials still have poor mechanical properties (specifically low elongation) and the improvements to the molecular weight and grafting chemistry have done little to advance these properties. This is critical to this technology being a viable membrane technology for PEM fuel cells.
- Investigators have demonstrated slight improvements in mechanical properties, but significantly more improvement is required.

Question 4: Collaboration and coordination with other institutions

This project was rated **1.8** for its collaboration and coordination.

- The collaboration with the Florida Solar Energy Center (FSEC) appears to be quite beneficial.
- It is positive that FSEC has generated some MEA data and General Motors has committed to testing some samples. However, most project work is still done at Case Western Reserve University (CWRU).
- This project would benefit from closer collaboration with a company that can test these materials in small MEAs to get initial stability data.
- The team is interacting with colleagues at CWRU and sending samples to FSEC for evaluation. There really needs to be substantial interaction with an MEA manufacturer or an original equipment manufacturer.
- The collaborations are not good enough for materials that are apparently successful. More characterization, such as small angle X-ray scattering, small angle neutron scattering, or other spectroscopic or morphological characterization would be helpful. Even the mechanical measurements are a bit unclear—therefore, collaborations are needed at this time. It is almost inevitable that problems will arise, as shown by the MEA testing. The relationship with the FSEC does not seem to be ideal.
- Besides testing at FSEC, there is no collaboration.

Question 5: Proposed future work

This project was rated **2.7** for its proposed future work.

- Focusing on composite membranes (supports or blends) and working to improve molecular weight, if possible, is the right path.
- The approach to increase the polyelectrolyte molecular weight and improve grafting is logical. The suggestion of putting the polyelectrolytes in an expanded matrix or cast with a reinforcing polymer is going to take significant resources and time, especially because these activities are a significant departure from the work that

has been done on the project. Considering the time left on the project and the other planned activities, it may not be possible to fully explore these options.

- Progress will be made toward improving mechanical properties while maintaining proton conductivity. Some of the work is toward low-cost production of the film, which is good. More careful thought should be put into how to make an optimized MEA.
- This project requires more collaboration and scaling up to get more material into other hands.
- It is not clear how the poor mechanical properties and brittleness are going to be addressed. This reviewer wants to know if this will be done through higher molecular weight and cross-linking.

Project strengths:

- Professor Litt's membranes are the most conductive at low relative humidity and 120°C.
- This ambitious, novel approach has provided excellent conductivity.
- This is an excellent application of well thought-out polymer chemistry.
- One strength of this project is its well thought-out plan for why the materials will work.
- The approach is unique among the membrane projects that have been investigated, and the conductivity performance is compelling.

Project weaknesses:

- This project has little collaboration with researchers who can help make the membranes more mechanically robust. Also, the materials are extremely brittle in water.
- It is not clear how the poor mechanical properties and the brittleness are going to be addressed. This reviewer wants to know if this will be done through higher molecular weight and cross-linking.
- The MEAs are not necessarily optimized, so the value of this activity is not as high as it could be.
- There is not enough collaboration with others, which would allow drawbacks to be discovered and steps taken to meet the challenges of these drawbacks.
- The chemistry (i.e., raw materials and reaction conditions) used to make the polyelectrolyte can be fairly expensive. The cost to process these materials into high-quality membranes may also be non-trivial. The mechanical properties (i.e., elongation) of these systems appear to be inherently low and are still a major problem. Even if the properties are improved to a point where they can be fabricated into MEAs without damage, they will still have to demonstrate durability during relative humidity cycling.

Recommendations for additions/deletions to project scope:

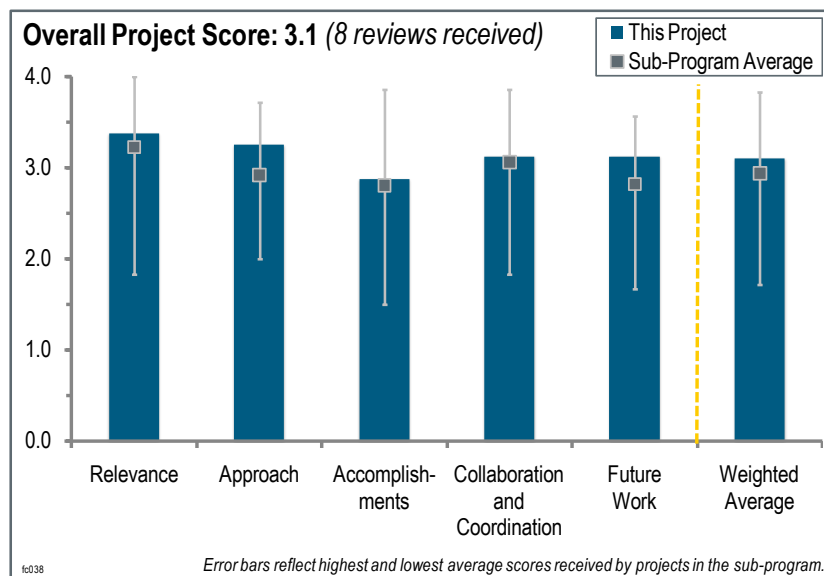
- The principal investigator is aware of the issues with the mechanical properties and has constructed his future work toward addressing it. No change in scope is needed.
- The investigators' primary focus should be to develop composite membranes by blending or using a mechanical support. They should also consider copolymerizing with non-functionalized monomers.
- The possible addition of a more flexible phase, through a co-polymer or a blend with another ionomer, may improve mechanical properties.
- The project team should concentrate on using the remaining resources to make the best possible PEM with this chemistry.
- The researchers should write a new proposal with a much larger team to do more extensive characterization.

Project # FC-038: Nanocapillary Network Proton Conducting Membranes for High Temperature Hydrogen/Air Fuel Cells

Peter Pintauro; Vanderbilt University

Brief Summary of Project:

The project objective is to fabricate and characterize a new class of nanocapillary network proton conducting membranes for hydrogen/air fuel cells that operate under high-temperature, low-humidity conditions with high proton conductivity, low gas crossover, and good mechanical properties. The 2010–2011 project goals are to: (1) evaluate two different nanofiber composite membranes, one of polyphenylsulfone (PPSU) nanofibers surrounded by perfluorosulfonic acid (PFSA), the other of PFSA nanofibers surrounded by PPSU; (2) begin electrospinning low equivalent weight PFSA (660 equivalent weight from 3M); and (3) continue to investigate electrospun fuel cell electrodes.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.4** for its relevance to DOE objectives.

- High-temperature, low relative humidity, low-cost, highly durable membranes are critical for the successful commercialization of fuel cell electric vehicles.
- The project is relevant to the goals of the DOE Hydrogen and Fuel Cells Program and will help meet the research and development (R&D) targets in the DOE Office of Energy Efficiency and Renewable Energy, Fuel Cell Technologies Program's *Multi-Year Research, Development, and Demonstration Plan*. The development of a low relative humidity operational membrane with high conductivity, low gas crossover, good mechanical properties, and dimensional stability is critical to the success of DOE's hydrogen research initiatives.
- The idea to use nanofiber materials in combination with state-of-the-art membrane materials is very good and fits very well with the DOE objectives.
- DOE is searching for an improved membrane that permits operation in hot and dry (e.g., low steam pressure) conditions. Such a material might provide lower costs and possibly enhanced durability. The topic of this investigation is to develop that sort of material. If successful, there could be cost and durability advantages.
- The project has been exploring in a novel way to make a "NanoCapillary" network polymer electrolyte membranes (PEM) for high-temperature hydrogen fuel cells. Eventually, the project may provide a membrane that outperforms traditional PEMs, though the challenge is to develop new processes and related equipment for large-volume membrane manufacturing.
- Improved membranes with higher performance, longer life, and lower cost are critical to achieving fuel cell stack and system targets.
- The project is well positioned to support the objectives of the DOE research plan, particularly for performance at low relative humidity.
- Stable, high-performing PEMs are critical for enabling automotive PEM fuel cell system commercialization.

Question 2: Approach to performing the work

This project was rated **3.3** for its approach.

- The initial approach of electrospinning fuel cell membranes is very unique and has the potential to provide a real breakthrough in membrane performance and durability. The principal investigator runs the project very well. The project is very well defined and managed.
- The approach of making a nanofiber supported membrane is a good approach to enhance the proton conductivity under low relative humidity conditions when the ionomeric matrix is relatively dry. The open nanofiber matrix has good potential to generate a support structure with high proton mobility.
- Electrospinning is a promising approach for making mechanically stable PEMs with highly conductive, low equivalent weight ionomers. The multiple matting methods provide several options for successful materials.
- The project has a very interesting and novel approach.
- The project features a truly unique approach that has generated good results among the different composite membrane technologies being explored.
- This approach offers opportunities to tune the ionomer fiber and inert polymer fiber, as well as address phase separation and surface segregation.
- The thesis of this endeavor is to form composite membranes derived using an electrospun PFSA polymer (the ionomer) and a second structural polymer physically mixed to form a composite. The premise is that a thin fiber of the ionomer will retain moisture (and thus conductivity) even under high and relatively dry conditions, and thereby permit adequate proton conductivity under these hot and dry conditions, but at a temperature where almost all water is gaseous. The second component is thought to provide structural strength. There appears to be some questionable logic here. There is no reason why the ionomer will exhibit different wetting tendencies when in fibrous form. If water is essential for proton transport, a “dry polymer” is not useful. Moreover, the total composite matrix necessarily involves a non-conducting (structural) polymer that must degrade conductivity, as only a fraction of the volume is the ionomer. Lastly, there is no reason to assume that the ionomer pathways will necessarily be continuous. Of course, if the reinforcing second component is degraded, by peroxide for example, the durability might be compromised.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.9** for its accomplishments and progress.

- The investigators have made good progress in the electrospinning manufacturing process, which is promising. They have also generated good results regarding the mechanical stability of the produced membrane materials.
- There has been very good achievement in reducing in-plane water swelling and improving life in relative humidity cycling tests. This novel and promising electrode development work has very good characterization data.
- There has been progress in electrospinning Nafion, which was accomplished with deposition of a second structural polymer to form a “mat.” There was no indication that H₂ permeation through the mat would necessarily be low. Considerable and very interesting results were shown of Pt loaded fibers. It was suggested that a useful membrane electrode assembly (MEA) could be formed by surface decoration of the electrospun fibers. Some of the progress is questionable. For example, conductivity was measured using a sample submerged in water. Soluble ionic impurities could influence those measurements.
- The investigators have demonstrated and achieved reasonable fuel cell results from low equivalent weight PFSA ionomer nanofibers surrounded by PPSU and inert PPSU nanofibers surrounded by low equivalent ionomer PFSA. Further exploring the surface properties of the two different composite membranes may help to make better MEAs. Investigators need to conduct more testing at lower relative humidity levels and higher catalyst loading to emphasize the membrane durability performance and eliminate the catalyst corrosion impact on the durability test.
- The accomplishments toward membrane development have been modest. With a polysulfone nanofiber supported membrane containing the 660 equivalent weight 3M ionomer, almost 78% of the conductivity, as compared to the cast membrane, was retained, while the in-plane and mass swelling were reduced. However, the team needs to work on low relative humidity fuel cell performance data to ensure the utilization of this membrane to meet DOE low relative humidity goals. The electrospun electrode structure represents little

deviation from the membrane development activities; however, the effect of such an electrospun low-loaded electrode in conjunction with an electrospun membrane is worth investigating.

- It was nice to see the cost assessment. The projected cost is above the DOE target, but it might be reduced with economies of scale. Membranes with low equivalent weight ionomers from 3M show promising conductivity, but still swell too much. The electrode development may be promising for improved durability.
- Unfortunately, the work on the membrane project has lagged since last year's Annual Merit Review meeting. Part of the reason may be due to Pintauro's move from Case Western Reserve University to Vanderbilt University, but part of the reason may also be his distraction by electrospun electrodes. While the electrospun electrodes are interesting, a lot of work remains on his original high-potential electrospun membrane work. Also, this reviewer questions if the cost estimates are really justified—especially for an eSpins cost analysis for an electrospinning membrane. The reviewer would like to see a graph explaining cost by volume.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.1** for its collaboration and coordination.

- This project's excellent collaborations include all of the necessary expertise.
- The partners complement the abilities of the principal investigator well and lend good support to the project. The coordination between the partners looks good.
- The team consists of good collaborators. One electrode manufacturer (3M), two automotive companies (General Motors [GM] and Nissan), and one national laboratory (Oak Ridge National Laboratory [ORNL]) is a good combination for a team. However, further collaboration with national laboratories may benefit the team. The team should also try to implement some of the testing protocols developed by their industrial partner, GM, to test newly developed nanofiber supported membranes and nanospun electrodes.
- The low equivalent weight PFSA ionomer was received from 3M. ORNL did some transmission electron microscopy. Professor Pintauro has made his membranes available for others to test.
- The project features very good collaboration with industry, but it may be improved by increasing collaboration with other research groups.
- The project should continue its good collaboration with 3M, Nissan Tech Center North America, and GM. However, the composite-membrane-making apparatus needs to be further explored and customized so that the project can collect more useful data using the resultant and well controlled membrane.
- 3M is the viable collaboration partner, but the relationship appears as mainly a vendor. GM and Nissan are both listed as partners, but both will only test interesting experimental articles. Working partnerships with polymer experts would have strengthened this team.
- Collaborations with outside partners thus far have mostly been comprised of discussions. Very few samples (if any) have been sent out for testing. Again, for such a high-potential project, the project team should have done more work with outside partners as well as more testing by now. The lack of sampling may be due to the relocation of Pintauro's laboratory, and hopefully will be rectified.

Question 5: Proposed future work

This project was rated **3.1** for its proposed future work.

- The project is well defined and has a lot positive potential. Now that the move to Vanderbilt University is complete, this reviewer is hopeful that new and exciting membranes will be made and sent out for validation by the various original equipment manufacturers.
- The extension of the project to demonstrate the performance and low relative humidity cycling goal for the 660 equivalent weight 3M ionomer containing nanofiber supported membrane is reasonable. The team should also focus on the dissolution properties of the low equivalent weight (660 equivalent weight) ionomer from such nanofiber supported membranes, which may affect MEA durability.
- It is a good idea to focus on the 660 equivalent weight ionomer from 3M. Decreasing the fiber diameter is also a good idea to get reduced swelling from the same mass fraction of the inert polymer. The electrode work, while encouraging, is beyond the scope of this project.
- The proposed future work is good, and should include testing at high temperature and low relative humidity levels.

- The future work is generally structured well and should advance the project. While interesting, the cathode catalyst work is outside of the scope of the project, and efforts should remain focused on the membrane.
- The future work extends this data set. The work will focus on the “short chain” 3M polymer and the performance and properties of electrospun fuel cell electrodes.
- The proposed future composite membrane work should be encouraged. The project should also focus more on the membrane surface property investigation and related MEA fabrication improvement. The proposed electrospun nanofiber fuel cell electrode work may need to be minimized, as the real fuel cell electrode has to handle higher current with a thinner electrode layer and lower catalyst loading, as well as readily processing.

Project strengths:

- The very unique technology of electrospinning is an area of strength for this project.
- Investigators have access to automobile test protocols and real-life drive protocols at their partners’ (i.e., GM and Nissan) test laboratories. The team also has a principal investigator with a solid understanding of the membrane field and related challenges in such membrane development work.
- The access to low equivalent weight ionomers for electrospun membrane development is a strength of this project. Professor Pintauro deserves credit for doing the cost assessment.
- This novel approach can be applied to membranes as well as electrodes.
- The technique used to make the composites is very versatile and can produce structures on a nanoscale.
- The project proposed a good concept to make new composite membranes.
- Electrospinning is an established commercial process. Even so, the rates of deposition are low and reproducibility is not excellent. MEA preparation has been focused on just one of many possible synthesis routes. It is important to search for alternatives for MEA fabrication because the current methods are probably not ideal. Scientists are learning much about electrospinning.
- The project has demonstrated very good results and has a very interesting manufacturing process.

Project weaknesses:

- The team is exploring the avenue of low equivalent weight ionomers, which is a common strategy that most researchers are pursuing. The team should think of an alternative strategy to circumvent the traditional approach to low relative humidity membrane conductivity. Low relative humidity ionomers may produce a good beginning-of-life performance, but they typically tend to dissolve over time during relative humidity cycling and affect MEA durability.
- Conclusions from tensile tests on PEM mechanical durability could be misleading. There is no chemical stabilization in the PEMs.
- Investigators have not yet shown performance characteristics of the novel composite membrane and MEA at 120°C and low relative humidity.
- There is no reason to assume that highly dispersed PFSA’s will exhibit useful conductivities when hot and dry. In fact, the ionomer surface will probably be more exposed to drying conditions than when it is in sheet form. Even so, if this approach is useful for conventional PEM fuel cell operation, it could be valuable. There are many applications for PEM fuel cells that work around 100°C. Adequate performance at elevated temperature is a plus, but the absence of that ability is not necessarily a reason to lose confidence in this work.
- The researchers need to deeply address the fuel cell membrane and electrode requirements, especially using larger MEAs.
- The lack of progress on the membrane project in the past 12 months—possibly due to the move or distraction by the electrode project—is an area of weakness.

Recommendations for additions/deletions to project scope:

- Membrane development should be the sole activity for this particular project.
- The investigators should run DOE accelerated stress tests for chemical and mechanical durability, as well as limit electrode work.
- The project team should measure conductivity and cell performance at 120°C and low relative humidity.

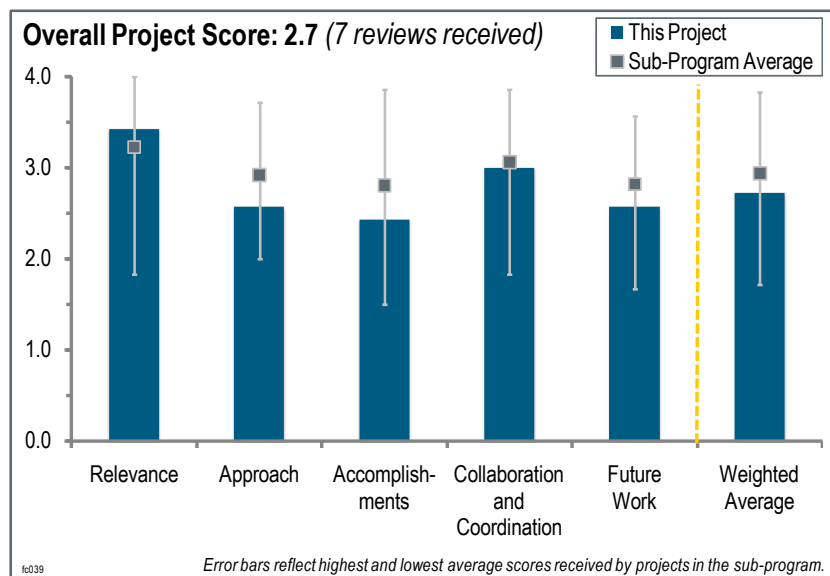
- The length of time used to compact and anneal the mats to make the membrane is a bit long, which could have a significant impact on the cost. This reviewer would like to see a study where this time is minimized.
- It makes sense to separate the two aspects of this invention. It would be interesting to make electrospun electrodes and electrospun membranes and then characterize them separately. Both could have value.
- The proposed electrospun nanofiber fuel cell electrode work may need to be minimized, as the real fuel cell electrode has to handle higher current with a thinner electrode layer and lower catalyst loading, as well as readily processing.

Project # FC-039: Novel Approaches to Immobilized Heteropoly Acid Systems for High Temperature, Low Relative Humidity Polymer-Type Membranes

Andrew Herring; Colorado School of Mines

Brief Summary of Project:

The overall objective of this project is to fabricate a hybrid heteropoly acid (HPA) polymer (poly-polyoxometallates [poly-POMs]) from HPA functionalized monomers with conductivity greater than zero S/cm (siemens/centimeter) at 120°C and less than 50% RH (relative humidity). The objective for 2010 was to optimize hybrid polymers in practical systems for proton conductivity and mechanical properties. The objective for 2011 is to optimize hybrid polymers for proton conductivity, mechanical properties, and oxidative stability/durability.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.4** for its relevance to U.S. Department of Energy (DOE) objectives.

- High-temperature membranes are crucial for the successful commercialization of fuel cell electric vehicles.
- The project addresses membranes at high temperatures and low relative humidity, which is relevant to DOE goals.
- The project is relevant to the DOE Hydrogen and Fuel Cells Program. The activities are aligned with the Program's goals. The development of a low relative humidity operational membrane with high conductivity, temperature stability, and a synthetically versatile membrane is critical to the success of DOE's hydrogen research initiatives.
- The project is very much in line with DOE objectives regarding improved membrane properties.
- The HPAs represent one of the few approaches for polymer electrolyte membrane (PEM) operation at "high and dry" conditions.
- The work is designed to achieve high conductivity at low relative humidity by using HPAs that are conductive dry. However, when water is present, conductivity versus relative humidity follows the same curve as the 3M fluorosulfonic acid polymers, showing only aqueous proton migration. That suggests that the conductivity should be proportional to the ion exchange capacity, as Hamrock demonstrated for the 3M polymers.
- There is interest in "high-temperature" PEMs, as higher temperature membranes might result in cost and durability improvements. Even so, the "high" temperature phosphoric acid (H₃PO₄, phosphoric acid fuel cell) and its recent derivative, phosphoric acid in a polybenzimidazole matrix, are already commercial. Therefore, a PEM membrane operable at 200°C is already well understood. Data from Fuji suggests robust and durable phosphoric acid fuel cell (PAFC) hardware is now commercial.

Question 2: Approach to performing the work

This project was rated **2.6** for its approach.

- The general approach has been good. Tethering HPAs to a polymer backbone is an excellent way to get around HPA dissolution issues.

- The fundamental approach is novel.
- The approach of making HPA immobilized membranes is reasonable for achieving DOE's low relative humidity membrane goals. This project is well designed and thought through. This project is well integrated with 3M's low equivalent weight perfluorosulfonic acid (PFSA) ionomer development for low relative humidity membrane applications.
- The use of trifluoro vinyl ethers as linking groups is a very nice approach toward improving polymer stability—much better than the vinyl monomers used earlier. The yields for critical reactions were well below 100%. Investigators did not demonstrate any attempt to determine the actual structures. There are too many uncontrolled parameters in systems that are very difficult to characterize. The X-ray data was interesting, but most of the terms were not defined, so the slides were meaningless to most viewers. It is unclear what was crystallizing and why it was doing so. Understanding this information would help scientists design better structures.
- Researchers have explored many different chemistries. These materials may break through the limitations of those based on the sulfonic acid (SO_3H) moiety. However, the investigators did not explain why these should be better. This reviewer is wondering if these materials allow higher acid content with lower swelling, or if they have stronger acidity or better morphology. In the end, it is still just an acid functionality and this reviewer is not sure why it should be pursued—other than for the beautiful structures that can be made. This work needs to be better justified.
- The HPAs are well understood, and have been explored as fuel cell membranes repeatedly during the last 50 years. They work well, but have proven to be brittle (i.e., easily fractured) and, when cold, they absorb water, swell, and dissolve. This project presents approaches that “immobilize” these materials.
- The idea to have more stable membranes with high conductivity is very good, but the results of the already ended project are not very promising.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.4** for its accomplishments and progress.

- The project started in 2006, and is now 100% complete. Researchers demonstrated a “hybrid HPA,” which achieved the conductivity target.
- The project successfully passed the initial conductivity milestone and the go/no-go decision point. The ultimate technical targets were not quite all achieved, but considerable understanding of the materials and synthesis methods was acquired and disseminated.
- Investigators have made a lot of progress from the project's inception, which was at a very low level. The latest-generation membrane almost achieves all of the key criteria necessary. A major drawback is the limited size and quantity of the membranes made thus far, and now the project is over.
- The accomplishments toward first-generation and second-generation membrane development were modest. This work demonstrated the feasibility of using immobilized HPA in an inert polymer or ionomeric matrix to enhance low relative humidity proton conductivity. The third-generation approach of immobilizing HPA by anchoring it with aromatic phosphonate groups with an inert hydrofluoropolymer backbone is interesting. The low relative humidity and high-temperature conductivity of third-generation material is impressive; however, it is close to the 825 equivalent weight PFSA ionomer. Moreover, the presence of the unsaturated $=\text{CH}$ -bond in the backbone is concerning due to its instability to peroxide degradation. The presence of HPA may induce some stability; however, that may not be enough for long-term durability under low relative humidity fuel cell operational conditions.
- There is no clear progress at the end of this project. None of the investigated HPAs were successful and competitive with existing membrane technologies.
- Much new and interesting work has been done, but the results show that the group is still far from overcoming the barriers. There is little chance that this approach, as projected, could reach DOE goals.
- After a long amount of time and cooperation with 3M, researchers have made little progress in generating a stable PEM.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.0** for its collaboration and coordination.

- Professor Herring's collaboration with 3M's fuel cell program is the project's greatest strength and was vital to the project's success. This type of interaction can be used as an example across all DOE programs.
- This project primarily involves the synthesis of new materials. The collaboration with 3M's Corporate Material Research Laboratory was very beneficial.
- The work with 3M has pushed the project well, including with new fluorocarbon backbones.
- The project has obviously benefitted by the collaboration with 3M, which included embedding the materials in its polymers.
- 3M collaborated in making membrane conductivity measurements. Other groups have agreed to test films and MEAs as they become available.
- Collaboration with industry (i.e., 3M) seemed to be acceptable, but more collaboration with other research groups might have been needed.
- The only collaborator is 3M, which conducts both synthesis and fuel cell testing on the laboratory scale. There could be more collaboration, including with a national laboratory such as Oak Ridge National Laboratory (ORNL), for conducting more analytical work on the HPA membrane. Although General Motors and Nissan have offered to test promising materials, this step cannot be reached until a promising material is discovered. The project team could have put more effort into understanding the membrane characteristics, which could have helped it find a promising material.

Question 5: Proposed future work

This project was rated **2.6** for its proposed future work.

- A no-cost extension will allow the Colorado School of Mines to perform nuclear magnetic resonance (NMR) characterization.
- This question is not applicable; the project is finished.
- The project ended in March 2011, so the team has very little opportunity to conduct any further research, except some NMR work that it proposed.
- This question is not application; the project has ended.
- The project is essentially done. The principal investigator would like to tether the HPA to a different scaffolding that retains more of the HPA, which is the next logical step.
- There is no future work proposed, as the project is concluded.
- This reviewer could not see reasonable future work.

Project strengths:

- This project has a strong partnership between the university and industry, which combines the out-of-the-box thinking of an academic with the practicality of industry.
- The team worked on a very unconventional approach of using inorganic HPA, and demonstrated the feasibility of using HPA for achieving good membrane conductivity at low relative humidity and high temperature. Incorporating HPA into an inert polymer to demonstrate better performance than 825 equivalent weight PFSA is a noteworthy accomplishment.
- It was a good idea to use HPA for new, high-conductive membrane materials.
- This concept is very interesting.
- This project explores an entirely new chemistry.
- Alternative membrane chemistry is interesting, as considerable work "making Nafion work" has occurred and improvements have been slow in coming. Getting the HPAs to perform is an idea that needed to be explored.

Project weaknesses:

- The initial progress was a little slow.
- The collaboration was not very extensive.
- The selected materials were not suitable for this objective.
- The project relied too heavily on conventional approaches of making functionalized monomers; consequently, the morphologies that could be made were limited. The project needed more insight on structural design versus low relative humidity conductivity.
- At the beginning of this project, it was well known that the HPAs would need to be stabilized as they are water soluble. At the end of the project, the researchers are still working on stabilization. There is no strong justification of why these materials should be better than traditional materials.
- The composite membranes—hydrocarbon polymers chemically bound to these heteropoly phosphate compounds—are sort of replicating Nafion. The intent is to build a structure that has conducting regimes separated by a polymer structure network. This approach, in a way, is no different than PAFCs, in which H_3PO_4 was imbibed in another matrix—historically chopped fibers (which are wet by H_3PO_4) of silicon carbide held together by polytetrafluoroethylene. This system works fine, but it would have been better to seek a homogeneous proton transport system. This reviewer wants to know if a fuel cell system can be built using HPAs that is designed to protect the membrane from wet, cold conditions. PAFC membranes become nonconductive at around 90°C. PAFCs are useful.

Recommendations for additions/deletions to project scope:

- None—the project is complete.
- This project has concluded, so there is no need to alter the scope.
- The involvement of a national laboratory, such as ORNL, with the availability of more analytical tools would have been beneficial for the team.

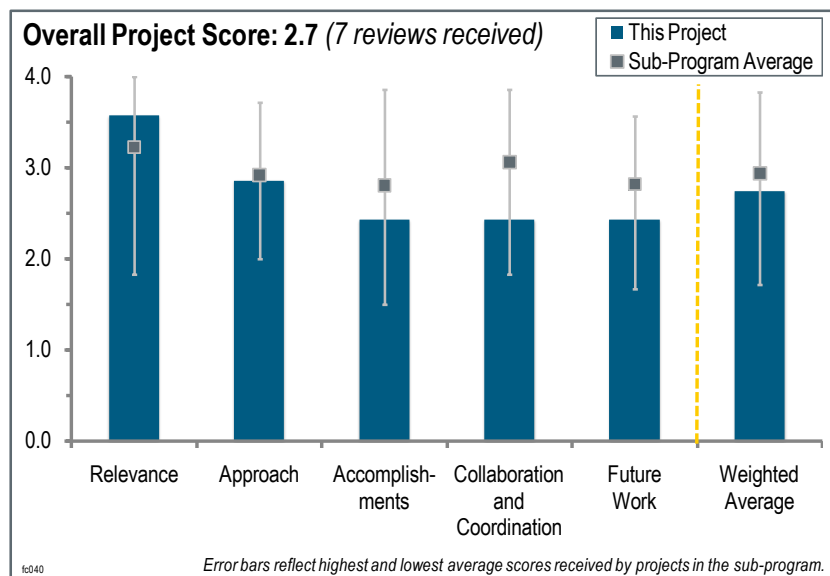
Project # FC-040: High Temperature Membrane with Humidification-Independent Cluster Structure

Ludwig Lipp; FuelCell Energy, Inc.

Brief Summary of Project:

The objectives of this project are to: (1) develop polymer electrolyte membranes (PEMs) with improved conductivity at up to 120°C, (2) develop membrane additives with high water retention and proton conductivity, (3) fabricate composite membranes, (4) characterize polymer and composite membranes, and (5) fabricate membrane electrode assemblies (MEAs) using promising membranes.

Question 1: Relevance to overall U.S. Department of Energy objectives



This project was rated **3.6** for its relevance to U.S. Department of Energy (DOE) objectives.

- The project's technical goals are directly aligned with the DOE Hydrogen and Fuel Cells Program goals.
- Stable, low-resistance PEMs are an enabler for automotive fuel cell system commercialization.
- Membranes with improved conductivity and durability are needed to meet DOE targets.
- The project has reached most of the Program's short-term objectives. However, commercial secrecy hinders good evaluation of the results.
- Development of a membrane with good conductivity at elevated temperatures (up to 120°C) and low relative humidity is desired for simplifying the fuel cell systems for transportation applications. Incorporating independent cluster structure in the membrane may be an interesting approach.

Question 2: Approach to performing the work

This project was rated **2.9** for its approach.

- Many important details were missing from the presentation, such as the precise nature and composition of the water retention additive and the protonic conductivity enhancer. This reviewer understands that it was difficult to assess the real impact of FuelCell Energy's (FCE's) work on the Program. Without knowing more about the membrane composition, it is difficult to assess the principal investigator's (PI) and the University of Central Florida/Florida Solar Energy Center's (FSEC) problems in making MEAs. It is unclear if MEA fabrication will be a serious issue that might kill this membrane.
- Making composite membranes with functionalities for water retention and proton conductivity enhancement is a promising approach. FCE has not included the support polymer as the initial plan indicated. Using a low, 650-equivalent weight ionomer is also the right direction, but the swelling of that material is a very big concern without the support polymer.
- The integration design process may be useful for understanding intrinsic perfluorosulfonic acid contributions to the membrane conductivity and conductivity enhancer impact, as well as a water retention additive function. The challenge is to correlate the different conductive mechanisms and the humidification effect.

- The project has overcome earlier problems. The presentation included vague verbal descriptions, meaning that the approach cannot be evaluated properly. What the materials are and how the films are cast and treated is unclear. The results are impressive, though superacid is needed to reach the goals.
- Investigators built a composite membrane with water retention and proton conductivity enhancement additives.
- The approach appears to be sound, but this reviewer worries about membrane swelling with such a high number of charges in the membrane, along with water retaining materials. The PI's approach must consider membrane swelling and shrinking and the effect of dimensional changes on membrane durability in an MEA.
- So little information was given that it is impossible to ascertain what the approach really is. People have certainly mixed the same set of components together.
- The project team provided very little information. It is unclear if this is a novel approach or just repeats work previously done by others. Many researchers have worked with these types of materials and gotten similar results.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.4** for its accomplishments and progress.

- The investigators met all of the project goals. The class of chemicals used as additives has been reported. The specific compositions remain undisclosed. Durability has not been established, but this was not a project target.
- It was nice to see the investigators disclose some information about the materials and share data (compared to disclosing nothing in 2010), although more details about ionomer, zeolite, and superacid would be appreciated. The conductivity is quite good, but those calculations relied on old data that was not collected in the past year. The method of area-specific resistance measurement is questionable where a large contact resistance is subtracted to get a small membrane resistance number. Although this is more of a question for Scribner, this reviewer cautions against reporting those numbers. There is still no proof of durability or effectiveness of chemical stabilization.
- The technical progress has been good. It appears that the PI has reached or surpassed the 120°C, 50% relative humidity conductivity target of 100 mS/cm (millisiemens per centimeter), but the presenter did not indicate which membrane surpassed this target and which fell below (at 86 mS/cm). This reviewer wanted to see more experimental data, such as conductivity versus relative humidity at different temperatures and membrane swelling versus relative humidity at different temperatures. The reviewer was disappointed that mechanical properties of the membrane were not discussed.
- The project has collected some conductivity, microstructural, and performance data. However, the investigators should address the impact of the superacid and zeolite on the interface of the membrane and electrode, as well as the microstructure changes before and after the life tests. The gradient of the additives in the membrane should be addressed other than with the pinhole tests.
- Some of the composite membranes shown have very good conductivity, but it is difficult to assess the significance of these results without knowing more about the material. Many water soluble composite materials have conductivity in this range, but the trick is to show high conductivity with a membrane that has good chemical stability and durability, and low swelling in water.
- Based on the statements, this reviewer would rate the accomplishments as “good.” However, when the statements are compared with the small amount of data presented, doubts emerge. The main questions are from slides 14 and 15. In slide 14, the PI states that the investigators have demonstrated long-term particle stability. However, the plot above has two very different curves, which are not labeled. One is very sharp with a number average diameter peak at 33 nanometers (nm), while the other has two maxima at 10 nm and 100 nm, with a number average diameter at 33 nm. According to particle diameter weight average, however, the particles have grown to approximately 100 nm. They are not stable and conductivity should decrease, based on the PI's earlier results. Slide 16 shows data at 120°C for the materials versus Nafion 212 membranes. While FSEC is listed on the graph, the curve for Nafion 212 is very different from other curves this reviewer has seen from FSEC. It dropped very rapidly above 100 mA/cm² (milliamps per centimeter squared), and was stopped at 400 mA/cm² and 580 mV (millivolts). The usual curve is 700 mV at 400 mA and .43 V (volts) at 1,000 mA. Their materials parallel their Nafion 212 plot as far as it goes, but are at .33 V at 1,000 mA—which is still good. The 80°C data for Nafion 212 is more normal, but the voltage still drops sharply above 1,000 mA/cm². If the investigators prepared the MEAs, including for Nafion 212, and sent them to FSEC for testing, this should be stated. If this is their data, that should have been made clear.

- The project showed no actual results that met conductivity targets—only “extrapolated” results. It is unclear what that means.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.4** for its collaboration and coordination.

- While most of the partners are unknown, the collaborations with the polymer and additive partners seem to be solid. Fuel cell data was collected at FSEC, and Oak Ridge National Laboratory did some nice microscopy.
- There has been some collaboration with FSEC on MEA fabrication and testing. On the overview slide, the PI indicates that there are collaborations and partners with regard to polymer synthesis and membrane fabrication and characterization, as well as additive synthesis and characterization; but these partners were not identified and their role in the project was not adequately explained.
- The investigators could get the materials they needed made by outside suppliers. They are working with FSEC to validate internal results.
- The key collaborating entities remain unnamed.
- This reviewer cannot assess the collaborations from the information provided.
- It is not possible to judge the quality of this team because the investigators will not share the identity of most of the participants. This reviewer questions if FSEC is a credible MEA maker.
- The collaboration did not provide either deep understanding or broader evaluation and characterization.

Question 5: Proposed future work

This project was rated **2.4** for its proposed future work.

- The proposed future work is adequate and reasonable, given the accomplishments in 2010–2011. More emphasis should be placed on MEA fabrication and membrane durability in an MEA. This reviewer would like for the PI to include a thorough cost analysis as a future work task.
- The project is basically over. FCE will continue collaborations, development, and testing to meet cost and durability targets, and to optimize the membrane-electrode interface.
- The proposed future work is noncontroversial.
- The future work should focus on reproducibility. However, the casting is only one of the factors. Currently, the results presented do not indicate that the investigators can control the dispersion of additives and related gradient change or the membrane surface and interfacial properties.
- The proposed future work does not contain any durability testing. Also, the investigators should do some preliminary cost assessment, especially considering that the lack of materials disclosure prevents an independent assessment. FCE should follow through on its plans to include a polymer support matrix.
- Investigators should show data on the stability of these membranes toward liquid water. This is a critical requirement.
- No details were given since the work is a company secret. However, the researchers are saying the right things.

Project strengths:

- One strength of this project is the use of low equivalent weight ionomers with chemical stabilization. The strong team provides materials to make highly conductive PEMs.
- The concept is interesting and the researchers have shown that it has good promise.
- A membrane has been fabricated that appears to meet the DOE 50% relative humidity, 120°C conductivity target.
- The investigators are strong on fuel cell fabrication and evaluation.
- The researchers seem to be able to produce membranes.

Project weaknesses:

- There is no durability data. The high swelling of membranes is likely to be an issue during relative humidity cycling. Other weaknesses include the lack of full disclosure of materials and the lack of cost projections. When the project ends, it is unclear how the community will benefit.
- Membrane cost and durability are important issues that were not addressed. The presentation was short on data and details. The composition of the membrane is unknown and there was no data presented for conductivity versus relative humidity, swelling versus relative humidity, or mechanical properties for different temperatures.
- The investigators need to work with partners to collect more characterization data and determine the crucial factors for reproducing and scaling-up the composite membrane.
- The “top-secret” approach impedes information flow. This reviewer does not believe that they have really met the targets as claimed. Otherwise, they would not have to resort to “extrapolations.”

Recommendations for additions/deletions to project scope:

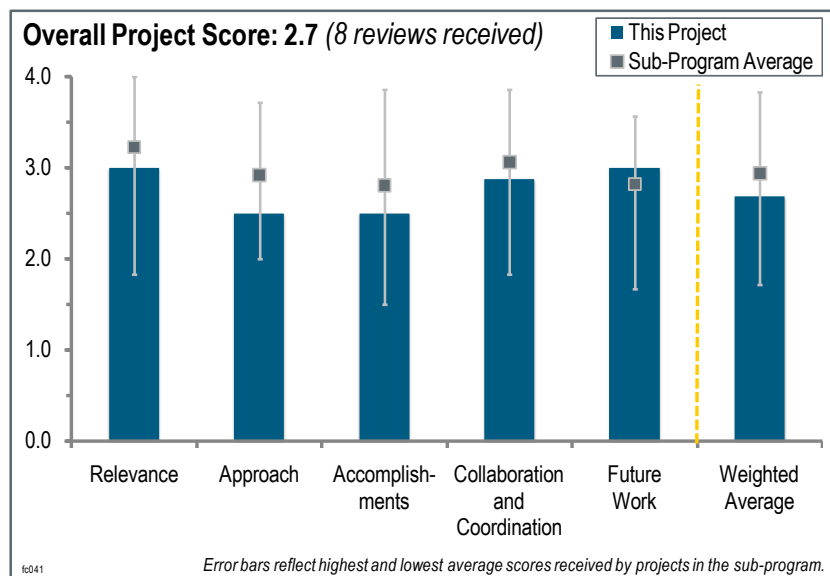
- None. The project is focused. This reviewer cannot recommend changes because there is not enough information.
- The project team should collect water isotherms and present swelling data. It should also follow through on the addition of polymer support and conduct the DOE recommended accelerated stress tests for chemical and mechanical durability.
- The researchers should perform membrane durability tests for relative humidity cycling, as well as determine and present the materials and fabrication costs of the new membrane. They should also provide more details at next year's Annual Merit Review regarding the membrane composition so that reviewers can better evaluate the potential of this new membrane material in fuel cells.

Project # FC-041: Novel Approach to Advanced Direct Methanol Fuel Cell Anode Catalysts

Huyen Dinh; National Renewable Energy Laboratory

Brief Summary of Project:

The overall objective of this project is to develop and demonstrate direct methanol fuel cell (DMFC) anode catalyst systems that meet or exceed the U.S. Department of Energy's (DOE) 2010 targets for consumer electronics applications. The specific goal is to improve the catalytic activity and durability of the platinum-ruthenium (PtRu) for the methanol oxidation reaction via optimized catalyst support interactions. A similar approach for oxygen reduction reaction (ORR) catalysis is advantageous for both DMFC and hydrogen fuel cells.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.0** for its relevance to DOE objectives.

- The proposed effort is targeted at DMFC anode performance and durability, which should help address the portable power lifetime goals. If the investigators can demonstrate improvements in performance for practical catalysts at a reasonable cost and production rate that can be implemented into a membrane electrode assembly (MEA), the work should address the cost targets.
- Improving the performance of the anode catalyst in DMFC systems is critical to achieving the DOE technical targets. With that said, it is not clear how this work will impact the effort to reach those goals. The investigators did not present any data on how this catalyst impacts the MEA performance (although MEA testing is planned for later this year).
- The objectives are consistent with required improvements in cost, performance, and durability.
- The project is directly relevant to DOE objectives, as it addresses three important limitations of state-of-the-art DMFCs: (1) the low catalytic activity of PtRu for the methanol oxidation reaction, (2) the low durability of the anodic PtRu catalyst, and (3) the high costs depending on the high loading of expensive Pt and PtRu-catalysts used.
- The project is relevant in that it focuses on an important aspect of DMFC technology (i.e., the anode catalyst). The DOE objectives are stated, but the milestones for the project are largely activities, rather than quantitative metrics that can be related to the DOE goals.
- DMFCs are part of DOE's strategy for the commercialization of fuel cells, but this project does not directly address the biggest problems with DMFCs—methanol crossover through the membrane and Ru dissolution from the anode, and the subsequent poisoning of the cathode by this Ru. Improving the evenness of the dispersion of alloy catalyst particles, which is the major advance promised by this project, is of more general potential in the DOE Hydrogen and Fuel Cells Program versus specifically in DMFC research and development. For ORR catalysts, it has proven more difficult to get even dispersion of Pt-alloy particles on corrosion resistant carbon (including Vulcan, which is more corrosion resistant than the Ketjen black or HSC [sic] on which alloys give highest activity) than it has been for pure Pt particles. Therefore, the methods of this project could be quite beneficial in combining high-activity Pt-alloy ORR catalysts with corrosion resistant carbon supports.
- Investigators did not conduct a cost analysis, which seems like a big omission. Surely a rough calculation would show whether this approach will meet the cost goals.

Question 2: Approach to performing the work

This project was rated **2.5** for its approach.

- The overall development approach seems to be quite sound. The work needs to continue toward measurements of catalyst performance and durability in a real MEA using a reasonable system with compatible operating conditions, including some tolerance for off specification operation such as low methanol concentration (partial fuel starvation) operation.
- The methods seem fine, but the logic behind them is problematic. If the PtRu is held on the carbon by the nitrogen groups on the surface, surely the nitrogen is protonated under normal operating conditions. This reviewer wants to know if the density functional theory (DFT) calculations address this issue, and if there is any basis in the literature.
- The authors followed a productive pathway to apply an advanced concept to practical carbon-black-supported catalysts. They first demonstrated their concept on highly-ordered pyrolytic graphite (HOPG), on which the ion implantation was easy, and then made the new equipment needed to do the ion implantation and sputtering coating on powders. This method is an ingenious and potentially productive approach to using line-of-site deposition techniques to coat all sides of powder particles. The use of DFT calculations to draw conclusions about the relative solubility of Pt and Ru from non-implanted versus implanted carbon supports is perhaps a bit questionable, as it appears to have been done by just calculating the energy to remove one atom (either Ru or Pt) from a particular four-atom Pt_2Ru_2 cluster. One would expect the dissolution energies to vary significantly with particle size, geometry, and composition.
- The approach to improve interactions with support has some merit, but it is unlikely to make significant gains due to the inherent instability of Ru in the DMFC conditions.
- It is not clear if a 20%–30% improvement in methanol oxidation reaction (MOR) half-cell activity will reduce the overall costs for a DMFC. A comparison of catalyst costs made by HOPG and sputtering methods for large quantities will be helpful.
- Doping of the carbon support with nitrogen appears to decrease the degradation of the electrocatalyst by nanoparticle migration and coalescence. However, one of the major mechanisms of loss of MOR activity is the leaching of Ru from PtRu alloy catalysts. The DFT calculations indicate that nitrogen doping will actually increase the tendency of Ru to leach from the alloy. The responses to reviewer comments are conflicting regarding the effect of nitrogen doping on Ru leaching.
- The main thrust of the project is the catalyst and support structure. A lot of experimental and theoretical effort has been spent on the HOPG system. This reviewer wants to know how relevant this material is to the types of carbon that are actually used as catalyst supports in the DMFC. The durability improvements realized for HOPG after ion implantation are not unexpected, given the nature of the material.
- This reviewer's chief complaint of the approach is that no MEA testing has occurred and the project is near completion. There is no time to learn from the MEA testing, which would have allowed critical information to be fed back into the development process.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.5** for its accomplishments and progress.

- The project has made good progress toward the catalyst goals, and continued testing in an MEA configuration is important. This testing will also help to understand the performance and durability performance benefits against the potential costs associated with the catalyst.
- The test data presented appeared to indicate improved catalyst activity. Again, the investigators did not present any data from MEA testing, so it is unknown how it will improve cell performance. Also, the investigators presented only minimal degradation information.
- The investigators have made good progress on understanding the process parameters to get either durability or performance improvements. The improvements are incremental, rather than significant, and may not justify the significant efforts on developing the processing parameter understanding. The balance appears to be off between optimizing processing parameters versus determining clear indications of potential improvements and the ability to meet targets. The stability of the Ru is a critical component, but the only stability test to date has

measured only the electrochemical surface area (ECSA) change. The investigators are already 80% into the project, and this is very late for not having this information.

- The researchers have made good progress in determining the effect of various deposition parameters on methanol oxidation activity in half-cell measurements. Though ECSA losses are lower for nitrogen-doped, carbon-supported materials versus undoped carbon support, the losses are still unacceptably high. The cell voltages are extremely low.
- The accomplishments of the project in terms of synthesis and physical characterization of the catalysts are good, but it is too late in the project for the electrochemical work to be in such an early state. Slide 13 claims a 20%–30% improvement in MOR activity for the implanted-support catalysts. Given the difficulties in measuring electrocatalytic activities, it is unclear whether such a gain is significant—one needs bigger effects to be sure. The conclusions from the DFT suggest that the net effect of the implantation may be negative for DMFCs, per the statement on slide 30 that “[Ruthenium] is more susceptible to preferential leaching from PtRu over the [nitrogen]-implanted carbon than over unmodified carbon”—a statement that appears consistent with the results for pyridinic nitrogen on slide nine. Slide 31 appears to give a contradictory statement—“[Ruthenium] is stabilized by the presence of [nitrogen]”—which probably refers only to the pyrrolic nitrogen on slide nine. The investigators appear to recognize at least part of the potential importance of their work to ORR (though the presenter did not mention the greater problems with dispersion of the Pt alloy than with pure Pt particles). However, the only ORR data presented (slide 24) shows an anomalously low ECSA and mass activity for Pt on the nitrogen-implanted carbon. That slide claims enhanced activity for Pt on the nitrogen-doped carbon, but only the specific activity, not the more important mass activity, is enhanced. On the positive side, the researchers do appear to have demonstrated improved durability against potential cycling for their MOR catalysts.
- A large effort has been made with nitrogen-doped carbon-supported PtRu catalysts regarding durability. Normally, MEAs with high-loaded PtRu catalysts are used in commercial DMFCs. It would be better to also compare the in-house catalyst with HiSpec 12100 from Johnson-Matthey Fuel Cells Inc. (JMFC). There is no information on how the nitrogen-doped carbon support will decrease the corrosion of Ru or Ru oxide from the catalyst. This is only done for Pt.
- Durability and performance have improved, but it is unclear why. This seems a little too much like alchemy. Also, the project is really not improving on the generally low performance of the anode in DMFCs.
- The new materials being developed here seem to have a similar performance to JMFC’s commercial JM5000 catalyst. There are no error bars associated with the numbers, and electrode preparation could introduce enough variability to make the differences small, if not insignificant. The micrograph on slide 14 shows only the catalyst after 5,000 cycles. It would be useful to compare these results with the original un-cycled sample, as well as the other catalysts studied.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.9** for its collaboration and coordination.

- The project demonstrates good collaboration between the participants.
- The project features good collaborations, although Mechanical Technology, Inc. does not appear to be very involved.
- The institutions and each of their roles were given. It appears that the group is working well together.
- The collaboration between the parties appears to be good.
- The project has lots of collaboration, but it seems that the presenter does not know what the collaborators are all doing.
- Though many collaborators were listed, it is unclear what they have contributed to the project in the past year.
- There is no indication of collaboration with BASF (the state-of-the-art catalyst is from JMFC).

Question 5: Proposed future work

This project was rated **3.0** for its proposed future work.

- The continued testing of the MEA in a practical fuel cell configuration to measure the durability and performance benefits under real DMFC operating conditions is important. This testing should be evaluated against the potential additional costs associated with catalyst synthesis and production at a reasonable scale.
- The MEA testing will be beneficial.
- The plans for finishing off the project are fine, and in line with past activities. The beam time at the Stanford Linear Accelerator Center will add more data. It is unclear whether this information is useful.
- It is very important to better understand these catalyst systems under fuel cell conditions. The composition of the catalyst after operation and voltage cycling needs to be determined.
- The future work plan is reasonable. Understanding the amorphous components of these materials is important, and will be addressed. X-ray diffraction studies only provide some of the story. The investigators have planned more MEA and DMFC performance studies with the new catalysts, which will be useful in determining the value of this approach.
- Establishing the “catalyst degradation mechanisms, e.g., extent of Ru dissolution and catalyst coarsening” should be the highest priority of the project, and will help determine if the nitrogen doping is preventing degradation of the chemical composition of the MOR catalyst. Testing full cells should also be one of the highest priorities.
- The project is largely completed, so a major shift in plans—such as directing the work to the stabilization of Pt alloy particles on corrosion resistant carbon for ORR—would be unrealistic, though it would improve the contributions of the project to the overall Program. The remaining work should be directed toward a continued search for large activity gains (not 20%), experimental quantization of the effects of nitrogen doping on Ru dissolution from PtRu, and a bit more exploratory work on ORR catalysts to see if a mass activity gain from nitrogen implantation can be demonstrated (larger advantages would be expected from supports more highly graphitized than Vulcan).
- There are milestones missing.

Project strengths:

- This project features good catalyst development and characterization, in particular the ex-situ characterization.
- The project is well structured, and has clear plans. The approach showed good increases in catalyst support interactions, as well as some improvements in activity, but not in the same design. The project has moved from model systems to viable project supports.
- The project partners have great experience and expertise in their respective fields.
- The deposition method appears to result in highly-dispersed nanoparticles.
- This is a good team that has the resources to perform the proposed work.
- The project features a rational pathway to improve the evenness of dispersion of Pt and Pt alloy particles on corrosion resistant carbon supports. Another project strength is the nice implementation of line-of-sight implantation and coating of powder supports. The project team demonstrated improvements of particle stability against voltage cycling.

Project weaknesses:

- The project needs to include more testing in an MEA using a set of reasonable characteristics for the MEA. The project team should also perform more testing using system compatible conditions.
- One weakness is the limited opportunity to learn from MEA testing and feed the data back into the development process. Also, there is minimal time to evaluate any degradation issues.
- The project is really empirical, and uses facilities that are in place but without sound rationales as to why they would work.
- One area of weakness of this project is the lack of understanding of how a catalyst system behaves in the MEA and under relevant conditions.
- One major degradation mechanism is Ru leaching from the alloy. Therefore, the Ru leaching should be measured—for example, with methanol stripping for both catalysts (commercial and in-house made) after 5,000

cycles. Methanol stripping experiments for the cathode would also be helpful to indicate leached Ru that is permeated through the membrane.

- The lack of focus on Ru leaching degradation is an area of weakness.
- The project needs a better focus, which works backwards from the DOE goals. It may have been covered in a previous review, but the amount of effort spent on HOPG studies is questionable. The project involves interesting work, but it may not have much relevance to real-world support materials that are used in fuel cells. Error bars and better statistics are needed to establish what is significant and what is not.
- One area of weakness is the dubious improvements versus the main catalyst durability challenge of DMFCs. Another weakness is the Ru dissolution from the anode and deposition on the cathode. The DFT results would need at least a study of the sensitivity of the conclusions to PtRu cluster size and shape before one could place any credence in them. The project devotes insufficient attention to ORR, where the benefits of this approach would likely be the greatest, even for DMFCs.

Recommendations for additions/deletions to project scope:

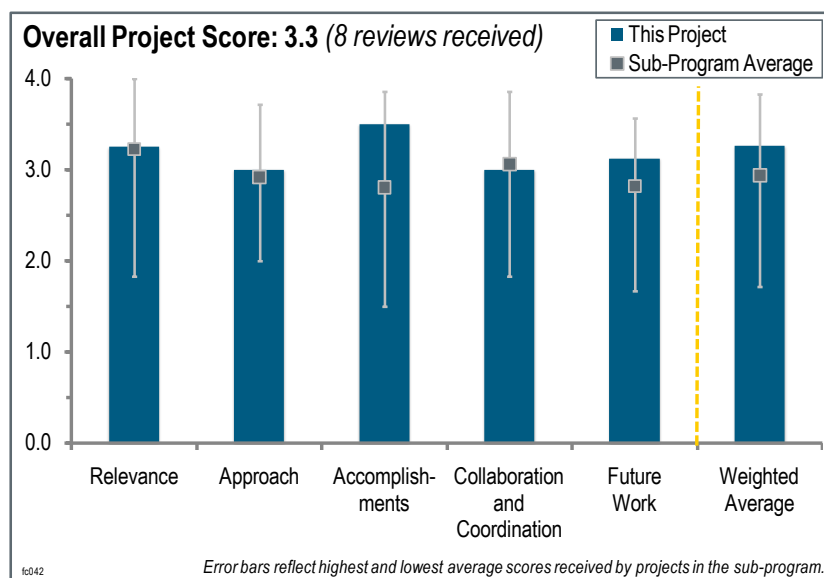
- Investigators should conduct an objective assessment of possible gains with continued optimization of processing conditions to determine the value of continuing the activity.
- Researchers should compare the in-house catalyst in real MEAs with commercial MEAs—for example, from JMFC—with improved PtRu catalysts.
- The project team should consider dropping implantation of other materials in order to concentrate more on the nitrogen implantation work. The team should also check the sensitivity of DFT conclusions to PtRu cluster size and shape, and consider MOR steady-state activity measurements in addition to cyclic voltammograms. Additionally, the researchers should do a bit more work on ORR, preferably with more supports that are more highly graphitized than Vulcan. While doing these studies with Pt-alloy (e.g., Pt₃Co) particles is probably not realistic in this project, one could get relevant information by using pure Pt particles, but then annealing to an alloy-formation-like 900°C in 3% H₂/N₂ to presinter the particles before starting to study the resistance of the catalyst to voltage cycling.

Project # FC-042: Advanced Materials for Reversible SOFC Dual Mode Operation with Low Degradation

Randy Petri; Versa Power

Brief Summary of Project:

The project objectives are to: (1) advance reversible solid oxide fuel cell (RSOFC) stack technology in the areas of endurance and performance through RSOFC materials development and reversible stack design; and (2) meet the following performance targets in a kW (kilowatt)-class RSOFC stack demonstration: (a) RSOFC dual mode operation of 1,500 hours with more than 10 solid oxide fuel cell (SOFC)/solid oxide electrolyzer cell (SOEC) transitions, (b) an operating current density of more than 300 mA/cm² (milliamperes/square centimeter) in both SOFC and SOEC modes, and (c) an overall decay rate of less than 4% per 1,000 hours of operation.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.3** for its relevance to U.S. Department of Energy (DOE) objectives.

- This project is unique within the DOE Hydrogen and Fuel Cell Program and addresses one of the major barriers of hydrogen-based energy storage systems—low capital utilization for systems that use separate fuel cells and electrolyzers. Success in this project could be a game changer for H₂-based energy storage systems.
- Understanding the potential of integrated reversible fuel cells is a relevant topic for DOE to explore. As an advanced concept, it fits within a small allocation of the overall Program budget.
- This project on RSOFCs for power generation and H₂ production fully supports the Program objectives.
- This is a feasibility study on the SOFC/SOEC concept, and leverages technology developed in the Solid State Energy Conversion Alliance (SECA) program.
- RSOFCs can integrate renewable production of electricity and H₂ when power generation and steam electrolysis are coupled in a system, which can turn intermittent solar and wind energy into “firm power.”
- The project represents good synergy between electrolysis and fuel cell operation, and should have application where renewable electricity generation can be coupled with H₂ generation to even out the diurnal cycle and intermittent nature of solar and wind power. A reversible SOFC/SOEC system should be a good fit for these applications. The project supports the Program objectives in a number of ways.
- The reversible fuel cell is an interesting concept, although it may not be viable to be used as suggested by DOE to support wind or solar energy storage. Demonstrating the possibility as soon as possible should be a priority. The SOFC/SOEC approach has been demonstrated by many developers, and Versa Power has made impressive progress on cell performance and longevity in both modes.

Question 2: Approach to performing the work

This project was rated **3.0** for its approach.

- The approach for cell component development and testing was well designed and performed. The cell testing setup that used full-size cells and stack component layers was a cost-effective way to demonstrate the operation

of all of the essential components of the stack. Another significant objective is the demonstration of the stack operation in switching between both modes. Researchers could have placed more emphasis on this objective.

- The investigators have identified technical barriers and are addressing the barriers adequately. They have developed a good mix of modeling and experimental approaches. There is noticeable progress in improving power density and degradation; however, much work is needed for commercial viability.
- Versa Power is focusing on developing high-performance and low-degradation RSOFC cell and stack technology that is critical for the reversible SOFC/SOEC system. These are the two most important activities that can be pursued at this time. Durability testing should follow these pursuits.
- Focusing on durability early on in the project makes good sense. The investigators should focus on achieving a fundamental understanding of the causes of degradation, and how to mitigate such losses as early and carefully as possible.
- The approach is sound, and uses a stage-gate go/no-go process. Down-selecting a set of materials seems to have paid off with better durability and performance. This reviewer would like to see more detailed analysis on degradation mechanisms. This analysis may be under way, but might involve sensitive proprietary data. The lack of details on decay mechanisms is acceptable as long as durability continues to improve.
- The project team has not identified any approaches to addressing the issue of RSOFC degradation under dual operating modes.
- The approach builds on a long history of development by Versa Power in the SOFC arena, primarily through the SECA program, and is to advance the stack technology through materials improvements. The approach has been largely successful, and Versa Power has met the quantified performance and durability targets set out in the beginning of the project. However, even though the project addresses the cost barrier, the presenter did not mention cost or targeted cost reductions in the presentation. It is not clear what performance and cost targets are required for commercialization, or how close Versa Power is to the targets.
- The targeted degradation rate of 4% per 1,000 hours seems rather high. This reviewer wants to know if there is a path forward to achieving a lower degradation rate.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.5** for its accomplishments and progress.

- The project exceeds the targets on performance, degradation, current density, and operating duration.
- The project team has met or exceeded project targets. The team has good test plans to demonstrate the results.
- The accomplishments since the last Annual Merit Review have been good. Several additional material sets have been developed and tested, and eight of the materials have met the target area-specific resistance value. The researchers have reduced degradation and surpassed their degradation target of less than 4% per 1,000 hours—achieving approximately 1.5% per 1,000 hours—in both fuel cell and electrolysis modes.
- It is good to see that the researchers met the endurance and performance targets. They appear to have made good progress in terms of developing an optimized material set for this challenging application. The reported absence of electrode delamination is very encouraging, as this has been a frequent issue in the past, at least for some developers.
- Versa Power made significant progress ahead of schedule, and has achieved the following:
 - Developed 11 types of RSOFCs.
 - Exceeded performance and degradation targets with two types of cells (i.e., RSOFC-4 and RSOFC-7).
 - Ran a steady-state single cell test of RSOFC-7 in electrolysis with a degradation rate of about 1.5% per 1,000 hours.
 - Ran a baseline 28-cell stack (kW-class) test in electrolysis for more than 1,000 hours at a degradation rate of about 1.3% per 1,000 hours.
 - Made significant progress with an 8,000-hour test in SOEC mode at 2.2% per 1,000 hours.
 - Achieved good ASR in the cells—around 0.2 ohms/cm² for both modes.
 - Achieved 500 mA/cm² in the cell.
- Versa Power has done an excellent job of selecting the materials and demonstrating their performance in the reversible mode. It is clear from the work that cells can operate in these modes with an acceptable lifetime.
- The investigators did a good job of getting to a stack test as early as possible to demonstrate decay and total performance. The asymmetric efficiency of fuel cells and electrolyzers makes one wonder whether there will always be an efficiency penalty for an integrated device. One might consider a series of trade studies so that the

cost benefit of an integrated system could be traded against the maximum allowable efficiency delta for integrated versus separate stacks in different application environments.

- This project has made significant progress on efficiency and durability. This reviewer would have liked to have seen more information on cost reduction—as that is a stated goal of the project—but the presentation did not include any specific information on the topic.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.0** for its collaboration and coordination.

- The project features collaborations with Idaho National Laboratory (INL) and Boeing.
- The reversible fuel cell and electrolytic cell concept is relatively new, so it is difficult to collaborate with too many organizations. However, there could have been more involvement with universities. The two collaborators that were selected were certainly some of the best U.S. entities. INL has been working for several years on high-temperature steam electrolysis with the advanced nuclear reactor projects, and has an excellent understanding of stack and systems requirements and issues. Boeing has demonstrated interest in SOFC technology and would most likely be the first to use SOFC/SOEC technology or a similar variation in its aviation products.
- The project team has identified Boeing and INL as partners, and is well coordinated.
- This project features collaboration with Boeing and INL.
- It looks like the project has good collaboration for rather esoteric applications. The investigators should consider partners for energy storage in areas of greater interest to DOE.
- Boeing collaborated on and funded initial RSOFC development work through its efforts and efforts funded by the Defense Advanced Research Projects Agency. INL is seeking to eventually integrate SOEC technology for H₂ production with the Next-Generation High Temperature Nuclear Reactor. Versa Power is a major player in this SOFC program.
- Boeing and INL are listed as collaborators. If the investigators intend to apply the project in the renewable energy field, they should involve an organization that focuses on the application. INL is looking to integrate the RSOFC technology with the Next-Generation High Temperature Nuclear Reactor, which will certainly not be near term.

Question 5: Proposed future work

This project was rated **3.1** for its proposed future work.

- The proposed future work exceeds the original project goals. Versa Power plans to complete the final project metric test with a kW-class RSOFC stack, and complete the additional cell development scope of advancing performance and degradation beyond the original project target.
- The most significant part of this project is demonstrating a stack of cells that can be reversibly operated in fuel cell and electrolytic modes. Versa Power should prove that the furnace can be essentially turned off during the stack operation. The SOEC can be run slightly above the thermal neutral point to keep the stack heated and giving up a little in efficiency. Showing that a stack can be cycled seamlessly between fuel cell and electrolytic modes is the most critical milestone of the project.
- The focus on integration with renewable energy makes sense.
- The proposed future work is logical and effectively planned.
- The future work plan is appropriate; certainly, the completion of the economic study is important.
- The future work makes sense as a progression of the development. In order to be commercially viable, the system would probably have to be scaled to the MW (megawatt) class. It is important to include that notion in the future funding of this project. It will also be interesting to see if the durability numbers hold up with a field installation. Durability in the laboratory is typically significantly better than in the field.
- The project is almost over—there is just enough time left to demonstrate a kW-scale stack that can operate in both modes. Future progress will depend on continued funding that may not be available. The technology could stagnate without further funding.

Project strengths:

- This technology is game-changing.
- Versa Power is one of the leading high-temperature planar stack developers, and has state-of-the-art SOFC designs and components. Versa Power has demonstrated that its cells and associated stack components can repeatedly cycle between fuel cell and electrolytic modes at full-scale with acceptable long-term operation.
- The focus on the demonstration of actual stack performance under reversal is an area of strength.
- The RSOFC technology being developed under this project has the potential to improve performance and performance stability for both power generation and H₂ production modes.
- This project features a good record of technical accomplishments and progress.
- The investigators have made good progress toward developing an optimized set of cell and stack materials.
- Strengths of this project include its dual mode operation and well defined quantitative milestones.
- The researchers will be able to use SECA advancements in this technology development, which is a significant advantage. Achieving 300 mA/cm² in both modes is another area of strength for this project.

Project weaknesses:

- Durability, cost, and scalability remain significant challenges that will require sustained development and early commercial success to overcome. The project would benefit from collaboration with a utility company, if that is an intended market.
- Versa Power should have demonstrated a stack of cells earlier in the project to demonstrate that ceramic technology can remain at temperature and cycle between the two modes. It is not certain that the high-temperature reversible concept will work for wind or solar energy storage.
- One area of weakness is this project's lack of partnerships in energy storage space or a clear path to demonstrate there.
- A better understanding of degradation mechanisms is needed.
- The project team has not given any indication of the performance, durability, or cost that will be required for commercialization.
- It would have been good to see some information regarding present and future cost targets for this technology. The path forward seems vague. It is unclear if the investigators are going to build a larger stack, develop a system (and if so, at what kW level), or pursue another option.
- The researchers have set a low bar for performance targets. The focus is on the stack and not a system.
- There seems to be more development on SOEC than on both SOEC/SOFC simultaneously.

Recommendations for additions/deletions to project scope:

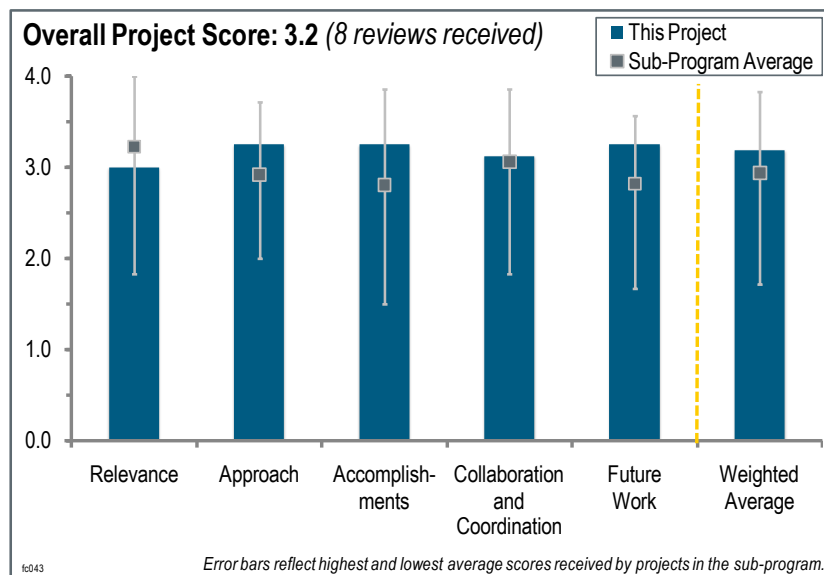
- The investigators should add a study to scope out a roadmap to a MW-class system suitable for deployment in utility applications.
- The high-temperature reversible SOFC/SOEC energy storage concept should be demonstrated as soon as possible.
- Researchers should consider quantifying or estimating the maximum allowable efficiency delta between an integrated and separate stack based on the assumption that the cost difference is 50%.
- The project team should clarify opportunities for further funding.
- The project should continue stack development. The investigators should test more SOEC/SOFC cycles.

Project # FC-043: Resonance-Stabilized Anion Exchange Polymer Electrolytes

Yu Seung Kim; Los Alamos National Laboratory

Brief Summary of Project:

The overall objective of the project is to demonstrate improved alkaline membrane fuel cell (AMFC) performance using novel polymer electrolytes and non-precious-metal catalysts. The specific objectives of this project are to: (1) develop anion exchange polymer electrolytes that have high hydroxide conductivity and stability in high-pH conditions, and (2) demonstrate improved single cell performance of solid-state alkaline fuel cells using polymer electrolytes and non-precious-metal catalysts.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.0** for its relevance to U.S. Department of Energy (DOE) objectives.

- The project is relevant to the objectives of DOE Fuel Cell Technologies Program's *Multi-Year Research, Development, and Demonstration Plan*, and the activities are aligned to DOE's Hydrogen and Fuel Cells Program goals.
- Improved membranes are critical to fuel cell stack performance, life, and cost.
- The study of anion exchange polymer electrolyte membranes (AE PEMs) for fuel cells is important because this under-studied component could meet DOE's need for performance and lower cost through non-Pt catalysts.
- The mechanism of conductivity in AE PEMs is still poorly understood, and the long-term stability of membranes and the electrode catalyses is not well known. This AE PEM work should be considered exploratory work. In the long run, AE PEM materials may not be able to satisfy DOE goals compared to more conventional, better understood materials such as proton-conducting Nafion®.
- Non-precious-metal catalysts and anion exchange polymer electrolytes may be required for future fuel cell development for a number of reasons. For example, faster kinetics of oxygen reduction reactions in an alkaline media allows the use of non-noble and low-cost metal electrocatalysts.
- Alkaline fuel cell development lags far behind polymer electrolyte membrane (PEM) fuel cells for DOE-relevant applications. This project has some relevance to the overall long-term goals of the Program, but this relevance falls far below that of PEM fuel cell work and has no connection to many of the near-term (2015) DOE targets for fuel cell performance.
- A clear vision of the potential application is required to set materials targets that will depend on power density, cost, efficiency, and durability needs. Rather than work toward a set of targets, this reviewer recommends focusing on a mechanistic understanding of alkaline fuel cell performance and durability.

Question 2: Approach to performing the work

This project was rated **3.3** for its approach.

- This is a good, systematic study of new materials for AMFCs. The target of a non-platinum-group-metal (non-PGM) catalyst AMFC is ambitious. This study of these novel materials gives insight into the critical issues in developing practical AMFCs.

- The project is well thought-out and planned very well. The researchers are investigating a variety of approaches, including different alkaline membranes and non-PGM catalysts. If the principal investigator (PI) wants to compare technologies (slide 4), he should add cost on the secondary Y-axis.
- The project team is pursuing some of the best strategies for improving the stability of the anion exchange membrane (its Achilles' heel), including fluorination of active carbon-hydrogen bonds on pendant cations and the use of polyphenylene frameworks.
- Los Alamos National Laboratory (LANL) is focusing on the right issues: improved conductivity, improved mechanical properties, and higher temperature operation for carbonate tolerance. More work is needed to prove the viability of non-Pt catalysts.
- The approach is to use a poly(phenylene)-based anion exchange membrane (AEM), which may have the potential to minimize the mechanical property degradation. Investigators also proposed the novel concept of using a perfluorinated ionomer as a spacer to stabilize the cation, though it is a challenging approach. The investigators should show good conductivity using the spacer concept.
- The PI's approach is very good, but not particularly innovative. The use of polyphenylene-based anion-exchange membranes for alkaline fuel cells is not new. Nevertheless, the PI did improve the performance (conductivity) and durability of his aminated poly(phenylene) membranes. The membrane water uptake for the high-conductivity films is substantial, and dimensional stability in a fuel cell may still be a problem. The PI's M-N-C catalyst work appears to be producing interesting results.
- The approach is to address the cost issue with PEM. Therefore, using polyphenylene or hydrocarbon as base materials is a good approach. Perfluorinated base materials are not going to address the cost issue. It is interesting to see that the team is trying to make a perfluorinated carboxylic acid-based AE ionomer to address the high cost of the perfluorinated sulfonic acid (PFSA) ionomer. The team should be aware of the fact that perfluorinated carboxylic acid polymers are more costly than their PFSA analog. Therefore, there is no point in using perfluorinated carboxylic acid for making an AE ionomer.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- Researchers have examined a number of approaches in some detail, and trends are emerging. Investigators have demonstrated non-Pt catalysts.
- Investigators have made very good progress toward the project objectives in terms of membrane development. The catalyst work is a bit more suspect. The PI ran fuel cell tests with Pt electrodes and oxygen; the use of both is undesirable. This reviewer wonders if there were problems making a membrane electrode assembly (MEA) with the PI's alkaline membrane and M-N-C catalyst.
- The project team has collected a lot of excellent data on both the membrane and catalyst concepts; however, some additional data need to be collected, such as from increasing the molecular weight of the HC polyelectrolyte and carrying out durability tests of the PFSA membrane material.
- Given the difficulty of the project, the team has made modest progress against the technical challenges. The M-N-C electrode development shows good promise. The team has not measured the performance of the M-N-C catalyst in an AEM fuel cell and has very little time to complete this task. The team should focus on fuel cell testing if it wants to complete the 500 hour durability study before the completion of the project in September 2011.
- LANL has demonstrated good performance at 80°C. Stability in NaOH is improved, but stability under fuel cell operation still needs to be proved. There is insufficient progress on membrane mechanical durability. The guanidinium-based perfluorinated ionomers have acid and cationic functionality—the impact of which concerns this reviewer.
- While good progress has been made in both non-PGM catalyst and membrane development, more progress on membrane stability is critical for success.
- The poly(phenylene) based AEM showed reasonable conductivity and relatively stable in NaOH. However, the perfluorinated ionomer spacer did not help to increase the conductivity and stability. The researchers need to perform more AMFC evaluation and explore a systematic characterization.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.1** for its collaboration and coordination.

- This is a difficult exploratory project that is being attacked by a group of qualified experts.
- There is clear, strong collaboration with Sandia National Laboratories (SNL) and the Jet Propulsion Laboratory (JPL).
- This project has a capable team.
- The project features relationships with industry, academia, and national laboratories. At this time, it appears that only SNL has provided insight. This reviewer looks forward to more concrete collaboration with industry and academia.
- The team has good collaborators and interactions with experts in the field. Because the work is focused on intermediate (10–50 kW [kilowatts]) power applications, the team should have partnered with a company that is experienced in the development of fuel cell portable power. Dr. S. Gottesfeld has significant experience in portable power fuel cells, and this reviewer hopes that the team is gaining good input through interactions with him.
- There are good interactions between LANL, SNL, and JPL. A closer collaboration with the non-national laboratory partners would allow the development of the most realistic targets possible.
- The contributions of the listed collaborators (i.e., SNL and JPL) are generally obvious (maybe a bit less so for the JPL connection). The input from others (noted as “Interactions” on the overview slide) is not apparent.
- The early-stage collaboration already showed some progress. Investigators should focus more effort on characterization and clarifying if the proposed mechanism will work.

Question 5: Proposed future work

This project was rated **3.3** for its proposed future work.

- The proposed work seems reasonable, and integrating the various membranes into MEAs for testing is critical. The presence of membrane in the catalyst layer and characterization of this work will be crucial for getting meaningful results in MEAs.
- The proposed future work is appropriate, given the budget and time.
- The proposed future work is strong, but it will really only take place at LANL. The project team should try to get collaborators more involved.
- The project is going to end in September 2011. The team should focus more on fuel cell testing using the products that have been developed so far.
- High molecular weight materials may not be sufficient for mechanical durability. The project team should consider composite structures. Researchers should only conduct MEA optimization if they develop sufficiently high-activity stable catalysts. LANL needs to down-select between Ag and M-N-C catalysts at some point.
- This is an ambitious plan for the short time remaining.
- Investigators need to prove if the perfluorinated ionomer spacer will work to increase conductivity and stability, fundamentally. They should also accelerate MEA fabrication and evaluation.
- Some of the future work tasks are of questionable importance based on results in the Program’s Annual Merit Review slides, such as the Ag catalyst work. Also, it is not clear why a perfluorinated polymer is needed in an alkaline fuel cell. Rather than develop new, alternative membranes to the PI’s aminated polyphenylene, work should focus more on MEA development and fuel cell performance and durability tests using hydrogen and air feeds (no oxygen).

Project strengths:

- The PI is a world-leader, and progress made thus far is very impressive and highly valuable. The proposed future work also covers all of the major bases.
- The team worked on the very challenging problem of anion exchange membrane and ionomer development. The team has gathered experienced scientists and laboratories for conducting such work. The team also has access to experts in respective areas through its interactions.
- The project represents an innovative approach to meeting DOE’s fuel cell needs.

- The reasonable air performance at 80°C is encouraging. The PI recognizes key issues that need to be addressed.
- This is a good, systematic study of new materials for AMFCs. The target of a non-PGM catalyst AMFC is ambitious. This study of these novel materials gives insight into the critical issues in developing practical AMFCs.
- The PI has made nice progress in making a more durable aminated poly(phenylene) membrane with good *ex situ* durability. Also, the PI's M-N-C catalyst work appears to be promising and moving forward.
- Anion exchange membrane electrolyte development is crucial for future fuel cell and catalyst investigation.

Project weaknesses:

- The project lacks more in-depth collaboration with industry.
- The team has put less emphasis on *in situ* qualification of its materials, and has instead focused more on *ex situ* qualifications. The team should put more emphasis on fuel cell testing to qualify its membrane, electrode, and ionomeric materials.
- The effects of long-term catalyst behavior are not under scrutiny. Still, these are early days, and the team probably will worry about long-term effects after some down-selection.
- The relevance of targets toward applications is unclear. The membrane materials are inherently brittle. The technology is only valuable if non-Pt catalysts can be used, which is unproven. The project does not have the resources to address the multiple challenges of this alkaline fuel cell system.
- Closer collaboration with the non-national laboratory partners would be beneficial.
- With a poly(phenylene) membrane that works well and a new catalyst (M-N-C) that shows promise, it is not at all clear why the PI is investigating poorly performing materials (i.e., fluorinated membranes and Ag catalysts). The project needs to focus more on examining working materials in an operating fuel cell. The researchers should stop looking for something better and prepare an MEA and alkaline fuel cell using the membranes and catalysts that now show promise.
- Progress is still too slow. The most important thing is to prove the concept at membrane, MEA, and fuel cell evaluation levels.

Recommendations for additions/deletions to project scope:

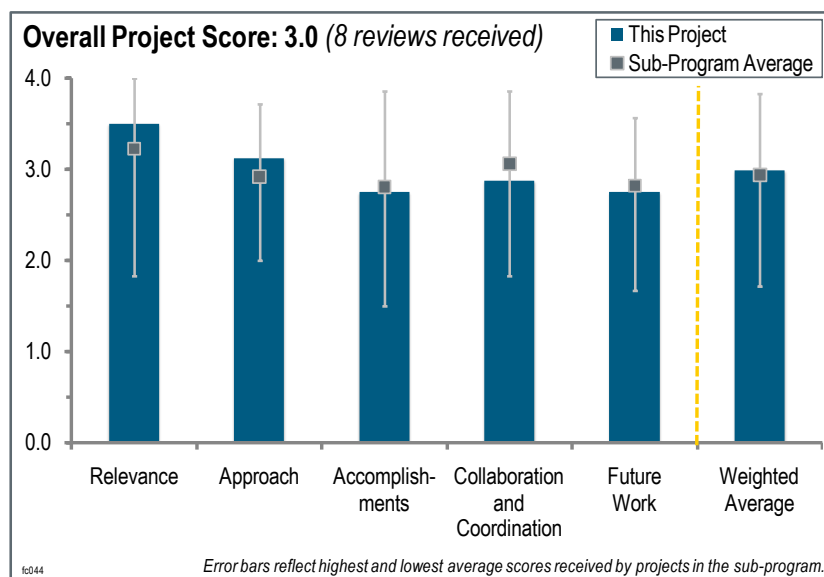
- The PI should pursue more interaction with industry and more durability studies on membrane and non-PGM catalysts (some are already planned).
- Polyphenylene and fluorinated polymers appear to be the most likely routes to increase stability. Catalyst testing in dilute alkaline media may be an easy way to screen for activity of electrodes. However, testing in concentrated electrolytes is more like the MEA environment, and this testing in concentrated electrolytes may be the way to find catalysts with more activity and stability for AE PEM fuel cells.
- The researchers should focus more on achieving a mechanistic understanding of membrane and ionomer/binder failure modes and performance limitations. They should not focus as much on meeting targets that are not tied to real applications.
- The project should have H₂ and air performance targets as well as H₂/O₂ targets, to address DOE goals more directly.
- The project team should stop working on Ag-based catalysts and perfluorinated membranes.

Project # FC-044: Engineered Nanoscale Ceramic Supports for PEM Fuel Cells

Eric Brosha; Los Alamos National Laboratory

Brief Summary of Project:

The overall objective of this project is to develop a ceramic alternative to carbon material supports for a polymer electrolyte membrane (PEM) fuel cell cathode. Ceramic supports must: (1) have enhanced resistance to corrosion and Pt coalescence; (2) preserve positive attributes of carbon such as cost, surface area, and conductivity; and (3) be compatible with present membrane electrode assembly architecture and preparation methods. Materials properties goals include: (1) high surface area; (2) high Pt utilization; (3) enhanced Pt support interaction; (4) adequate electronic conductivity; (5) resistance to corrosion; (6) a synthesis method or procedure amenable to scale-up; and (7) reasonable synthesis costs.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.5** for its relevance to U.S. Department of Energy (DOE) objectives.

- This project addresses the DOE goal related to improving the durability of PEM fuel cells.
- The search for novel materials that could potentially positively impact the current technical barriers is vital. This reviewer is happy that this small project was initiated.
- Alternative catalyst supports do have a very high interest, and therefore this reviewer believes that this project fits perfectly with DOE objectives regarding matching 2015 targets.
- This project is an effort to address DOE Hydrogen and Fuel Cell Program goals by using stable supports for catalysts.
- The corrosion of carbon supports for Pt electrocatalysts is well-known to be a failure mode in the operation of automotive fuel cells. While carbon supports can be made more corrosion-resistant (e.g., more graphitic), some corrosion is still caused during certain operating modes (e.g., freeze start and start-up and shut-down) despite the best system mitigations and purge strategies. This project attempts to address carbon corrosion by displacing carbon for a ceramic support. The project is relevant to the DOE efforts to help automotive fuel cell stack technology achieve 5,000 hours of real-world drive cycle durability.

Question 2: Approach to performing the work

This project was rated **3.1** for its approach.

- The approach is well-designed and focused on achieving DOE goals.
- The scope is not overstretched, yet considers a small number of disparate approaches to assess a broad field of possibilities. The flexibility of the approach to accommodate lessons learned (e.g., the inclusion of carbon) is a great asset.
- This project shows a good approach, as it is not easy to find alternative catalyst support materials. The investigators have done a good job so far, especially regarding evaluating different alternatives.

- The principal investigator (PI) has identified a nice class of materials. This reviewer liked the fact that the PI swiftly acted on underperforming systems with a no-go decision.
- The approach has focused on metal nitrides and oxides that are considered cheap, electrically conductive, and amenable to high surface area fabrication. The main questions with the approach center on the prospects for durability, particularly with respect to either corrosion or passivation. It is unclear if MoN_x will convert to MoO₃, or if NbO₂ will convert to non-conductive Nb₂O₅.
- For some materials, the project overachieved on surface area. Investigators have introduced a novel route for producing high surface area MoN_x, which appears promising. Some praise is deserved for executing no-go decisions on materials that were not promising (e.g., hexaborides and Pt/NbRuY₂O₇).
- The project suffers from very poor electrochemical characterization. Los Alamos National Laboratory (LANL) needs to improve its rotating disk electrode (RDE) methodology significantly and should try to follow literature approaches. The data look poor. There is too much emphasis on ex situ X-ray diffraction (XRD), which is not a particularly useful tool for oxygen reduction reaction (ORR) catalysts.
- The materials selection and considerations seem a bit unfounded—any magneli phase of TiO₂ will form a hydrous Ti oxide scale on its surface, and any electronic conductivity in the bulk will be irrelevant.
-

- The inclusion of an industrial partner is important to this project. It is a shame that a fuel cell company is not also involved, which would take the forthcoming materials to the next level of evaluation.
- This project may not have industry cooperation, but the investigators are doing well regarding collaboration with other institutes and universities.
- Two major types of collaborators are missing from the project: a stack original equipment manufacturer (OEM) or integrator, and a membrane electrode assembly (MEA)/catalyst supplier. The project needs to be directed toward standard RDE for Pt-containing catalysts, which OEMs will often do. A catalyst supplier would suggest surface techniques for understanding the lack of activity, not bulk techniques such as X-ray diffraction (XRD). Catalysts from UNM do not appear to play a major role in the project. Oak Ridge National Laboratory appears to have provided some valuable insight on what the Pt morphology is not, but the actual morphology is not clear. The role of SDC is also not clear.

Question 5: Proposed future work

This project was rated **2.8** for its proposed future work.

- The proposed plans address overcoming barriers.
- The project is appropriately focused at this stage, and things are going well. This reviewer is pleased that there is a plan to evaluate the chosen materials in fuel cells.
- The researchers are on the right path, so the proposed future work is also on the right track.
- The PI should focus more on electrochemical evaluation. While microstructure characterization is essential, it will be difficult to judge these materials without electrochemical characterization.
- Fuel cell testing of Pt/MoN_x should wait until activity has been established for the catalyst. The future work should particularly focus on using the standard perchloric acid technique for RDE, and then on using surface characterizations to understand why activity may be low. The aerosol Pt/MoN_x work at UNM should probably leverage what is learned from the investigations of the polymer-assisted-deposition-derived material's low activity.

Project strengths:

- The project is well-managed—the investigators met all of their goals on time. The team makes fast progress toward understanding the key properties of the support and made go/no-go decisions in a timely manner.
- This is a small project with clear focus and objectives. It includes the necessary skills, especially the scale-up expertise of the industrial partner. The flexibility of the project to down-select avenues of exploration and incorporate new ones that have revealed themselves in the learning process is true research.
- Very good analytical tools are available.
- The use of alternative materials is an area of strength of this project.
- Strengths of this project include its good team and collaborations, methodical approach, and swift go/no-go decisions.
- One strength of this project is the researchers' effort to look at alternative materials for ORR catalysts.
- The materials meet high level requirements—Mo nitrides and Ti oxides can be conductive, cheap, and made at high surface area. Another strength is the project team's ability to use familiar experiments to draw conclusions. LANL has used both XRD and TEM to draw conclusions about the conversion of carbon to Mo₂C, the structure of Pt, and Pt domain sizes. A final strength is the project team's ability to quickly eliminate materials from contention; the project has already made numerous no-go decisions.

Project weaknesses:

- The project is focused more on materials physicochemical characterization than on electrochemical characterization. Investigators need to report the Pt electrochemical surface area and electrocatalytic activity toward the ORR to understand the potential of a new catalyst. The conclusion about platinum-support interaction is not confirmed by the results of the electrocatalytic activity toward the ORR.
- This project is simply too small. It does not need to be bigger, just longer. A fuel cell partner would provide an advantage in getting the MEA structure optimized with these new materials, and would open the door for discussions between the materials producer and the end user.

- Mo may be inherently unstable. Electrochemical characterization, which is the most crucial aspect, is minimal to nonexistent.
- The project features very poor electrochemistry, and an overemphasis on *ex situ* materials characterization.
- Despite enhancing the ability to perform electrochemistry, the project is still not performing RDE according to standard techniques described in the literature. The use of XPS or, perhaps, X-ray absorption spectroscopy would benefit the project. OEM or MEA/catalyst supplier partners would assist in driving the material development toward targets.

Recommendations for additions/deletions to project scope:

- This reviewer would like to see this project extended with cost to continue the exploration of novel catalyst support materials and the understanding of the materials discovered in the project.
- The hydrophilic and hydrophobic properties of the support need to be characterized.
- The investigators should improve the project's electrochemistry. They should also rethink materials selection or justification with consideration of the actual ORR environment.
- Investigators should use surface techniques to examine the Pt oxidation state, Mo oxidation state (it is unclear if there is still Mo₂N on the surface), and the lack of oxygen reduction activity. The project team should add collaborators from within the fuel cell industry, and wait to conduct fuel cell testing until activity is shown for the MoN_x catalysts.

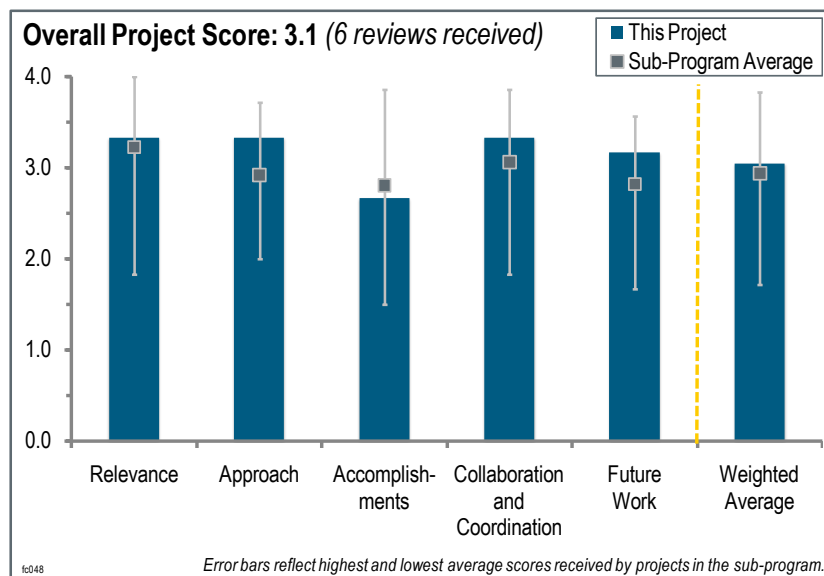
Project # FC-048: Effect of System and Air Contaminants on PEM Fuel Cell Performance and Durability

Huyen Dinh; National Renewable Energy Laboratory

Brief Summary of Project:

The overall objective of this project is to assist the U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program in meeting cost, durability, and performance targets in fuel cell systems. The effort is focused on system-derived contaminants. Project objectives are to: (1) select relevant balance of plant (BOP) materials based on physical properties and functionality; (2) develop *ex situ* and *in situ* test methods to study system components; (3) benchmark testing protocols and equipment among the different institutions; (4) screen BOP materials, identify and quantify system-derived

contaminants, and determine their effect on membrane conductivity and catalyst performance; (5) identify and select model species for further study; and (6) develop gates and strategies for selecting materials for in-depth analysis and durability testing.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.3** for its relevance to DOE objectives.

- This project is very relevant—this type of work is going to become even more important as fuel cells move toward commercialization and low-cost BOP systems are developed.
- Looking at system requirements in terms of their impact on the fuel cell is a critical activity to support fuel cell commercialization.
- BOP and system-derived contaminants have been shown to affect fuel cell stack durability. As cell and stack costs are reduced, BOP cost accounts for an increasing fraction of the total system cost, so cheaper BOP components are desirable. BOP components' stability and durability as well as the impact of degradation products on cell and stack performance are not understood well. This project intends to elucidate these issues.
- This project indirectly addresses durability and cost barriers by assessing the impact of system contaminants. A critical tradeoff may be between the cost of a fuel cell stack versus the cost for BOP components and system designs that avoid contamination or aspects of these items that cannot resolve the issues in the absence of stack improvements.
- As the project states, numerous elastomers, lubricants, structural materials, component degradation products, and other species can exist within the fuel cell system and cause degradation of the catalyst, membrane, or other stack components. In principle, so long as the species chosen for study are widely applicable—and not just specific to one system manufacturer—the project should maintain relevance for all customers. It is not entirely convincing that the fuel and air components of a fuel cell system will see some of the contaminants studied here. For example, the Zytels are often used in radiators or cooling systems. If there is a transfer between coolant and a reactant circuit, Zytel contamination will be overwhelmed by other failures.
- The evaluation of the impact of various system-derived contaminants on polymer electrolyte membrane (PEM) fuel cell performance is an important topic. My concern is that the study is system-specific and may only be relevant to the General Motors (GM) system, and not to the community at large.

Question 2: Approach to performing the work

This project was rated **3.3** for its approach.

- The researchers developed a solid approach, including a very thorough analysis and selection of materials, parametric studies to determine the effect of containments on fuel cell performance and durability, the identification of poisoning mechanisms and mitigation strategies, and even the development of a predictive model.
- The project team employed a good approach of focusing on a few key contaminant source areas as a starting point, and did a good job of determining criteria for selecting those areas. There is a good balance between model compound assessment and practical materials assessment. The project also features a strong initial focus on benchmarking methods prior to conducting detailed studies, and well-defined metrics for gate points in the project.
- The approach covers many required aspects of an impurity project, such as drawing up the list of possible impurities, selecting those worthy of study, figuring out particular components that cause degradation (if multicomponent mixtures are involved), and running ex-situ and in-situ tests to determine performance and material response to contamination. The approach does not explicitly mention whether specific vehicle or system data could be provided that would identify the presence of an impurity in a stack fluid input. It would also be interesting to know the concentration of the impurity during the particular operating conditions where that concentration would be at its highest. Without knowing relevant concentrations, in-situ testing may overestimate the resulting degradation.
- The researchers identify which compounds to test based on GM's experience, the wetted surface area, the total volume in the system, and the proximity to the membrane electrode assembly (MEA). The combination of ex-situ and in-situ evaluations can help to identify functional groups that cause cell degradation, which will guide future BOP materials selection.
- The overall approach is very good. Including another original equipment manufacturer (OEM) could possibly ensure broader relevance. The project is open to input from others, so this should be encouraged.
- The various laboratories appear to be focusing quite a bit of effort on obtaining the benchmark MEA performance. This duplication of effort may be diluting the impact and rate of progress of the entire project. Better partitioning of the effort along lines of expertise may speed up progress.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.7** for its accomplishments and progress.

- The project team has demonstrated good reproducibility of results for benchmark samples between laboratories. The team has also achieved good down-selection of materials systems to study based on strong criteria for likely impact in terms of contamination. Additionally, investigators have made good progress in benchmarking methods between various laboratories. Much of the initial project period has focused on establishing and validating analytical methods and demonstrating reproducibility across the different collaborator sites. This reviewer is looking forward to seeing study results at next year's DOE Hydrogen and Fuel Cell Program Annual Merit Review.
- The researchers have established analytical techniques and procedures, as well as reproducibility between participants. They have also conducted in-situ and ex-situ tests on some model compounds.
- This project features great development of analytical techniques and verification of reproducibility between three different laboratories. It includes nice studies of real-life containments on fuel cell performance, analyzing both membrane resistance and electrochemical surface area, measuring multiple doses, and attempting recovery.
- The investigators established reproducibility of test methods. The project has a good approach to establishing model compounds, etc., and is not just working with trade names. Ex-situ impacts on membrane conductivity and catalyst activity also need to be conducted via cyclic voltametry, along with recovery potential.
- The test stand validation among the project partners appears to be excellent, although some data from the University of South Carolina (USC) (on GM-assembled cells) and the National Renewable Energy Laboratory (NREL) (on NREL-assembled cells) appears missing. The researchers have done excellent work figuring out the leached components from the materials of interest and their effect on PEM conductivity and Pt contamination. Questions remain, however, as to how the dosages (or monolayers) and infusion levels relate to

concentrations that would be seen in a fuel cell stack. This reviewer wants to know if the model degradation species would make it from where the structural polymer (e.g., Zytel) is to the stack inlet, and in what concentration.

- The project is approximately 30% complete, and investigators have just completed the benchmark MEA performance at the three organizations. This should have been higher on the priority list or limited to one or two organizations in order to speed up overall progress. This reviewer expected to see more in-cell performance data and studies of the impact of more contaminants by this stage in the project.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.3** for its collaboration and coordination.

- This project features a strong team with complementary expertise. The tasking for each member within the project is clear.
- An automotive OEM is providing real-world insight into the compounds to be studied. The cell testing and H₂ quality expertise are strong.
- This project has good collaborations, especially with an industry partner such as GM to help define and select the most critical materials for testing.
- The project features a good set of collaborators, including industry fuel cell system and component developers, a university, and national laboratories. The project appears to be well coordinated and have good communication.
- The project has kept the main collaborators well organized. Testing among GM, NREL, the Hawaii Natural Energy Institute (HNEI), and USC is evident in the data. The roles of 3M and the Colorado School of Mines (CSM) appear to focus on membrane degradation, which does not seem to have been a major aspect of the project thus far. HNEI's work on the effects of silicone is interesting, and it would benefit the project to have deeper reporting on this topic.
- The accomplishments by GM, Los Alamos National Laboratory, and USC are evident. It is not evident what the other organizations (i.e., the University of Hawaii, USC, 3M, and CSM) have accomplished this year.

Question 5: Proposed future work

This project was rated **3.2** for its proposed future work.

- The future plans align well with overall project goals and intermediate gate decision points.
- The proposed future work is logical and methodical.
- The investigators have a good plan for future work. The durability testing will be especially interesting.
- The proposed future work is well laid out and has clear benefits for the industry. It will be important to maintain focus and activities on the analysis, characterization, mechanisms, and models.
- The proposed future work is vague in terms of the number of leachants to be studied.
- The plan does not indicate how the membrane degradation byproducts work will be incorporated into the remainder of the project. It is unclear if the "In-Situ Screening of BOP Leachants" has already begun. The Zytel study appeared to be the beginning of such an effort. The future work slide, however, indicates that this work-stream has not yet begun. The time provided for validating the experimental methods should be well worthwhile. Hopefully, validation overlap with in-situ screening tasks will not cause excessive iterations in testing.

Project strengths:

- This is a relevant and well-executed project that utilizes expert collaborators and produces results that are useful to the fuel cell community.
- The project has a very relevant, very structured approach that includes looking at mechanisms and models, and will produce practical and valuable knowledge.
- One strength of this project is GM's participation and background knowledge regarding system materials and potential contaminants. The diversity of the team is another area of strength.

- The presence of an automotive OEM is an area of strength. Without GM on this project, the entire effort would be impractical. GM is necessary to identify what materials actually exist in an automotive fuel cell system. Another strength is the presence of partners with deep fuel cell testing experience. GM, HNEI, 3M, and USC are all extremely capable of executing the testing. A final area of strength is the solid methodology toward understanding the impact of a given level of contamination. Given an identified contaminant and a certain level, this project is very capable of testing a fuel cell with contaminant feed, understanding whether the cell recovers under particular conditions, and performing in-situ diagnostics and post-mortem measurements to understand failure mechanisms.

Project weaknesses:

- The specificity of the project to the GM system is an area of weakness.
- One weakness is the lack of system and vehicle data on what contamination actually enters the stack. There may be fairly good reasons for why such information is not available. However, if a particular contaminant does not enter the stack or enters at extremely low concentrations, the testing in this project should either not be done or, at the least, should not be performed at an exaggerated concentration. Another weakness is the disconnect between membrane degradation byproduct activity and the remainder of the project. If the plan was not available at the time the slides were submitted, it is understandable that this disconnect exists. However, the project may already have plenty to do without studying membrane degradation byproducts, and it may be wise to leave this task out.

Recommendations for additions/deletions to project scope:

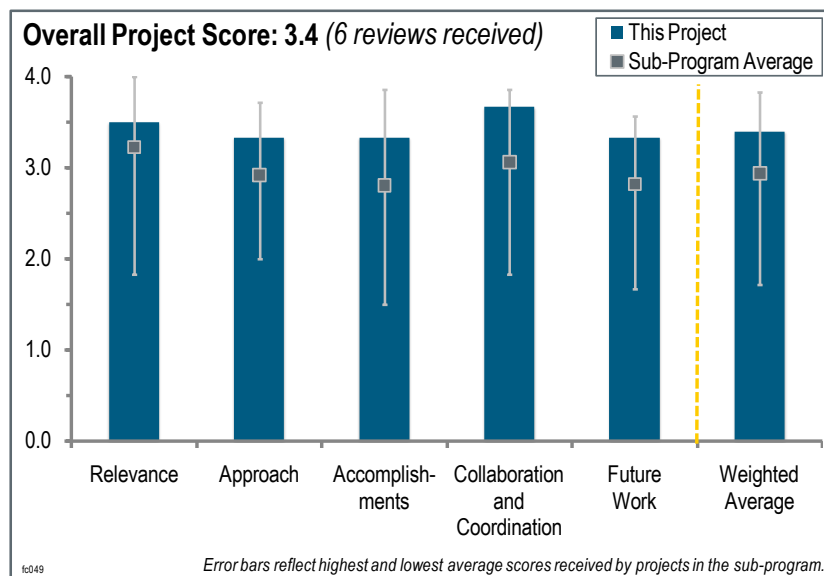
- The project team should indicate how resources can be devoted to the membrane degradation byproduct activity. There may be some chance that this task should be dropped in favor of investigating other system contaminants. It may be interesting to understand whether contaminants change gas diffusion layer/microporous layer surface energy. For in-situ screening, it would be good to report the level of contamination versus that which could be expected under certain vehicle operating modes.

Project # FC-049: Development of Micro-Structural Mitigation Strategies for PEM Fuel Cells: Morphological Simulations and Experimental Approaches

Silvia Wessel; Ballard

Brief Summary of Project:

The objectives of this project are to: (1) identify and verify catalyst degradation mechanisms, including Pt dissolution, carbon-support oxidation and corrosion, ionomeric thinning and conductivity loss, and mechanism coupling, feedback, and acceleration; (2) correlate catalyst performance and structural changes, including layer and unit cell operational conditions, catalyst layer morphology and composition, and gas diffusion layer properties; (3) develop kinetic and material models for aging, including macro-level unit cell degradation models, micro-scale catalyst layer degradation models, and molecular dynamics degradation models of the platinum-carbon-ionomer interface; and (4) develop durability windows for operational conditions and component structural morphologies and compositions.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.5** for its relevance to U.S. Department of Energy (DOE) objectives.

- This study of catalyst durability at low Pt loading is relevant to DOE fuel cell objectives.
- The project is relevant to automotive and stationary fuel cells, and addresses key barriers defined in the DOE Office of Energy Efficiency and Renewable Energy, Fuel Cell Technologies Program's *Multi-Year Research, Development, and Demonstration Plan*.
- The project supports fuel durability, which is one of the most critical objectives of the DOE Hydrogen and Fuel Cells Program.
- This project is an important bridge between the many catalyst efforts and reality.
- Working on understanding and improving durability while maintaining costs is obviously important to the overall objectives of the Program. It is not clear how the project is differentiated from or integrated with the other DOE projects on this topic.
- This project's objectives are to determine cathode electrocatalyst degradation mechanisms, correlate catalyst performance with structural changes, develop models for catalyst aging, and address other aspects that affect fuel cell durability.

Question 2: Approach to performing the work

This project was rated **3.3** for its approach.

- The approach focuses on degradation of the cathode catalyst and catalyst layer, an area where degradation effects are substantial. The project also features modeling with an extensive experimental component for validation. The modeling includes statistical sensitivity, which is not usually included and adds value.
- The project includes model development, experimental testing, and model validation. The researchers are using a comprehensive set of materials and structural parameters to help develop the models. The test plans included statistical sensitivity measurements and analyses, with up to 10% variability in each input parameter.

- The technical approach used in this project is adequate and in accordance with the set project objectives.
- The approach is fairly standard, and uses empirical data to validate developed models. The project partners seem to be doing the validation studies while Ballard is focused on model development. The approach uses a systematic variation of membrane electrode assembly (MEA) component parameters in looking for incremental improvements in performance over extended cycle times.
- The statistical approach using modeling is very nice. However, the molecular dynamics (MD) simulation work may or may not be good. The project lacks a description of the detailed approach, which is very important with this method. Also, the neutron imaging looks completely gratuitous to the main line of work. It is difficult to see how it all fits together. Overall, the approach is pretty much an unfocused hodge-podge.
- There seems to be a lot of work at very different length scales, but it is not exactly clear how well these various efforts will be integrated. The goal of developing a predictive MEA degradation model using a multi-scale approach seems good, but the presenters did not explain how molecular dynamics would be integrated with a one-dimensional model and a two-phase microstructural model.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- Investigators presented a wealth of test data and fits with model results, covering several different parameters. While some of the conclusions would have been expected, such as low surface area carbon catalysts being less susceptible to corrosion than medium and high surface-area carbon catalysts, others were less intuitive. For example, no additional significant degradation (i.e., carbon corrosion) was caused by spiking the upper voltage limit to 1.3 V (volts) (from 1.0 V) to simulate shut-down or start-up. The presentation also included results on the work of the various project team members, such as MEA water content, the effect of carbon type and catalyst heat treatment, and ionomer loading in the cathode.
- The investigators have made very good progress, and have demonstrated good agreement between generated models and experimental results.
- This project's progress seems reasonable to date—approximately one-third of the way through the three-year project. Thus far, the model validation with the experiments appears acceptable. The investigators have emphasized the experimental work, with much of the model development remaining to be done. The scheduled go/no-go decision will occur in about five weeks—hopefully more modeling results will be available then. The progress to date appears acceptable and pretty much on schedule.
- This project has shown good progress to date. Investigators have begun studies on the effects of carbon type, upper potential limit, and ionomer loading on durability. The researchers need to improve the model fit at high-current density.
- The project features very good communication of issues with the catalysts. The water-related work is a puzzle, and seems like a disproportionate amount of effort for the problem at hand. Surely neutron imaging is not necessary to determine if there is water build-up as the life tests proceed.
- The model results do not appear to correlate well with the actual performance data. It appears that the researchers are spending more time on statistical analyses and sensitivity studies than actually getting the model to accurately describe performance.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.7** for its collaboration and coordination.

- There is good interaction among the project partners—the collaborators are doing most of the experimental and characterization work. The interactions with the DOE Durability Working Group are positive.
- There is good collaboration among the partners, which include a fuel cell manufacturer, a national laboratory, and universities. The project team is also collaborating with other projects through the DOE Durability Working Group.
- The project lead is a major fuel cell developer. Team members include four universities and a national laboratory, resulting in a good mix of academic, research, and commercial organizations. In addition, the project participates in the meetings and discussions of the DOE Durability Working Group.
- The project activities are well coordinated among the team members.

- The investigators need a better understanding of how outputs from the model at one scale will match to inputs of the model at a completely different scale.
- The partners include several of the usual suspects who show up on every project. Not much insight can be gained from some of the work.

Question 5: Proposed future work

This project was rated **3.3** for its proposed future work.

- Implementation of the two-phase flow microstructural model will be key to improving the model fit at high current density. The experience of the team members at Michigan Technological University will be beneficial in this area.
- The planned future work is consistent with the project approach. The activities will include continued model development, materials characterization, experimental testing, and model validation.
- The proposed future work is well planned to advance the project's progress and is phased in a logical manner. It has appropriately incorporated go/no-go decision points and risk mitigation strategies.
- The plans for future work are reasonable, assuming a positive go/no-go decision in June.
- The investigators need to make sure the model is robust and can produce better predictions. They also need to include a go/no-go decision point for continuation with the microstructural model. Researchers should consider requiring more focus on the integrated model. The ultimate outcome of the MD model is unclear.
- It is unclear how models will be "validated". Neutron imaging does not seem to validate very much. The best parts of this work are related to catalyst dissolution, yet this is de-emphasized in the future work.

Project strengths:

- The project's strong team is an area of strength.
- This project has a strong team. Ballard brings a wealth of in-field experience regarding fuel cell durability.
- Strengths of this project include its strong team and comprehensive work plan.
- Highlights of this project include its multipronged approach to exploring key failure modes and its good experimental capabilities.
- This project logically combines modeling and experiments for critical issues, such as catalyst degradation and water management.
- This project's good raw data, statistical analysis that meets a real need, and catalyst evolution understanding are all areas of strength.

Project weaknesses:

- One reviewer felt there were no weaknesses.
- There was some discussion that the agreement between the model and experimental test data on cell voltage versus cathode Pt loading (the left plot on slide 10) was not as good as it should have been. This should be re-examined.
- The researchers need to further refine the base model before trying sensitivity studies and statistical analysis. They also need to better integrate the various project elements.
- The tools for analyzing water aspects seem way overblown for the level of detail needed.

Recommendations for additions/deletions to project scope:

- The investigators need to make sure the beginning-of-life model gives a good fit to the data from various MEAs (e.g., loadings and ionomer level) along the whole range of the current-voltage curve before progressing to degradation.
- The project team should incorporate the membrane thickness degradation into a model.
- The researchers should drop neutron imaging from the project.

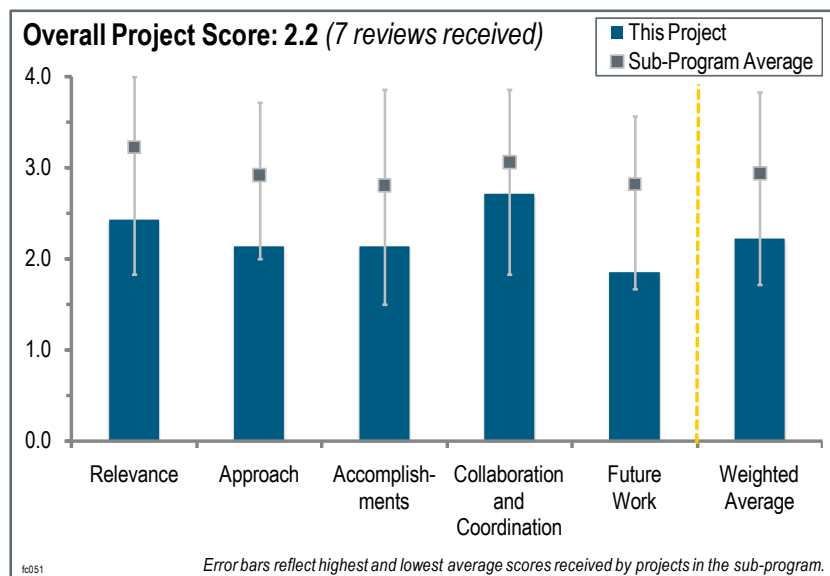
Project # FC-051: Fuel Cell Testing at the Argonne Fuel Cell Test Facility: A Comparison of United States and European Union Test Protocols

Ira Bloom; Argonne National Laboratory

Brief Summary of Project:

The overall objectives of this project are to: (1) develop standardized test procedures for evaluating different stack technologies; (2) characterize stacks and systems in terms of initial performance, durability, and low-temperature performance; (3) adapt the Fuel Cell Test Facility hardware and software as needed to accommodate the unique needs of different technologies; and (4) address barriers regarding durability and start-up time.

Question 1: Relevance to overall U.S. Department of Energy objectives



This project was rated **2.4** for its relevance to U.S. Department of Energy (DOE) objectives.

- It is critical to assess and benchmark state-of-the-art fuel cell technology, and to develop and verify standard protocol to do so.
- This project is useful for generating reliable procedures for benchmarking fuel cell technologies at the international level. It also gives DOE an independent assessment of fuel cell development.
- The primary goal is to look at how European Union (EU) laboratory results compare to U.S. results. This laboratory-to-laboratory variation assessment is interesting, but well off the critical path to success of the DOE Hydrogen and Fuel Cells Program.
- The testing being done at several locations is of value for calibrating test stations; however, it does not offer significant value on its own and should be a minor part of the project. Performance and durability test procedure improvements could reduce the error and time required to characterize a technology.
- The project is largely driven out of DOE headquarters; hence, it appears to naturally align with the Program's plans.
- It is not clear whether a "state-of-the-art fuel cell" will be available for this work or whether test conditions will be those used by industry as up-to-date hardware and operations are proprietary. Equipment that will be available will probably have been extensively tested by the supplier or provider if it is planned for commercialization, precluding any value in testing it at the Argonne National Laboratory (ANL).

Question 2: Approach to performing the work

This project was rated **2.1** for its approach.

- The principal investigator (PI) appears to approach the various tasks with a focus on delivering quality work and using the appropriate level of resources.
- The technical approach used in this project is adequate. It intends to standardize the fuel cell test procedures to objectively evaluate a variety of stack technologies to address the main commercialization barriers: durability and performance.

- The approach is reasonable, but this reviewer would like to see a more in-depth assessment of how the industry should be testing stacks to improve throughput and maintain accuracy. One of the objectives is to “provide the DOE with an independent assessment of state-of-the-art fuel cell technology,” but the stack concepts sampled are not extensive. This reviewer would rather see the results of several fuel cell stack concepts broken down by segment (e.g., automotive, backup, materials handling, combined heat and power, etc.).
- Through no fault of the PI, a fuel cell was selected that has nearly no relevance to stationary or especially transportation fuel cells. As such, it only shows that there is little laboratory-to-laboratory variation in making the same measurement in conditions of little interest.
- In terms of durability testing, the accelerated aging tests that are described as part of the activity and would truly represent aging mechanisms for a stack require a considerable amount of understanding of the conditions responsible for aging and degradation. This would be a huge task for this effort to have any value. Each supplier should already have this information on-hand, and this would not require duplication at ANL.
- Testing one stack is hardly “an independent assessment of state-of-the-art fuel cell technology.”

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.1** for its accomplishments and progress.

- Given the level of funding allocated, the project appears to deliver reasonably good value. Support of the standards activities seems good and should be highlighted a bit more for the purposes of the reviewers.
- The investigators have shown results for the comparison of the two protocols; however, they have not shown the durability data results they have generated so far.
- Tests replicated in the EU and at ANL showed 2% variation in data. However, due to strict operation limits of the stack chosen in Europe (not by the PI) the range of the test was very limited and the conditions were mild. There was, accordingly, no ability to compare protocols in the United States and the EU to see if the results are comparable or similar.
- Not too much can be expected in terms of stack testing because of the costs involved and the small amount of funds available. “Several fuel cell stacks and systems” and “most fuel cell test objects...” (slide 4) does not provide much information regarding what has been tested. The presentation did not include very much description of the 10-kW (kilowatt) polymer electrolyte membrane stack to correlate the data to stack evaluation.
- The results reported for the past year are essentially a beginning-of-life characterization test for one 10-kW stack. It is hard to imagine this taking one year and anywhere near \$300,000–\$500,000.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.7** for its collaboration and coordination.

- It was good to see the collaboration with JRC/IE for the EU protocol data.
- The project activities are well coordinated among the team players.
- This project attempts to connect several institutions worldwide. This is the strength of this project. If organizations around the globe can assess technology and share data, this would offer insight to the industry.
- The collaboration is suitable.
- The collaborations seem adequate relative to testing protocols, but they could probably be improved by working closer with the Los Alamos National Laboratory (LANL) group that performs similar functions. The collaboration necessary for standards development is significant and appears to be well done.
- There is not much evidence of significant partner involvement.

Question 5: Proposed future work

This project was rated **1.9** for its proposed future work.

- The plans largely continue on the current trajectory. As a service program, it is a bit difficult to assess the plans.
- This reviewer did not see any future work planned.
- The investigators intend to conduct aging tests, again over limited process conditions.

- It is not clear what the proposed future work is. There is no specific slide or bullet point that explains it.
- There are not enough funds to assess fuel cell technology for freeze characterization adequately.
- The presenters did not display any proposed future work, as there is none evident, or provide any planned expenditures.

Project strengths:

- It is important to assess and benchmark state-of-the-art fuel cell technology and develop a standard protocol to do so. The comparison between EU and DOE protocols was interesting.
- The support of the standards initiatives was an area of strength for this project.
- This project's strength is the ability of the international fuel cell community to work together on the standardized fuel cell testing procedures.
- This project is a good education tool on performance testing. The worldwide collaboration is a benefit.

Project weaknesses:

- The minute differences in the test results are not worth further discussion.
- The coordination with LANL on test methods could probably be improved.
- It is not clear how the standardized procedure will be generated and whether different procedures will be made for different fuel cell applications.
- If a test station is calibrated properly, there should be no significant difference in performance. Too much focus was put on calibrating tests between locations. Durability is harder to understand. The accelerated stress tests developed by DOE and other organizations are a good starting point; however, when testing on the stack or system level, the design of the stack or system has a huge influence on the stressors and degradation mechanisms. This might cause confusion and make a comparison between stack concepts difficult.
- The value of fuel cell stack testing is not obvious. It seems that the original equipment manufacturers and stack suppliers should be capable of evaluating their products, or potential products, if there is a market for them, as the market should dictate the stack performance.
- The researchers did not accomplish much, given the time and money spent. The results boil down to a beginning-of-life characterization test of one 10-kW stack.

Recommendations for additions/deletions to project scope:

- It would be nice if this activity would ultimately lead to a "global" test protocol.
- DOE should, through the Durability Working Group, establish an extensive set of parametric studies to evaluate the effects of different aspects of cyclic tests. ANL could take part of that work, which would facilitate better proactive task management and better use of the intellectual assets currently assigned to this project.
- Investigators should use an automotive fuel cell that can work in many circumstances and do this sort of work on EU tests methods and on U.S. test cycles. This would allow them to learn something worth knowing.
- The test procedures include only H₂ so far, and should also include reformat.
- With the remaining funds, the researchers should focus on assessing the performance and response of commercial and state-of-the-art stacks and systems. For durability assessment, they should focus on small-scale tests and work with industry partners to compare this to stack, and stack and system durability data.

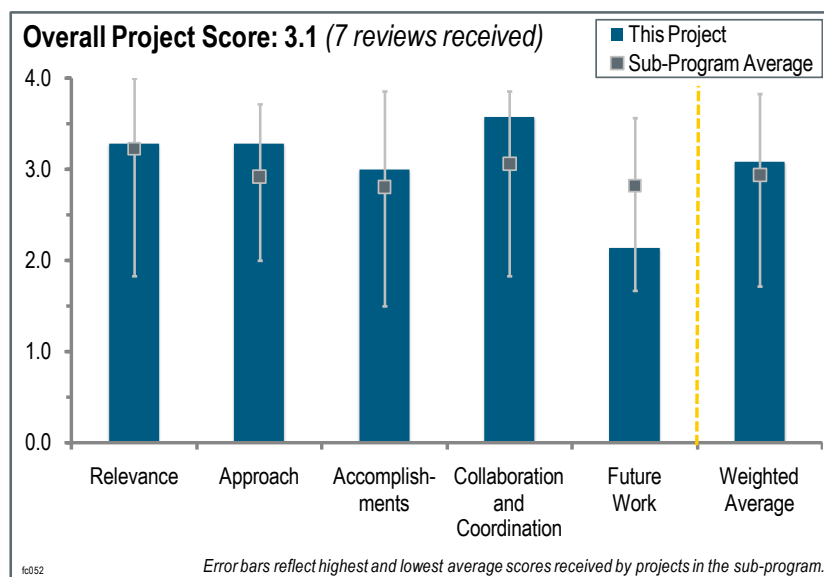
Project # FC-052: Technical Assistance to Developers

Tommy Rockward; Los Alamos National Laboratory

Brief Summary of Project:

This project involves Los Alamos National Laboratory's (LANL's) technical assistance to fuel cell component and system developers as directed by the U.S. Department of Energy (DOE). This project is expected to include materials testing and participation in the further development and validation of single-cell test protocols with the Fuel Cell and Hydrogen Energy Association. The project also includes offering technical assistance to Working Group 12, and the USCAR's U.S. DRIVE Fuel Cell Technical Team.

Assistance provided includes pursuing focused single-cell testing to support the development of targets and test protocols, and participating regularly in working and review meetings.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.3** for its relevance to DOE objectives.

- The objectives of this project are needed and support the fuel cell industry. The investment by DOE in fuel cell technology at LANL can be validated by this project's ability to assist industry, universities, and other national laboratories.
- LANL supports many individual projects and the DOE Hydrogen and Fuel Cells Program in general with cell testing and technical advice.
- The project seems directed to key objectives.
- This project is quite relevant because it provides valuable testing and evaluation services, which not all organizations can access. Furthermore, it engages the national laboratories and their expertise and facilities.
- The relevance of this project is entirely dependent on the relevance of the projects it assists. Assistance on neutron imaging to provide a low coefficient of thermal expansion holder may possibly benefit many projects. The use of a rotating disk electrode (RDE) to examine catalyst durability is an active topic of debate within the research community. This may or may not emerge as a relevant effort. Nafion® and Aquivion are fairly old perfluorosulfonic acid technologies. The reasons for comparing these materials are not quite clear. Assistance to catalyst and membrane electrode assembly (MEA) developers with low resources is relevant. This includes RDE, X-ray fluorescence (XRF), and cell measurements.
- This project is essentially driven by DOE headquarters, and so it is assumed to be driven by the *Multi-Year Research, Development, and Demonstration Plan*. It does seem a bit service-oriented, which seems strange given the intellectual capacity of the team involved.
- It is hard to evaluate this project relative to others and DOE objectives because it primarily supports other researchers. However, from the information the researchers have provided, it looks like LANL is providing a worthwhile service, and the overall strength of the Program is better for having this opportunity.

Question 2: Approach to performing the work

This project was rated **3.3** for its approach.

- The approach to individual issues described seems to be effective for developing an understanding of the issues.
- The approach is excellent. It covers most of the important tests, in particular electrochemical tests. This reviewer's only suggestion would be to add mechanical property testing capabilities. Working with the DOE working groups is excellent.
- LANL focuses on testing single cells for LANL and other DOE-funded developers, as well as on developing and evaluating testing protocols.
- The approach in this project is to help other projects. To that extent, this project has certainly been helpful to a wide breadth of researchers, although some degree of discrimination among efforts may be worthwhile. For example, a Nafion® versus Aquivion comparison may not contribute much toward future developments. The catalyst evaluation methods appear to have revealed weaknesses in MEA fabrication. For example, in the work with Pacific Northwest National Laboratory (PNNL), a cyclic voltammogram (CV) contains a slope indicative of electrical shorting. The choice of membrane and the cell assembly are presumably decided by LANL, which poses the question of whether LANL cell assembly methods are appropriate.
- The development of accelerated stress tests (ASTs) has aided fuel cell development, and the establishment of standard procedures has been a benefit.
- The team seems to have a focused list of priorities, primarily durability and cost, that makes good sense.
- It is clear that various researchers and commercial entities utilize the good technical capabilities at LANL to answer relevant questions about fuel cell technology.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- LANL has compared various membrane materials and started work on testing protocols. LANL has also been instrumental in developing hardware and techniques for materials characterization and in-cell water studies. LANL is supporting the Hydrogen Engineering Storage Center of Excellence by testing the effects of potential storage impurities on the fuel cell.
- The project generally seems to be reasonably effective at moving toward and understanding the objectives.
- The facility is available and up and running.
- The principal investigator (PI) appears to make good assessments of tactical issues that are proposed. The thematic inconsistency causes the depth of each study to be somewhat limited.
- The standardization of rotating ring disc electrode (RRDE) techniques furthers understanding of fuel cell catalysts and permits a sharing of data that were measured using a well-understood procedure. Experiments to validate stress tests are beneficial.
- The slide comparing Nafion® to Aquivion indicated that the materials were similar, but there were no conclusions on the slide.
- Making available scanning XRF to small companies is a great benefit for emerging companies.
- Making the fuel cell holder for neutron imaging for the National Institute of Standards and Technology (NIST) demonstrates cooperative interaction of federal resources, which benefits all.
- The XRF scanner appears interesting, but it may be more interesting to see if it can be validated against another technique (e.g., a destructive inductively coupled plasma quantification of Pt loading).
- The PI described an AST approach for non-platinum group metal (non-PGM) catalysts, but has not shown the results.
- It would be interesting to see how the new neutron imaging holder works over a range of temperatures compared to other holders—in other words, to see if the benefit of the holder can be shown.
- It would be good to show the diborane contamination results versus a cell run without diborane over the same period of time, to understand whether the difference is statistically significant.
- The project has produced interesting results that are not particularly coherent, but this reviewer would not expect them to be coherent.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.6** for its collaboration and coordination.

- The efforts with the other national laboratories are impressive. The development of state-of-the-art characterization equipment for fuel cell developers is a welcome activity for the national laboratories. DOE and LANL should be commended for this activity.
- LANL supports and collaborates with a broad array of developers (e.g., stack, materials, and H₂ storage).
- LANL seems to involve good collaboration with a wide range of institutions for finding the key issues, and seems to be well directed toward finding the issues to investigate.
- All of this project's collaborations are good.
- This project features good examples of working with large and small companies. Good support is noted for the Durability Working Group.
- Unlike other years, a list of collaborators was not shown on this year's poster. It is well-understood that the project does not exist with collaborators, but it would be good to see the breadth of partners.
- Other organizations with similar projects, such as NIST, have shown the ability to discriminate among proposals for work. This project would do well to do the same. Some of the efforts presented do not necessarily promise to advance the state of fuel cell technology or help others do the same. If there is not enough proposed work, then the question must be raised as to whether enough investigators are aware of the services, or whether others have sufficient capabilities and are not in need of LANL.

Question 5: Proposed future work

This project was rated **2.1** for its proposed future work.

- This reviewer recommends adding non-electrochemical test methods and procedures. The reviewer also recommends using ASTM test methods when possible, in addition to the accelerated test protocols being utilized.
- No future work slide was presented. It is presumed that similar valuable work will continue.
- The future work is unclear, although this could be due the nature of the project definition.
- It seems that future work will continue to be driven by requests and that there is not a very proactive plan.
- This reviewer would like to understand better what the process is for granting technical assistance by LANL. There does not need to be a formal review process, and work should not be slowed up to add bureaucracy, but information on what the decision process is or which projects should be given priority would be helpful.
- The presenters did not show a future work slide. Presenters did give some indication that there will be further development of RDE-based ASTs for non-PGM catalysts, but did not entirely present the plan for this.
- The overview says the activity is ongoing, and the percentage complete is not applicable. The fiscal year 2011 budget is \$570,000, but it is unclear what is going to be done.

Project strengths:

- LANL has proprietary access to many developers' technologies. This collaboration helps LANL interpret and understand the results in a broad context.
- The project seems to have a wide range of participants to address the issues in the fuel cell.
- Strengths of this project include the significant fuel cell knowledge at LANL, national laboratory engagement, independent site, and excellent test facilities.
- The collaboration is good and the technical caliber of the team doing the work is quite high.
- This project features excellent outreach and collaboration; timely results, which allow individual researchers to get results quickly without having to build or duplicate facilities; and an excellent technical team.
- The history at LANL in conducting fuel cell work is decades old and the institutional knowledge should still remain quite significant. Additionally, some measurements shown could be of great use, for example, the neutron imaging holder and the scanning XRF. Another strength is the project's wide range of collaborations with many organizations.
- The project strengths are the experience and technical expertise developed at LANL.

Project weaknesses:

- Visibility is not a weakness, but should be promoted sooner rather than later to the entire fuel cell community.
- Sharing more results would benefit the industry as a whole, and DOE-funded projects in particular.
- The reactive nature of the project limits the depth of each study undertaken.
- By its nature, the project is ad hoc and cannot fit with strategic objectives. It is more related to the tactical aspects of ensuring that other work is completed in a timely manner.
- The reporting of results is an area of weakness. This year the project team has either not reported results of some of its efforts, or reported results without indicating the significance of the results for its collaborators. The number of efforts that the project is associated with does not seem as large as in past years. Judging by CV presented in association with the PNNL project, cell assembly techniques may allow for electrical shorting.

Recommendations for additions/deletions to project scope:

- The investigators should add more testing capacity and capabilities.
- With respect to the Durability Working Group, achieving detailed parametric understanding of accelerated test conditions would be very useful, and is a serious undertaking.
- Additions and deletions are entirely dependent on the collaborators who approach LANL. LANL could consider performing some outreach (beyond just short courses) to the newly-funded DOE projects to ensure that newer projects understand what is available at LANL and that they take full advantage of it.
- The publication of a booklet describing the technical assistance available would be beneficial.
- This reviewer suggests that the researchers make the data accessible to the general community, when possible, through an online database.

Project # FC-054: Transport in PEMFC Stacks

Cortney Mittelsteadt; Giner Electrochemical Systems, LLC

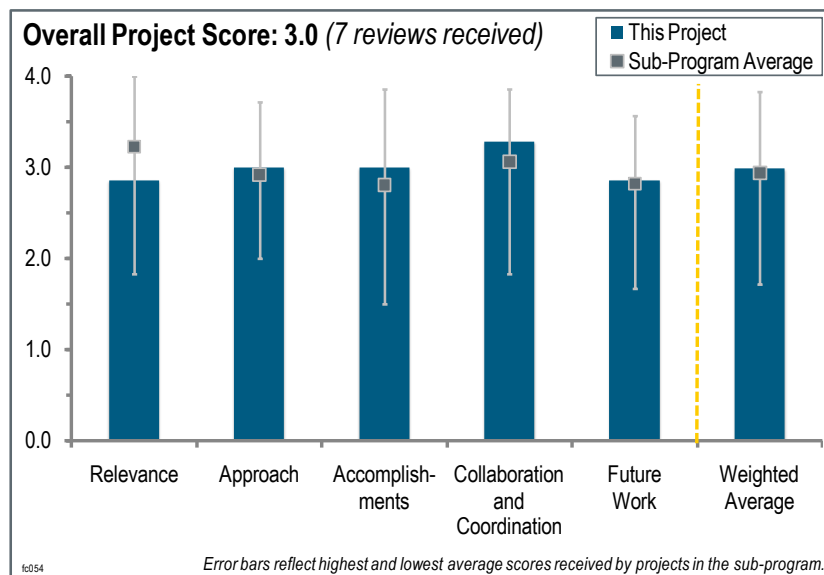
Brief Summary of Project:

The objectives of this project are to: (1) study transport phenomena in proton exchange membrane fuel cells including diffusivity, electro-osmotic drag, water uptake, and conductivity and (2) develop a transient three-dimensional model.

Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **2.9** for its relevance to U.S. Department of Energy (DOE) objectives.

- Transport is a critical issue for optimization and control of polymer electrolyte membrane (PEM) fuel cell stacks. This team has an appropriate focus on fundamentals, characterization, and modeling, and integrates these efforts to further understanding of overall fuel cell performance. The project has a good focus on key issues, and an appropriate work breakdown.
- This project will—and already has—improve fundamental understanding of water transport in PEM fuel cells, which will help improve performance and cost.
- This is a good project concerning understanding fuel cell behavior under sub-zero temperatures.
- Water transport in fuel cells is an important aspect, especially in the context of starting and stopping and operation under cold or wet and hot or dry conditions. This project is reasonably well aligned to the DOE goal; however, the ultimate goal of this project is unclear. If it is the delivery of a PEM fuel cell model, more details on how it is different from other water transport models and the benefits of this model to the DOE Hydrogen and Fuel Cells Program would help.
- The project addresses the water and thermal management aspects.
- This project's objectives contribute more to improving component performance than to overcoming stack operation. Both are, of course, linked, but it is difficult to see how the outcome of this project will be linked to stack operation.



Question 2: Approach to performing the work

This project was rated **3.0** for its approach.

- This project represents very good work. It has challenged some commonly held understandings and achieved some very nice work in getting to the root of some key issues, particularly regarding membrane transport and evaporation.
- The project includes some excellent experimental techniques to measure fundamental properties in new ways that enhance understanding. The project also includes some modeling to maximize the understanding obtained from the experiments.
- This is a good, comprehensive study on fuel cells at low temperature. Scientifically, it is an excellent approach. Technically, more factors should be considered; however, it is difficult to compare model and real factors.
- This project has a well thought-out approach, especially regarding identifying deficiencies in the literature of some of the critical and necessary experimental parameters (e.g., diffusivity), and measuring them accurately with well-designed experimentation. The researchers should publish these results in peer reviewed literature as

soon as they are completed. Testing the model with varying materials sets—especially materials with similar conductivity but very different diffusivities—is a very good idea.

- The project features a good combination of ex-situ and in-situ testing and modeling. It is not clear what operating conditions the focus is on. The integration of the components, where cell testing occurs, and how the new materials integrate into components are also unclear.
- The approach assumes that improved membrane measurements will be critical for advancing membrane technology. There is also a need to understand that membranes are inherently heterogeneous and subject to inelastic deformation with stress, which will modify internal geometries and alter transport rates. The result is that membrane characterization is necessarily challenging, and there needs to be far more profound thinking than is presented in this project in order to reach the stated goals.
- The presented approach is not complete. The researchers should integrate extension to the stack level, taking into account the temperature and relative humidity distribution along the different cells composing a stack. The researchers should also address stack operating mode (e.g., dead end, recirculation).

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- The investigators have conducted very good research to understand water behavior under low temperatures for different components of the PEM fuel cell.
- The measurement of the diffusion coefficient is impressive. The agreement between steady-state and transient measurements is very good.
- This project has already yielded some interesting and useful results. In addition, the investigators have established some capabilities that will certainly generate some additional useful results in the future.
- The researchers have made good progress so far, with the project being 48% complete. They have done some excellent work on the diffusivity. The segmented cell work may provide a low-cost alternative. However, the data should be compared with commercially available systems that have more segments (e.g., Baltic fuel cell), even though they might be more expensive. The project team should use more realistic diffusion media (with microporous layers) in the modeling (slide 14).
- There is extensive literature on correcting the diffusional gradients in the diffusion setup. Eliminating the inerts means that the water activity is at unity, which has been shown to have no interfacial resistance (similar to liquid water). Similarly, water uptake at unit activity is fast, so there are no long time constants. This is not the correct test, as the relative humidity is not changing. This reviewer recommends doing it with different water volumes. This reviewer wants to know if the water uptake really matters in the membrane surface, because there is ionomer around in a cell. The project team has made good progress on current distribution studies. The material studies are progressing with some promising results.
- The researchers have obtained interesting technical results this year, but none in clear relation with the project title (i.e., transport in PEM fuel cell stacks). This is the case for the experimental data as well for the model, which concerns single cells. Comparisons between the experimental and model results were not obvious at all.
- The experiments seem simplistic. Investigators did not mention reversible and irreversible structural changes, or developing a set of standard “preconditioning” protocols so that all membrane samples can be compared fairly. For example, the fluid network in perfluorosulfonic acids (PFSAs) is physically altered when the polymer is stressed and the geometries of the flow channels change.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.3** for its collaboration and coordination.

- The partnership appears correct. The project comprises all of the necessary expertise, from industrial to academic.
- The project has very good collaboration, and a very strong team with 3M and the United Technologies Corporation.
- The project features a good set of collaborators, including material suppliers, experimentalists, and modelers. The roles of the collaborators are clearly defined.

- The project has a great team that includes institutions with complementary capabilities and appears to be well connected and coordinated.
- The project features good individual efforts. This reviewer is looking forward to see how this works going forward. The reviewer suggests interactions with the LANL durability effort to the degree that mass-transport effects play a role in degradation.
- The project has a good team and collaboration, but it is unclear how material transfer occurs.

Question 5: Proposed future work

This project was rated **2.9** for its proposed future work.

- The future work is reasonable and achievable.
- The proposed future work is very good. The project team should keep up the good work.
- The following future work is very important and valuable for the Program: “Confirm model with performance, current distribution and water collection results.” The data generated from this project should be published or shared with other DOE projects, especially modeling projects. The properties of the baseline material sets should also be published. Extending testing to a more realistic automotive platform is also very useful for the Program.
- This reviewer looks forward to this project producing additional useful results.
- The future work is simply a continuation of previous work.
- It would be good to get capillary pressure and thermal conductivity data for the gas diffusion layer (GDL) materials. Model validation will be key.
- The proposed work is too focused on material selection and therefore appears to not be in accordance with the original targets dealing with understanding transports in stacks.

Project strengths:

- The competence of the different partners and the characterization techniques developed and applied during the first year are areas of strength for this project.
- This project’s strengths include the diffusion measurements and the refined understanding of Nafion transport. The resolution of the MacMullin number is very important.
- Investigators have done excellent experimental work in determining fundamental membrane-related parameters.
- This project features excellent and innovative experimental capabilities and results. It has an excellent team with the ability to provide and evaluate interesting materials and improve fundamental understanding. The investigators are transparent with their results.
- Giner Electrochemical Systems, LLC has identified quality partners.
- The researchers have done a good job regarding the characterization work and key parameters.

Project weaknesses:

- There was no discussion about the critical task of preparing samples with known pedigree and history, or showing the reproducibility between batches of “duplicate” samples. There was also no discussion about heterogeneous sample properties—for example, a Nafion membrane with “fish eyes”—and the way physical properties vary along the x, y, and z coordinates and with testing history and time.
- The work does not clearly relate to the announced technical barriers to overcome. The work done can be seen as a new “component study” among current and previous projects.
- The cell-level current distribution modeling seems to be less of an advancement than other areas. Current distribution cell work has been done before, but this could be refined to be helpful.
- More details are needed on how this model differs from those in literature and other DOE funded projects.
- As in all heterogeneous catalytic reactors, mass and energy transport are critical and a good reactor design needs to reflect those parameters. Today fuel cells are working well; so much of this necessary understanding has already been achieved. Refinement can certainly be useful and new measurement tools can help. The tasks proposed here are small steps forward that are probably necessary. However, this project seems to propose only small steps.
- There are no set targets for the material development tasks. The project appears disjointed.

Recommendations for additions/deletions to project scope:

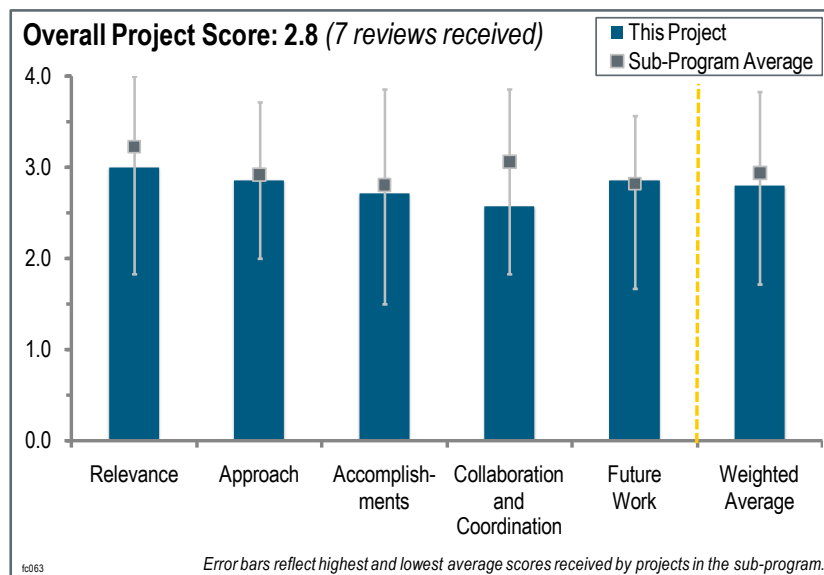
- The investigators should keep up the great work.
- The investigators should reorient the work in order to address the following specific stack transport issues:
 - The effect of temperature and relative humidity distributions depending on the cell position in the stack.
 - The effect of the stack operating mode (e.g., dead end and recirculation).
 - The effect of the stack design (e.g., membrane electrode assembly and bipolar plate design).
 - The effect of load cycles and start/stop cycles.
- This reviewer wants to know if it would be possible to extend some of these experiments to properties of the ionomer in the catalyst layer.
- There are many people doing fuel cell development. There needs to be thought about starting a project that builds a lexicon of terms and measurement techniques for membrane characterization, probably done with European Union and Asian partners. This could be done under the International Organization for Standardization auspices, and needs to include quality polymer suppliers. People need to think about how membranes and GDL properties influence energy transport, especially heat. There needs to be some good measurement technology for stress and creep and all of those dimensional parameters. This reviewer wants to know if there are acoustic emissions during fuel cell operation. Useful efforts need to focus on all transport properties. Water transport is a place to start and indeed considerable progress has been made in that area. The essential information is the understanding of the overall dynamics of fuel cell operation, of which water is just one part.
- The investigators should do more experiments at conditions relative to liquid water conditions (e.g., diffusion coefficients). There was mention of cold operation, but it is not reflected. The researchers also need to do more model validation.

Project # FC-063: Novel Materials for High Efficiency Direct Methanol Fuel Cells

Chris Roger; Arkema

Brief Summary of Project:

The objectives of this project are to: (1) develop ultra-thin membranes having extremely low methanol crossover, high conductivity, durability, and low cost; (2) develop cathode catalysts that can operate with considerably reduced Pt loading and improved methanol tolerance; and (3) produce a membrane electrode assembly combining these two innovations and having a power density of at least 150 mW/cm² (milliwatts/centimeter squared) at 0.4 V (volts) and a cost of less than \$0.80/W (watt) for the membrane and cathode catalyst.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.0** for its relevance to U.S. Department of Energy (DOE) objectives.

- Producing improved membranes with increased stability and selectivity is an important objective in the effort to increase durability and to reduce costs for direct methanol fuel cells (DMFCs).
- Improving DMFC performance and decreasing Pt loading are highly relevant.
- The project concentrates on membrane and catalyst development and offers clear, quantitative metrics for these materials. The investigators should indicate how these materials' metrics map to DOE's ultimate performance goals, including cost.
- The project is directly relevant to DOE objectives: it addresses two of the most important limitations of state-of-the-art DMFCs: (1) the loss of efficiency due to low fuel utilization as a consequence of high methanol crossover, and (2) poor cathode performance, although high loading of expensive Pt catalyst is used (usually 1.5–5.0 mg/cm² [milligrams/cm²]). Considering Pd as a low-cost alternative to Pt is questionable given that today the prices differ only by a factor of two.
- This project features good relevance in terms of addressing goals, but it is keeping too much information confidential and not providing sufficient information to allow the reviewer to judge, particularly with respect to the membrane support structure. There are several other research groups doing similar work that share details about the level of and mechanism behind the membrane performance. This project should be more open about its technical rationale. Nano-Pd may not be a helpful catalyst.
- The project focuses on DMFCs, but DOE targets for portable power are not presented and it is not completely clear that the defined targets will allow DOE targets to be approached.

Question 2: Approach to performing the work

This project was rated **2.9** for its approach.

- This project features two approaches to decrease methanol crossover: (1) using a polymer blend to decrease permeability and (2) decreasing the methanol oxidation kinetics on the cathode while not sacrificing the overall oxygen reduction reaction (ORR) kinetics. Both of these approaches are reasonable and worthy of investigation.
- The overall approach seems to be sound regarding the membrane selections. It is unclear if the additives provide a benefit compared to the base membrane alone, and it would be useful to understand the cost tradeoffs both in

production and materials of including the additives. The catalyst approach seems to be good for improving the cell voltage, but system effects of venting methanol may need to be considered in the tradeoff.

- Basically, the project is structured well, with two completely independent experimental approaches (membrane development at Arkema and cathode catalyst development at QuantumSphere [QSI]). The joint membrane electrode assembly (MEA) development is to be started after the first half of the project. The Arkema membrane approach of decoupling the membrane requirements (a polyvinylidene fluoride [PVDF] network as a mechanically robust, inert matrix filled with a polyelectrolyte) is neither completely new (Arkema has been working with similar membranes for PEM fuel cells a couple of years ago) nor unique (e.g., the development of dimensionally stable membranes at Giner Electrochemical Systems, LLC within this program [FC-036]). Nevertheless, this membrane approach is considered to be an auspicious option that might also be implemented technically within a few years. The success of the membrane development will depend on the degree of homogeneity of the PVDF matrix that can be achieved during its production. It is not clear how investigators plan on achieving this feat. If QSI believes that Pd-metal catalysts can outperform Pt/C (there are a few indications in literature that this might be the case), then Pt/C should be omitted completely for cost reasons. If QSI aims for a cooperative effect of Pt and Pd, alloy catalysts should be prepared and used. The addition of inorganic additives to tune the properties of DMFC membranes in terms of conductivity and permeation rate has been studied intensively in recent years with significant improvements achieved in only a few cases.
- Increasing membrane selectivity is fairly common using hydrocarbon membranes and, building on the blend approach Arkema has demonstrated, it seems likely that the investigators can perhaps slightly improve membrane properties using blends. The use of composite membranes containing inorganic additives has been studied for DMFCs, with results consistent with those presented, suggesting that no improvement in selectivity is achieved. Cathode catalysts do suffer from methanol-crossover-related mixed potential effects, but these are minimized by working at reduced crossover rates, and a screening of Pd alloys will not yield an improvement over current approaches.
- The approach is based on different PVDF grades for the membranes, plus an acidified silica particle filler to generate selectivity. There seems to be a lot missing in terms of measurement of properties. It would be very useful to conduct dynamic mechanical analysis in methanol/water mixtures, among other substances. Thermomechanical analysis swelling data, and other data, would be very useful to provide.
- The approach is somewhat Edisonian in that many materials are screened for desirable characteristics. That being said, the investigators have developed membrane and catalyst materials that appear to be promising. Materials cost and scale up is being addressed.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.7** for its accomplishments and progress.

- Investigators have prepared and characterized a large number of materials, and have been able to evaluate the performance of promising materials in MEAs. ORR catalysts show improved tolerance to methanol. Very little work on durability has occurred, but it is planned.
- The approach seems to be good, and the team has made good progress against the materials goals. The MEA performance—in particular the durability of MEAs and the membrane under long-term testing in practical fuel cell conditions—will be important.
- This project has shown good results so far with promising membrane materials, though the only three materials that meet the methanol permeation milestone have rather high areal resistance, and only two of the 11 materials identified as candidates have conductivities within a factor of two of the milestone. The project has produced impressive cell performance results in 10-M (molar) methanol, but has not demonstrated a significant benefit of adding functionalized silica. The methanol tolerance of Pd-containing catalysts looks promising so far, with good progress made toward achieving milestones. Durability data on these new materials is needed and is addressed in future work.
- The project has made very good progress toward performance goals, but progress toward understanding the mechanism has not been shared.
- A large number of membranes have been synthesized and screened for conductivity and permeation properties. Eleven of them are considered “high potential candidates,” and two of them nearly fulfill the requirements of the first membrane milestone. Some work on the scale-up of membrane production at Arkema has been performed with a membrane developed for H₂ applications. There is no indication that this has been repeated

with one of the “high potential candidates.” Arkema reports low electro-osmotic drag coefficients, methanol crossover current densities, and membrane resistance values for its novel membranes when used in MEAs. However, these values are obtained after calculations and corrections, and in comparison to an MEA based on a 7 millimeter thick perfluorosulfonic acid (PFSA) membrane not specified in terms of its equivalent weight. This makes the assessment of the results very difficult. The DMFC performance of MEAs with the new membrane (and commercial electrodes) is very good. However, the comparison with the data of the PFSA-based MEA is irrelevant as long as it is not further specified. Excellently high-surface areas have been obtained at QSI for a wide range of Pd black and Pd alloy catalysts. Unfortunately, no information on the homogeneity of the alloys is given. A mixed catalyst (Pt/C plus nano-Pd) was found to have a higher mass activity for the ORR in the presence of methanol than Pt/C during rotating disk electrode measurements. This is explained by the fact that nano-Pd is “methanol-tolerant.” Scale-up of catalyst production seems to be in time. Although MEA development has been started at QSI, no results have been shown.

- Membrane and fuel cell performances today show limited promise, but inorganic composite and cathode catalyst approaches do not. The project suffers from its poor ability to make relevant comparisons. For example, the data on slide eight is graphed in an inconsistent manner compared to literature as it should be in more consistent units (ideally independent of thickness) and not in semi-log form. Selectivity values of materials should also be given with some information shared about different ionomers used versus different blend concentrations and processing conditions. Additionally, fuel cell comparisons are performed under un-optimized conditions, making comparisons between materials more complicated and unfairly weighting the importance of methanol crossover. Comparisons with Nafion should be replaced with literature references of hydrocarbon materials that have already demonstrated far superior performance.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.6** for its collaboration and coordination.

- The collaboration with QSI is valuable because the project would be much weaker without the catalyst component.
- The collaborations are stated, but there is not a lot of detail on how efforts are coordinated.
- The project is essentially three disconnected projects at this point—independently investigating PVDF blends, inorganic composites, and cathode catalysts. There is no synergy except for that Arkema's ionomer is used in the inorganic composite. A DMFC system integrator should be involved.
- There is no indication for cooperation with external partners, although this would certainly be necessary for a thorough characterization of the novel materials.

Question 5: Proposed future work

This project was rated **2.9** for its proposed future work.

- The overall approach to the future work looks good. Increased MEA testing to evaluate the membrane and catalyst durability under practical conditions will be important and should proceed as soon as possible.
- The choices of milestones and the detailed plans for future work are logical.
- Future work is being planned in accordance with the milestones and will address appropriate issues such as durability, scale-up, and performance.
- The proposed future work presents a good path toward achieving the overall project goals.
- The cost analysis emphasis is good, although it is unclear that it properly focuses on system-wide costs. For cathode catalysts, the down-select criteria are not presented and the importance of durability tests and their use in MEA testing is not clear. A 50% reduction in Pt by itself is a poor metric, as it needs to have a proper baseline and cost analysis associated with its use to be meaningful.

Project strengths:

- This project has a good team with appropriate skills. The membrane approach has good potential for lower crossover and improved durability. The project features sound membrane development with evaluation of

membranes that are scalable and reproducible. There is good evidence of the capability to scale the catalyst materials if they show appropriate performance characteristics.

- This project features a good basis of materials available.
- The project partners have great experience and expertise in their respective fields. The project could continue successfully even if one of the experimental approaches should fail (using commercial membranes or electrodes as a fallback solution).
- The project is focused on critical components of the DMFC and has established quantitative metrics for improved materials.
- Overall the project is interesting and relevant.
- The project builds on well established Arkema PVDF blends and explores a class of ionomers that has shown improved selectivity.
- Arkema's extensive experience with PVDF-based polymer blends is an asset to this project.

Project weaknesses:

- While a methanol-tolerant cathode catalyst may improve the voltage performance of the fuel cell, there may be some concern that operating with a crossover that is too high will cause too much unburnt fuel to be vented from the fuel cell. This should be considered in the overall evaluation of the fuel cell performance, as it affects DOE system-level goals. Additionally, the use of lower stoichs on the anode should be considered at the stack level, as stoichs potentially affect the level of CO₂ removal and lead to anode problems (fuel starvation) that should be considered during the testing.
- There has not been enough basic understanding revealed and the rationale for using Pd is not clear.
- Catalyst optimization will probably be experimentally-driven. The project does not have any partners or cooperation to elucidate the role of the individual catalyst components (Pt/carbon and Pd-metal). The absence of an MEA specialist might slow down the MEA optimization (variation of the ionomer content in the electrodes and manufacturing technique (catalyst coated membrane [CCM] versus gas diffusion electrodes [GDE])).
- The critical path to meeting the DOE goals needs to be clarified. The reviewer asks if stacks will be fabricated and tested at some point, and if so, who will do the fabrication.
- Comparisons are made between Nafion and PVDF blends based on non-Nafion, but it is not clear how the blend membrane performance compares to the ionomer by itself at different levels of sulfonation. It is unclear at this time that blending offers an advantage, although it may. The extent of an advantage, if any, is critical to this project's relevance.
- This project is missing an analysis of anode effects. Even if optimizing an anode is outside of the defined scope of the project, the anode still affects the components that are the focus of this project (e.g., Ru crossover through the membrane to poison the cathode).

Recommendations for additions/deletions to project scope:

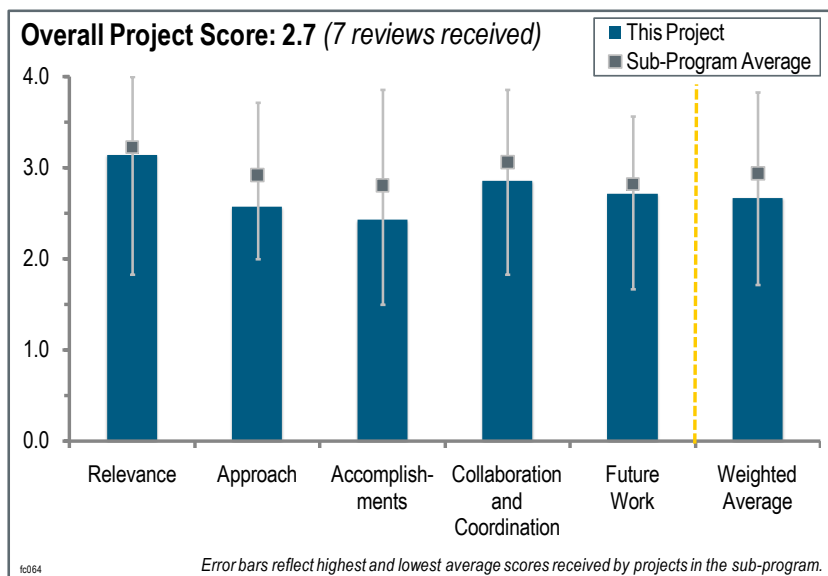
- Investigators should consider increasing MEA testing for durability and performance under fuel cell conditions and conducting an evaluation of the effects of any unburnt fuel and other fuel cell contaminant releases from the cathode exhaust.
- The project team should not look at the high methanol concentrations, but instead focus on 1–3-M methanol
- Investigators should focus only on blends and comparisons to the ionomer family used in the blends using reference materials that have shown high selectivity in the literature. The project team should stop working on inorganic additives and cathode catalysts.
- Durability studies should include the effect of Ru crossover (assuming Pt/Ru anodes are used).

Project # FC-064: New MEA Materials for Improved DMFC Performance, Durability, and Cost

Jim Fletcher; University of North Florida

Brief Summary of Project:

The overall objectives of the project are to increase membrane electrode assembly (MEA) functionality and internal water recovery, which facilitates system simplicity and increases power and energy density, as well as reduces costs to address the U.S. Department of Energy's (DOE's) consumer electronics goals. The specific objectives of this project are to: (1) improve the performance and durability of the University of North Florida MEA design to increase power and energy density and lower costs, (2) develop commercial production capabilities to improve performance and lower costs, and (3) increase catalyst stability and lower loading.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.1** for its relevance to DOE objectives.

- This project's water barrier layer idea is excellent for addressing DOE Hydrogen and Fuel Cells Program goals. The development of better catalysts is also very relevant. The project leverages previous PolyFuel membrane technology to provide approaches to costs.
- This project is very relevant to DOE objectives, particularly cost and durability.
- Improving direct methanol fuel cell (DMFC) performance and durability and simplifying balance of plant (BOP) is relevant.
- The project is relevant to DOE objectives because it addresses the most important limitations of state-of-the-art DMFCs: (1) low catalyst stability and high degradation rates and (2) poor water management and low energy density due to a complex water management system. It is difficult to understand how the catalyst loading is reduced to decrease the costs.
- The DOE goals are clearly stated, as well as progress toward these goals. The investigators are to be commended for their honesty in providing realistic numbers, given the fact that they fall short of the DOE goals. However, it is unclear how the investigators plan on meeting the DOE goals.
- The project does a good job of documenting performance as of 2008 and comparing that performance to DOE and project targets. The project targets are often far below the DOE targets.
- DMFCs are considered an important part of DOE's fuel cell commercialization plan, and this project addresses important issues in DMFC development such as BOP, mass and volume, and the deleterious transfer of Ru from anode to cathode, though it does not appear to address the methanol crossover issue.

Question 2: Approach to performing the work

This project was rated **2.6** for its approach.

- This project features a very good approach involving systems and scale-up factors. Degradation problems are a major issue, but this project appears to provide sensible approaches to understanding and solving this problem.

- Modifying water transport characteristics is a good approach to simplifying the system and improving performance. Increasing Ru stability in the anode is critical to improving durability.
- The barrier layer and anode catalyst work are well aligned to address the barriers. The work on methods that may be applied to commercial processes is also positive, even if it may be a bit outside of the scope of the DOE barriers.
- The system design approach to avoiding active liquid water recovery is a key aspect of enabling DMFC fuel cell systems and has been a problem for companies for some time. It is not clear that this project does anything but continue along similar lines of the approach applied by PolyFuel (as well as MTI and others), for which a much larger investment had been made without achieving commercial viability. Much of this approach is dependent on a barrier layer that presumably needs to allow proton conductivity while preventing water transport, as these two processes tend to be coupled and similar approaches have been employed. It seems unlikely that the approach will be sufficient to enable commercialization.
- The approach appears to be made up of several independent activities. There are no quantitative metrics for the materials properties that are being developed or critical path to meeting the DOE goals.
- This reviewer could not understand the approach to reduce the MEA costs by increasing the durability. There is no experiment comparing the performance with high- and low-loaded anodic catalyst layers. The ultra-stable anode catalyst is compared with the E-TEK Pt/Ru catalyst, which is not state-of-the-art and no longer commercial. Durability is shown in continuous operation mode—normally this should be measured in cycling on-off cycles, focusing on the Ru-degradation.
- This concept avoids the BOP required to capture product water from the cathode by using a barrier layer on the cathode that forces the water back out through the anode. This means that the cathode is always operating in a fully-flooded condition that significantly reduces the already too-low power density of the standard DMFC. This issue should have been raised early in the presentation, but it did not appear to be raised at all until the question-and-answer section. While reductions in BOP mass and volume could perhaps compensate for the additional volume and mass in the stack forced by the flooded operation of the cathode, the project needs to address this trade-off more directly and openly, as well as in a quantitative manner.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.4** for its accomplishments and progress.

- Good progress has been made in this project. The degradation issue is a slight setback, but that possibly was to be expected. Catalyst scale-up from Northeastern University (NEU) is a critical item, and quality control needs to be improved.
- Within the limitations imposed by the flooded cathode, this project appears to have made fairly good progress in implementing the concept. Constant-operation durability is good, and the investigators conducted good detective work on the durability problems associated with discontinuous operation that seems to be pointing the way toward a solution to this problem. Progress with the ultra-stable anode from NEU in the past year seems to have been limited to a slight scale-up in synthesis that is still inadequate for the needs of this experimental program, meaning that a complete retuning of the synthesis was deemed necessary.
- The exact positioning of where the project is with respect to the milestones is a little unclear, but the project progress on catalyst work and barrier layer scale-up is significant. Some progress was made on MEA development and durability, although the durability of the barrier layer looks like it impeded things slightly.
- There is no comparison with milestones inside this project. There should be a water flux (water permeation) measurement through the MEA and there should also be a total system verification that the system water is running autonomously over a certain time. Some information about the Recovery Act project H2RA-004 (conducted by the same principal investigator) would be helpful.
- An improvement in anode catalyst performance by Johnson Matthey is a meaningful advance to the field, particularly with decreased Ru dissolution. However, it is unclear that novel catalyst development at Johnson Matthey is part of the project, as little information other than performance was given and no detail regarding catalyst development was presented. Very few documented results were presented.
- The decrease in Ru dissolution with ultra-stable anode catalysts is valuable, both in terms of preserving anode performance and reducing Ru crossover to the cathode. However, this appears to be the same work that was presented last year. The effect of this project on cell performance is hard to gauge. Some performance results were provided, but the operating conditions were not clear. The project is oriented toward improving

performance by changing water management characteristics, but the presentation lacks a clear indication that performance has indeed been improved within the review period. The durability results are encouraging.

- It is unclear how the catalyst development efforts will impact the project. Apparently, a commercial catalyst was used for the MEA performance and stack data, but it was not clear from the presentation. There are no metrics for the new materials being developed. This reviewer wants to know what catalyst performance metrics will be required to meet DOE's goals, and what the overall plan is for introducing these materials into MEAs and stacks. There is a serious degradation problem that has yet to be solved.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.9** for its collaboration and coordination.

- This project features good collaborations. The source of the PolyFuel membrane is not specified—presumably the investigators have leftover material, but it is unclear who will make the membrane later. The project team is well balanced, and the role of Johnson Matthey toward commercialization is good.
- There is good cooperation with the external project partners. Johnson Matthey's position is not clear. It is unclear if there is any catalyst development occurring besides the development involving the barrier layer coating.
- The role of each of the partners is well-defined. Collaboration and coordination between the partners appears to be going as outlined in the project plan.
- Johnson Matthey is a strong addition to this project. It is unclear if the University of North Florida (UNF) would be in a position to commercialize this technology or if another DMFC systems developer should be involved.
- Johnson Matthey and NEU are good collaborators for this work. With several catalyst developers involved, the University of Florida's role in catalyst development is unclear.
- The partners appear to be working well together. During the preparation of the slides for this presentation, somebody should have raised the issue of discussing the negative consequences of flooded cathode operation.
- It is not clear how the collaborations and coordination efforts are working together toward meeting the DOE goals.

Question 5: Proposed future work

This project was rated **2.7** for its proposed future work.

- The proposed future work is well focused on the issues that need to be solved.
- The choice of milestones and the detailed plans for future work are logical.
- The future work proposed is good, although it should be more clearly tied to milestones and deliverables. Some mention of how UNF will meet the go/no-go decision for 500-hour durability at 60 mW/cm² (milliwatts per centimeter squared) should have been included.
- The plans to surmount the durability problems associated with discontinuous operation are good. The project needs to face the issue of whether the smaller, lighter BOP of this approach (though the airflow must be increased) is favorable to the smaller, lighter stack of conventional approaches. Therefore, a detailed engineering comparison would seem to be an appropriate part of future work.
- The first order of business should be to determine the cause of the degradation. A clear plan with quantitative metrics and intermediate milestones is needed, as well as a path to reach the quantitative performance goals.
- The future work, much like the project, has several disconnected thrusts. It is exceptionally broad, focusing on seven different bullets in slides 21 and 22, and includes aspects from barrier layer optimization to stack testing. A focus on scale-up production capability demonstrates a significant void in the catalyst team and approach.

Project strengths:

- The project partners have experience and expertise in their respective fields.
- Using the barrier layer is an interesting concept that can potentially simplify the system and reduce its cost.
- The investigators have managed to assemble a DMFC system that has reasonable performance when operating continuously.

- One area of strength for this project is the inclusion of Johnson Matthey—a leader in current DMFC catalyst materials.
- This project is an innovative idea for improving water management, and has good catalyst development partners.
- This project features an interesting concept to reduce DMFC balance of plant size and mass. The project has good, continuous-operation durability and has made good progress on the barrier layer, including work to address apparent leachates that produce the poor discontinuous operation durability of the cathode catalyst.

Project weaknesses:

- The project might need some more chemical help on the degradation. The identity of the supplier of membranes is not clear.
- Normally, the DOE lifetime goal for portables is 5,000 hours, not 2,500.
- As the principal investigator outlined in the talk, the durability of the barrier layer is a concern and needs to be fixed.
- The power from the new systems is not as high, but the overall power on the system may be about the same because the water recovery components will not require power.
- There is a lack of science to help advance the topic for the community. The focus on the liquid barrier layer is very product specific, and it is unclear if improvements or advances that would make the product commercially competitive are possible. The project is exceptionally dependent on a single architecture that has been explored in detail without meeting commercial requirements. This is in contrast to an active system such as the one provided by Smart Fuel Cells, which has commercially available products today based on an active water recovery system.
- Areas of weakness include the lack of focus and lack of reporting on MEA or system performance improvements yielded by the component development efforts.
- Inadequate attention was given to the negative impacts of cathode flooding on stack performance. Little progress has been made to date on incorporating the NEU anode catalyst, which is purported to provide improved durability and reduced Ru transfer to the cathode.
- The project lacks focus.

Recommendations for additions/deletions to project scope:

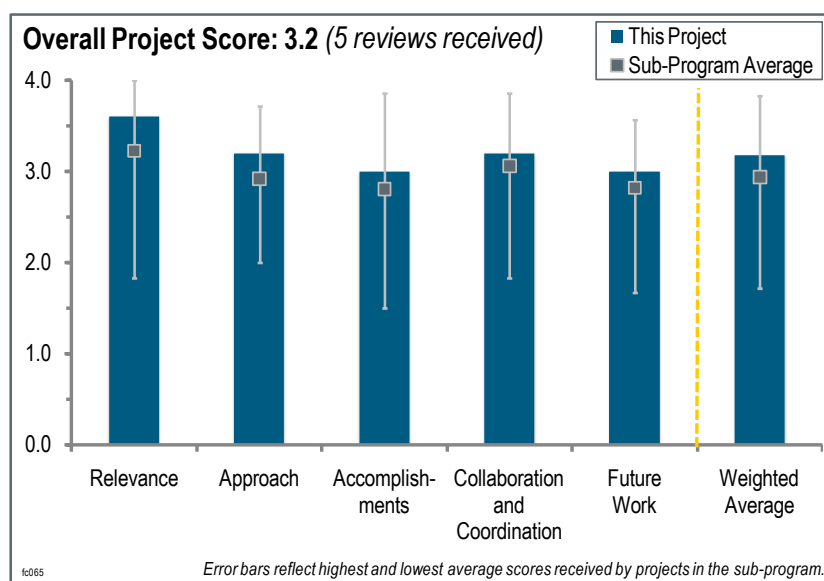
- None. The principal investigator mentioned that the project team is focusing on addressing the durability of the barrier layer.
- The project team should show more performance data under real operation conditions to demonstrate that the system is running water autonomously and with low degradation rates (the approach is to reduce costs by increasing lifetimes). Some information about the Recovery Act project H2RA-004 would be helpful.
- The overall objective of this project is to simplify the BOP for the DMFC. The investigators should demonstrate how this approach compares to commercial and emerging DMFCs. They could compare their system as it exists today and with expected future improvements with more conventional DMFCs. If they do not solve the degradation issue, the project should not be continued.
- The project team should focus on cost analysis to see if even the best imaginable barrier layer would enable commercial competitiveness.
- The investigators should conduct a detailed engineering system analysis of this approach versus standard DMFCs that must recover water from the cathode air stream.

Project # FC-065: The Effect of Airborne Contaminants on Fuel Cell Performance and Durability

Jean St-Pierre; Hawaii Natural Energy Institute

Brief Summary of Project:

The main project objective is to identify the currently unknown effects of many airborne contaminants on membrane/electrode assembly materials, and mitigate resultant adverse impacts such as hindering system performance and durability. Specific objectives for this project are to: (1) characterize, analyze, understand, and prevent the harmful effects of airborne contaminants that have the potential to reduce the performance or durability of polymer electrolyte membrane fuel cells; and (2) disseminate this information in a useful form to industry and other end users.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.6** for its relevance to U.S. Department of Energy (DOE) objectives.

- This project focuses on identifying airborne impurities that might degrade performance in stationary applications. This could be important to the ultimate end user for systems operating in an industrial or hostile environment.
- The project is relevant to the objectives of the DOE Office of Energy Efficiency and Renewable Energy, Fuel Cell Technologies Program's *Multi-Year Research, Development, and Demonstration Plan*. The activities are aligned to the overall DOE Hydrogen and Fuel Cells Program goals. A thorough understanding of the effect of airborne foreign contaminants, and hence developing a mitigation strategy to prevent the adverse effects of these contaminants on membrane and electrode assembly materials, is crucial to achieving high efficiency and durability in fuel cells. This study will also lead to the development of necessary analytical techniques that can be broadly used by the fuel cell industry.
- It is very likely that the initial operation of fuel cells will be in markets where additional incentives will be offered for clean air vehicles. Therefore, an appreciation for how airborne pollutants may affect fuel cell performance is critical.
- This work has good relevance to the Program objectives. The results can apply to both transportation and stationary applications.
- The level and type of work described in this project are necessary to help evaluate the degree of difficulty presented by the wide range of air and cathode operating conditions that fuel cell vehicles will face. This is important in determining the effect of contaminants on platinum-group catalysts at lower loading in the cathode, as well as identifying the degree of importance of implementing fuel cell protection from different contaminant species.

Question 2: Approach to performing the work

This project was rated **3.2** for its approach.

- Considering four broader milestones—such as (1) contamination studies, (2) real-world operations and mitigation strategies, (3) model development and applications, and (4) outreach to the industry and research communities—is an appropriate approach to address this problem. The go/no-go points are also aligned with the project.
- Overall, the approach is good. The investigators picked contaminants after a literature search based on what had not been previously studied and also to represent classes of molecules likely found in air. The screening study was done at 45°C to enhance adsorption to the catalyst; however, this neglects the fact that oxidative cleavage may occur at hotter and drier conditions, so fuel cell performance degradation that may occur at realistic operating conditions may not be captured. The group needs to collaborate with The National Center for Atmospheric Research (NCAR) or the U.S. Environmental Protection Agency (EPA) to ensure that all likely containments are identified, as crucial reactivities may be missing.
- The approach is thorough—187 airborne, 68 indoor, and 12 roadside contaminants were identified. The Hawaii Natural Energy Institute consulted with numerous organizations involved in the development of fuel cell systems when paring down the list. Fundamental models will be developed to predict the effects of various levels of contaminant on fuel cell operation and lifetime. Care is being taken to not duplicate other ongoing contaminant studies.
- Conducting a survey of potential impurities and creating classes of those impurities is tolerable; however, surveying current users to determine any operational problems they may have encountered might be a better approach. An alternate approach would be to consider potential stationary applications and identify potential airborne impurities that may exist in each application's operating environment, and then study their impact on fuel cell performance. It was noted that impurities, such as ammonia and sulfur compounds, were excluded because they have been extensively studied elsewhere.
- The elements of the investigation and the breakdown of activities within each element seem to be based on past experience that was developed while exploring the effects of H₂ fuel contaminants on fuel cell operations. However, there needs to be a closer tie between the learning in task 1.3 and the mitigation activities in task 2.2. The two should not be described as independent and unrelated tasks, but information developed in task 1.3 should serve as the base from which the task 2.2 work proceeds. Also, the extension of modeling (task 3) single species to include mixtures should involve consideration of the analyses of chemistries involved in the functional group's effect on the cathode degradation.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- Identifying 187 airborne contaminants, 68 indoor pollutants, and 12 roadside species that may have potential adverse effects on fuel cell performance is an outstanding accomplishment. Studying some of the down-selected contaminants to understand their adverse effects on fuel cell performance and their respective recovery behaviors is also a great accomplishment.
- Because of the strength of the team and the plan description, the progress has been quite good. Continuation into the next phase of activity should incorporate the identification of contaminant species in the cathode effluent in order to help the project team understand the mechanisms involved in performance degradation causes.
- The group has completed an initial screening study and is making good progress. There needs to be a discussion of errors. The fuel cell testing must be completed at more realistic operating conditions that reflect today's state-of-the-art stack operation and any future projected operating conditions. Common containments such as isoprene, particulate matter (< 10 microns), and others should also be considered in collaboration with an expert in air pollution.
- Thirteen contaminants were selected and tested since the last Annual Merit Review, which is a pretty good accomplishment. Two methods for ranking the severity of the contaminant effect have been developed. Each method leads to a value for a parameter called a selection criterion. The rationale for choosing between the two

criteria is not clear. The connection between the numerical values of the selection criteria and fundamental understanding of the mechanisms is not obvious. Recovery exceeding the initial loss in performance was observed in some cases; this is not explained.

- This project is only one year into its four-year plan and seems to be off to a slow start, perhaps due to its systematic approach to impurity selection. Mitigation studies will be the important result of this project—given that the right impurities are chosen.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.2** for its collaboration and coordination.

- United Technologies Corporation (UTC) and Ballard should be able to provide valuable input when determining which impurities should be studied.
- Involving two stack manufacturers, UTC and Ballard, in the project will help the team validate their learnings and hence validate the mitigation strategies from the project directly in real-world situations. The collaboration with the Center for Clean Energy Engineering at the University of Connecticut is also good, as this group has many years of experience in fuel cells.
- Numerous collaborators were mentioned—the most significant appears to be through a subcontract with Ballard that involves helping to identify contaminants for testing.
- This project features good team composition. As the project progresses, however, it is imperative that the cohesion continues in order to keep activities from stove-piping and benefit the overall understanding of how to mitigate the range of contaminants' deleterious effects on vehicle fuel cells.
- The interest group has a good mix of industry. Specific collaboration with original equipment manufacturers beyond the fuel cell technical team should be pursued. There must be collaboration with the EPA or NCAR to get state-of-the-art information on airborne pollutants beyond what is available in the literature.

Question 5: Proposed future work

This project was rated **3.0** for its proposed future work.

- The future work on the completion of the screening of selected contaminants and down-selecting four contaminants for detailed studies is aligned with the project goal.
- The plan for future work is good. The project team will complete the screening of contaminants, decide between selection criteria, begin modeling activities, and investigate the cause of the excess recovery.
- The fiscal year 2011 work looks to be a good continuation. Operating conditions designed to exacerbate bad chemistry should be pursued, especially if they are likely to ever occur in a fuel cell. Investigators should measure more post operation diagnostics.
- The logic of progression in this work is proper, but the learning from the baseline work should be highlighted throughout the remaining tasks to help transition from modeling to experimental work. The frequency of discussions among the team should be described.
- Mitigation studies regarding impurities that are shown to degrade performance should start as soon possible, or quantification of the extent of performance degradation should be determined. Modeling may be complex and difficult, and ultimately may be of only limited value. Finding a working cure for poor performance is more important.

Project strengths:

- This project features good project partners.
- The major strength of the team is the involvement of Ballard and UTC, which can validate the contaminant effect and mitigation strategies from this project in their stack studies. The stack validation will provide more confidence for the findings than if they were validated in the laboratory-scale, single cell configuration.
- This project is filling in the gaps of airborne pollutants that need to be understood before widespread application of fuel cells can be adopted.

- This project features thorough identification of contaminants through collaborations with other investigators.
- This project has a good proposal plan and team identified to pursue this task.

Project weaknesses:

- The approach is perhaps too academic—a more empirical approach to solving poor performance due to the identified impurities might accelerate progress in this project.
- The involvement of a national laboratory would have provided more material analysis and characterization capabilities for the team.
- The lack of real fuel cell operating conditions is an area of weakness.
- There was not enough description in the presentation concerning the test cell hardware and fuel cell material set being employed. The rationale for using the selection criteria method is not clear.
- The investigators need to tighten up the integration of test analyses and the subsequent use in modeling, particularly in defining the effects of mixtures of contaminants.

Recommendations for additions/deletions to project scope:

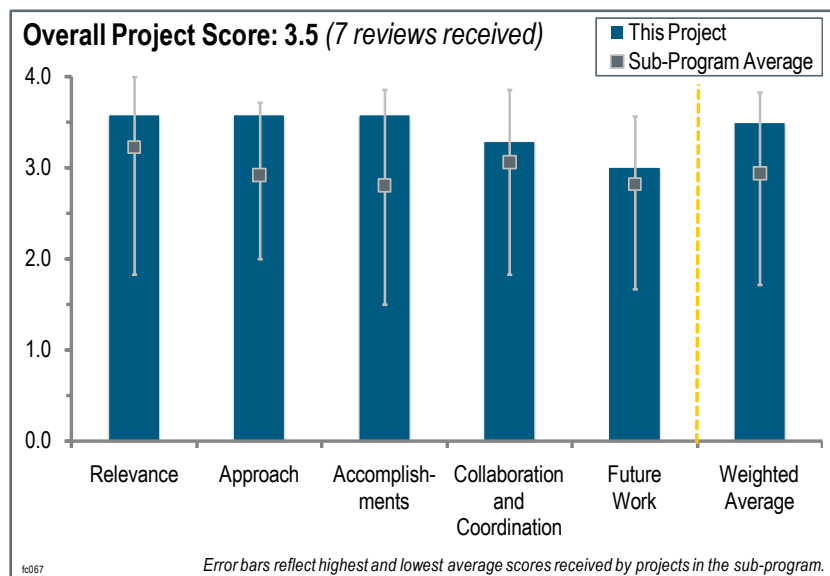
- The fuel cell should be cycled repeatedly to failure, and post diagnostics of membrane electrode assemblies should be accomplished. The project should also include a greater number of hotter and drier testing conditions.
- Investigators should consider using recovery protocols to help determine fuel cell tolerance to contaminants. They should use contemporary material sets for the electrodes and membranes as much as possible to get a true picture of the effects of contaminants on current systems. The project team should also look for salts, soots, and sulfates to add to the list of contaminants. With regard to water washing away impurities from the air, investigators could talk to Savannah River National Laboratory and Lawrence Livermore National Laboratory to prevent repeating something that has already been completed. The researchers should talk with the other contaminant projects regarding cation modeling, as there is potential for overlap.
- Updating the efforts in this task with work in the freezing efforts may provide insight into defining options in mitigation considerations.

Project # FC-067: Materials and Modules for Low-Cost, High Performance Fuel Cell Humidifiers

Will Johnson; W.L. Gore

Brief Summary of Project:

The objectives of this project are to demonstrate: (1) a durable, high-performance water transport membrane; and (2) a compact, low-cost, membrane-based module utilizing that membrane for use in automotive, stationary, and/or portable fuel cell water transport exchangers. More efficient, low-cost humidifiers can increase fuel cell inlet humidity, reduce system cost and size of balance of plant (BOP), improve fuel cell performance, potentially decrease size of fuel cell stack by running under wetter conditions, and improve fuel cell durability.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.6** for its relevance to DOE objectives.

- Developing low-cost balance of plant (BOP) components is critical to achieving fuel cell commercialization. Even if long-term prospects are to increase operating temperature and decrease relative humidity, the current systems cannot avoid humidifiers.
- Humidification has been and continues to be a significant problem with fuel cell system development, and represents a substantial issue for both initial system cost and ongoing service costs. The focus of this work to identify a lower cost and higher performance membrane humidification system to address these practical needs.
- Because humidifiers will be part of first-generation fuel cell systems, this research is appropriate and relates to DOE goals.
- This project makes an important contribution to the competitiveness of fuel cell systems.
- A membrane humidifier is important in the near term for all fuel cell applications because it provides wetter conditions which can improve both performance and membrane durability. A low-cost humidifier can contribute significantly toward the projected high-volume cost targets; however, the cost impact might not be that significant in the near term (low volumes), when membrane cost is the major factor. Defined DOE targets for BOP components (performance and cost) can be useful in better evaluating these projects. However, the principal investigator has done an excellent job trying to determine the performance and cost targets for this project.
- This project represents a very important device for fuel cells.
- Current and near-term polymer electrolyte membrane (PEM) systems need humidification, and the membrane technology promises worthwhile improvements. It is unlikely, however, that the overall membrane humidification approach will meet the more stringent operational requirements for the automotive market, particularly low power operation, which generally drives air stoich up and thereby reduces cathode exit water concentration.

Question 2: Approach to performing the work

This project was rated **3.6** for its approach.

- Gore's approach to the project work is well conceived to address current real-world issues. The two-fold approach, addressing both the micro and macro levels of the membrane and the module simultaneously, is appropriate for arriving at a low-cost/high-performance system. The investigators have done good work.
- The approach will contribute to overcoming some barriers. The module design should have started earlier, as it may greatly impact the overall humidifier performance.
- This project features a good, logical, and comprehensive development plan.
- The project has done an outstanding job of identifying key areas and concentrating on what it will take to commercialize this technology. Achieving materials identification using a fast screening test followed by detailed testing (both performance and durability) on promising materials is a very good approach. The go/no-go decision before beginning any scale-up tasks is also very good.
- Using dPoint from the heating, ventilation, and air conditioning industry is good. It would have been good to see more connection with micro-studies, scale-up, and prototype work.
- The approach to the materials section of the project is well defined and easily followed, and the testing at the system level with dPoint is very good. However, the development of the cost model is more obscure and more difficult to assess.
- Because Gore can leverage its experience with both ionomer materials and mechanical reinforcement, the approach (working with a non-composite membrane, then a composite membrane, and then a laminate membrane) appears feasible and likely to succeed. However, the slides could demonstrate the logical flow of materials design and testing a little more clearly. For example, the phrase "Utilize unique, high performance, GORE™ Humidification Membranes" on slide seven is unclear. This makes the starting point for novel exploration of materials unclear. However, based on slide eight, the wide variety of novel materials being pursued becomes apparent.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.6** for its accomplishments and progress.

- Investigators have made very good progress based on the expected project timing and testing goals. Testing the materials at a system level provides a good sense and direction of the performance of the materials in real-world systems conditions.
- It is clear that a wide range of materials have been screened for good performance, and some promising candidates have been identified for further improvement and incorporation into modules. The best Gore laminate structures appear to have high water permeance and promising durability, although this reviewer is not sure how to interpret the meaning of some of the less encouraging "hot soak durability" results on slide 11 in terms of device performance. As shown on slide 12, such laminate structures would result in large cost savings, and provided the durability is sufficient, it seems that Gore is on track to meeting the overall project goals. The module design aspect of the project has also been substantially advanced.
- It appears that the project is on track.
- The technical accomplishments are good and in line with the project targets and timeline. The investigators developed and used an interesting material characterization, but the main drawback is that only relative data is provided. It is therefore difficult to check if the water transport targets will be met. The first durability test performed at 65°C indicates that, as it could have been expected, module design has a strong incidence on the performance evolution. Therefore, module design should have started earlier. Moreover, it is unclear why the endurance test has not been carried out at 80°C, corresponding to the project target. Degradation mechanisms have to be better understood. Module design should have been better reported, in particular the advantages and the drawbacks of the adopted cross-flow design.
- Investigators have evaluated multiple materials and structures. Gore has great access to applicable materials. The water soak test could be too hard on membranes—they could be subject to a more severe swelling and structure change in the liquid soak than would occur in contact with water vapor.
- This project has made good progress to date. The project has screened several materials and identified the M311 material as a good candidate. The future tasks should focus on the durability of this material and, more

importantly, the polytetrafluoro ethylene (PTFE) microporous layer that is on both sides of this material. The design of the dPoint system needs to be improved in order to actually see a difference in the performance of different materials with different water permeances. This should be accomplished before extensive durability testing occurs.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.3** for its collaboration and coordination.

- The coordination level is appropriate for the scope of the project. The interaction is limited mostly to dPoint Technologies.
- The collaboration in this project appears to be effective and efficient. A new academic partner may need to be introduced for the degradation mechanism's comprehension.
- This project features excellent collaboration with dPoint, which resulted in good test results at the system level. Question remains with respect to whether the selected materials can be used in other system geometries (such as tubes), or if they are only appropriate for a flat plate system geometry.
- The membrane group and integrators seem to be focused on their own objectives, which is understandable for this sort of project. The concern is whether the integrators' objectives will meet the automotive needs.
- Having W. L. Gore provide the materials and dPoint Technologies design the module is a good fit. Guidance from General Motors (GM) is good, and will help the project team design the humidifier based on customer requirements.
- There should be more activity on device prototypes from dPoint, who should develop and present a scale-up approach.

Question 5: Proposed future work

This project was rated **3.0** for its proposed future work.

- There is agreement with the proposed future work; nevertheless, the following additional actions should be taken:
 - Achieve understanding of the degradation phenomenon of the membrane
 - Present clear and quantitative values to be compared with the announced targets
 - Perform durability tests at 80°C for automotive and about 60°C for stationary applications, taking into account application lifetime expectations
 - Perform module tests corresponding to a power polymer electrolyte membrane fuel cell system. Start and stop cycles should be integrated to test the robustness of the membrane and module assembly regarding temperature and relative humidity cycling
- There should be a more detailed description of the cost model that was developed as well as comparisons to incumbent technologies.
- The intent of the future work plan is clear—complete durability testing (e.g., hot soak, relative humidity cycling, and contamination), down-select final material, refine module design, and build one full-scale module—but the type of durability testing that is suggested may not identify all the device issues far enough in advance. Given that (1) so much material progress has already been made (slides 9–10), and (2) the somewhat lower performance of the best materials in terms of hot soak testing (slide 11), the behavior of the materials under more realistic conditions should perhaps be evaluated before the actual module is built at the end of the project. “Rapid prototype modules” are listed on the timeline slide (slide 5), but durability testing is not mentioned.
- Original equipment manufacturer specifications for operating conditions should be evaluated closely. The approach may be more appropriate for stationary installations than automotive applications, unless the entire operating envelope is shown to be feasible. The issue is not the quality of the work or product, but rather the physical limitations in water transfer rate from the hot exhaust stream to the pressurized inlet stream. Even if the humidifier is extremely large, equilibrium would be approached across the membrane, and that may not always be sufficient.

- The project features good proposed work on both the membrane and module sides. The project should focus a little more on durability of the down-selected membrane in practical conditions, not just the accelerated test at 90°C–95°C.
- A little more about the project's future direction should have been presented.

Project strengths:

- This project features a good partnership between a material developer and a component developer.
- The collaboration with industry partners is good. The hardware developed and tested during the project provides good correlation between analysis tools and hard data.
- There is a good match between the previous experiences and skill sets of the partners and the project goals, which has allowed for rapid achievement within the confines of a two-year project.
- The project is focused on an important aspect of fuel cell systems, and has a very structured and systematic approach.
- This project has strong technical competence in membrane technology, development path, and testing.
- W.L. Gore's material suite that can be applied to this project is an area of strength.
- This project represents a very important unit operation.

Project weaknesses:

- This is not a real weakness, but it would be good to see more of the system cost from both Gore and the primary subcontractor, dPoint. The target cost of \$150 is good, but it is unclear how the project team is doing in terms of achieving that target.
- Quantitative project objectives are given for automotive applications, but these fuel cell components should also be applicable to stationary applications. Thus, lifetime objectives should be changed from greater than 5,000 hours to greater than 40,000 hours. The cycling effect expected from starting and stopping fuel cell systems is not currently taken into account.
- Some more realistic durability testing earlier in the project would be desirable.
- An overall justification for automotive use was not presented.
- More work is needed on the design by dPoint Technologies.

Recommendations for additions/deletions to project scope:

- Investigators should add durability testing in device-like conditions.
- It would be good to look at more diverse applications (in addition to automotive and GM), including stationary fuel cell manufacturers.
- The project team should produce full-scale prototypes as soon as it can—there are new challenges and failures ahead. Uncovering those failures will help move the technology forward.

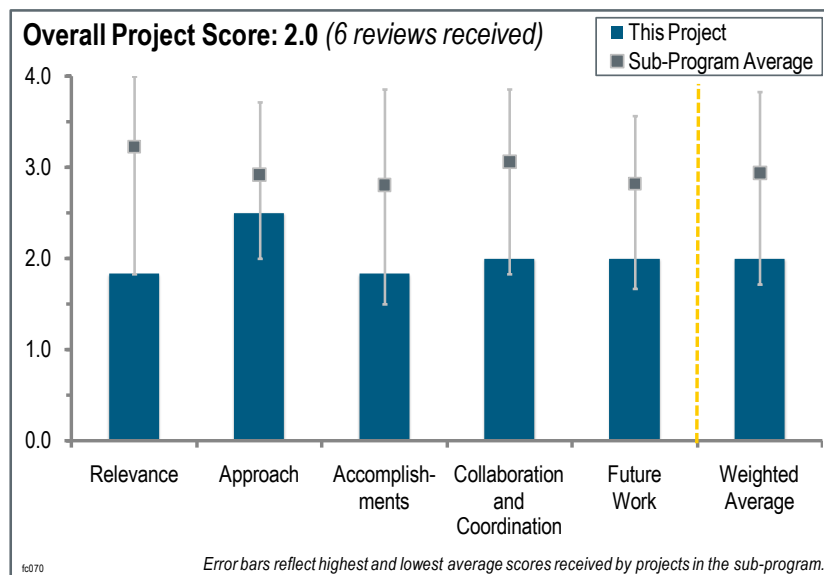
Project # FC-070: Development of Kilowatt-Scale Fuel Cell Technology

Steven Chuang; University of Akron

Brief Summary of Project:

The overall objective of this project is to develop a kW (kilowatt)-scale coal fuel cell technology. The results of this research and development effort will provide the technological basis for developing MW (megawatt)-scale coal fuel cell technology. Objectives for 2011 are to: (1) develop the process for fabricating large-scale fuel cell components by tape casting and screen printing and (2) test the long-term durability of fuel cell components.

Question 1: Relevance to overall U.S. Department of Energy objectives



This project was rated **1.8** for its relevance to U.S. Department of Energy (DOE) objectives.

- This project is an example of long-term, high-risk research and development (R&D) that, if successful, could impact the DOE Hydrogen and Fuel Cells Program goals. However, this project is very long term.
- DOE is interested in using coal as a potential fuel for fuel cells, but only at large output powers. The Program in particular is not interested in systems operating on coal at the kW level.
- Efficient, direct coal-consuming fuel cells do not seem to be consistent with the Program goals. However, the work with anodes involving directly converting carbon to electricity does seem interesting and relevant.
- The poster states that a coal-fueled fuel cell “provides a smooth transition from a fossil-fuel economy to a hydrogen-based economy”—this makes no sense, it is still fossil-fuel based. This project provides little relevance to the Program. The development of solid oxide fuel cells (SOFCs) for MW-scale utility power generation is not relevant to the Program.
- This project is relevant because it is a fuel cell technology that employs coal to generate electricity; however, it has no particular relevance to the Program because it has no connection to H₂ production or use from a coal source.
- This project does not approach DOE research, development, and demonstration targets or suggest that these targets could be met with this approach. The project is more closely related to the goals of DOE Office of Fossil Energy than the Office of Energy Efficiency and Renewable Energy.

Question 2: Approach to performing the work

This project was rated **2.5** for its approach.

- The approach is to feed coal into the anode of a conventional SOFC and react the coal with oxygen in the SOFC to produce CO₂ and CO, as well as electrons that can run through a load back to the cathode.
- The concept of scaling from the kW level to the MW level is questionable. The approach of using low-ash carbon sources seems to avoid one of the major objectives of this project—fly ash removal.
- The approach is consistent with past R&D and attempts to address cell and chemistry advancements and operational issues. The approach is sound, yet the plan calls for increasing cell size and sealants, among other qualities, and it is too early for the latter initiatives. More work seems to be needed on the process, improving the power density, and the cell itself before increasing the size. No voltage versus time data was presented. It is

important for investigators to understand the chemistry and address cell limitations before they work on a scaled version.

- The principal investigator should focus on anode development and try to avoid any other unnecessary distractions, as this seems to be the key technology barrier.
- The low cell efficiencies, difficulties of dealing with a solid fuel, and issues associated with high concentrations of contaminants with known detrimental effects leaves the project with significant weaknesses.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **1.8** for its accomplishments and progress.

- There is very little in the public domain regarding direct solid carbon conversion in fuel cells, so any data produced adds to the knowledge base. The impacts of CO₂ concentration on power density and differences between two different cokes have been presented.
- Progress has been acceptable, but slow. There are many issues to address related to the hardware, process, and overall electrochemistry. The power density improvements have been positive, but more needs to be understood regarding the cell chemistry (i.e., electrochemistry) perspective and also regarding the materials stability.
- Progress appears to be very slow, with only one year left for this six-year project. The dependence of an SOFC's performance on CO₂ injection is well known and appears to only be a diversion for this project. Many of the tasks proposed for this project have not been addressed, such as fly ash removal and coal injection.
- The results obtained thus far seem quite limited. It seems that investigators have put forth a lot of effort to get experiments up and running and establish methods, among other activities. The longest running tests are still very short, so it is hard to establish whether the concepts are valid. The effort on interconnect and seals seem to duplicate other SOFC efforts and would be better addressed via collaboration.
- The basic feasibility of the approach appears to have been demonstrated, but many issues must be resolved for this fuel cell technology to achieve practicality. For example, there are major issues with solids feeding and removal from the anode, as well as removing and sequestering the carbon-based gases that are produced. There are also issues with fuel cell degradation due to impurities in the coal.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.0** for its collaboration and coordination.

- The project has good collaboration with an end user, which is impressive. However, it would be far more efficient if this was an anode project in collaboration with an expert SOFC company that already has or can adapt its seal and interconnect technology to address identified issues.
- The collaboration seems acceptable, but narrow. It is not clear what the Coal Development Office brings to the (technical) discussion. There could be a broader set of disciplines involved. The question of scalability is important and needs to be addressed, but it is too early to make such an assessment because the technology is not yet developed. Therefore, it is too early to address collaborations on this subject.
- There is limited collaboration, but the mechanism and effectiveness of the interaction is unclear. Only a few areas are mentioned under collaboration for each of the project partners.
- The collaborative activities are not well described.
- It is not clear if the collaborative partners have contributed anything to this project.

Question 5: Proposed future work

This project was rated **2.0** for its proposed future work.

- The proposed future work is reasonable, but it is unclear how it will be accomplished with no funding.
- Factoring in the funding and lifetime of the project, the proposed future work is based on what would be needed if everything was developed on schedule. As stated in this review, there are still fundamental factors that must be addressed. There needs to be more effort on stability. A one-month operating time is not enough to discern the reliability, stability, robustness, and degradation mechanisms.

- Based on the progress so far, it is doubtful that the proposed work can be accomplished in the remaining one year of the project.
- The proposed future work includes scale-up of the technology, but the technology does not appear ready for that step. A lot of work to address stability and usefulness with real coal samples would be needed before scale-up is warranted.
- The project is far from commercial relevance and the future work is focused on more advanced issues such as stack design and long-term testing. The results to date do not validate these efforts and increased fundamental understanding is required.

Project strengths:

- The principal investigator is good and seems to be addressing the right topics. The project is being performed in an academic environment, where it belongs at this stage.
- This project features good collaboration with an end user.
- This project represents an interesting fuel cell concept of using coal as the fuel.

Project weaknesses:

- This project has made slow progress, and lacks focus on stated goals, objectives, and tasks.
- This project's plan and expectations are too ambitious. There has been inefficient engagement with other SOFC experts.
- This project does not appear to have any relevance to the Program. Areas of weakness for this project include low power density, low efficiency, and low durability.

Recommendations for additions/deletions to project scope:

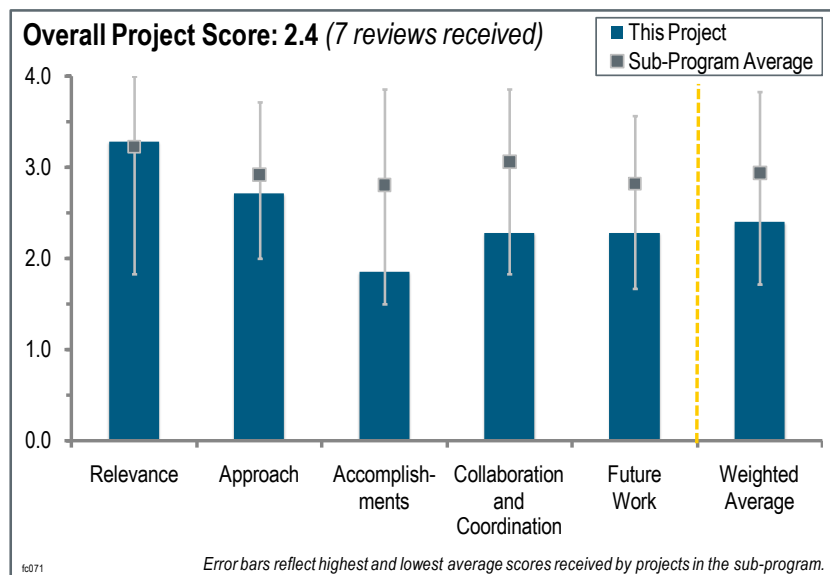
- Investigators should focus on the cell performance and modes of operation, not ancillary themes, and produce statistically significant cell data under a much broader set of conditions.
- This project should add an SOFC partner to assist in addressing inefficient resolution of interconnect and seal problems.

Project # FC-071: Alternative Fuel Membranes for Energy Independence

Kenneth Mauritz; University of Southern Mississippi

Brief Summary of Project:

The objective of this project is to engage in fine molecular and morphological tailoring and evaluation of novel, low-cost hydrocarbon fuel cell membranes that possess high temperature performance and long-term chemical and mechanical durability in polymer electrolyte membrane (PEM) fuel cells. This effort will support the U.S. Department of Energy's (DOE) Hydrogen and Fuel Cells Program by developing high-temperature, low relative humidity, and high-proton conductive membranes for use in PEM fuel cells. The project is focused on alternative materials with performance up to 120°C at low relative humidity.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.3** for its relevance to DOE objectives.

- All aspects of the project are relevant.
- This project supports the development of high-temperature (120°C), low-cost (hydrocarbon), and low relative humidity membranes for PEM fuel cells.
- This project focuses on fundamental and elegant polymer chemistry to develop phase-segregated hydrocarbon membranes that meet DOE targets.
- This project addresses the DOE barriers of performance and durability.
- Improved membranes are critical to fuel cell stack performance, life, and cost.
- The project shows innovation in polymer synthesis, but it is not always clear how the new syntheses meet DOE needs for advanced membranes.
- New polymer materials are being developed. In general, chemistries that are known to have some fuel cell durability are explored.

Question 2: Approach to performing the work

This project was rated **2.7** for its approach.

- The synthetic approaches pursued in this project are novel and hold promise. This project features a well thought-out approach that includes membrane synthesis, membrane electrode assembly optimization, and durability testing.
- As shown in the review, this project is almost entirely synthesis of new hydrocarbon membranes, specifically block copolymers of N,N-diisopropylethylammonium 2,2-bis (p-hydroxyphenyl) pentafluoropropanesulfonate. To better understand the novelty of this approach, the investigators need to consult similar work in the public domain, for instance, the work conducted at Virginia Tech. The synthetic work and characterization is good, but there seem to be the following three separate projects: (1) making polymer backbones with acid groups (2) making polymers with acid (phosphonic acid) and base (triazoles), and (3) making structural segregated

materials. The project team should focus and choose what is best from the three approaches that have been pursued to date.

- Some of the approaches are good, some are not as good. For instance, approaching ionomer development by combining elements from perfluorosulfonic acid polymers (PFSA) with those from sulfonated poly (arylene ether sulfones) and enhancing phase segregation (subtask 2.1) is a good idea. Using stronger acid groups than those used in most hydrocarbon-based membranes may make this approach viable. On the other hand, others have tried similar approaches to subtask 2.2 without success. In particular, there was a recently DOE supported project attempting the use heterocyclic nitrogen-containing bases and acid groups in the same polymer to achieve high conductivity in a low-relative humidity environment.
- The approach is to attempt to improve the performance of block copolymers by several synthetic strategies. The approach of looking at pendant triazoles has been tried without success by other groups. Similarly, phosphonic acid derivatives have not been successful in previous efforts by others. Attempting to place perfluorosulfonate hydroquinone blocks in block copolymer appears to be the strategy with the most likelihood of success.
- Too many chemistries are proposed with marginal explanation on their promise (e.g., better acidity, high functionality). The investigators are attempting to find a known block copolymer morphology, which has seen little success in the past.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **1.9** for its accomplishments and progress.

- A number of materials were synthesized and nuclear magnetic resonance (NMR) and atomic force microscopy (AFM) of the materials were presented. No conductivity, stability, or gas permeability results were presented other than the mention of “poor conductivity” at 120°C. The project is nominally in its third year and 80% complete, yet there was a complete lack of real data from the materials synthesized—conductivity, conductivity dependence on relative humidity or temperature, water uptake, stability and durability, gas permeability, and fuel cell testing were all absent from the presentation. It is impossible to tell whether any progress has been made without any performance data.
- There is a very high risk of not achieving a viable membrane for testing in a fuel cell by the end of the project.
- Each of the three projects is progressing, but at this point, none can provide a practical membrane for a fuel cell. Perhaps progress would be improved by focusing only on one approach. The investigators have not made membranes with improved properties compared to current hydrocarbon or fluorocarbon membranes.
- This project features very good polymer synthesis work, but improved conductivities at high temperatures and low relative humidity have yet to be shown.
- The project is nearly completed, but investigators have pursued too many paths. The project team has accomplished some good synthetic work, but the conductivity results to date have not been impressive.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.3** for its collaboration and coordination.

- The team seems enthusiastic and well knit, and shares resources and methodology on the “three approaches” in this project.
- There is little inclusion of other institutions, but there are several partners in the synthesis that work together really well. Obtaining some industrial support would be preferred, but perhaps this is premature. If successful, the project team would have to seek a scale-up partner.
- This project does not have much collaboration, but not much is needed for a project that is principally focused on synthesizing new ionomers. Some industrial collaboration may be helpful to steer material sets toward those most likely to be relevant for fuel cell applications.
- Collaboration with the Illinois Institute of Technology (IIT) is mentioned, but there is no collaboration apparent or presented, including in the publications. There is no collaboration with any industrial developer.
- Collaborations are limited—IIT is the only collaborator.
- The few outside collaborations reflect the focus on polymer synthesis.
- The only partner listed is the principal investigator. The project team could greatly benefit from working with someone who regularly characterizes these membranes.

Question 5: Proposed future work

This project was rated **2.3** for its proposed future work.

- This project has a good plan, but little time is left for tasks three and six.
- The proposed future work continues to be more synthesis of new materials, with some characterization, although the project team did not mention what kind(s), and fuel cell performance. With only 20% of the project left, it seems like the project needs to wrap up the synthesis work and characterize and present the results of the materials made to date for value to come out of this work.
- All milestones are delayed to the end of the project, and this reviewer does not see a path to the fuel cell evaluation part of the project.
- The team seems to pursue addressing multiple approaches, while down-selecting might be a better idea. For instance, the pursuit of the second approach, making a polymer with an acid and a base, would be a good idea.
- The investigators do not have much time left, but they need to focus on one system and begin relevant characterization as a PEM. They still are planning a lot of new synthesis.
- The future work lists alternative synthesis ideas, which look reasonable, but it is unclear what alternative pathways exist if the proposed approaches fail.

Project strengths:

- The polymer chemistry is the strength of this project.
- This project team features good synthetic skills.
- This project represents very interesting novel polymer synthesis and characterization.
- Some new and interesting chemistries are proposed in this project, including some PFSA chemistries that have not been explored.
- This project features some good ideas, including several potential pathways to prepare high-performance membranes.

Project weaknesses:

- There is a lack of data on the synthesized materials in this project. If the data exists, it needs to be presented to understand any progress. The only characterizations presented were NMR and AFM, which were presented without a correlation to what they mean regarding how the membrane materials might work.
- There appears to be a desire to keep trying new synthesis without any thought of the timeline. The technical issue is the production of good membranes with these polymers, yet little work is being done to fix that problem. Down-selection of polymers and a focus on their modification to enable membrane fabrication would have allowed fuel cell testing to start in parallel to further synthetic work, which would have been positive. The project team's effort is diluted by pursuing multiple approaches.
- It is not clear how high-conductivity membranes with good stability can be achieved during the short time remaining in this project.
- One weakness of this project is the lack of PEM characterization, especially conductivity versus relative humidity. This project took too many synthetic pathways.
- The project team has not explained why some of the approaches are likely to succeed when previous similar work has not, as evident from work reported in the public domain.

Recommendations for additions/deletions to project scope:

- This project needs to present data.
- This project is near completion without delivering a membrane for fuel cell evaluation. If the project team can make a down-selection of the best available polymer now and move toward membrane fabrication, a one-year, no-cost extension to enable the fuel cell work to occur would allow for greater project completion.
- The investigators should streamline the work in approaches one and two (especially two). Morphology control (approach three) can be pursued if it is needed later.

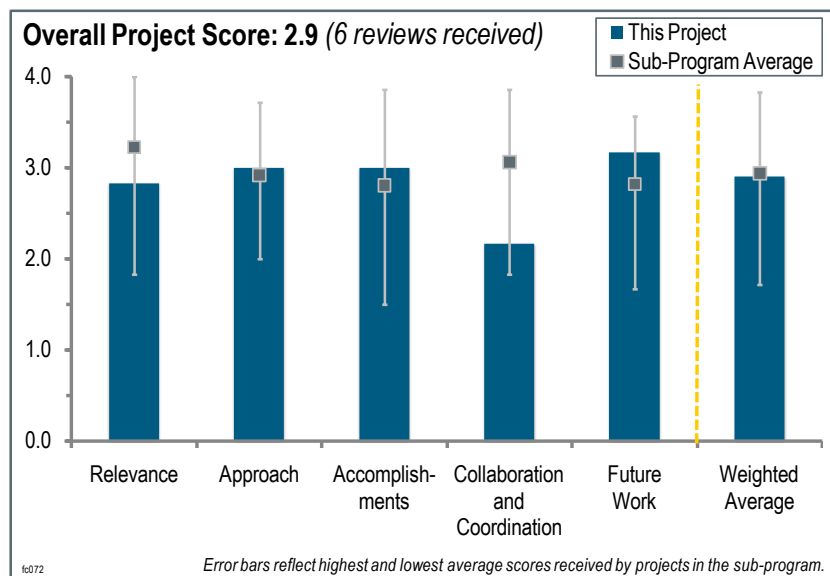
- This reviewer would suggest the work focuses on the fluorinated sulfonic acid type block copolymers. The reviewer would stay away from the triazole systems, as work at Lawrence Berkeley National Laboratory suggests that this system is not productive.
- Investigators should show results of the benchmark Nafion tests.
- The project team should choose one membrane and work with someone with PEM experience to help evaluate it.

Project # FC-072: Extended Durability Testing of an External Fuel Processor for SOFC

Mark Perna; Rolls-Royce Fuel Cell Systems (US) Inc.

Brief Summary of Project:

Overall objectives for this project are to: (1) conduct long-term tests in relevant environments for the three fuel processor subsystems that support operation of the 1 MWe (megawatt-electric) solid oxide fuel cell (SOFC) power plant; (2) determine the long-term performance of key components such as catalysts, sorbents, heat exchangers, control valves, reactors, piping, and insulation; (3) evaluate the impact of ambient temperatures (hot and cold environment) on performance and component reliability; and (4) determine system response for transient operation.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **2.8** for its relevance to U.S. Department of Energy (DOE) objectives.

- The Rolls-Royce Fuel Cell Systems (RRFCS) SOFC power plant concept—through its high-efficiency, negligible air emissions and potential fuel flexibility—directly supports the DOE Hydrogen and Fuel Cells Program’s mission to reduce petroleum use, greenhouse gas emissions, and air pollution and to contribute to a more diverse and efficient energy infrastructure by enabling the widespread commercialization of hydrogen and fuel cell technologies. The RRFCS SOFC power plant concept for stationary power supports the Program’s goal to advance fuel cell technologies through research, development, and validation efforts, to allow them to be competitive with current technologies in cost and performance, and to reduce the institutional and market barriers to their commercialization.
- This work on the durability of the various fuel processing subsystems aligns well with the DOE research, development, and demonstration plan objectives.
- Fuel processors for stationary fuel cell applications are definitely relevant to DOE goals and objectives. The project addresses the durability, performance, start up, and energy and transient operation of SOFCs. Specifically, the project is concentrated on evaluating the fuel processing subsystem performance for distributed generation systems. The fuel processing subsystem is an extremely important part of the overall stationary fuel cell power system because, in addition to pipeline gas, other feed stocks such as biogas may be used. Successful operation requires less than 100 parts per billion (ppb) of total sulfur in the fuel.
- High-temperature stationary fuel cell systems are typically fueled with natural gas. Decoupling fuel processing from the fuel cell stack provides an added degree of freedom, particularly if the total system still offers high operating efficiencies. The objective of this project is to test and verify the desired durability of an external fuel processor using catalytic partial oxidation reforming. The process includes feed gas desulfurization and yields a reformat that is suitable for use in an SOFC (i.e., contains less than 100 parts per billion [ppb] of sulfur; tests showed less than 10 ppb sulfur, which is below the detection limit of the sulfur analyzer).
- The project’s aspects are in the objectives of the Program.

Question 2: Approach to performing the work

This project was rated **3.0** for its approach.

- The approach is to evaluate the three major components of the subsystem—the synthesis gas, the start gas, and the desulfurizer subsystem. The project team operated and determined the performance of these subsystems individually for various times and under various environmental conditions. Each subsystem was evaluated for varying times from 200 hours to 8,000 hours. The desulfurizer subsystem will run around the clock in unattended mode for up to 8,000 hours by the end of the project in December 2011. Post-test inspections and analyses were or will be performed. Ten start-up cycles were performed. One goal was a target performance of less than a 10% reduction in H₂ over catalyst life.
- The project involves three separate subsystems: a start gas subsystem, a synthesis gas subsystem, and a desulfurizer subsystem. Each subsystem is separately designed, installed, and tested for operation and durability, including a target number of start-stop cycles. Separate test plans have been developed for each of the three subsystems. Post-test analyses will include physical and chemical analyses of catalysts and sorbents, as well as determination of wear and damage to the subsystem hardware and plumbing.
- Long-term testing in a practical environment (outdoor) is an excellent approach for durability evaluation. The principal investigator does not clearly explain why such a low sulfur target has been selected for SOFC systems.
- This project addresses technical barriers associated with durability, performance, and start-up and shut-down time and energy/transient operation. This project does not seem to present any new and novel natural gas fuel processor technology. It is not clear which desulfurizer sorbent technology is used in this project.
- Natural gas desulfurization has been investigated for a long time. This project uses sorbents at temperatures of around 200°C to remove the sulfur-containing materials before feeding the natural gas into a reformer.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- Various tests were completed as planned.
- All milestones have been completed or are on schedule.
- The project is in the durability testing phase and is on schedule for completion on December 31, 2011. Data for the desulfurizer test shows good stability for more than 3,500 hours (8,000 goal).
- The experimental test was well done and very simple.
- Synthesis gas subsystem testing was completed in April 2010 (results were presented at the 2010 AMR). No results of the post-test analyses were presented, however. The desulfurizer and start gas subsystems have been installed and successfully operated in the outdoor test facility. The desulfurizer subsystem has been successfully operated for 3,600 hours by mid-April 2011, which is on the way to completing the target 8,000 hours of testing by the end of the project. The start gas subsystem is ready to begin durability testing.
- This project started in August 2008 and will conclude in December 2011. It is only 58% complete, and there is some concern about whether the remainder of the work will be completed by the end of the project. The project team has not run the processor subsystem with a fuel cell. The plan, following the conclusion of the project, is to send it to the United Kingdom, where it will be run with a 1-MW SOFC at the Rolls Royce's facility. Results showed very good stability in the outlet, retaining better than 90% of the hydrocarbons with low sulfur concentrations (approximately 10 ppb).

Question 4: Collaboration and coordination with other institutions

This project was rated **2.2** for its collaboration and coordination.

- This project has two partners—the Ohio Department of Development and Stark State College. Stark State provides student interns, many of whom are subsequently hired by the Rolls Royce Canton facility.
- The Ohio Department of Development provided funding (\$3 million) through Ohio's Third Frontier to expand the Fuel Cell Prototyping Center located on the campus of Stark State College.
- There appears to be no collaboration with fuel cell developers or system integrators.
- This project needs to have more inputs from the RRFCS SOFC activities.

- The primary technical collaboration is the involvement of students and facilities from the Stark State College Fuel Cell Prototyping Center.
- No collaborations seen.

Question 5: Proposed future work

This project was rated **3.2** for its proposed future work.

- The proposed future work is planned in a logical manner.
- The planned future work should lead to orderly conclusions in 2011.
- The project concludes in 2011, during which durability testing, post-test analysis, and reporting will occur.
- Investigators will perform post-test analysis of all three subsystems from the second quarter through the fourth quarter. Durability testing of start gas and desulfurizer subsystems will be completed by the third quarter. The project team will then issue its final report.
- The planned future work (through the end of the project, scheduled for December 31, 2011) is to complete the test plans and post-test analyses of the three subsystems, and to document the results.
- Desulfurization is well developed in the natural gas industry. There were not any unique achievements or future accomplishments from this project.

Project strengths:

- This project involves relevant development and testing of a desulfurizer subsystem.
- The subsystem has been run under varying environmental conditions from -20°C to 40°C in an outdoor test facility. The project team performed 10 start-ups. The system can ramp up within one minute.
- Project strengths include the successful installation of the three subsystems, completion of the durability testing of the synthesis gas subsystem, initiation of the durability testing of the other two subsystems, and demonstration of operation under extreme outdoor weather conditions (ambient temperature of -23°C).
- Conducting long-term tests in outdoor facilities to evaluate the durability of various fuel processing subsystems is a strength of this project.

Project weaknesses:

- There are none.
- This project has not been run with a fuel cell.
- There has been little coordination with SOFC development activities.
- There are no novel items in the project.

Recommendations for additions/deletions to project scope:

- The project team should integrate and run the desulfurizer subsystem as a complete system with a fuel cell.

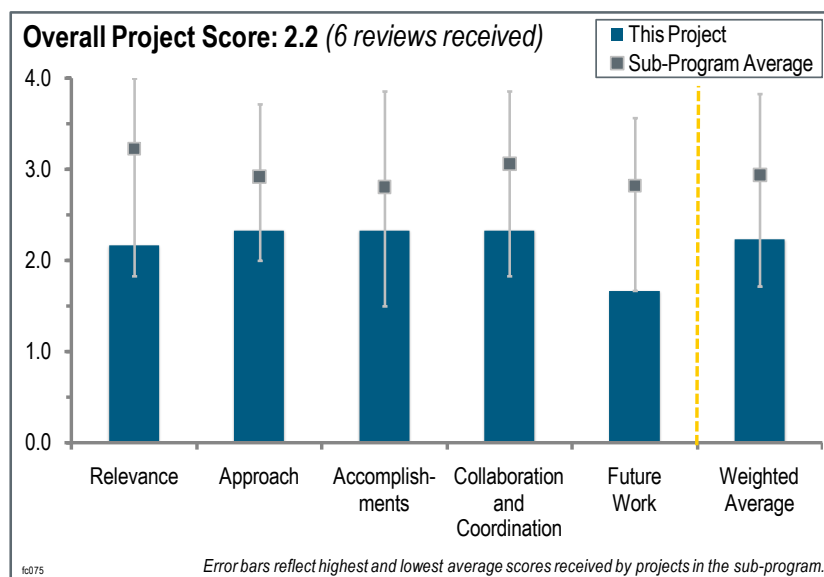
Project # FC-075: Fuel Cell Balance of Plant Reliability Testbed

Vern Sproat; Stark State College

Brief Summary of Project:

The overall objectives for this project are to: (1) develop testbeds to address the challenge of improving durability and reliability of non-stack fuel cell system components (balance of plant [BOP]); (2) develop a test plan to address the candidate BOP components and basic testbed design for long-term operation; (3) use collaborations with component manufacturers to develop and enhance final product performance; (4) develop statistical models for extremely small sample sizes while incorporating manufacturer validation data for future evaluation of candidate components, (5)

conduct real-time, *in situ* analysis of critical components' key parameters to monitor system reliability; and (6) use the testbeds to enhance the education of the technical workforce trained in polymer electrolyte membrane fuel cell system technology.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **2.2** for its relevance to U.S. Department of Energy (DOE) objectives.

- The commercial success of fuel cells depends on BOP as much as on the stack itself. It is good to see effort being put into these components and, more importantly, into educating our next generation of fuel cell engineers.
- The contractor has developed useful and well designed test facilities for transient and endurance testing of BOP components. The facility has also generated educational value, contributing to the trained workforce that will be needed in the future. It is, however, rather difficult to predict the type and quality of data to be generated in the test facilities, as there is no comprehensive test plan presented. Details and a more formal test plan layout—including error analysis and uncertainty, as well as design of experiments considerations—would strengthen the remainder of this project. Overall, this is a useful project and it should certainly be continued.
- While BOP reliability and cost issues are important to overall cost reduction and reliability, there is such a breadth of equipment and suppliers that a small program like this cannot adequately address them. The manufacturers should provide this type of testing, and most of them do. The degree of education that this provides to the overall DOE Hydrogen and Fuel Cells Program is also questionable. This is a small project that caters to a small number of students. There does not seem to be much potential for the project to continue after DOE funding expires.
- This project does not support the Program's research and development objectives. Having students build three different testbeds for testing BOP components does not add value to the Program. Better value would have been derived if an appropriate choice of BOP components was made, relevant to the Program's needs, for testing.
- This project has essentially built three test stands. Test stands are commercially available. The project does not appear to address any targets of the Program.

Question 2: Approach to performing the work

This project was rated **2.3** for its approach.

- The test system appears to have been designed with flexibility in mind. The analytical approach of Weibull/Weibeyes is sound. Additional environmental stressors should be added to make the testbed really valuable.
- This project may need to be better aligned with original equipment manufacturer (OEM) requirements. A better option would be for OEMs to share which BOP components they are interested in, and for this organization to test the high-priority ones.
- The approach slide lists six different objectives; however, the project has primarily focused on just objectives one, two (building a testbed), and six (education of students). If the project had actually concentrated on the third (consult manufacturers of relevant BOP components and test those components), fourth (statistical analysis), and fifth (determine failure modes of critical components) objectives, this project would have been of value to the Program. Moreover, the education component needs to involve actual testing of fuel cell related components.
- Most of this project has involved designing and building test stands. There are industrial suppliers that do this. There is no new development in the approach.
- The scope is too broad to be effective. The test rig design appears to be competent. The use of students to do the work has limited the effectiveness for the overall Program.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.3** for its accomplishments and progress.

- The systems have been built and operated. The sheer number of components and manufacturers is impressive.
- Impressive progress has been made in developing the testing capability.
- It seems like very little was accomplished last year. The 2010 report states that: “This year [two] Test-beds have been assembled and the third is under development. Several test parts have been identified, looking for others to test.” Most of the testbed work was already performed, and there should have been materials testing data available for presentation in this Annual Merit Review (AMR). The only new technical accomplishment for 2011 is presented in slide 12 and concerns material testing. Slides 7–11 are identical from the 2010 presentation, with the photographs in slide 7 newer. Slide 13 states that the pump that was selected in 2010 (slide 12) has been discontinued. Slide 14 is similar to slide 13 from 2010. Slide 15 is similar to slide 14 from 2010. Therefore, the accomplishments and progress in this project over the past year are very disappointing. This is unfortunate, given the project is supposed to end in July 2011 (75% complete).
- This reviewer realizes that student participation is one of the goals of the Program. It would be beneficial to bring in external help to initiate sensor testing. Students may be better involved in operating test stations than building them. Once test stations are online and running, time can be used to gather statistical information. The project started in 2008, so more data than what has been collected was expected by now. Stations can be built and commissioned quickly during the first 12–18 months, so data gathering should have commenced.
- The investigators identified a pump in fiscal year 2010, but did not procure it and now it is discontinued. Thus, they are using a lower capability blower. This is not much progress. Three test stands were built, but component testing was marginal. There are no results presented to guide developers.
- Rendering the testbeds operational was time consuming so limited testing has been completed.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.3** for its collaboration and coordination.

- Lockheed Martin has participated at a significant level.
- There appears to be good collaboration with Lockheed Martin. The level of educational cooperation is not clear—while there is a regional educational network in place, the presentation does not give any specifics about what the project has contributed. It is not clear how the data gathered from the test program is going to be disseminated to the fuel cell community.

- Collaboration was described on slide 16. This project features an outstanding survey of potential component suppliers.
- The only real collaborator is Lockheed Martin. Listing approximately 65 companies that supply parts does not constitute active collaboration. The project should focus on automotive OEMs and other funded DOE projects to identify the relevant BOP components and get them reliability tested. Components that are part of the air handling and humidity control systems are particularly important.
- While this project's collaboration appears to include a long list of parts suppliers, it is more important that the organization collaborates with fuel cell OEMs. It is unclear if this occurs. It is important that sensor and component testing is performed on the sensors that are most likely to be needed for systems.
- There are no collaborations other than with Lockheed, and that collaboration is unclear. The project team lists approximately 60 collaborators, which is simply untrue. Buying a Swagelok fitting does not count as collaboration. The investigators also list a number of educational institutions with no demonstration or description of the collaboration, or any proof it existed beyond perhaps a phone call. There is nothing believable about any of these collaborations.

Question 5: Proposed future work

This project was rated **1.7** for its proposed future work.

- The Weibull/Weibeyes analysis and conclusions regarding which components represent the highest risk for the industry should be published.
- The project has the right overall goals in sight. It is unclear if the project is progressing on a timely schedule.
- The proposed future work has no specifics other than to "test parts."
- Testing needs to be prioritized. There is not enough time remaining to accomplish any meaningful testing. The commitment of Stark State to make this project a core component of its curriculum is unclear.
- The plan may be good, but it has not been articulated in sufficient detail. It would help if the team would present the expected issues, how these issues will be realized, and how the results will be measured and quantified.
- The future work for this AMR is identical to that from the last AMR.

Project strengths:

- This project features a practical approach, and provides useful testbeds for industry.
- The test rig design appears to be competent and well executed.
- This project features well designed testbeds and a strong educational component.
- BOP component reliability is actually an issue that is of value to the Program.

Project weaknesses:

- No environmental (temperature especially) factors were considered as stressors. These will significantly affect the reliability of components.
- This project has been too slow in getting the test rigs operating and actually generating data. There has been no identification or focus on critical balance of system components that should be tested and would contribute to the overall Program. The student-orientated focus is not particularly effective.
- The plans for the final stage use of the test facilities is an area of weakness.
- The progress in this project has been minimal and this project has provided very little value to the Program.
- Testing of durability as mean time between failure is going to be challenging on a small number of stations. This reviewer thinks that it needs to focus on "new" BOP components instead of off-the-shelf ones. For example, testing pressure sensors will not yield novel results for a well established technology. However, if the investigators test new blowers or humidifiers, they are likely to find meaningful failure mechanisms that would impact the Program. Again, they would need closer collaboration with system OEMs to achieve this.
- This project has done nothing to help any technological developments and has no apparent plans to do anything useful.

Recommendations for additions/deletions to project scope:

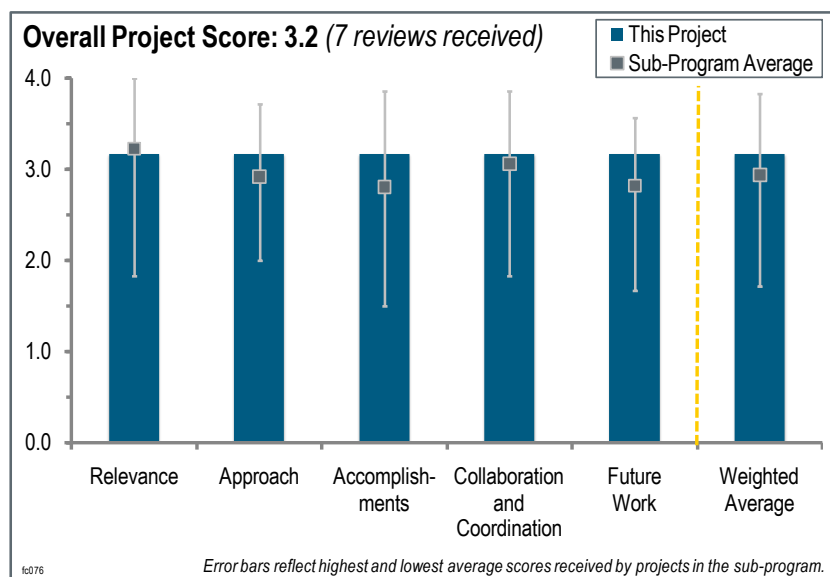
- The investigators should add a thermal chamber to one rig for environmental testing.
- The investigators need to focus testing on a few critical components that have been identified as needing more reliability testing. They should also develop a feedback mechanism to get the results out to industry.
- The project team should talk to automotive and stationary OEMs and consult the investigators from other funded DOE projects to identify relevant BOP components to test. The project team should then test these components for the long term and report on their reliability.
- The project team should focus less on off-the-shelf technology and more on the “new” BOP components of fuel cell systems (e.g., humidifiers, air blowers, and ejectors). While three-dimensional drawings of piping and instrumentation are a great way to package systems, they do not require the level of detail that was expanded here. The purpose of using compression fittings is their versatility. Investigators could have spent more time building and less time drawing.
- This project should be ended.

Project # FC-076: Biomass Fuel Cell Systems

Neal Sullivan; Colorado School of Mines

Brief Summary of Project:

The overall objective for this project is to improve the durability and performance of solid oxide fuel cell (SOFC) systems while lowering costs. Task one is to develop SOFC materials for robust operation on bio-fuels, including integrating barrier-layer technology into tubular SOFC geometry and nickel-free, perovskite-based anode supports. Task two involves fuel processing of bio-derived fuels, including developing fuel-reforming strategies for anaerobic-digester-derived biogas and decreasing the cost of fuel-processing balance-of-plant hardware. Task three includes modeling and simulation to: (1) develop chemically reacting flow models of fuel-processing hardware; (2) conduct thermal modeling of hot-zone system components; and (3) use system modeling to explore tradeoffs in biogas-processing approaches.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.1** for its relevance to U.S. Department of Energy (DOE) objectives.

- The project goal is to improve robustness of hydrocarbon- and biomass-fueled SOFCs and systems. This project is relevant to the DOE Hydrogen and Fuel Cells Program goals of increasing the durability and performance (efficiency and transient response) of SOFC systems while lowering costs.
- Utilizing biofuels as a feedstock for SOFCs is critical to becoming oil independent. The micro-channel reactor is a key and novel contribution to meeting this objective.
- Fuel cells operating on renewable fuels would definitely be aligned with the Program goals. The desired MW (megawatt)-scale system still seems to be geared to distributed generation, so an appropriate cost target should be developed. It is unclear if the cost of a tubular stack will be competitive.
- The technologies under investigation support a range of future applications and SOFC system designs. With the greater focus relative to 2010, the project should have a high impact.
- The poster should do a better job of relating the work performed to specific DOE targets. The approaches presented do address DOE objectives, particularly in the area of stationary power.

Question 2: Approach to performing the work

This project was rated **3.2** for its approach.

- The project consists of three tasks. The approach in the first task is to develop materials and architectures to improve SOFC durability for operation with biomass-derived fuels. The approach in the second task is to develop biofuel processing strategies for optimal compatibility with SOFCs, and low-cost ceramic micro-channel reactive heat exchangers for fuel reforming. The approach in the third task is to provide computational fluid dynamics modeling support for tasks one and two.
- The project approaches the objective from multiple angles, including material selection and process development with the support of simulation and modeling tools.

- The approach had a lot of detail, but it was not clearly communicated in the file.
- This approach is a good mix of fundamental analysis, experimentation, and design.
- The project has responded to concerns raised last year about being too broad and has sharpened its focus on key aspects such as the micro-channel reactor.
- The team appears to use the correct tool for the job when it comes to a particular problem, and ANSYS is one such modeling tool. However, integrating the different tasks into a bigger, more meaningful whole seems unclear.
- The approach is generally effective and improved compared to last year, when the work was even less focused. Still, the project has several aspects that are not particularly synergistic when considering the development of tubular cells and anode studies, and contrasting those with ceramic heat exchangers.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.1** for its accomplishments and progress.

- The project team has synthesized perovskite ($\text{Sr}_{0.8}\text{La}_{0.2}\text{TiO}_3$ or SLT) barrier layers and has integrated with and used CoorsTek tubular SOFCs (task one). The team has also developed kinetic models, conducted experiments to validate the models, and used them to guide the definition of external-reforming operating windows (task 2). Investigators determined the electrochemical performance of SOFCs with catalytic partial oxidation with oxygen and steam reforming of biogas (task two), and fabricated and determined the performance of ceramic (alumina) micro-channel heat exchangers (task two). Additionally, the project team developed FLUENT and CANTERA models for micro-channel reactive heat exchangers, a control model for dynamic-load following, and a system-level model for thermal integration (task three).
- The combined use of FLUENT and CANTERA provides an excellent modeling solution. The heat exchanger design thoroughly addresses some of the major challenges in the development.
- The realignment of effort is bringing positive results.
- This project features good results on simulated biogas operation, interesting ideas about thermal integration, and good use of analytical tools.
- This project has made impressive progress overall.
- With relatively limited funding, the team has made meaningful advances in several areas. The work in biofuel reforming and ceramic heat exchangers is particularly interesting.
- This reviewer would like to see a timeline for the progress made and the future tasks.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.1** for its collaboration and coordination.

- This project makes very good use of partnerships with SOFC companies and avoids reinventing the wheel on stack design issues.
- This project features especially effective small business collaboration with the heat exchanger fabricator.
- CoorsTek is a partner and supplies SOFCs and materials for the project.
- The collaboration with CoorsTek is clear. Engaging additional partners (e.g., national laboratories) would enhance the model verification process and the process optimization.
- There is very close collaboration with CoorsTek. There does not appear to be any significant collaboration with other outside resources.
- CoorsTek is an excellent partner, but the collaboration is only between the Colorado School of Mines (CSM) and CoorsTek.

Question 5: Proposed future work

This project was rated **3.1** for its proposed future work.

- This project is on an excellent path.

- The proposed future work is generally reasonable and focused on furthering three thrusts in the areas of SOFC materials, biomass reforming, and modeling.
- This plan is logical and based on the results of modeling work.
- Investigators have identified appropriate next steps.
- The proposed future work includes defining next-generation SOFC materials and architecture (task one); depositing ceria-based catalyst supports in micro-channel reactors and improving sealing approaches (task two); and improving and advancing component, controls, and system models (task three).

Project strengths:

- Strengths of this project include academic knowledge of SOFC materials and processes (CSM) and the industrial expertise of CoorsTek, which is the largest ceramic company in the United States.
- Areas of strength for this project include its strong modeling and design capability and collaboration with CoorsTek. The project is focused on the micro-channel reactor, which is a key component.
- The partnerships are well chosen and utilized, and there is good use of analytical modeling.
- The potential to improve the heat exchange/catalysis unit seems high. The contributions in modeling and component design are significant.
- Integrating modeling is key for a deeper understanding and production of efficient systems.

Project weaknesses:

- This project has a broad scope with diverse tasks.
- Some outside collaboration with a national laboratory in conducting design reviews might be helpful.
- This project lacks specific application related targets, such as cost.
- This project was initially too broad, but the principal investigator has responded well and focused efforts.
- The durability of ceramic heat exchangers was reported as a weakness last year and has not yet been investigated. The ability of these materials to withstand thermal stresses and long operating conditions with high durability need to be demonstrated.

Recommendations for additions/deletions to project scope:

- This reviewer had no recommendations.
- The project team should enhance the validation of the modeling results through experiments. The intense thermal gradient on the plates could lead to mechanical deformation or fatigue issues—investigators should study these phenomena through modeling.
- Researchers should confirm that the cost of such a design is compatible with the desired application via should-cost analysis.
- Within “Future Work: Task 3b,” thermal modeling of balance-of-plant hardware seems to stand alone. Effort could be diverted to further narrow the project’s focus and accelerate efforts on thermal modeling.

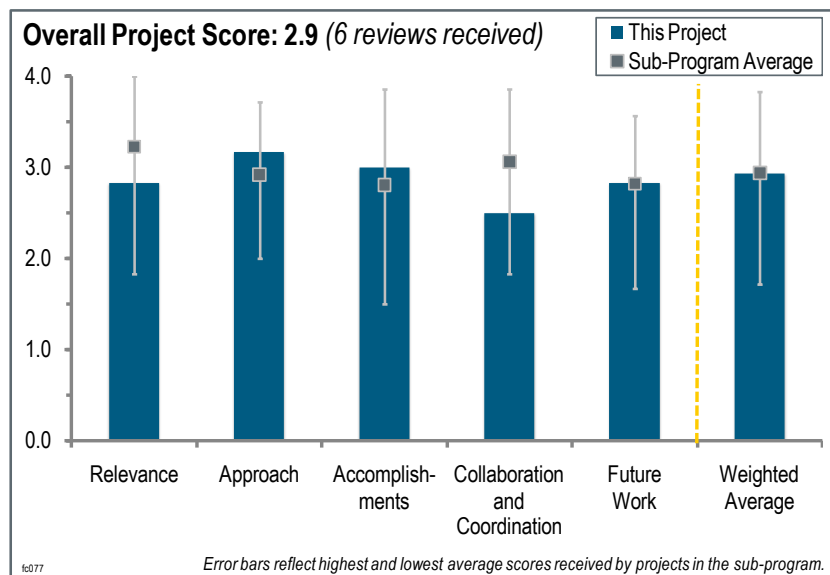
Project # FC-077: Fuel Cell Coolant Optimization and Scale-Up

Satish Mohapatra; Dynalene

Brief Summary of Project:

The objectives of this project are to (1) optimize and then scale-up the process of making Dynalene fuel cell coolant with a great deal of reproducibility using 100-liter (L) batches of nanoparticles on a pilot plant scale, (2) determine the effects that various parameters have on the size and charge density of the particles, (3) optimize the two-step filtration process, and (4) develop a quality control procedure.

Question 1: Relevance to overall U.S. Department of Energy objectives



This project was rated **2.8** for its relevance to U.S. Department of Energy (DOE) objectives.

- The development of a durable fuel cell coolant that does not suffer from performance degradation over time is very relevant to the DOE Hydrogen and Fuel Cells Program goals and objectives.
- This project is very relevant and supports the Program objectives. It could have commercial impact upon successful completion of scale-up activities.
- This project is highly focused with extremely clear and definite objectives that, if met, should enable significant advances in one key barrier for automotive and long-term stationary fuel cell applications.
- This project addresses the requirements for a better thermal management system for low-temperature automotive fuel cells. It appears to be meeting key requirements for corrosion inhibition and suppression of shunt currents. Developers appear to be keyed in to key requirements (e.g., low viscosity, good thermal properties).
- This project complements Small Business Innovation Research (SBIR) funding by enabling the scaling up of the synthesis process that provides an alternative glycol-based coolant. The coolant is not a critical part of DOE research, development, and demonstration objectives, but the project does have value. The emulsion polymer anionic resin nanoparticle approach is certainly novel.
- The main driver for this work seems to be the desire to eliminate the coolant de-ionizing filter, which, while it may provide value to original equipment manufacturers (OEMs), is not a significant enabler for automotive fuel cell commercialization. This reviewer questions whether DOE should support this work when critical enabling areas are underfunded.

Question 2: Approach to performing the work

This project was rated **3.2** for its approach.

- Dynalene has taken a straightforward, focused approach in developing a fuel cell coolant that maintains low electrical conductivity along with high corrosion resistance. The project has a very targeted objective and the approach directly supports that objective.
- The approach appears to be a wonderful, practical use of aqueously dispersed nanoparticles to achieve a bulk, ion-scavenging medium, as opposed to the usual filtration or surface adsorption device approach. The advantages are immediately obvious and the effectiveness is surprising.

- This project features a good approach to scaling up—going from 100-milliliter (ml) system, to a 500-ml system, and then a 10-L system. The project appears to have a good process-engineering approach to scaling up, with an eye for reproducibility.
- Using conventional water-glycol mixtures with a proprietary nanoparticle additive package simplifies the cooling loop in fuel cell systems by eliminating the deionizer column. This project builds on the success achieved in the SBIR program, during which the additive package was developed and patented. The additive is able to maintain electrical conductivity at the low levels required for fuel cells for the life of the stack (5,000 hours). The approach in this project is to scale-up the process, using an understanding of the mixing and stirring process on product performance in order to optimize the process. The cost target (\$10 per gallon) is to be equivalent with conventional glycol-based automotive coolants.
- The scientific work is proprietary and cannot be disclosed here. Therefore, the approaches used cannot be assessed, but progress has certainly been made.
- The materials are not designed to run hotter than 80°C, which makes them unacceptable for automotive applications. Nanoparticles may settle and clog stack channels and manifolds. The approach inherently reduces coolant conductivity.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- This project had made excellent progress toward developing a coolant that meets or exceeds operational lifetime requirements. The project has identified and developed a nanoparticle-based additive that allows standard coolants to meet operational requirements relating to electrical conductivity and corrosion resistance. The improved corrosion resistance will permit use of stack materials with reduced costs. In addition, in-house processes have been scaled-up for production of the additive, and additive production has been increased to pilot-plant scale.
- The investigators have made progress toward the stated objectives and are on track to achieve the objectives in August 2011, when the project ends.
- It is important to see that the investigators have identified a recipe for nanoparticle size and surface charge density. It is also good to see increased yield and optimized process control variables.
- The project team has achieved significant accomplishments since the last Annual Merit Review. The team finalized the recipe for the nanoparticle size and charge density, as well as optimized the production process for 10-L batches and 55-gallon drums and supplied the coolant to fuel cell developers. Mixer speed, location, and number of impellers; timing and rate of shot addition; and filtration were all optimized to produce the final product specifications.
- Progress has been straightforward and on-track with the project team's schedule to meet the targeted volume scale-up. The project was given a rating of three only because it addresses only one barrier, but the progress has been very good. The process issues the investigators have been studying and scaling up are fraught with complex mechanisms that could easily confuse many research efforts. The principal investigator and the project team appear to have a deep understanding of the processes involved.
- Dynalene has successfully scaled-up its nanoparticles and coolant to pilot scale. However, the material does not meet the needs of OEMs because it does not run at a high enough temperature and is more expensive than conventional coolant, plus investigators have yet to prove that settling is not an issue.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.5** for its collaboration and coordination.

- Lehigh University has to be one of the leading institutions anywhere to study and apply the fundamentals of emulsions to new material systems. This project features a good mix of applied process development at Dynalene and basic nanoparticle work at Lehigh.
- The collaboration with the university is very close and effective. Involving a fuel cell company in the evaluation of the coolant would have been good.
- Collaborations with appropriate university researchers is ongoing. Due to the proprietary nature of this project, which is directed at developing a commercial project, further collaborations appear to be difficult or not needed.

- It is not clear how much feedback commercial partners or stack developers offer. The investigators appear to have a good relationship working with Lehigh.
- Lehigh University is the only collaborator mentioned. Lehigh's role in the project is not indicated, but presumably it is in the development of the nanoscale additive package. As more of the coolant is placed in the hands of fuel cell developers, some additional collaborations might be anticipated.
- It is not clear what the team from Lehigh does, and there are no other collaborators.

Question 5: Proposed future work

This project was rated **2.8** for its proposed future work.

- The project is more than 75% complete and plans for completion of the remaining tasks are reasonable.
- The proposed future work is sufficient to reach the stated objectives at the end of the project.
- The project appears to be on track to achieve all of its goals.
- The project is nearly complete. The process will be scaled up to 100-L nanoparticle batches in the remaining time. This translates to about 5,000 gallons of finished product, which is enough for several developers to substantially test the material.
- Dynalene is not addressing the primary issue of needing to run at higher temperature with its materials. Without that, the process development and quality control efforts may be irrelevant.
- Scale-up is important. There should be thermal management system data, particularly regarding long-term stability. It is unclear if there is a plan to address calendar life.

Project strengths:

- The focused, directed approach is an area of strength. The project has resulted in a patented, commercial product that will be available to fuel cell manufacturers and operators.
- This is a small, very focused project. The necessary skills were included in the project and it has all worked well.
- This project features a good approach to scale-up and process control. It could produce some promising properties if the project team can achieve durability when producing at the larger scale.
- The project is focused on a single objective with good prospects for success. Success will be measured by how many fuel cell developers adopt the coolant.
- This project features a novel technology that is narrowly focused on key issues with good understanding of the application needs and basic materials requirements.
- Nanoparticles do seem to scavenge ions and reduce corrosion. There is a clear path for large-scale manufacturing.

Project weaknesses:

- One reviewer felt this project had no weaknesses.
- No fuel cell partners were involved in the project, although they are testing the product outside of the project. If they had been included, their testing data would have proved the efficacy of the technology.
- The risks of settling and blockage were not satisfactorily addressed. There is a cost penalty of adding nanoparticles. The coolant does not operate over the desired automotive temperature range.
- Not much of the general approach of the results can be shared with the larger Office of Energy Efficiency and Renewable Energy effort.
- It is not clear how many fuel cell developers have tested the coolant and how many are interested.

Recommendations for additions/deletions to project scope:

- This project should be allowed to be completed.
- The most important addition would be to develop materials design for operation up to preferably 120°C, or at least 105°C.

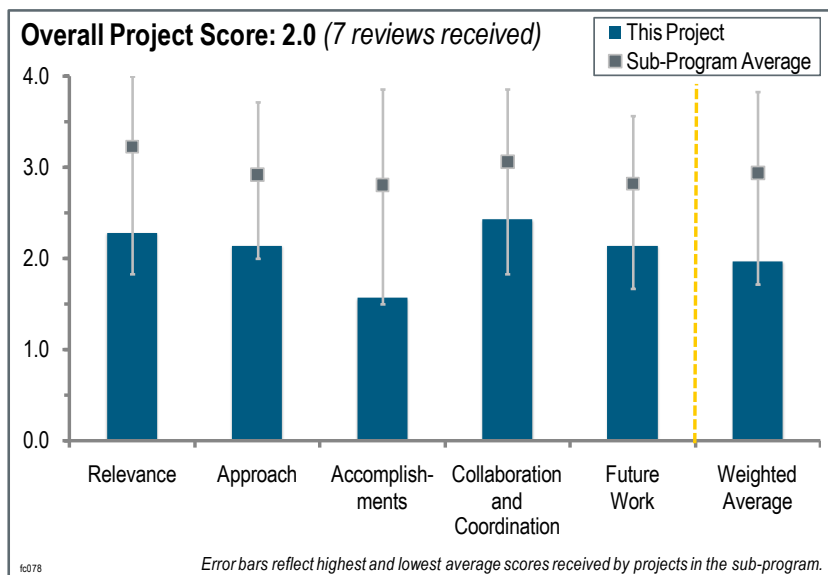
- The investigators should also test for settling and clogging of stack coolant channels, and look to increase conductivity.
- The investigators should obtain more stack and thermal management system data from power plants.
- The project team should plan for more widespread distribution of the coolant to the fuel cell community.

Project # FC-078: 21st Century Renewable Fuels, Energy, and Materials Initiative

Joel Berry; Kettering University

Brief Summary of Project:

The overall objectives for this project for 2010–2011 are: (1) developing an improved high-temperature fuel cell membrane capable of low-temperature (less than 100°C) starts with enhanced performance; (2) developing a 5 kWe (kilowatt-electric) novel catalytic flat plate steam reforming process for extracting hydrogen from multi-fuels, and integrating the process with high-temperature fuel cell systems; (3) developing an improved oxygen permeable membrane for high power density lithium-air batteries with simple control systems and reduced cost; (4) developing a novel high energy yield agriculture bio-crop (Miscanthus) for alternative fuels with minimum impact on the human food chain; and (5) expanding a math and science alternative energy educator program to include bio-energy and power.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **2.3** for its relevance to U.S. Department of Energy (DOE) objectives.

- This is probably the most ambitious project and includes high-temperature membranes, multi-fuel reformers, ceramic lithium-air batteries, development of renewable biofuels, and an education program on alternative energy.
- The project addresses a range of objectives, and it is unclear how many of the objectives address the DOE Hydrogen and Fuel Cells Program goals. A high-temperature membrane for fuel cells is a good objective.
- This project addresses only two of the relevance items listed by the authors: (1) development of an improved high-temperature fuel cell membrane capable of low-temperature starts (less than 100°C) with enhanced performance, and (2) development of high power density lithium-air batteries with simple control systems and reduced cost.
- This project has several distinct objectives, only some of which relate to the Program. The objectives include the following:
 - The relevant development of an improved high-temperature fuel cell membrane capable of low-temperature starts (< 100°C) with enhanced performance.
 - The relevant development of a 5-kWe novel catalytic flat-plate, steam-reforming process for extracting H₂ from multi-fuels, and integration of the process into high-temperature fuel cell systems, but with no clear indication if the barriers would be addressed.
 - The not-relevant development of an improved, oxygen-permeable membrane for high power density lithium-air batteries with simple control systems and reduced cost.
 - The not-relevant development of a novel, high-energy yield agriculture bio-crop (Miscanthus) for alternative fuels with minimum impact on the human food chain.
 - The not-relevant extension of the math and science alternative energy educator program to include bioenergy and power.
- Portions of the project are outside the scope of the Program. Development of a lithium-air battery and development of an agricultural bioenergy crop are not relevant to the Program.

- This reviewer does not believe these ideas are critical to the Program, though they do align with DOE objectives.
- This project is scattered. It includes membrane development, catalytic reforming, an oxygen permeable membrane, biofuels, and education on bioenergy. Only one portion relates at all to the Program.

Question 2: Approach to performing the work

This project was rated **2.1** for its approach.

- The approach on the high-temperature membrane is good. Substituted silsesquioxanes have shown some promise as a proton conducting enhancing additive in previous work in other membrane systems. The project needs to show more details of the water-gas shift work. The other work is not relevant to the Program.
- This project features a good approach, but some aspects of the work are omitted, such as electrodes for the high-temperature membrane or how the reformer will be able to process multiple fuel sources.
- This reviewer only saw results for the first three tasks.
- With only minimal data presented in any category (the categories are quite diverse and difficult for one person to referee), it is quite difficult to assess whether the approaches are likely to result in improvements. Moreover, the rationale for why the approaches, especially in the absence of data, should result in better outcomes versus the state-of-the-art approaches was not clearly presented. The testing procedures used to obtain conductivity and assess battery performance should be expanded.
- The feasibility of this project is questionable, considering it has four targets in the space of one year.
- This project includes a lot of scattered effort. It is unclear how the objectives and effort move toward the DOE goals.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **1.6** for its accomplishments and progress.

- Some accomplishments were obtained, but not a lot of data was presented. They casted a total of three membranes, which is poor progress. Other projects have casted that many and more in one day. No information is presented about the performance of these membranes, such as conductivity, water take-up, gas cross-over, or dimensions/uniforming of the casting. There is no valuable information presented. Investigators used a multi-meter to test their lithium-air battery, which is not an adequate testing method. Their battery could only hold voltage for approximately 16 days with no load. They present zero details on what the catalytic flat plate reformer looks like, what it is made from, what the catalysts are, what the operating procedure is, or what the model is composed of; nor is any prior work by the other developers mentioned.
- From the presented data, it is unclear how the project fits together and moves the effort forward. The development of the membrane is a good objective, but the level of progress beyond coating the three control membranes as described is unclear. The conductivity data for the membrane used for the lithium-air battery looks good, as expected for a polymer compared to a ceramic, but there is no data on the durability under storage or operation ion performance in a lithium-air battery. The data on the bio-ethanol production did not clearly demonstrate the benefits of the approach.
- For the high-temperature membrane, the investigators did not present any performance or characterization data, or a proof-of-principle of the primary stated objective of a start temperature $< 100^{\circ}\text{C}$. For the reformer, there was no cross reference of actual performance to DOE targets. The reformer does not show multi-fuel capability. The presentation did not list any accomplishments for renewable bio-fuels (no progress) or the Alternative Energy Education Program, although the author states those two project areas are 40% and 50% complete, respectively.
- There were limited results reported. Of the work presented, task three (the lithium-air battery) lacked experimental detail. Task two (the reformer) has been done better by many others. Task one (membrane conducting below 100°C) was interesting. Demonstrating a system that integrates the three pieces would be a good goal for the future.
- The results presented only represent two of the four research categories. The main results are from the catalytic flat plate reformer modeling. There are some interesting results here, but it is unclear if this flat, low-temperature design would be able to deliver the throughput required for stationary power application at a

sufficiently low cost. As for the lithium battery materials, no baseline materials are provided to compare the conductivity and open-circuit voltage values. More importantly, the most important data to obtain and present for such materials would be the voltage-capacity curves, number of discharge cycles, etc. The only result presented for the membrane section was a statement that some casting procedures for the control materials had been refined. For such a short project (one year), work with the novel materials would be expected by now. There were no results presented for the biofuels portion of the work.

- Results for the water-gas shift and membrane portions are insufficient to judge progress. The project has not included any conductivity or mechanical properties measurements for the membrane, and includes only a computational fluid dynamics analysis for the water-gas shift reactor.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.4** for its collaboration and coordination.

- There was collaboration with the other institutions listed.
- The work that was presented is mostly being done at Kettering, and the team seemed enthusiastic. The effort and progress of the work being done at Saginaw Valley State University was not evident in the poster or reported by the presenters.
- Partners are contributing, but because the parts of the project do not relate to each other, each individual principal investigator is essentially stand-alone.
- Interaction with a national laboratory or industry would have been beneficial.
- Project partner collaboration was not evident from the progress achieved in this project.

Question 5: Proposed future work

This project was rated **2.1** for its proposed future work.

- The membrane future work seems to be the most noteworthy. Reformers are becoming mature, especially for sulfur-free fuels. The battery work is interesting, but was not clearly presented. Integrating these three components seems like a worthy task.
- The proposed move to test the materials is good, but extensive durability and performance data is needed for the proposed activities under practical operating conditions.
- It is unclear if the remaining tasks will be completed, given the remaining time and budget for this project.
- The present work is insufficient to build on, and the rationale for the overall work is poorly described. Also, the future work is specified for fiscal year (FY) 2011 through FY 2012, but the project is supposed to end in June 2011, according to the slides. It is unclear which date is correct.
- The project is supposed to finish in June 2011, yet future work is listed for 2011–2012 and probably includes enough work to last through 2012.

Project strengths:

- This project covers virtually all areas of concern for DOE—high-temperature membranes, reforming, fuel generation, lithium-air batteries, and education.
- The membrane work is an area of strength. The authors should focus on proving that their modification can produce a high-temperature polymer electrolyte membrane that can operate stably between 100°C–120°C.
- The institutions involved will expose students to energy science. The topics chosen are very relevant to the Nation's energy needs.
- There are no strengths to this project.

Project weaknesses:

- This project has a wide range of focuses, and the future approach to move toward practical solutions was unclear in the presented slides. It is also unclear what technical barriers the proposed work addresses.
- The project is too diverse and appears to be ignoring some of the initially stated objectives.
- No real bio-derived fuel work was apparent during the presentation.

- The purpose of joining together so many unrelated tasks is puzzling. The chosen research tasks in every area should be compared explicitly to the state-of-the-art, both in the review slides and in the laboratory setting.
- A large portion of the work is not directed toward the Program goals and objectives.
- There are no results for task five, the educational program. More results would have been obtained if the scope was narrower. Four major tasks in diverse areas in one year is too broad.
- Most of this project is irrelevant to the Program. There is little or no progress to date. The project is not exploring any novel concepts related to fuel cells.

Recommendations for additions/deletions to project scope:

- There should be more focus on real testing of the proposed materials to show any potential benefits.
- Investigators should reduce the project to the most promising two areas, such as high-temperature membranes and reforming. They should delete the lithium-ion battery, renewable fuels, and education aspects.
- Further development of the high-temperature membrane so it can start below 100°C seems to be a worthy goal. Integrating the three components (fuel cell, reformer, and battery) to make a complete power source seems like a worthy task.
- In the future, fewer, directly related topics should be included in a single project. Breaking this project into four projects for membrane, reformer, battery, and biomass topics is suggested.
- The project team should delete the work on the lithium-air battery and biofuel, and concentrate on membrane development and the water-gas shift reactor.
- This project should be ended. At a minimum, the irrelevant sections should be deleted or funded from an appropriate funding source, which the Program is not.

Project # FC-079: Improving Fuel Cell Durability and Reliability

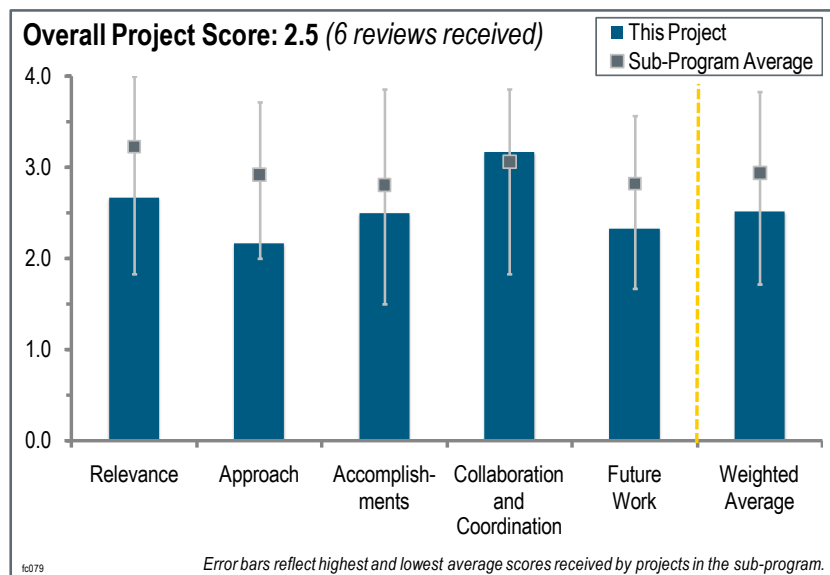
Prabhakar Singh; University of Connecticut Global Fuel Cell Center

Brief Summary of Project:

The objectives of this project are to: (1) develop an understanding of the degradation processes in advanced electrochemical energy conversion systems; and (2) develop collaborative research programs with industries to improve the performance stability and long-term reliability of advanced fuel cells and other power generation systems.

Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **2.7** for its relevance to U.S. Department of Energy (DOE) objectives.



- The project is relevant to the objectives of the DOE Office of Energy Efficiency and Renewable Energy, Fuel Cell Technologies Program's *Multi-Year Research, Development, and Demonstration Plan*. The activities are aligned to the overall DOE Hydrogen and Fuel Cells Program goals.
- The relevance of this project is very broad. It covers almost every element of advanced electrochemical energy conversion systems, including advanced fuel cells and other power generation systems.
- The project's stated objective to improve fuel cell reliability and durability is certainly relevant to DOE goals and objectives for fuel cells.
- Most project aspects support the Program objectives. Some project aspects, such as solar energy harvesting, are not directly related to the Program.
- This project has many diverse subprojects, a few of which are unrelated to fuel cells and hydrogen, such as solar, power electronics, and fluidized catalytic cracker modeling. The relevant subprojects cover the gamut of polymer electrolyte membrane fuel cells, phosphoric acid fuel cells, molten carbonate fuel cells, solid oxide fuel cells, and fuel processing. The subprojects address durability, performance, and manufacturing.
- The project objectives as stated are extraordinarily broad and diverse. The tasks, by default, address materials and systems issues that deal with the durability, cost, and performance of the various fuel cell technologies that the industrial partners are involved in. However, no one success in any of the 10 areas would likely enable commercialization success of that application.
- The high-level objectives of this project are good; however, the subtasks cover a variety of technical areas and the actual execution seems to be ad-hoc.

Question 2: Approach to performing the work

This project was rated **2.2** for its approach.

- Every approach in this multiple-project program seems reasonable. The approach does not qualitatively address any particular DOE targets.
- The approach for each subproject is rational and generally defined or supported by an industry partner that helps solve a specific issue. Not all subprojects have quantitative technical targets, but rather are aimed at understanding mechanisms and structures for the purpose of improving cost, performance, or durability.

- The approach involves soliciting proposals from individual professors who are, in turn, required to secure an industrial partner for their proposed subproject. Projects that have been selected are cross-cutting over several fuel cell types. The funding levels for the individual subprojects is not indicated.
- The approach of considering five different programmatic tasks and related subtasks in a two-year project seems very ambitious. This project is focused on the development and validation of the mechanistic understanding and subsequent creation of novel, cost-effective materials to mitigate degradation processes, which is supposed to happen through the collaborative programs between industry and university. Two years seems to be very little time for placing, executing, and completing such a broad collaborative project.
- The overall scope is incredibly broad for this project's relatively small amount of funding and time. It is very difficult to understand how this level of funding could be leveraged into any significant advances in any of the diverse set of five tasks identified, which cover fuel cell systems, fuel processing, advanced materials, H₂ storage, and solar energy/waste water treatment.
- The core of this project is unclear. The subtasks cover a variety of technical areas and fix durability problems. The approaches for each task do not seem to be systematic, and actual execution seems to be ad-hoc. Engineering methodology could be applied to improve the approaches and develop a more systematic approach.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.5** for its accomplishments and progress.

- Progress has been made toward identifying and synthesizing new materials that have the potential to address some technology problems.
- The prime contractor (the University of Connecticut) has pulled together a large team, as proposed in the proposal. The team has accomplished good progress in all of the tasks and subtasks in collaboration with eight industrial partners. The progress concerning biomass cleanup (desulfurization) for energy conversion is impressive.
- The progress is good considering the relatively short time spent so far. The biomass desulfurization subproject and the enzyme-based sulfur removal subproject present results, but do not provide a comparison to state-of-the-art materials. The total project is \$2.5 million. There is no task budget breakdown to compare progress to the budget.
- Work has begun on the selected subprojects and preliminary results have been reported in some cases. But with only 14 months left on the project schedule, much work is left to be done. The subproject selection process seems to have been time consuming.
- Technical accomplishments are on par with expectations for project funding, which is applied to 10 very diverse application areas and is one-third completed. In that sense, the work is probably utilizing its funding as planned. However, the accomplishments appear to be routine results of the application of common analytical tools (e.g., scanning electron microscopy), low level chemical engineering models, and simple laboratory bench testing of gases. The rate and depth of progress proposed in the objectives is inconsistent with the resource levels expended and the results obtained.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.2** for its collaboration and coordination.

- There is well coordinated collaboration between industry and the University of Connecticut Global Fuel Cell Center.
- Each subproject has a cost-sharing industry partner, which include fuel cell manufacturers and system integrators.
- The team has done a good job of engaging eight industrial partners. This will help in the assessment of the technology development from this project for direct practical applications.
- The large number of proposed collaborators is a strength of this project. However, the diversity of the proposed collaborators is a weakness, as there is no synergism among them and they simply dilute the small level of efforts that the principal investigator's institution can provide. It was not clear exactly how the principal investigator interacts with the various collaborators on a weekly basis.

- Each subproject has an industrial partner. Partners will have to play critical roles if meaningful results are to be achieved in these subprojects. There was no indication of the partners' actual degree of involvement in the subprojects.

Question 5: Proposed future work

This project was rated **2.3** for its proposed future work.

- The future work described in all of the tasks is clearly aligned with the project's proposed work. So far, the progress seems to align with the project timeline.
- Plans are focused on advancing the project, and do not specify barriers or go/no-go decisions. The proposed future work for each particular subproject is too detailed.
- Most of the subprojects have a duration of one year, so there is not much time left for additional effort. The future work for the relevant subprojects is reasonable and should provide some meaningful results for the industry partners.
- Plans for future work were vague and general, and lacked specifics. With only 14 months left in the project schedule, it is evident that most of the subprojects will not be completed in time. There is no indication of plans to continue work past the end of the project.
- The continued focus on so many diverse areas will only result in preliminary results with superficial benefits to any one area. If that is the intent, for later prioritization, then that may be acceptable. However, the stated goals and objectives strongly declare that much more will be accomplished than will actually be possible.
- The subtasks cover a variety of technical areas and fix durability problems. The approaches for each task do not seem to be systematic, and actual execution seems to be ad-hoc. Engineering methodology could be applied to improve the approaches and develop a more systematic approach.

Project strengths:

- The project brings together expertise in fundamental science and technology, and helps students understand the real impact of scientific research on technology.
- The major strength of the team is the involvement of different industrial partners with a wide breadth of experience. These partnerships will support technology developments in respective areas of the project.
- This project covers a variety of fuel cell technologies and provides technical solutions.

Project weaknesses:

- Some of the aspects of the project do not directly relate to the Program. Particular DOE targets are not addressed.
- Some of the subprojects, such as subproject 3.1, which deals with solar cell development, do not seem to relate to fuel cell reliability or durability.
- The involvement of many different organizations may make project management very difficult. It is hard to coordinate between multiple partners when the project has a timeline of only two years. There is no room for any incremental delay in any tasks or subtasks—a brief delay due to any unforeseen reason may jeopardize the overall project.
- The project is too diverse and the set of objectives and topics is unrelated.
- The lack of systematic problem solving is an area of weakness.

Recommendations for additions/deletions to project scope:

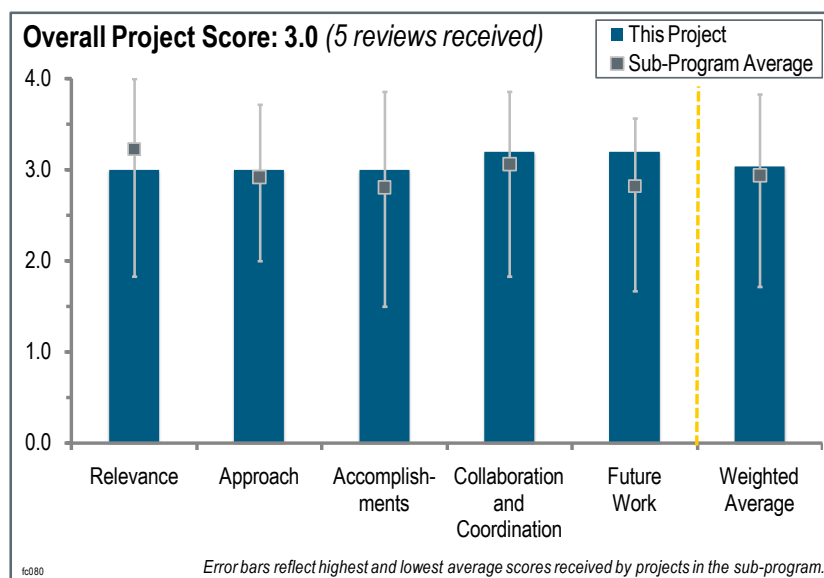
- The project team needs to specify a particular target for each project of this multiple-project program, as well as specify how the specified target is different from the state-of-the-art system.
- Investigators should prioritize and focus on just one or two of the 10 or so tasks, and try to have a more in-depth and significant impact on solving the critical gaps of those key areas.
- The subtasks cover a variety of technical areas and fix durability problems. The approaches for each task do not seem to be systematic, and actual execution seems to be ad-hoc. Engineering problem solving methodology could be applied to improve the approaches and develop a more systematic approach.

Project # FC-080: Solid Oxide Fuel Cell Systems Print Verification Line (PVL) Pilot Line

Susan Shearer; Stark State College

Brief Summary of Project:

The Rolls-Royce Fuel Cell Systems (U.S.) Inc. (RRFCS) 1 MW (megawatt) solid oxide fuel cell (SOFC) power plant concept is designed for base load stationary power generation applications. This project provides the test system necessary for long-term operation of the fundamental building block of the RRFCS 1-MW fuel cell plant—the fuel cell stack block—at full system operating conditions. Objectives for this project are to: (1) complete the electrical and mechanical build and commission test of the Stack Block Test System (SBTS); (2) provide a block-scale test system for the active fuel cell tubes produced by the Print Verification Line (PVL); and (3) perform initial commissioning tests to qualify SBTS operation and control.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.0** for its relevance to DOE objectives.

- This project's goal is to develop fuel cell technologies for early markets such as stationary power. The project provides a block-scale test system for active fuel cell tubes produced by a PVL. Accelerating the commercialization and deployment of fuel cells is a primary goal of the DOE Hydrogen and Fuel Cells Program. The project tests a PVL for anode and cathode electrodes on fuel cell substrates. One of the project's goals is to create the basis for future manufacturing decisions.
- Large-scale SOFC systems are important to the Program objectives.
- The RRFCS SOFC power plant concept for stationary power supports the Program's key goal to "develop fuel cell technologies for early markets such as stationary power (primary and backup)." This project provides the test system necessary for long-term operation of the fundamental building block of the RRFCS 1 MW fuel cell plant—the fuel cell stack block—at full system operating conditions. The investigators completed the SBTS and created or retained more than five jobs in Ohio. The supply chain benefited from procurements used in the fabrication and building of the SBTS.
- This project supports the Program objectives. However, the project scope is limited to completion of the commissioning of the SBTS.
- The project addresses durability and cost barriers, although the latter was not claimed in the presentation. It makes a valuable contribution, as the whole power plant is covered by Rolls-Royce.

Question 2: Approach to performing the work

This project was rated **3.0** for its approach.

- The approach is to address manufacturing issues through the PVL and assembly, and move to test systems from cell to block scale. Three tasks are included: control the software and human/machine interface; complete the stack and component wiring and install the stack instrumentation; and perform mechanical commissioning to

exercise all components and control loops, except those associated with stack electrical power. This project also utilizes student interns in a training program, creating an educated workforce that will be needed in the future. This project is a joint project with project FC-072. A goal is to do long-term durability testing (5,000 hours), which was not accomplished in this project.

- This project focuses on completing the control and electrical systems for the SBTS.
- Module testing is absolutely needed for development. It is unclear what percentage of the effort is covered by DOE funding, and whether RRFCS will fund its own test station development if it is seriously pursuing this development.
- The technical approach is good, though it is hard to evaluate because little data on progress is provided. With the information provided, it is hard to assess whether the project is well on track.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- All tasks described are 100% complete. The investigators accomplished about 200 hours of operating time at temperature.
- The tasks were completed as planned.
- The system development in this timeframe shows good technical accomplishments for the time.
- SBTS operation was successful at the end of 2010. The project team demonstrated SBTS mechanical performance over the required range of fuel cell operating temperatures, pressures, and anode/cathode gas compositions. Investigators also installed a stack prototype for anode/cathode flow circuitry, but it is not electrically connected. The team achieved about 200 hours of operating time at temperature in long-term operation in 2011, and demonstrated the control and safety system hardware and software up to powered stack operation. The SBTS is ready for powered stack operation in 2011.
- The work on the balance of plant and the electrical control of the plant seems to be making good progress; however, no proof was provided. Data should have been provided to verify the activities on the manufacturing line cell and stack performance. Although mechanical performance was mentioned as the first and foremost barrier for this project (and it indeed is for all of the SOFC projects), investigators did not provide any proof of improved mechanical durability through long-term tests, thermal cycling, or other customized mechanical tests for cells and stacks.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.2** for its collaboration and coordination.

- Collaborators include RRFCS, the Ohio Third Frontier program, and supply chain companies.
- This project featured good collaboration.
- This development was performed almost exclusively in collaboration with RRFCS. This allowed meaningful development without exposing Rolls-Royce's confidential design information.
- RRFCS seems to make a solitary development on the materials' and stack level. There are appropriate alliances for the plant development.
- This project featured collaboration with RRFCS and the Ohio Third Frontier program.

Question 5: Proposed future work

This project was rated **3.2** for its proposed future work.

- No future work has been proposed because this project is complete.
- This project was completed, so no proposed future work is required.
- The project is finished. It was rated well in order to finalize the review.
- This project ended in March 2011.
- More feedback on how this system will be used would be useful. Although no funding was mentioned, benefits of the system will be expected in testing and validation of modules. It is unclear if this development can benefit other stack development efforts, and possibly other original equipment manufacturers.

Project strengths:

- Students at Stark State College are trained in the technology, and many are hired by the Rolls-Royce Canton facility following graduation.
- This project focuses on completing the commissioning of the SBTS, and all of the tasks were completed as scheduled.
- The project was executed in a timely manner, with talented staff.
- This project is developing a whole system at a very relevant power level of 1 MW.

Project weaknesses:

- This project has no weaknesses.
- This project had a relatively short testing time. There are no obvious future benefits in expanded manufacturing capabilities.
- The systems development is way more advanced than the fuel cell stack development. The presenters mentioned very limited lifetimes of about 8,000 hours upon request in the discussion. The project team did not provide in writing any information on life expectancy of a stack. A life expectancy of 16,000 hours was mentioned in the discussion as a goal for an early-market introduction stage. Investigators did not provide any valuable proof of durability in case of thermal cycling. Economically, a lifetime of 40,000 hours is generally considered the lower limit for market introduction, and 80,000 hours of operating time is considered a reasonable target. Therefore, the RRFCS target does not seem ambitious enough. Most of the valuable information was acquired in the discussion. The papers presented did not provide enough information to assess the project.

Recommendations for additions/deletions to project scope:

- The project is complete. This reviewer has no recommendations for follow-on work.
- RRFCS might be advised to work on the degradation of the single cells, which apparently show 2% power loss in 1,000 hrs; longevity; and mechanical fatigue owing to cycling.

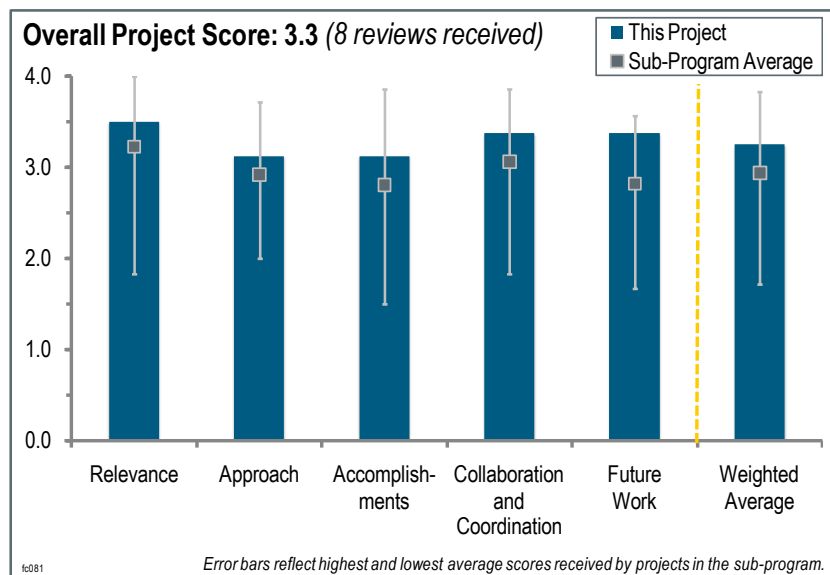
Project # FC-081: Fuel Cell Technology Status - Voltage Degradation

Jennifer Kurtz; National Renewable Energy Laboratory

Brief Summary of Project:

The objectives of this project are to: (1) benchmark (measure) state-of-the-art fuel cell durability; (2) leverage analysis experience to utilize analysis methods, experience, and data from fuel cell field demonstrations and laboratory and field data comparisons; and (3) collaborate with key fuel cell developers, including providing feedback, determining factors affecting fuel cell durability, and studying the differences between laboratory and field durability.

Question 1: Relevance to overall U.S. Department of Energy objectives



This project was rated **3.5** for its relevance to U.S. Department of Energy (DOE) objectives.

- This project is very interesting because it evaluates the current state-of-the-art technology.
- Data analysis and sharing assist the community to move forward with development.
- This project provides a means for valuable data gathering from field and laboratory tests for both individual customers (if they do not have the internal resources) and the fuel cell community at large.
- This project concerns analyzing data that has been accumulated at the National Renewable Energy Laboratory (NREL) Hydrogen Secure Data Center pertaining to the performance and durability of real-world, fielded fuel cells. The HSDC receives data both voluntarily from fuel cell providers and as a requirement for certain DOE-funded technology validation projects. This project distills this voluminous information into an accurate, fact-based assessment of the state of the technology with respect to fuel cell durability. Importantly, it also provides benchmarks of the durability differences between pampered, laboratory-based units and units fielded for different applications (e.g., automotive, backup power, or forklift).
- The Technology Validation sub-program is very important from the standpoint of confirming the results of the vehicle demonstration project through independent analysis. As the vehicle demonstration program winds down, the project is shifting to collecting, analyzing, and reporting data from fuel cell forklift operations and perhaps stationary fuel cell installations. Independent analysis is important to DOE to gauge the state of the technology and the effectiveness of its research portfolio.
- This approach is a great way to share information and data in a non-proprietary manner.
- Real-world durability demonstrations are very important to the overall DOE Hydrogen and Fuel Cells Program. However, with limited data, this project may misrepresent the state-of-the-art technology.
- It is useful to find and document the state-of-the-art technology, but this project does not seem well poised to move that forward, which is the true objective.

Question 2: Approach to performing the work

This project was rated **3.1** for its approach.

- The approach taken by NREL is very good. Maintaining control over sensitive information through the secure data room is essential to ensure participation by competitors in the fuel cell technology development. Allowing companies to review the composite data packages before they are released also provides an assurance that

sensitive information will not be revealed. The data products also provide information for the general fuel cell community to gauge the technology status and focus areas for further research and development efforts.

- This successful approach includes both a skilled analysis of fuel cell performance and the diplomacy needed to build confidence with potential suppliers of voluntary data.
- This project's approach is appropriate. More focus should be placed on achieving comparability of data among different data providers. Effects like cycle characteristics, environmental conditions, and system architecture should be explored more thoroughly.
- This project covers many kinds of fuel cells. The data comes from operation, but the conditions may not be well defined for transient information. The investigators intend to have more robust information on how data was taken, but cannot yet accomplish that feat.
- The approach is interesting. The team should really break the data out according to fuel cell type (solid oxide fuel cell [SOFC] versus polymer electrolyte membrane [PEM]) fuel cell, and separate and label vehicle, laboratory stacks, and module data. This reviewer realizes that the investigators group them together because the individual sample populations are small, but the duty cycles for each group are so different that they really should not be compared. The team should also use the data generated from vehicle stacks to develop a drive-train protocol for fuel cell vehicles for use in other DOE projects.
- Providing operating windows (control parameters) that span the data would be helpful. Narrowing these windows to include certain percentages of the data would help one evaluate the performance and durability results.
- The data analysis tools are great. The reviewer wants to know if there is any way to push them out to industry.
- Limited information can be gathered from the consolidated data presented. A clear separation should be made between projected life results and actual life results. Steady-state laboratory tests do not need this degree of analysis, and can be omitted.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.1** for its accomplishments and progress.

- DOE can use the consolidated data from the field results to report the status of technology for various applications.
- Accomplishments throughout the course of the validation program have consistently been very good. The progress in fiscal year (FY) 2011 may not be as significant as in previous years because of funding limitations. The presentation was unclear regarding which composite data products (CDPs) were and will be completed in FY 2011. Extending the durability projections into other applications—namely, backup power, forklift, and stationary power—is good and should be continued as more operating hours are accumulated in these near-term applications.
- This project features nice data analysis. The reviewer has also seen some of the reports online. The service and product offered by this project are very helpful.
- The initial results from this project are shown on slides 8–12.
- The project team developed a degradation curve based on the data in the voltage versus current graph. The team also extrapolated degradation to expected life, and correlated it with the generation of technology.
- The team has made significant progress on data analysis—hopefully more stacks will be provided in the future for analysis in the project.
- Defining some standard polarization curves for which developers could provide comparison data could increase interest in providing data.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.4** for its collaboration and coordination.

- Collaboration with partners that provide data to the project is critical for success, and NREL did an excellent job of engaging several players in the industry. It is recommended to continue expanding the partners' network in order to gain more statistical confidence in the data presented.
- Collaborations are extensive, with most developers—at least those in North America—willing to share their data with NREL for the purpose of producing the composite data packages.

- This project features excellent collaboration. Jen and the team's integrity (and the data center's security) help make it easy to share proprietary information.
- Collaboration is an important part of this project, as it relies on convincing fuel cell providers to voluntarily submit data. While the current collaborators were not disclosed in order to protect proprietary information, the presenter hit the right notes to demonstrate that the project has been modestly successful to date in convincing companies to participate (8 out of 22 contacted), and that diplomatic efforts were ongoing to convince others to participate as well.
- Investigators shared results with the voluntary data providers.
- This project has eight partners across several industries.

Question 5: Proposed future work

This project was rated **3.4** for its proposed future work.

- The future work plan is good, especially the study of the differences between laboratory and field durability projections and performance. Availability of funding may impede progress.
- Future work is aimed at increasing statistical confidence by increasing the breadth of statistical samples (more partners) and filtering data to eliminate noise coming from environmental factors.
- The project team should focus on analyzing the data to see how lifetime results depend on operating conditions, duty cycle, ambient conditions, etc.
- The future work outlined by the principal investigator was on slide 14. The most important future work of this project is perhaps the continuity of the project itself. Fuel cell durability (more specifically, the lack of durability with respect to targets) is one of the greatest impediments to the widespread adoption of this technology. It is vital to the Program to have reliable, fact-based assessments to demonstrate the maturation of fuel cell technology. Reliability claims should reflect the broad, real-world experience of users rather than cherry-picked anecdotal examples.
- The future work is suited to the task the investigators set for themselves.
- The project team should keep adding to the data pool—it can only help.

Project strengths:

- This project's strengths are its relevance to the Program and its usefulness to the fuel cell community.
- Providing a single consolidated comparison of life data and projections as well as conducting comparative analyses of different applications and laboratory data versus field data were areas of strength for this project.
- As the advertisements for an aerospace company say, this project is "turning data into knowledge."
- This project features a wide spectrum of uses and makers.
- This project represents a great concept regarding understanding what is really happening in the field.
- This project features well-established and accepted protocols for handling sensitive information. The project team displayed a strong willingness to tailor analyses to meet requests for specific comparisons or information.
- This is a great way for the fuel cell industry to collaborate.

Project weaknesses:

- The data presented could have been influenced by external factors, making comparisons quite difficult.
- The project should not lump together different technology types (i.e., PEM fuel cells and SOFCs). A linear decay assumption for projected life may not be sufficient for all stacks or systems.
- While the data sources are rather broad based, it is still not a completely representative sample of the current technology.
- Data conditions may vary and add to uncertainty.
- Execution relies on data, and developers gain no utility from comparing data at widely different conditions.
- One area of weakness was the unwillingness of some developers to share data sets with DOE. The presentation was unclear as to which CDPs were completed since the last Annual Merit Review.

Recommendations for additions/deletions to project scope:

- The project team should increase its efforts to understand the “stressor” pertinent to a specific cycle in order to achieve comparability among different datasets.
- Steady (single point) tests should not be included. The investigators should correlate results with operating conditions and duty cycles.
- If a regular and controlled test could be done on scheduled or timely occasions, the data would greatly improve. The project team needs to complete its intention to subdivide the data by test conditions or type as soon as practicable.
- The investigators should continue with the acquisition of data from non-automotive and near-term applications, particularly durability data for incorporation in lifetime projections.
- Adding tools for the industry to this project would be great.

Project # FC-083: Enlarging the Potential Market for Stationary Fuel Cells through System Design Optimization

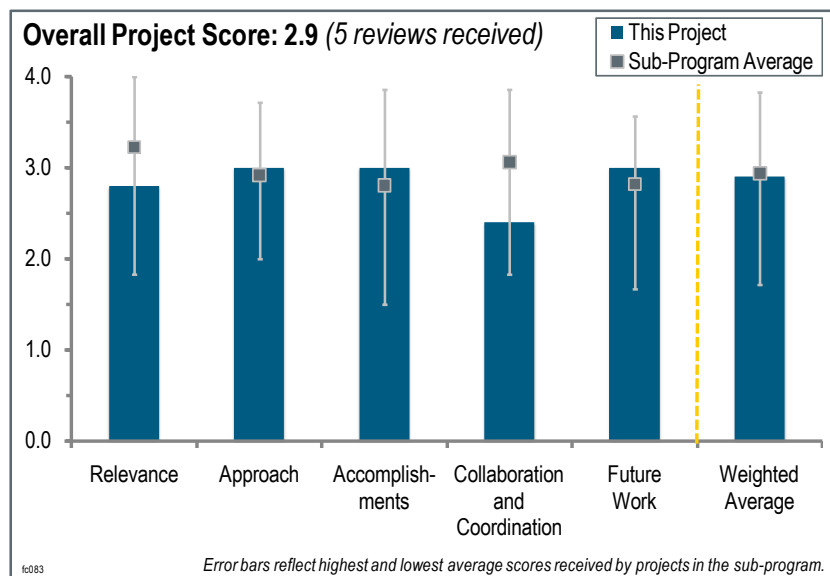
Darlene Steward; National Renewable Energy Laboratory

Brief Summary of Project:

The overall project objective is to determine optimum fuel cell types, sizes, and control strategies to meet economic and environmental goals. The project will model fuel cells in realistic combined heat and power (CHP) applications to provide guidance for designing and manufacturing stationary fuel cells.

Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **2.8** for its relevance to DOE objectives.



- The project is relevant to DOE's goal of developing fuel cells for CHP applications. There are many technical barriers to commercializing fuel cells for CHP, including cost and durability. This project is indirectly relevant to overcoming these barriers.
- This project is relevant to market transformation activities and market development. The compilation of building load data and other building attributes would be useful. Depending on how the model is developed, it could be a very useful tool for fuel cell developers who are interested in the CHP market.
- This project directly supports the DOE Hydrogen and Fuel Cells Program (although perhaps it is not critical, as some of the fuel cell manufacturers are doing similar analyses). This project seems to fit better with the Analysis subprogram (based on the barriers addressed) or the Market Transformation sub-program (based on the project title and objectives), rather than the Fuel Cell sub-program.
- The purpose of the project is a bit muddled. The purpose of the presentation seemed to be to develop guidance to determine the selection of a stationary CHP fuel cell to match the characteristics of a particular building, climate, and application (e.g., in-building loads). Slide four indicates that the purpose is to assist manufacturers by determining a suite of standard types and sizes of units that would meet market needs while lowering costs by having a well chosen set of standards. The title suggests that the purpose is to engineer design optimizations, such as "energy control strategies" (slide 14), that can deal with transients and outages (slide 13). On balance, it seems that the first two purposes dominate, and that this project has more to do with marketing than developing or improving the technology. For this reason, this reviewer does not believe this project to be "critical" to the Program. Furthermore, while this project may be an interesting intellectual exercise, the reviewer does not believe that it can be fully successful as a marketing tool unless one can somehow map the actual product (specific, marketed fuel cells) onto the model fuel cells developed for this project.
- A detailed list of the requirements for various buildings with an assortment of tenants in various climates is not critical to the Program. However, some additional details are warranted, at least for a few target building applications.

Question 2: Approach to performing the work

This project was rated **3.0** for its approach.

- This project's approach is to develop a high-level model to capture the interactions between the building loads and the fuel cells. The approach is clearly spelled out, and relies on literature review to gather data and models.

- This project is being approached as a modeling and simulation problem. While appropriate, this reviewer does not understand why this is being managed from the Fuel Cell sub-program, rather than the Systems Analysis (AN) sub-program. The reviewer's first reaction was that this project could have duplicated work that AN had already done, but that seems to have been an unfounded concern. Indeed, the author (Steward) has a major role in developing the Fuel Cell Power Model within AN, and can leverage that knowledge into this work. However, there are also some similarities with the work reported at the 2010 Annual Merit Review by Mahalik (AN-003/2010) and Greene (AN-004/2010), specifically regarding the H2A model tri-generation fuel cell system. Overall, the project seems to be going well as a modeling exercise, and the types of information being gathered seem appropriate.
- It is not clear how the CHP market will be segmented in the model, or how the optimum fuel cell type and size will be determined. The outcome could depend significantly on how the market is segmented (e.g., by electrical demand, heat demand, or cost of electricity). It is not clear what input is being used for determining fuel cell performance, efficiency, and cost, or if the end user will input these variables into the program, which would probably be the most useful method. It is also not clear what data will be used to validate the sub-models. The reviewer is not aware of CHP demonstration projects in the United States of a large enough scale or breadth of scope to produce data for model validation.
- Given the degree of market readiness and market penetration, it seems more logical to initially include molten carbonate fuel cell (MCFC) systems rather than solid oxide fuel cells (SOFCs).
- This project appears to be starting from scratch, whereas a lot of what is required is already available.
- Building databases exist—a good place to start might be DOE's Innovation Hub for Energy Efficient Buildings (<http://gpichub.org/>). Additionally, fuel cell system models exist, including CHP and combined cooling, heat and power (CCHP) system models.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- This project is at an early stage, but has made excellent progress in a short period of time.
- This project appears to have made good progress, but probably could be even further along if existing building and fuel cell resources were utilized.
- This project has just started. Much of the initial effort has focused on literature review and designing the graphical user interface (GUI). At this point, many of the modules shown in the GUI screen layout for system setup are conceptual in nature.
- The primary accomplishments mentioned were “screen design” and “screen layout.” However, these are only cosmetic. The depth, substance, and accuracy of the models constructed are far more important, and it is too early to make judgments on those.
- The project is early in the scope and has no real accomplishments yet. Investigators have developed several sub-models, but no results have been shown.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.4** for its collaboration and coordination.

- This project seems synergistic with other modeling (analysis) projects within the Program, and could benefit from more interaction with them. Also, the project is supposed to develop “energy control strategies,” but the “Smart Grid” project offers tangible substance for that goal and the relationship between these two efforts is unclear. Directed Technologies, Inc. (DTI) is identified as a partner, which could be useful to the extent that this project is meant to minimize costs (bullet two on slide four).
- Collaborators include the University of California, Irvine; the Colorado School of Mines; and DTI. The roles of the University of California, Irvine and the Colorado School of Mines are unclear. Collaboration with some fuel cell manufacturers targeting CHP applications would be beneficial (e.g., Idatech, Intelligent Energy, etc.).
- This reviewer did not see any plans to collaborate with actual fuel cell manufacturers, who have done a lot of this kind of work already. They should at least be reviewing the assumptions and results of the project.
- This is a single-institution project. The project lists three other institutions as reviewers and partners.

- The investigators definitely need to collaborate more with others working on distributed generation, CHP, and building technologies.

Question 5: Proposed future work

This project was rated **3.0** for its proposed future work.

- This project has a good, well-structured work plan.
- The planned future work consists of model validation in the first and second quarters of fiscal year 2012, and model application after that.
- Validation is a key component of future work, and validating fuel cell performance and cost models is necessary. Cost estimates have significant error bars. Validating manufacturing and cost models will be difficult.
- This project needs more emphasis on comparisons with other alternatives—not just other CHP alternatives, but also the current status quo, which is not CHP. This study will not reveal the key barriers to CHP commercialization if it completely ignores the present alternatives.
- The proposed future work (slide 18) is appropriate, although it is not entirely clear what the purpose or focus of this project is supposed to be.

Project strengths:

- The approach and plans are well laid out. The scope of the project is very broad, as it includes different types of fuel cells, refrigeration cycles, electric generators, energy storage systems, and even renewables.
- This is an in-depth modeling project that could be useful for planning and forecasting purposes.
- The willingness to start from scratch is an area of strength. The principal investigator will fully understand her work.

Project weaknesses:

- At best, the project will produce a high-level model. The model will likely be more suitable for policy studies than providing guidance for designing and manufacturing fuel cells. DOE may consider moving this project to the System Analysis team because it also lists 4.5.B, 4.5.D, and 4.5.E as the barriers being addressed.
- This project has an unclear focus and utility.
- If the model is not made publicly available, its utility will decrease.
- Starting from scratch and not taking advantage of what already exists are two areas of weakness.

Recommendations for additions/deletions to project scope:

- DOE may consider moving this project to the System Analysis team because it also lists 4.5.B, 4.5.D, and 4.5.E as the barriers being addressed.
- Adding fuel cell manufacturers to the team would be beneficial.
- Given the degree of market readiness and market penetration, it seems more logical to include MCFC systems initially, rather than SOFCs.
- The project team should make the model publicly available. To be useful, it has to be used by potential fuel cell users.
- If this work continues beyond fiscal year 2011, it should be integrated with the DOE Office of Energy Efficiency and Renewable Energy's Building Technologies Program.

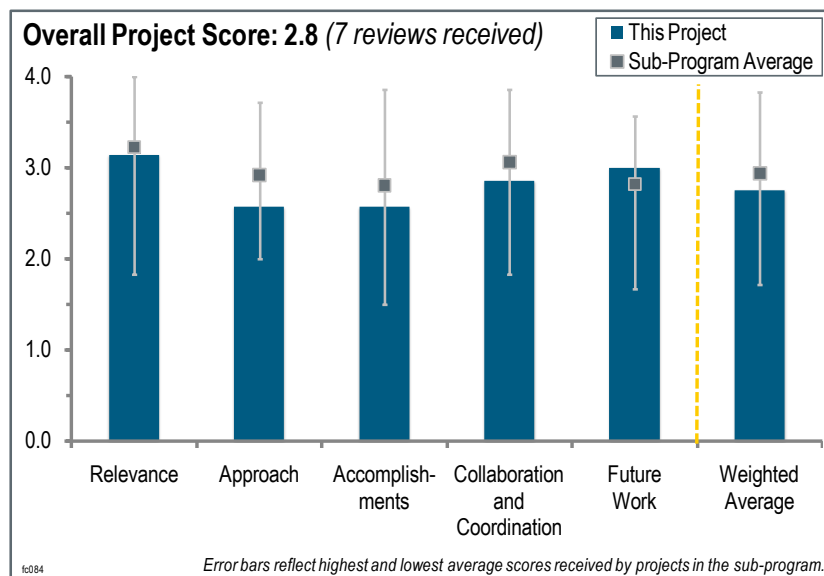
Project # FC-084: WO₃ and HPA Based System for Ultra-High Activity and Stability of Platinum Catalysts in PEMFC Cathodes

John Turner; National Renewable Energy Laboratory

Brief Summary of Project:

The overall objective of the project is to improve the electrocatalyst and membrane electrode assembly durability and activity by using platinum/tungsten trioxide (Pt/WO₃) and heteropoly acid (HPA) modification to approach automotive polymer electrolyte membrane (PEM) fuel cell activity (a four-fold increase) and durability targets (5,000 hours/10 years).

Objectives are to: (1) enhance Pt anchoring to the support by suppressing loss in the Pt electrochemical surface area (ECSA) under load cycling operations and enhancing electrocatalytic activity; and (2) lower support corrosion through increased durability under automotive startup or shutdown operation and suppressed Pt agglomeration or electrode degradation.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.1** for its relevance to U.S. Department of Energy (DOE) objectives.

- By reducing the cathodic over-potential, the project addresses one of the key issues to decreasing the Pt loading of PEM fuel cells—goal C. Better platinum-anchoring to the support serves goals A and C. Goal A is pursued through reducing agglomeration and support corrosion.
- Putting Pt on an oxide is a good approach to reducing corrosion and taking advantage of metal support interactions.
- Fuel cell durability is a cost issue, and this work addresses electrode issues that impact cost and performance.
- This project attempts to address multiple barriers (activity and durability) with a single technology development—an alternative catalyst support based on WO₃. This objective is good if it can be done. The target for catalyst support durability listed on slide five is the old target of 100 hours at 1.2 V (volts). The new target is 400 hours and the investigators should use this goal.
- High activity, robust catalysts are critical for enabling fuel cell system commercialization. Eliminating carbon from electrodes could eliminate the need for operational fixes for start/stop degradation.
- This project addresses the durability of fuel cells, and specifically addresses catalyst support durability.
- There is little indication here, or in the literature, that this route will achieve significantly greater oxygen reduction reaction (ORR) mass activity, although durability may be enhanced.

Question 2: Approach to performing the work

This project was rated **2.6** for its approach.

- Tungsten oxide supports have promise. The suggested accelerated stress test cycles only between 1.0–1.6 V. When looking at alternative supports, it is important to include cycling to lower potentials as well as high potentials in the accelerated stress test. Platinum-support interactions are important in determining agglomeration and Pt mobility on the surface. These interactions will depend on the oxidation state of the Pt

particles, and going from oxidized Pt at open circuit voltage and higher potentials to reduced Pt at lower potentials (during operation, which will occur between each start-stop cycle) will change these interactions. In addition, there may be some reduction of the support, which would influence these interactions as well, especially in systems such as the tungsten oxygen system where substoichiometric oxides exist and tungsten bronzes could form. The difference in Pt deposition observed between WO_3 and substoichiometric tungsten oxides (WO_x) suggests that there are differences in Pt- tungsten oxide interactions with different tungsten oxide oxidation states, and that cycling to lower potentials may be important.

- The approach to use support materials other than carbon for Pt is straightforward. Tungsten oxide is one of the materials of choice.
- Literature suggests promise with these proton conducting materials, and this work builds on that established scientific base.
- It is good that the approach focuses on one material system, thereby building some in-depth understanding about this new support system. However, there may not be enough fundamental characterization of the subsequent catalyst particles and interfaces with the supports.
- A literature search suggests that this approach (using WO_3 as a support) is reasonable. It is not obvious how the HPA helps prevent corrosion or anchor Pt to the support. The approach includes hybridization with HPA on carbon, which would assumedly still have corrosion issues.
- The approach to do a metal oxide support seems reasonable. Prior researchers have had similar DOE projects that have been judged very critically. The investigators should look at the work done by Adzic on Pt-niobium dioxide (Pt-NbO_2)—it is not clear why this work was not mentioned as part of the background.
- There is no fundamental understanding of why an interaction between Pt and WO_x may enhance the ORR activity of the cathode. There is some science missing here. Perhaps some modeling would be useful.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.6** for its accomplishments and progress.

- This project has really only just started, but progress has been good. It is good that baseline performance has been established early.
- The project is just completing its first year and has made a good start to date. Investigators have made tungsten oxide supports with various geometries and developed alternative methods to measure electrochemical area that will not be affected by the formation of tungsten bronzes.
- Investigators presented considerable work that speaks to possible new supports that might reduce the rate of Pt migration. Much progress was reported in the fabrication of unique structures, namely Pt loaded on tungsten oxides or on heteropoly acids (phosphates). There were indications of enhanced stability, but that was based on comparisons of data obtained with classical carbon support materials.
- This project has reached a very modest number of milestones for one year's effort, though it is only 13% complete. It appears from the data that Pt still catalyzes the corrosion of WO_3 with similar oxygen evolution reaction currents as from polycrystalline Pt. This data suggests that support corrosion may still not be improved over Pt on graphitized carbon. This data also suggests that the investigators should compare corrosion resistance to Pt on graphitized carbon instead of Ketjen Black (KB) which is the least stable carbon, in order to see if their approach can significantly improve the state-of-the-art materials.
- Investigators have made some Pt dispersed on WO_3 using Atomic Layer Deposition. The corrosion appears better than KB and similar to graphitized KB. The limited cyclic voltammogram data is confusing. Investigators did not present any activity or fuel cell data. There is no stability to the voltage-cycling data to validate the premise that Pt is anchored to WO_x support.
- Investigators seem to have made a lot of progress on making the catalysts, but there is no real information on the catalyst performance. It is impossible to evaluate the results from the electrochemical performance on page 18. This reviewer wants to know what the loading and rotation rate are. This team should know how to report electrochemical performance.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.9** for its collaboration and coordination.

- The consortium is well balanced between industry giving advice and research groups performing the tasks.
- The National Renewable Energy Laboratory (NREL) has subcontracted two local Colorado universities to assist in the fabrication work. 3M and Nissan are also advising NREL.
- NREL has put together a potentially strong team. To date, there is no evidence of contributions from 3M or Nissan.
- Involving industrial partners at different levels is useful. Perhaps the principal investigators might consider contacting Global Tungsten Products as key WO_x experts.
- The principal investigator has just two significant collaborators—the University of Colorado, Boulder and the Colorado School of Mines—that contribute work to the project. This collaboration may be adequate for focusing on just one material system, but there would appear to be a significant opportunity for more fundamental characterization of the materials that the investigators are generating.

Question 5: Proposed future work

This project was rated **3.0** for its proposed future work.

- The plans for future work align with the project objectives and build on the initial work.
- The topics mentioned are very important and the future work is sound as outlined.
- NREL was correct to question the stability of tungsten oxides—compounds that form with variable stoichiometry. Indeed, various forms of WO_x will demonstrate different electronic conductivity. This clearly is a very complicated system, and worthy of additional study. Building PEM electrodes using the ink approach is also an essential task. There are many variables in making useful catalyst formulations.
- The project is still fairly new—the investigators need to get some credible electrochemical results.
- The future work seems well focused on two important issues. However, in contrast to trying to get even smaller Pt particles for higher surface area (which will only exacerbate the Pt dissolution phenomena from high-voltage cycling), investigators should consider coating extended films on the WO_3 supports. Coating will generate a more stable form of Pt with higher specific activity to compensate for any lower ECSA. The addition of carbon to increase the conductivity of WO_x seems counterproductive from a durability standpoint. It would be useful to look at the projected costs of the catalysts if they need special WO_3 supports with HPA functionalization.
- The details of the future work are ill defined. Statements such as “Continue to achieve better control of Pt nucleation and dispersion” give no indication of how this achievement will be accomplished or how difficult it will be. There is also no indication of electrode optimization methods. The work on “structure activity relationships” is also unclear.

Project strengths:

- The partners and collaborators represent a wide range of expertise. The approach is rational.
- The expertise of the collaborators in the main pathway of the approach.
- Tungsten oxides should be more stable than carbon at high potentials, and the presence of HPA should provide a proton conduction path.
- This project features a good experimental basis and industrial advice, so the work is correctly focused.
- This project features a very strong materials component with synthesis. Introducing new fabrication methods is another area of strength for this project.
- The principal investigator is excellent.
- A potentially strong team exists, assuming everyone contributes. There is potential for WO_x to be a decent corrosion resistant catalyst support. The project also features excellent materials processing and characterization capabilities, including electrochemical and fuel cell testing.

Project weaknesses:

- This project has no major weakness.
- There is perhaps not enough fundamental characterization of the catalyst particles' structures (e.g., surface facets) and their interfaces with the WO_3 supports.
- The focus on HPAs for enabling proton conduction is an area of weakness. The project team should have considered making electrodes with conventional ionomers. Also, there is a lack of electrochemical data to this point.
- The electrical conductivity of the most stable tungsten oxide support may not be adequate. The presence of added carbon may lead to degradation at high potential.
- There remains some real question about the hydrolytic stability of heteropoly acids. Historically, these compounds have been successfully used for useful proton conductors. However, they are fragile, and when cold and wet, they imbibe water, swell, and dissolve. The thought to bond a crystal of these materials as a way of increasing durability seems unproven. It is perhaps possible to keep part of the phosphate complex attached, but there is no reason why the majority of the particle would not be addressed by a reaction with water. Slowing the wetting process by surrounding the particles with a hydrophobic shroud might be possible, but slowing is not stopping.
- This project features some ill conceived ideas about metal-support interactions and the role of electronic conduction in supports. There was also a very poor representation of electrochemical data from a strong team.
- There is no fundamental science explaining why this project should succeed, so it will be difficult to understand the failures that necessarily occur in research.

Recommendations for additions/deletions to project scope:

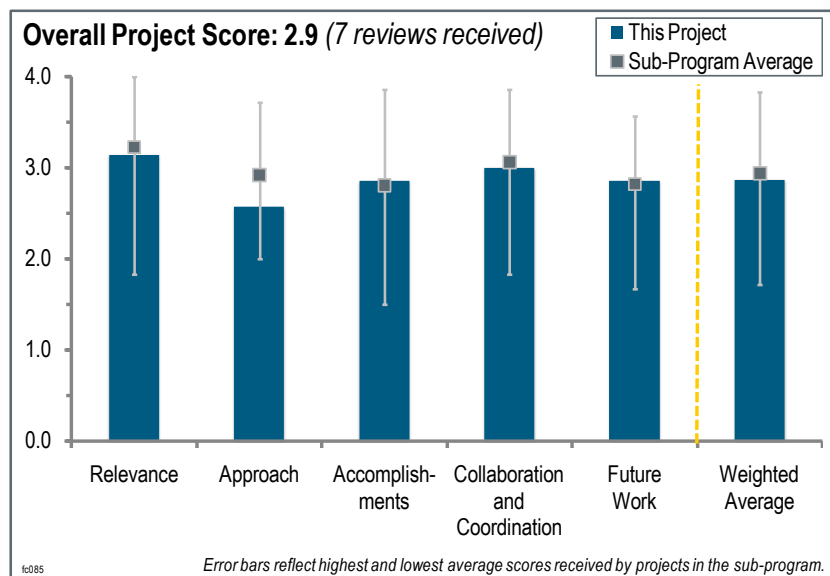
- This work is fundamental in nature. The issue is to show that WO_x has utility (e.g., is durable, offers suitable performance), and then proceed to make that performance adequate to address DOE targets. Some issues, such as Pt particle size, are not central. Instead, the utility of WO_x as a support should be the focus.
- This project has only just started. The scope is perhaps ambitious. Investigators should focus on the Pt/ WO_x work and not consider the complexity of the HPA until much later.
- If WO_3 nanorods and other shapes can be easily generated, they should be looked at as supports for extended thin catalyst film coatings, either in this project or in the other NREL project, FC-007.
- The near-term focus should be on getting acceptable activity with WO_x supported catalysts. This should include running standard DOE voltage-cycling tests (spanning the Pt oxide transition region) to validate that Pt anchoring is in fact occurring. Intentionally adding carbon seems counterintuitive. The project team should also look at electrodes containing traditional electrolytes, such as Nafion.
- The project should include cycling to lower potentials in durability testing.
- This fascination with the electronic conductivity of oxide supports, as given in this presentation and others at this review, makes no sense. If WO_x can be doped to have a conductivity of 10^{-1} S/cm (siemens/cm), it is still four to five orders of magnitude lower in conductivity than carbon. The researchers should focus on making nanomaterials and increasing the contact with carbon so that the conduction occurs at the relevant boundary with the Pt/ WO_x , instead of making the catalyst conductive. They should not bog themselves down with the electronic conduction issue.

Project # FC-085: Synthesis and Characterization of Mixed-Conducting Corrosion Resistant Oxide Supports

Vijay Ramani; Illinois Institute of Technology

Brief Summary of Project:

The objectives for this project are to: (1) develop and optimize non-carbon mixed conducting materials with high corrosion resistance, high surface area (greater than 200 square meters/g [gram]), and high proton (greater than or equal to 100 mS/cm [millisiemens/centimeter]) and electron (greater than 5 S/cm [siemens/cm]) conductivity; and (2) concomitantly facilitate the lowering of ionomer loading in the electrode with enhanced performance and durability, by virtue of surface proton conductivity of the electrocatalyst support.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.1** for its relevance to U.S. Department of Energy (DOE) objectives.

- The topic is important for successful completion of fuel cell catalysts development. The proposed work is relevant to the goals of the DOE Hydrogen and Fuel Cells Program.
- Corrosion-resistant catalyst supports would improve durability and simplify automotive fuel cell systems.
- Improved stability supports provide a path to increased durability of the catalyst and thus the membrane electrode assembly (MEA).
- Carbon corrosion is a well-known failure mode that threatens automotive fuel cell stack durability. Although system mitigation strategies can be found and more types of graphitic carbon have been used, carbon still experiences corrosion due to the stresses generated by certain vehicle operating modes. As this corrosion may limit stack lifetime, a project seeking to replace carbon supports is relevant.
- The durability of cathode materials and other fuel cell components is very important in the quest to fully implement polymer electrolyte membrane (PEM) fuel cell technology. One way to avoid the corrosion of cathode supporting materials is to develop and optimize non-carbon mixed conducting materials with high corrosion resistance, high surface area, and high proton and electron conductivity. As the principal investigators propose, one way to achieve this would be to use conductive metal oxide supports such as silicon dioxide (SiO₂) and Ru dioxide (RuO₂). However, in order to justify the use of these oxides, the principal investigators need to discuss the cost issues associated with preparing and synthesizing these oxides, as well as advances of these oxides relative to inert support developed and used by 3M.
- The proposed development of corrosion resistant supports for polymer electrolyte fuel cell electrocatalysts is a relevant topic for the Program. However, as demonstrated in the ongoing 3M projects, the 3M nanostructured thin film support eliminates the issue of support stability.
- Replacing carbon support materials is an important project; however, there is little evidence that replacement with metal oxides will be an acceptable approach. Objectives include high corrosion resistance—for a metal oxide, the corrosion resistance must almost be absolute unless the metal is a valve metal that is not easily deposited on the Pt catalyst.

Question 2: Approach to performing the work

This project was rated **2.6** for its approach.

- This seems like a good approach to developing and testing the supports. The principal investigators seem to be on a path to improving the measurement techniques.
- Enhancing the proton conductivity of certain oxides and combining those oxides with electronically conducting ones is a reasonable approach, although many questions regarding functionalization have to be answered. Using RuO₂ as a model system may not help much for several reasons. Its conductivity cannot be matched by other oxides, and it is not stable at high potentials.
- Ex-situ characterization seems to be lacking, other than transmission electron microscopy (TEM). Cost needs to be addressed, especially with Ru and complicated synthesis. The test cycles need to be widely vetted. It is not clear whether the mixed system will eliminate the need for ionomer in the catalyst layer. There is no discussion of interface issues between the membrane and the mixed conducting electrode/support structure.
- This is a relatively good approach, mostly focusing on classical synthesis and testing protocols. The principal investigators should consider implementing a surface science approach to characterize the nature of oxides under operating conditions, utilize TEM for monitoring Pt size and agglomeration, and develop methods for monitoring Pt dissolution.
- The approach may overcome some barriers, but may also introduce other issues. The choice of materials is questionable. The presentation lacked the rationale behind why lower ionomer content in the electrode layer are necessary—it implied that Pt utilization is decreased due to a lack of proton conductivity to the catalyst sites. Depositing electronically conductive oxide nanoparticles on nonconductive oxide supports to impart electronic conductivity to the support raises questions about the stability of the electronic conductivity, as nanoparticles are known to migrate and agglomerate in the fuel cell environment. Also, RuO₂ may not be a good choice as the electron-conducting component, as RuO₂ is known to be unstable from electrolyzer O₂ evolution electrocatalyst development efforts. In its screening tests, the project team should measure the stability of the proton and the electronic conductivity of the material as a function of potential cycling, and not just the surface area or weight loss of the materials. The project should also attempt to deposit on the supports in the screening stages and before optimization of the composition, as Pt is known to accelerate the oxidative corrosion of materials. The stability screening without the presence of Pt may not be valid to the actual stability of the supported catalyst.
- The material describing what is required in a catalyst support leaves out one criterion—cost. The project investigates the use of a series of supports based partially on Ru, so this is an interesting omission. The ionomer reduction efforts are intriguing, but raise the question of whether the incumbent ionomer or the support material sulfonation is better from the perspectives of cost, performance, and durability. As the proton conduction function remains, ionomer reduction itself is not unconditionally advantageous. The project is to be commended for seeking surface area, conductivity, and durability at the same time, rather than prioritizing one property over another.
- It appears the project has arbitrarily picked RuO₂ as a material, and, other than the SiO₂, no other oxides are under consideration. RuO₂ is not fully stable. This reviewer believes the researcher should have checked the Pourbaix for solubility values.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.9** for its accomplishments and progress.

- Given that the project has just started and the subcontract with Nissan has just been established, the progress is good and the initial conductivities of the materials are promising.
- Investigators at the Illinois Institute of Technology (IIT) have done a good job confirming that their materials can achieve surface area and electrical conductivity targets without catalyzation. While the proton conductivity is lower than hoped, a cell test would be the best way to confirm whether or not proton conductivity is sufficient. Durability should be confirmed after catalyzing the materials, and preferably in an operating fuel cell. The materials have still not been catalyzed.
- For a project that has been ongoing for less than one year, the accomplishments are good. The stability of RuO₂ at 1.8 V (volts) is highly surprising, unless it is covered by SiO₂. This result should be supported by additional

evidence. Using double layer charge for estimating the oxide surfaces area is not a reliable method. Surface charge transfer processes often occur that cannot be separated.

- The project is only a few months old. The preliminary conductivity and durability data on individual materials shows promise. However, some requirements have not yet been met (e.g., stand-alone proton conductivity and Brunauer-Emmett-Teller [BET] surface area).
- The initial progress seems good, but the work seems to be in the early stages and testing in real fuel cell systems is important.
- The project started in the middle of 2010, so one cannot expect significant progress to be made in such a short period of time. The initial results suggested that most of the proposed milestone were met for oxide synthesis and testing. In the second part, the principal investigators have focused on benchmarking and testing Pt catalysts supported on carbon. This “benchmarking,” however, might not be relevant for catalysts that are synthesized with other chemical methods, and this should be clearly stated in the future presentations.
- BET surface area has reached its target and, the project has achieved conductivity of 24 S/cm. Progress is being made on improving proton conductivity. The stability of the RuO₂ has not been properly assessed, and the researcher does not appear to have an approach to successfully stabilize RuO₂.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.0** for its collaboration and coordination.

- It appears that the collaborators were well selected, and have the right possibilities and expertise.
- Nissan North America brings substantial knowledge and expertise in support requirements, testing, and data interpretation.
- Incorporating an automotive partner should ensure that the support and the MEAs made with the support are tested under system applicable conditions.
- This proposal is nicely coordinated, and all partners are involved in the realization of the project.
- It is difficult to judge the efficacy of the collaboration with Nissan, as the Nissan subcontract has just been established.
- Every catalyst project usually needs at least two forms of collaboration—a stack original equipment manufacturer or integrator and a place for materials characterization. This project has done an excellent job of the former, but the latter is still missing. Nissan appears well prepared to perform both rotating disc electrode (RDE) and fuel cell testing to determine catalyst stability. When failure modes are detected in either RDE or fuel cell testing, there will be a desire to see what happened with the catalyst and the support. Measurements using X-ray photoelectron spectroscopy or x-ray absorption spectroscopy would help to uncover oxidation states or adsorbates, and microscopy would help identify composition and particle sizes. Collaboration on materials characterization will be necessary.
- This project represents a disappointing program from Nissan. There was no detailed discussion. It is unclear what industry perspective means. There was no definition of benchmarking. The project defines durability as a test for oxidation of carbon, and does not define a durability test for an oxide.

Question 5: Proposed future work

This project was rated **2.9** for its proposed future work.

- The proposed future work is appropriate. The task of catalyzing the supports should be moved up in priority and supersede the support-only durability measurements of weight loss after cycling in the aqueous environment.
- The major problem with oxide supports is the deposition of Pt to obtain an active and stable catalyst. This question has not been addressed. Researchers also failed to discuss how and where Pt nanoparticles will be formed—at one oxide or both.
- IIT will address the targets not yet met through synthesis and precursor modifications. Both uncatalyzed and catalyzed testing of the mixed system will be initiated.
- At this point, the fuel cell community should be much more aggressive in resolving durability issues. This is certainly true for the proposed future work of this project. The principal investigators must focus more on understanding, not only testing.

- Greater priority should be given to catalyzing existing materials as opposed to increasing proton conductivity. The proton conductivity targets are more subject to debate than some of the other targets, so it would be interesting to see what the existing material conductivity would be capable of facilitating. However, before that can be done, the material must be catalyzed and deemed worthy of MEA testing through RDE screening. Future work pertaining to continued adjustments in $\text{SiO}_2/\text{RuO}_2$ is worthwhile. Investigators may wish to consider whether they have the resources to contain a non-platinum-group-metal materials development task.
- This project does not address finding a replacement for the RuO_2 , which will have some dissolution. The researcher has or will reach a stumbling block.

Project strengths:

- This project is a good idea to address oxide functionalizations.
- This project features a good team, as well as a sound approach and plan to move to practical MEA testing and operating conditions.
- The durability of cathode materials is very important in the quest to fully implement PEM fuel cell technology. Obviously, the principal investigators are highly experienced in synthesizing and testing conductive oxide cathode supports.
- Strengths of this project include its novel ideas and collaboration with Nissan.
- The original equipment manufacturer collaboration is a strength of this project. Working with Nissan will help to ensure that relevant RDE and fuel cell testing will be incorporated into the project. The project team established high surface area and electrical conductivity. Many novel catalyst support efforts struggle with a tradeoff between surface area and electrical conductivity, but this project does not appear to have this problem.
- Increasing the proton conductivity appears reasonable.

Project weaknesses:

- Having electrocatalysis as part of the project is an area of weakness.
- The principal investigators must use more sophisticated methods for surface characterization of both conductive oxide supports and Pt catalysts. They must also use different analytical methods to monitor Pt dissolution and TEM technique, and to follow surface agglomeration. Discussion is needed about the effect of support on water managements.
- Two areas of weakness for this project include the choice of materials that have questionable stability at high potentials and the composite approach toward achieving the desired functions of a catalyst support.
- Regarding materials characterization collaboration, failure modes will arise in the course of running RDE and fuel cell voltage cycling. It would be good to have someone like Oak Ridge National Laboratory or a university on the project to do TEM or other characterization. This project also has a weakness in terms of platinum group metals (PGMs) in the proposed new materials. Ru is a PGM and could possibly increase in cost with commercialization. Ideally, there would not be a PGM in the material. Regarding the emphasis on proton conduction over advancing with other project workstreams—while ionomer reduction may have cost benefits, it still remains to be seen whether this is true. In the meantime, catalyzation of the materials to see RDE results and possibly fuel cell results might be worthwhile.
- The choice of RuO_2 is very poor. It is unclear why this was done.

Recommendations for additions/deletions to project scope:

- The project team should minimize activities on model systems and address the electrocatalysis part of the project. An expert on oxide electrochemistry would be a good addition to the team.
- The principal investigator should justify why the project will use the $\text{SiO}_2/\text{RuO}_2$ type of catalysts. The principal investigator must also consider some other conductive oxide supports.
- One recommendation would be to judge the stability of the supports by the stability of proton and electron conductivity as a function of cycling, not just by surface area measurements. The project team should also make catalyzing the supports a higher priority task.
- The project team should add materials characterization collaborators who would help to understand the failure modes that will eventually be discovered by the project. As it is, it would be interesting to know what the particle size of RuO_2 is on SiO_2 , and whether the particle size of RuO_2 matters toward other properties. A cost

analysis may sound premature, but it would be interesting to identify the tradeoff between removing the ionomer from the catalyst layer and sulfonating the support materials. Even a very cursory analysis might be useful in this regard.

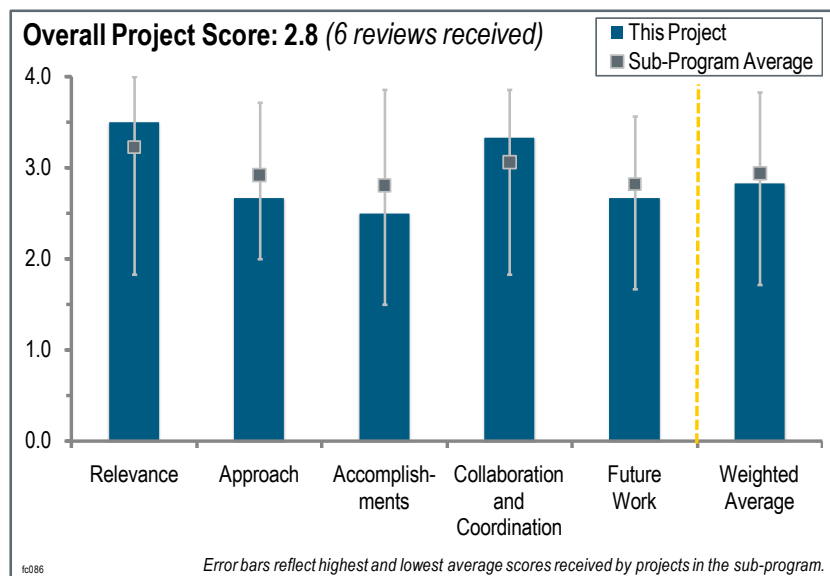
- The project team should identify a replacement for RuO₂.

Project # FC-086: Development of Novel Non-Pt Group Metal Electrocatalysts for Proton Exchange Membrane Fuel Cell Applications

Sanjeev Mukerjee; Northeastern University

Brief Summary of Project:

This project will develop new classes of non-platinum-group metal (non-PGM) electrocatalysts that will meet or exceed U.S. Department of Energy (DOE) 2015 targets for activity and durability. The DOE activity targets are 130 A/cm² (amps/centimeter squared) in 2010 and 300 A/cm² in 2015. This will enable decoupling polymer electrolyte membrane (PEM) technology from Pt resource availability and lowering membrane electrode assembly costs to less than or equal to \$3/kW. The science of electrocatalysis will be extended from current state-of-the-art supported noble metal catalysts to a wide array of reaction centers.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.5** for its relevance to DOE objectives.

- The development of non-PGM electrocatalysts is a goal of major importance for sustainable use of fuel cells in energy conversion and generation.
- The project addresses the DOE Hydrogen and Fuel Cells Program goal of lowering the cost of PEM fuel cells and increasing durability by replacing Pt with non-precious metal catalysts.
- The development of non-PGM catalysts is of paramount importance for the realization of PEM fuel cell technology.
- The development of active, durable, and inexpensive electrocatalysts is critical to the Program, and is one of its highest priorities.
- This project seeks to enable non-precious catalysts in PEM systems, thereby potentially addressing the largest cost limitation of fuel cell systems.
- On the relevance slide (slide three), the investigators claim “greater independence to poisons which typically effect of [sic] Pt and Pt alloys (i.e., S, CO, etc.)” However, they postulate that the active sites in their catalysts are the metals, and that S and CO will coordinate to Fe, Co, or Cu in these transition metal complexes and still poison these catalysts.

Question 2: Approach to performing the work

This project was rated **2.7** for its approach.

- Using triazol molecules, attaching various ligands, and using metal-organic frameworks (MOFs) and open framework templated structures are all promising approaches for developing non-platinum group electrocatalysts.
- The project seems feasible. The approach is focused on overcoming barriers.
- The novelty of the materials and how the synthetic approach differs from what has been tried previously by other organizations are unclear. The approach on slide nine is very similar to work published by the University

of South Carolina, in which investigators looked at the addition of melamine and urea to carbon. Triazoles have been studied by Gewirth and others. Argonne National Laboratory has looked at several MOFs previously. The approach to prepare an open framework templated structure through preparation with silica nanoparticles, pyrolyzing with transition metal salt, then etching the silica away with hydrofluoric acid is likely to provide an open framework, but it is not clear if it would be cost effective. Characterization by extended x-ray absorption fine structure analysis and infrared (IR) spectroscopy and modeling efforts are good. Efforts that are more directed along these lines and at characterizing active species and determining the active sites rather than making “new” or more active catalysts of a similar nature to those already prepared would be more beneficial to the community.

- The proposed approach is truly a surface science approach that utilizes various methods to design the active centers for breaking oxygen molecules without peroxide formation. In combination with powerful x-ray spectroscopy and computation transport and reaction dynamics, this project offers catalyst designs based on understanding and implementing fundamental knowledge in catalytic activity on non-PGM catalysts.
- The approach is good, but it is not novel and has been or is being pursued by many other groups. It is unclear what is unique about this project or why the materials developed in this project would be an improvement on what has already been developed in this class of materials.
- The approach to improving the performance and durability of non-PGM catalysts is based on exploring very broad classes of organic frameworks, polymer composites, and controlled ligand environments. While much background is given and the focus on trying to increase fundamental understanding is sound, details were not presented regarding specific approaches and why they would be likely to result in increased performance and durability to the level of system viability.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.5** for its accomplishments and progress.

- Good progress has been made over the short time period. The activity of some electrocatalysts for the oxygen reduction reaction (ORR) is quite promising, although it is much lower with air. There are many open questions, in particular regarding the mechanism of the reaction, the nature of the oxygen interaction with the active sites, and the nature of the site itself. Investigators should pay attention to the large IR drop with these catalysts. Identifying oxygen adsorption using IR spectroscopy is a difficult task.
- This is a new project. Some progress was demonstrated. The oxygen reduction currents and onset potentials for oxygen reduction for the triazoles are so low that it is not even valid to say that one metal center has higher “electrocatalytic” activity than another. A kinetic current cannot be extracted from the Michigan State University (MSU) rotating disc electrode (RDE) data for the metal-N-C catalysts (quoted as 2.11 A/cubic cm) because the requirements for valid extraction of kinetic data from the RDE are not met (i.e., constant and theoretically expected diffusion-limited current reached at low voltages). Given the poor performance of the MSU materials in the RDE tests, the fuel cell performance with cathodes comprised of the MSU catalyst are unexpectedly high—an explanation is needed. The reviewer wants to know if there is any chance that Pt has crossed from the anode to the cathode through the thin membrane.
- As this is a relatively new project, accomplishments and progress are modest, as expected. The results presented from MSU using melamine are interesting; however, they also revealed a potentially critical flaw in non-precious catalyst systems for many applications when examining their durability under start-stop conditions.
- This project represents a fair performance, but it is not as good as others in the field. The volumetric activity is well below that reported by Los Alamos National Laboratory.
- No significant progress has been made toward DOE targets. The volumetric activity measured in fuel cells is a factor of 10 lower than the DOE target. The high activities calculated from RDE curves are not real because the thin-film limit is not fulfilled at loadings that are that high. The Tafel slopes are not meaningful because kinetic currents cannot be extracted from RDE curves if the thin-film limit is not fulfilled.
- Although the principal investigators presented an impressive collection of results, there are many points that are puzzling and must be resolved in order to progress toward objectives and overcome DOE barriers for non-PGM materials. For example, it is unclear why deactivation of non-PGMs is so fast at very low current densities in fuel cell testing. Furthermore, it is difficult to believe that it is possible to see oxygen on both Pt and non-Pt materials. If oxygen can be detected, it is unclear why some of the reaction intermediates cannot be detected.

This proposal is nicely coordinated and all partners are involved in the realization of the project. Other remaining questions include the following:

- The concentration of the three-dimensional elements
- The role of the three-dimensional elements in the oxygen reduction reaction
- What researchers can learn from density functional theory (DFT) calculations if so little is known about the stability of metal centers

Question 4: Collaboration and coordination with other institutions

This project was rated **3.3** for its collaboration and coordination.

- Good collaboration can be expected from this team, as it includes experts in specific areas.
- The collaboration between the partners appears to be good. The collaborators include original equipment manufacturers, catalyst developers, national laboratories, and universities.
- This collaboration is indeed outstanding—there is appropriate collaboration among the eight partners.
- This project features a large number of collaborators and evidence of significant input from about half of them (i.e., Nissan, MSU, and the University of New Mexico).
- There is good collaboration between Nissan and the universities. The roles of the University of Tennessee and BASF are unclear.
- The involvement of BASF helps provide industrial input; however, the role of BASF in the project is unclear from the presented materials.

Question 5: Proposed future work

This project was rated **2.7** for its proposed future work.

- This is a good project regarding learning about the fundamentals of oxygen interaction with these catalysts.
- The proposed future work is well designed and builds on previous experience and knowledge of the principal investigators. The investigators should focus more on collecting reliable experimental results before applying computational methods.
- The proposed future plans are focused on overcoming barriers, and do not specify when go/no-go decisions will be made for different groups of catalysts. The templated, self-supported non-PGM catalysts do not seem very promising.
- The proposed future work appears to be directed at improving transport in the catalyst layer and the gas diffusion media. The work on the gas diffusion media appears to be out of the scope of a non-precious metal catalyst project. There is enough work to do determining and optimizing the active site. Optimizing transport in the catalyst layer could be beneficial, but the work related to optimizing transport in the gas diffusion media does not belong here and detracts from the effort of developing a non-PGM catalyst.
- The proposed future work as described in the presentation was broad and general. There were no specifics regarding how the project intends to increase the ORR activity of the various classes of materials or what criteria would be used for the go/no-go and down-select decisions.
- Two of the five bullets in “Future Work” were focused on mass transport. While mass transport is certainly a potential issue in non-PGM catalysis, the focus on this work is premature until materials that meet performance and durability needs are found.

Project strengths:

- This project’s strengths include its highly qualified team and good resources.
- This project features a well balanced combination of experimental and theoretical components. It uses insights from modeling when designing catalysts and catalyst layers.
- This project features strong characterization techniques.
- The proposed approach is good, and the principal investigators have a great opportunity to resolve many puzzling issues related to the importance of non-PGM catalysts in PEM fuel cell technology.
- This project has a multifaceted, multi-technique approach with a team comprising many experts in the field.
- This project’s strong team is built on the experience of the academic participants in the area.

Project weaknesses:

- The approach may be too broad, although this research is at an early stage.
- The DFT modeling needs to be complemented by validation in experimental systems. It is unclear why the expertise of the team members (i.e., Dodelet and Zelenay) who were successful in the synthesis of non-PGM catalysts for fuel cell application cannot be used to find a fast solution for optimizing the structure of the catalyst layer and making a proper choice of materials.
- The principal investigators should be more critical in discussing and designing the experiments. In particular, the principal investigators must be more careful in assigning the active centers for cleaving the oxygen molecules and focus more on stability of these sites.
- Down-selecting to a particular subset of the materials proposed should occur early on in the project. The number of materials classes proposed for investigation is too extensive for the short duration of the project.
- The approach is broad, and there is a lack of clarity regarding the studies to be performed and how they might lead to improvement. There is also a lack of clear metrics to evaluate materials and manage the direction of the project going forward.

Recommendations for additions/deletions to project scope:

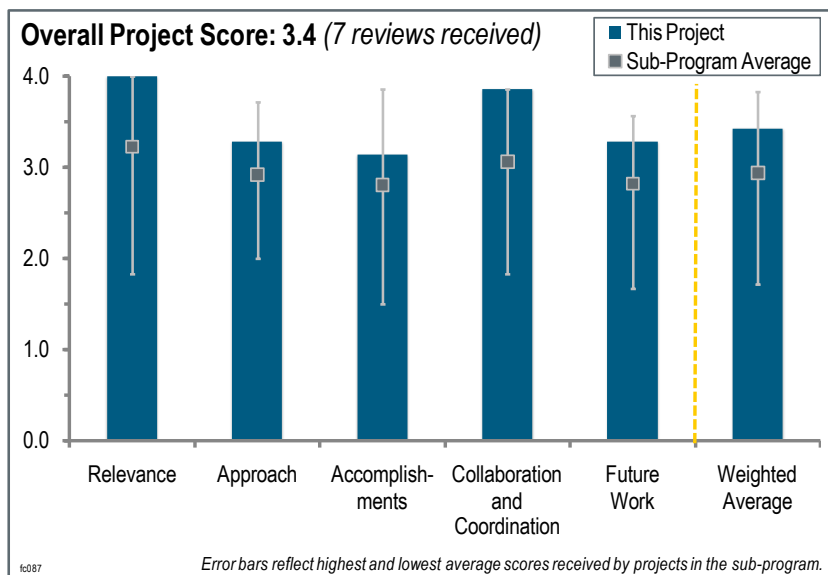
- It would be interesting to synthesize graphene with incorporated non-PGM catalytic centers to validate DFT modeling.
- Investigators should delete the proposed future work on improving mass transport in the gas diffusion media, and increase efforts in characterization and attempts to determine the real nature of the catalytically active site.
- This project is in an early stage, so reviewers should wait until the next review before recommending any changes in the direction of such complicated systems.
- Investigators should remove the emphasis on mass transport issues until adequate durability is demonstrated, and provide downscoping criteria for different catalysis approaches.

Project # FC-087: High-Activity Dealloyed Catalysts

Fred Wagner; General Motors

Brief Summary of Project:

The overall objective of this project is to reduce catalyst cost while achieving the required durable performance, allowing fuel cells to become economically competitive with other power sources. Specific project objectives are to: (1) demonstrate reliable oxygen reduction reaction kinetic mass activities greater than the U.S. Department of Energy (DOE) target of 0.44 A/mg (amps per milligram) of platinum group metal (PGM) in hydrogen/oxygen fuel cells; (2) demonstrate durability of the kinetic mass activity against DOE-specified voltage cycling tests in fuel cells; (3) achieve high current density performance in H₂/air fuel cells that meets DOE heat rejection targets and PGM-loading goals of less than 0.125 g PGM/kW (grams PGM/kilowatt) and less than 0.125 mg PGM/cm (milligrams PGM/centimeter); (4) scale up to full-active-area fuel cells, to be made available for DOE testing; (5) demonstrate durability of high current density performance; and (6) determine where alloying-element atoms should reside with respect to the catalyst-particle surface for best durable activity.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **4.0** for its relevance to DOE objectives.

- The evaluation of Pt alloy catalysts in fuel cell tests is critical for the DOE Hydrogen and Fuel Cells Program.
- The focus on catalyst performance and durability is critical.
- This is a very relevant project that focuses on ways of reducing catalyst cost and tries to understand how activity is increased to compensate for the decrease in Pt. The second and equally important goal of durability is also relevant.
- The project seeks to explore the effect of de-alloying in bimetallic catalysts. Given that this process might take place in any electrocatalyst that contains a combination of noble and non-PGM elements, a detailed understanding of de-alloying-induced catalytic enhancement is of critical importance. Importantly, the project aims to scale-up the most promising systems in order to demonstrate a novel platform for utilization of Pt alloys in polymer exchange membrane fuel cells.
- This project addresses the most critical fuel cell research and development material issues—those of catalyst performance, cost, and durability.
- Low-loaded, high-performance, durable cathode catalysts constitute the primary topic of concern for the commercialization of fuel cell vehicles. This project is directly focused on all of the targets associated with fuel cell electrocatalysts (i.e., loading, mass activity, and durability). Furthermore, this project is also focused on the manufacturability of the catalysts, as well as performance stability at high current densities.
- This project represents an important bridge between fundamental science and applied realization.

Question 2: Approach to performing the work

This project was rated **3.3** for its approach.

- The approach is good. Elucidating the behavior of de-alloyed Pt electrocatalysts in fuel cell tests provided necessary information for further research planning. Comparing rotating disk electrode (RDE) and membrane electrode assembly (MEA) results is helpful to understand the difference between these sets of data, which is often observed. Several powerful characterization methods have been employed.
- The approach is well planned, and includes all of the critical scientific elements as well as evaluation in fuel cells.
- The approach for the most part is very good. The project's approach may elucidate some of the questions regarding the concept of strain-induced reactivity, which is potentially very useful. The approach includes modeling activities and surface characterization measurements, in addition to straightforward MEA evaluations, promising to provide a better understanding of the concept.
- The approach is based on previously established electrochemical methods of de-alloying Pt trimetal (PtM_3) (where M is another metal such as Cu or Co) bimetallic catalysts. By focusing on scaling-up catalyst production, the project considers balanced electrochemical studies between RDE and MEA, as well as ex-situ and in-situ characterization of de-alloyed catalysts. However, selection of catalysts is rather narrow—a new generation of stable catalysts is needed.
- The approach looks to further investigate PtM_3 alloys and the durability and activity enhancements possible from their use. To date, Cu and Co have been investigated and the presentation suggests that Ni will perhaps be next. These alloys have all shown promise and have remaining unanswered questions, and therefore merit further investigation.
- Extending the approach beyond the in-situ electrochemical de-alloying process, developed by Peter Strasser of the University at Houston, is a major step in the right direction. The essential aspect of the approach is that it could be possible to locate base metal atoms in a nanoparticle so that the Pt compression is preserved, while base metal atoms are not allowed to migrate to the surface of the particle and dissolve. There is no data that says it cannot be done, but given the impracticality of a reproducible, atomic-scale design of nanoparticles from a high-volume fabrication process, one has to concede that there is considerable risk inherent in the approach.
- It is good to see the emphasis on scale-up, which will be necessary. The activity/durability trade-offs are often underrated in importance, but are treated properly in this project. The project features a nice combination of applied and fundamental work.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.1** for its accomplishments and progress.

- An outstanding amount of work has been accomplished over a short period. Identifying serious problems associated with the use of Pt-transition metal alloy electrocatalysts was very useful.
- Considering the recent start of the project, the accomplished set of results is very promising. The DOE mass activity target is being achieved, while durability studies confirmed that additional improvements are necessary. The de-alloying process was probed and confirmed under manufacturable conditions.
- While this is a relatively new project, it has made significant progress, suggesting that previous work was conducted. The move toward PtCo_3 from PtCu_3 as the precursor is important.
- Chemical de-alloying of PtCu_3 to generate higher mass activity has already been achieved. The project team found that excessive remaining Cu compromised high current density performance, and that the Pt-Cu material showed lower mass activity after cycling versus the Pt-Co material. The project team also discovered “Swiss Pt Particles” from observing the microscopy of de-alloyed Pt-Cu and Pt-Co. Multiple cores in a particle is suggested as a concept that might promote durability. Generally speaking, the project has uncovered some trends that were expected (e.g., the superiority of Pt-Co to Pt-Cu), while uncovering some unexpected atomic segregation within alloy nanoparticles. It remains to be seen whether the multiple cores concept improves durability.
- The accomplishments and progress appear good, but with pre-existing data available. With a delayed start, the project should be very productive when everyone on the team is fully up to speed.

- This is a new project, so progress to date has been modest, as expected. The activities of the catalysts presented are appropriate for further study. The findings of multiple cores versus single cores and the impact on durability yield some insight into the importance of catalyst particle morphology on durability. Johnson Matthey (JM) scaling-up the catalyst synthesis of select catalysts to 100 g batches is a valuable addition to this project, as that allows for significant testing and validation.
- Investigators were very honest about early progress and presented a realistic view. The investigators were not hesitant about admitting their failures.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.9** for its collaboration and coordination.

- A very strong team is gathered on this project and collaboration appears to be excellent.
- All of the key elements are here and functioning well together.
- This project features an excellent team. Everyone is appropriate for the tasks.
- The team comprises well established experts who offer a wide range of expertise that is critical for the success of this project. Task coordination between the participants is efficiently determined, and the role of each subcontractor is clearly defined.
- The team is excellent and—although it is early in the project—seems very well coordinated on the tasks, almost certainly due to pre-existing collaboration.
- In other catalyst projects, a materials characterization partner has been identified; this project has three—the Massachusetts Institute of Technology, Northeastern University, and George Washington University. At present, the work of these three institutions has not been heavily reported, but they do factor into the future work. The plans to use JM as a scale-up partner are fairly self-evident. The contributions of the Technical University of Berlin (TUB) and the University of Houston were widely attributed throughout the presentation.
- This project features good collaboration, drawing all needed expertise and funneling up fundamental science toward applied realization.

Question 5: Proposed future work

This project was rated **3.3** for its proposed future work.

- This proposed future work features an excellent plan with a high probability of success.
- The future work is well planned and the roles and tasks for each participant are well laid out. The future work is very well conceived, as nearly all possible directions have been considered. The goal is to achieve reliable mass activities, as opposed to one measurement, which should serve as paradigm to catalyst projects. Both small-scale (TUB) and large-scale (JM) volume fabrication of catalysts are accounted for in the future work. Synthesis parameters related to annealing, depositing, and de-alloying are being thoroughly investigated. Material characteristics such as particle size, morphology, compositional maps, adsorbates, and Pt coordination are all covered in the work plan. RDE and fuel cell testing are well covered by General Motors (GM).
- The project team has planned too much work related to solving serious drawbacks of some systems that were identified in the current work. For instance, the dissolution of Cu and Co affect fuel cell membrane and anode performance. Preventing double-positive Cu ions from reaching the anode does not seem to have a simple solution, and growing “multiple cores” to minimize the quantity of dissolved transition metal provides no guarantee of stopping dissolution.
- The plans for the rest of 2011 are good and detailed, with the plans for 2012 ambitious in their detail.
- The future work is targeting mass activities above 0.44 A/mg of Pt by employing several catalytically active systems. Particular attention is placed on durability and proper characterization, including local atomic coordination.
- The future plans are not very clear. For example the criteria and approach for the selection of the two materials for the next step and for which TUB will screen potential choices are unclear.

Project strengths:

- The research team consists of experts in the number of relevant areas. The project’s resources are good.

- The project has a relatively specific focus, a good balance between fundamental scientific investigation and technology validation, and a good blend of partnership skills.
- The project is a continuation of strong earlier work. The project team is strong and appropriate.
- The project features a rational, well-balanced approach aimed at tackling the most challenging issues in fuel cells including decreasing the total Pt loading while improving performance; utilizing a wide range of characterization tools that are capable of providing atomic level insight into catalyst properties; and scaling-up the most promising catalysts. This project features a strong team (and principal investigator) that is investigating demonstrated high activity materials.
- One area of strength is the experience of this project's team members. The team involved constitutes some of the most experienced in the fuel cell electrocatalyst research community including GM, Northeastern, TUB, and JM. The project also features an appropriately diverse list of expertise including a scale-up partner, an original equipment manufacturer (OEM), a materials characterization partner (or three), and a materials development partner. The project also features a strong OEM perspective, which implies the perspective on automotive customer requirements is built into the project.
- This project's strengths include the integration of applied and fundamental research. The project has a goal-driven focus with the right fundamental work leading to practical realization, ensured by the project's organization.

Project weaknesses:

- The team has selected too many systems with major problems for future studies in addition to selecting new alloying transition metals.
- There is only one go/no-go decision point after the second year, which is concerning. The team should make a decision on whether to pursue a Cu- or Co-based system at the end of 2011.
- The concept by strain-induced reactivity enhancement is unclear, and possible corrosion issues are of additional concern. However, this is what research is about.
- The project offers optimization of catalysts that have already been examined in the past. The main concern is the lack of control of critical parameters such as particle size and the composition of de-alloyed particles. Based on the presented results (slide nine of the presentation), the principal investigator should consider not pursuing further study of the Pt-Cu system. Planned work does not seem to properly address Cu dissolution and contamination of the anode as well as the cathode electrodes. Developed models with a Pt-shell structure over Cu (III) do not reflect the case depicted on slide 14, and will not solve the problems reported on slide 9.
- The project focuses on a few promising catalyst compositions that have seen a fair amount of investigation. While the approach discussed should lead to an increased understanding of the system and influencing factors, it is far from certain that this will translate into improved performance and durability. As alloys have been studied for some time, it seems that the project's effort is more of an incremental advance as opposed to a breakthrough. Another weakness is the high risk of activity and durability not being both met. Although the proposed research contains some novel concepts on how to prevent the leaching of base metals from fairly unstable Pt alloy nanoparticles, the fact remains that base metals are being expected to stay stable at potentials of known instability, which introduces risk as to whether durability could be achieved under drive cycle conditions.
- This reviewer found no weaknesses.

Recommendations for additions/deletions to project scope:

- No recommended additions or deletions are being made, consistent with the praise given in this review for the future work. There is some temptation to suggest eliminating the Pt-Cu work. However, a revisit of Pt-Cu by TUB is likely containable within the resources and worthwhile.
- Focusing on a smaller number of systems is recommended.
- Investigators should insert a decision point on whether a Cu- or Co-Pt system should be pursued beyond 2011. The principal investigator should go beyond Pt-Ni and Pt-Co systems.
- In general, the team should provide clarification on the selection of materials to be investigated.

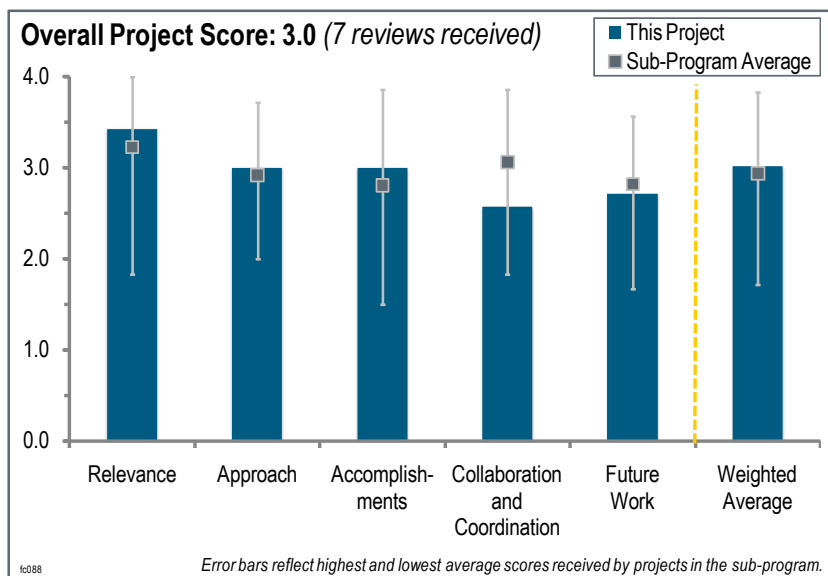
Project # FC-088: Development of Ultra-Low Platinum Alloy Cathode Catalyst for PEM Fuel Cells

Branko Popov; University of South Carolina

Brief Summary of Project:

The overall objectives of this project are to develop: (1) low-cost and durable hybrid cathode catalysts (HCCs) consisting of non-Pt/C composite catalysts (CCCs) and low platinum group metals (PGMs) for oxidation-reduction reactions; and (2) triplatinum-metal (Pt₃M) on activated graphitic carbon (AGC) catalysts. Specific objectives are to: (1) achieve kinetic mass activity in hydrogen/oxygen fuel cells that is higher than the U.S. Department of Energy (DOE) target of 0.44 A/mg (amps/milligram) of platinum group metal (PGM) and demonstrate durability of the

kinetic activity (per the DOE cycling protocol) between 0.6 and 1.0 V (volts), (2) demonstrate high current density performance and durability in hydrogen/air fuel cells to meet 2015 DOE targets, (3) define the parameters that control the number of non-metallic catalytic sites on CCCs and optimize the procedure for the formation of more active leached Pt-alloy HCCs, (4) define the parameters that control the activity of leached Pt-alloy catalysts deposited on AGC support, (5) develop corrosion resistant hybrid supports such as Ti dioxide (TiO₂)-CCC and AGC, (6) develop a facile scale-up synthesis procedure for the developed catalysts (at least 100 g [grams]), and (7) construct a short-stack (50 cm², up to 10 cells) and evaluate the performance under simulated automotive conditions.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.4** for its relevance to DOE objectives.

- This project is focused on the generation of a low-cost, durable HCC consisting of non-Pt CCC and Pt₃M/ AGC catalysts. Both activities support the goal of lower cost, higher durability, and higher performance polymer electrolyte membrane fuel cells.
- Improved low-cost catalysts are an important part of increasing membrane electrode assembly (MEA) performance and durability. This fits well with the overall DOE Hydrogen and Fuel Cells Program objectives.
- This project addresses the key barriers of fuel cell cost and durability.
- The project is focused on reducing the Pt content in fuel cell catalysts by introducing Pt alloy on AGC support and developing highly active corrosion resistant systems. The concept of the project is well placed and addresses the most critical needs in future development of fuel cell catalysts.
- Highly active, robust catalysts are critical for enabling fuel cell system commercialization.
- The stated objectives align fairly well with DOE objectives.
- This project proposes to address the three main catalyst related barriers. In reality, it probably focuses on meeting cost objectives through reduced Pt use more than it is able to address performance or durability.

Question 2: Approach to performing the work

This project was rated **3.0** for its approach.

- The project has a sound approach to materials development and evaluation.

- The main strategy of this proposal is novel and interesting as it attempts to combine aspects of dispersed Pt and non-PGM catalyst concepts.
- The approach combines advances made in non-PGM catalyst work with advances made in Pt-alloy catalysts to make a hybrid catalyst with higher activity and durability. The investigators are performing appropriate testing.
- The project is based on a novel approach regarding HCC development that employs a synergistic effect between Pt-alloys and non-Pt CCCs. In addition, other corrosion resistant hybrid supports are considered, such as TiO_2 on carbon composite material. These novel systems are supposed to enhance catalytic performance and durability during fuel cell operation. In addition, the project team will develop scale-up synthesis protocols for the most promising catalysts.
- The approach is novel, but it may be too focused on improvements at low current/power density, as shown on the polarization curve of slide 15. Cost is most effectively addressed by operating fuel cells at higher power density. Reducing Pt loading and improving performance and durability at high power are most important.
- The approach follows three separate paths. The first is to develop a hybrid catalyst—a non-Pt, carbon-based composite catalyst with active catalytic sites. The second is to develop an AGC supported Pt-alloy catalyst. The third is to develop a corrosion resistant catalyst support based on TiO_2 .
- The benefits of including an ultra-low Pt catalyst will likely be reduced by high mass transport losses due to the thick electrode layers made necessary by the lower activity CCC catalyst. Fe in the catalysts is a concern as a likely source of initiating membrane degradation. Corrosion resistant supports may be required for CCCs, but investigators did not present any evidence showing that oxygen reduction reaction (ORR) activity can be met with TiO_2 -CCC.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- The project is making good progress against the materials development goals.
- The data shows significant improvement and progress toward targets. The project team has achieved an initial mass activity that is $> 90\%$ of the target value with three types of catalysts, one of which showed less than 40% loss in mass activity after cycling per DOE protocol.
- Considering the early stage of the project, the reported accomplishments are very encouraging. The project team has almost achieved the DOE mass activity target, and durability studies confirmed that additional improvements are necessary. HCCs showed slight improvement in catalytic activity over conventional Pt/C catalysts, and durability studies also demonstrated some improvement in the electrochemical surface area losses.
- Investigators presented a good deal of information supporting this project's progress toward meeting DOE catalyst targets, but when the material is employed in an actual cell (slide 15) the internal resistance corrected performance is very poor. This gap was questioned by one reviewer during the session and the principal investigator's response was less than adequate.
- H_2/O_2 fuel cell data is encouraging, and activity losses after 30,000 cycles are reasonable. The specific activity results are questionable because the reference Pt/C values are high. The H_2/air data was initially encouraging, but significant improvements are still needed. No progress has been shown on corrosion resistant supports.
- The accomplishments to date appear very promising for the first and second approach. Nothing was mentioned of the third approach, and it is assumed to not have been as successful. The first two approaches have resulted in higher performance. Durability and cost have not been discussed.
- The HCC approach is showing very good values for mass activities in MEA measurements. The durability targets appear to be met or nearly so for all three approaches. The project should be commended for working at 0.1 mg/cm^2 levels regarding Pt on the cathode, which is near the ultimate target. A more serious concern is that the high current density performance under hydrogen/air is not being met, perhaps due to the excessive thickness of the HCC electrodes for the loadings used. The data on slide 15 would suggest that at 1.5 A/cm^2 , the resistance corrected potential is about 200 mV (millivolts) lower than is required to meet the DOE MEA power density requirements.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.6** for its collaboration and coordination.

- The collaboration is with an original equipment manufacturer (OEM) and a second university. Testing at the OEM should ensure that relevant operating conditions are examined and sufficient durability testing under relevant conditions is performed.
- The project team has participants with academic and industrial backgrounds, some of whom are well established experts in fuel cell catalysis. The coordination between the lead institution and the subcontractors is very well placed, and the role of each subcontractor is obvious.
- Seemingly all of the information presented was generated and coordinated by the University of South Carolina (USC). The presentation should include better representation of Hyundai Motor Company and Yonsei University contributions.
- The USC project is very strong, but the university is essentially working independently from a materials characterization and development standpoint. This may be partially addressed in the future by more involvement by Yonsei University.
- The collaboration appears to be limited to the project partners. This should not be surprising. Development of a novel catalyst could be lucrative.
- The project has not presented any evidence of collaboration to date, although the future plans include collaborative work.

Question 5: Proposed future work

This project was rated **2.7** for its proposed future work.

- The proposed work appears to be a rational extension of the project.
- The proposed work fits well with moving toward the proposed objectives.
- The plans for future work are a logical extension of the current work that is designed to meet the DOE targets.
- The future work aims to further improve mass activities and durability. Particular attention is placed on the scale-up synthesis of the most promising systems. The proposed future activities represent a rational approach in the synthesis and characterization of hybrid catalysts.
- The project is focused on achieving 0.44 A/mg at 0.9 V—which is the DOE target, but is not the way to reduce cost. The project must demonstrate applicability and reasonable performance at higher current density to bring any value to the community.
- The future work is consistent with advancing and firming up catalyst materials that have good ORR activity and durability. The OEM will require at least 625 mV at 1.5 A/cm², so addressing the mass transport properties of the electrodes may need to be an added focus. Not addressing this until the final year may not be sufficient—resolving this issue should be pursued earlier. As activity or durability catalyst optimization will not address this issue, achieving the first three milestones of the project will not address the fourth one.
- The project team did not describe a clear plan to improve H₂/air performance in order to meet DOE targets. Also, the principal investigator should include potential cycling tests of HCC and Pt-alloy/AGC catalysts. The planned work on corrosion resistant supports is unclear.

Project strengths:

- This project features a novel approach.
- The strengths of this project appear to be the research and analytical skills of the researchers.
- This project has a strong team with a good development plan.
- This project features a novel approach that synergistically combines useful properties of multiple approaches.
- The principal investigator offered samples for others to evaluate for themselves. Base alloy catalysts show reasonable activity and stability. Fuel cell testing is done in a H₂/air cell.
- Investigators have developed catalysts with activity that is > 90% of the DOE target value. The project has a strong team, including an OEM.

- The project features a novel approach in catalyst synthesis aimed to address high Pt-loading in fuel cell stacks while improving durability. The multidisciplinary team is capable of performing the proposed activities. The reported mass activities are in line with DOE targets.

Project weaknesses:

- The project lacks a strong theoretical approach. The project should address the practical concern of applicability beyond 0.3 A/cm².
- The project report does not discuss the TiO₂ work or the costs of the new catalyst. A near-term cost estimate of the newer catalyst as a percentage of the existing catalysts would be useful.
- It is unclear how the high current density air performance will be met, and it is likely the CCC may actually hurt that effort. The dependence on Fe in Pt alloys is also an area of weakness.
- The initial testing of catalyst performance is done by rotating ring disc electrode (RRDE) in sulfuric acid, which is known to suppress activity. It is strongly recommended that RRDE should be performed in perchloric acid in order to diminish the anion effect. It is not clear how the total content of Pt can be efficiently controlled, including the other critical parameters such as alloy particle size distribution, composition, structure, and shape. The proposed future work does not seem to address these issues. The observed enhancements are not discussed in terms of the mechanism for improvements in mass activity and durability.

Recommendations for additions/deletions to project scope:

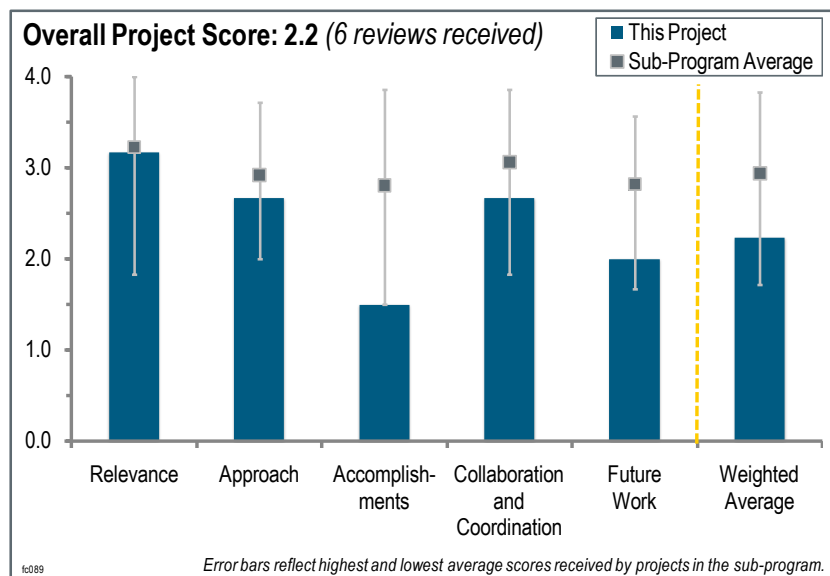
- The scope should include reporting the causes for low, high-current performance, and adjusting the plan to address those issues.
- The project team should further present work on corrosion-resistant catalyst support development, based on TiO₂. Even if the outcome is not successful, a lot can be learned. The investigators should focus on mass transport loss reduction in H₂/air systems and stability under potential cycling, which will be the two biggest challenges with this concept. Also, they should focus on alloy catalysts without Fe.
- The principal investigator must further explain why the systems they are studying are more promising than conventional bimetallic systems studied so far.

Project # FC-089: Analysis of Durability of MEAs in Automotive PEMFC Applications

Randy Perry; DuPont

Brief Summary of Project:

This project addresses several areas that intend to fill gaps in the understanding of cell performance degradation. The objectives of this project are to: (1) develop or confirm accelerated tests designed to separate individual degradation mechanisms; (2) develop an overall degradation model that correlates the stack operating conditions to degradation of the membrane electrode assembly (MEA); and (3) develop MEAs with a design lifetime target of 5,000 hours with less than or equal to 7% degradation and that show a clear path toward meeting 2015 U.S. Department of Energy (DOE) technical targets.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.2** for its relevance to DOE objectives.

- The key topics focus on durability as one major issue/barrier for fuel cells in automotive applications. They address establishing DOE's 2015 technical targets and the need to understand the correlation between degradation in accelerated stress tests (ASTs) and actual automotive operation.
- Understanding cell degradation mechanisms, especially in automotive applications, is important. The delivery of an improved durability membrane is critical.
- The objectives are clearly aligned with the need for high durability in automotive fuel cells. The collaboration with Nissan ensures industry relevance. The work at both the single cell and stack levels significantly increases the project's relevance. The interaction of degradation mechanisms is critical and not often studied.
- Analysis of the degradation mechanisms of polymer electrolyte membrane fuel cell components and the extent of their contribution to the overall cell performance degradation is an important research area for the DOE Hydrogen and Fuel Cells Program.
- This project supports fuel cell durability—one of the critical Program objectives.
- The project is focused on automotive fuel cell durability, which is perhaps the most demanding application for establishing fuel cell durability. A durability model would be helpful to the community, although it needs to be amenable to various flow field designs, operating modes, ranges of operating conditions, and some changes in material sets. Focusing on new materials is consistent with the mission of the Program. Ideally, those materials should be capable of meeting other targets such as cost and performance. The development of new ASTs is not necessarily consistent with DOE goals because DOE already has recommended ASTs that are designed to address failure modes individually. Hopefully the investigators will provide some justification for the new ASTs.

Question 2: Approach to performing the work

This project was rated **2.7** for its approach.

- The approach includes a combination of experimental data, development and selection of material, and modeling. Identifying and separating the individual effects listed by the presenter by cell and ex-situ tests is very ambitious and very useful for the model development. Integrating the degradation model into an existing model is also very ambitious and very beneficial. Understanding the differences between small-scale, single-cell behavior with serpentine flow fields and full-scale single cells with straight channels is of tremendous interest for correlating the typical academic research to automotive applications. Spatial measurements would help researchers understand these differences. The project approach appears straightforward and well planned.
- Comparing Nissan's stack degradation behavior to DuPont's ionomer, electrode, and MEA degradation experience will help establish the validity of accelerated testing and elucidate interactions between individual degradation mechanisms. This information will guide the formulation of more durable materials for cell and stack testing under automotive conditions.
- The technical approach used in this project is adequate and well defined to support the project objectives.
- The approach has some significant risks that may impact the ultimate success of the project and the resulting value. The duty cycle for the historical data must be analyzed and representative stressors must be determined to compare against the AST stressors. The same or similar MEAs run under the load cycle must also be run under ASTs to compare failure fingerprints and determine the acceleration factor. Both of those aspects are critical for establishing correlations and relationships. As shown on slide three, there are three objectives to the project, and without the above actions, the first two objectives of this project are at risk. The work focused on developing new MEAs to meet targets is valuable, but it may not be realized without realizing the first two objectives of the project. In addition, the project's value to others will be minimized if clear relationships are not established. The modeling approach cannot be properly assessed because very little information was included. The study of interactions, including between different types of degradation for a given component, and between different components, will be an important and valuable aspect once the single individual mechanisms have been studied. The details on the measurement techniques for membrane degradation, including peroxide and reactive O₂ species production, should yield valuable results and will provide important input to the modeling activity. While the effect of having different flow fields and the resulting effect on the degradation is important, the approach will only provide value if the degradation effects can be linked to MEA conditions that result from the flow fields. For example, the water content and temperature in the electrodes and membrane for different flow fields will need to be identified. Investigators will also need to explain how this effect will be fed into the MEA design. It is not clear why performance at one ampere per cm² would affect open-circuit voltage degradation. Rather, a relationship between the voltage stress under operation and degradation could be explored.
- As described in the presentation, the approach is vague. It is not obvious how the degradation mechanisms will be resolved.
- The approach mentions that the test methods used in this project will be compared to the tests that have been done at Nissan. However, the question and answer session revealed that Nissan used different MEAs than those that will be chosen for this project. This will make test validation difficult, if not impossible. The milestones and go/no-go plan makes it very clear that MEA selection will precede stack testing. It does not appear that Nissan's prior materials are being taken into account. In general, there is no plan for how to relate the stresses of an AST to the events that occur during the proprietary load cycling.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **1.5** for its accomplishments and progress.

- Progress has been made in the preparation and selection of catalyst coated membranes. However, accelerated tests and durability modeling are behind schedule.
- The project is only about 5% executed and most of the effort has been spent getting contracts in place. Very little actual work has been performed toward the project goals.
- The project is only six months old. Contractual issues with the partners have delayed the project. DuPont estimates that the project is only 6% complete.

- It is too early in the project to assess progress (6% complete), and there have not been any significant accomplishments to date. The project is behind due to contractual issues. Clear planning was not communicated in the presentation.
- There has been little progress in this project due to difficulty establishing the subcontracts and non-disclosure agreements.
- The project's progress so far is limited to membrane synthesis and an equipment upgrade to the test stands—there is no substantial data to critique. Poor progress has been made on finalizing contracts with subcontractors.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.7** for its collaboration and coordination.

- The collaborators consist of three companies and one academic institution. The required competencies for the project are covered well.
- An automotive original equipment manufacturer (OEM) is playing a major role. 3M is providing nanostructured thin-film (NSTF) materials as well as insight into NSTF behavior and the role processing parameters have on durability and performance.
- This project features a good team. Subcontracts still need to be established, which may have held up progress on communication between partners.
- The project has a good team, though the contribution and coordination of the team members is unclear at this time due to difficulties in establishing the partnership.
- The collaboration is not completely clear because the project recently started. The Nissan-DuPont collaboration seems good, but the involvement of the other team members so far is not clear.
- In principle, the partnerships should be decent because there is an OEM (Nissan), a characterization and modeling partner (the Illinois Institute of Technology), and a source for an alternative catalyst layer (3M). However, not all of the legal arrangements have been completed for all of these partners, which is holding the project back. It is not clear which tasks in the Gantt chart coincide with which partners. For example, it is unclear where 3M fits in. It does not appear that there has been an opportunity to run round-robin testing between the partners responsible for fuel cell testing.

Question 5: Proposed future work

This project was rated **2.0** for its proposed future work.

- The future work proposes essentially to start the project using the suggested approach.
- The future work is the published schedule, with approximately a five-month delay. It is not clear if the project will make up any lost time.
- The future work describes what remains of the project; however, it needs to be more explicit about what the work tasks will be in the next year. For example, it would benefit the project to describe which MEAs will be tested using DOE ASTs. The future work section could include a flow chart diagram to clearly indicate where data inputs and outputs exist and what other parts of the project will benefit. The plan for how the degradation model receives inputs should be clearly identified.
- Details of the plan, model approach, interactions to be studied, and industrial results to be compared against are not clearly outlined.
- The proposed future work as described in the presentation is vague.
- The proposed future work is incomplete and obscure. The project team needs to share more details about performance and durability criteria, potential risks, and risk mitigation strategies.

Project strengths:

- The project features a very sound comprehensive approach and modeling supporting experiments.
- The project has excellent materials and system team members in DuPont and Nissan.
- This project features an excellent team.
- The experience of DuPont, 3M, and Nissan—who have conducted a considerable amount of fuel cell research—is an area of strength for this project. DuPont and 3M have experience leading successful DOE-funded projects,

while Nissan has been a solid subcontractor on numerous projects. The OEM and characterization presence is another area of strength. Most projects require partners that include both a stack OEM/integrator and a characterization house. This project has both, and the OEM is capable of providing stack designs.

- Having Nissan onboard to provide the historical duty cycle and failure data is critical to the project. DuPont has a strong understanding of membrane degradation and performance mechanisms, which will be important to the success of the project.

Project weaknesses:

- The schedule delay has been a weakness for this project.
- A serious weakness of the project is the potential use of proprietary MEA and duty cycle designs for the historical data. This may result in very low value to the resulting models. The approach to modeling has not been well outlined, and the probability of success cannot be predicted.
- The project plan is too scattered and vague at this point. Given that this is only a three-year project, the plan should be fairly clear by now.
- The present state of execution is an area of weakness for this project. The project team needs to get matters settled with the partners and begin generating data. The project team also needs to fully plan out what materials sets will be used when, and take into account the existing baselines from Nissan. No clear link exists between AST stress and durability cycling.

Recommendations for additions/deletions to project scope:

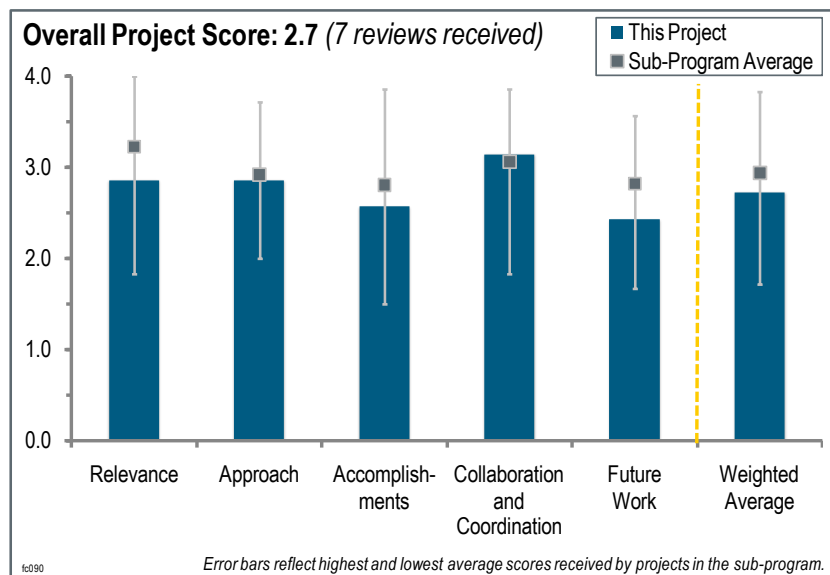
- Spatial effects may have to be considered when comparing the different cell geometries and flow fields.
- The duty cycle for the historical data must be analyzed and representative stressors must be determined to compare against the AST stressors. The same or similar MEAs run under the load cycle must also be run under ASTs to compare failure fingerprints and determine the acceleration factor. Investigators should link degradation effects to MEA conditions that result from the flow fields, such as the water content and temperature in the electrodes and membrane for different flow fields. Researchers could also explore a relationship between the voltage stress under operation and degradation.
- This project should have more efficient collaboration between team members.
- The project team should run ASTs with the same MEAs as those with which Nissan has experience. For modeling purposes, the researchers should begin planning how durability cycle events will relate to the stresses in ASTs.

Project # FC-090: Corrugated Membrane Fuel Cell Structures

Stephen Grot; Ion Power

Brief Summary of Project:

The overall objective of this project is to pack more membrane active area into a given geometric plate area, thereby achieving both power density and Pt utilization targets. The project's objectives are to: (1) demonstrate a single fuel cell (50 cm²) with a two-fold increase in the membrane active area over the geometric area of the cell by corrugating the membrane electrode assembly (MEA) structure and (2) incorporate an ultra-low, Pt-loaded corrugated MEA structure in a 50 cm² single cell that achieves the U.S. Department of Energy's (DOE) 2015 target of 0.2 g Pt/kW (grams platinum/kilowatt), while simultaneously reaching the power density targets of 1 W/cm² (watt/cm²) at full power and 0.25 W/cm² at one-quarter power.



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **2.9** for its relevance to DOE objectives.

- The project is relevant to the objectives of the DOE Office of Energy Efficiency and Renewable Energy Fuel Cell Technologies Program's *Multi-Year Research, Development, and Demonstration Plan*. The activities are aligned to the overall DOE Hydrogen and Fuel Cells Program goals. This project brings in an unconventional approach of using corrugated MEAs to access higher active areas and thereby harvest higher power density from a defined stack volume.
- This project's relevance to DOE objectives consists of its pursuit of increased power density (the demonstration of a two-fold increase in the active area over the geometric area of the cell) and the use of an ultra-low, Pt-loaded corrugated MEA.
- The lower cost plates and gas diffusion layers (GDLs) address the DOE goals. GDL cost, in particular, is a critical item that needs to be addressed.
- This project's corrugated membrane structure fits very well with the DOE objectives to reduce costs.
- This task addresses the cost and durability of proton exchange membrane fuel cells. It also proposes a new architecture for fuel cell stacks. It is clear that many stack designs are moving forward right now. Therefore, it is appropriate to continue the "hunt" for a design of the heterogeneous reactor known as a fuel cell stack, with some novel designs.
- This is a modest project with an interesting possible impact on cost.
- The principal investigator (PI) claims that costs can be lowered by using a corrugated system in which twice the amount of surface area can be used in a cell repeat unit, thereby decreasing the number of repeat units and cost. However, each repeat unit is now much more complicated. Also, the PI seemed unaware that a cooling channel is automatically created by the combination of the anode and cathode plate in current bipolar plates. This system would need a separate cooling channel, adding another cell component. The PI argues that this configuration allows lower noble metal loading, but also assumes the exact same MEA performance. This is not true. With this assumption, a flat plate and a corrugated system will optimize to the exact same place. The only merit of this system is if more surface area can be fit in the same volume and built with lower costs. The first system that the investigators are building has a very large pitch and they seem unaware of current stack buildups, so volume

savings are highly unlikely. Cost savings also seem highly unlikely because the corrugated parts and welds seem harder to make than stamping a sheet.

Question 2: Approach to performing the work

This project was rated **2.9** for its approach.

- Making corrugated three-layered MEAs, and thereby corrugated five-layered MEAs including the GDL, is a good approach to condense larger active surface area within the confined two-dimensional space. Moving from two-dimensional, flat MEA geometry to corrugated MEA geometry provides a four-fold improvement in Pt utilization, and thus a four-fold reduction of Pt loading for achieving the same power density. The approach clearly addresses the high power density barrier and is well integrated with the DOE goal.
- This represents a novel construction. Slide six should clearly show that the loading is 0.1 A/cm^2 (amps/centimeter squared), with respect to cell area. However, if the performance indicated can be achieved with one-fourth (not one-eighth) of the loading of the control ($0.4 \text{ mg [milligrams]/cm}^2$), this is a significant accomplishment.
- This project features a very interesting and uncommon approach.
- The project team is capable of doing the work. Ion Power can certainly make the MEAs, and it already has a first prototype.
- The tasks are related to rather fundamental stack design. The targets may eventually be addressed with this stack, should it prove successful. However, the designs shown are preliminary and many of the important design elements apparently were not considered.
- It is not clear whether this structure will meet many requirements, such as thermal and electrical contact issues, providing sufficient pressure drop, etc. However, it is nice to see DOE take a little risk instead of spending millions on yet another permutation of certain national laboratories doing a bunch of characterization. At least the PI is trying to innovate.
- If the structure shown by Ion Power really provides double the packing density of a conventional stack, the approach is good. However, this claim is questionable. It appears that two conventional cells can fit in the same space as one “double area” corrugated cell. If this is correct, this would mean that the effects of corrugating and doubling the cell are equivalent, and there is no benefit to a “corrugated cell.”

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.6** for its accomplishments and progress.

- Investigators made good progress during the first six or seven months of this project.
- Significant work has been devoted to the development of a corrugated GDL made of an expanded Ti metal screen. However, a pristine Ti screen GDL does not perform well and a Pt-coated Ti screen demonstrated higher power. The Pt loading on the Ti GDL was not given in the presentation and should be included in the total Pt loading when calculating the effective Pt utilization benefit for corrugated MEA technology.
- Two things were demonstrated: (1) 0.05 mg Pt/cm^2 and (2) a Pt-coated Ti screen GDL with good conductivity (the uncoated version showed poor conductivity).
- This project began in September 2010, but progress has been delayed for various reasons. The PI claims a 10% “finished” status. The only data presented were a few polarization curves collected using “Pt-coated titanium screens”; however, a variety of component vendors have been identified.
- These are the early days for the project. Most of what was presented is essentially the motivation for the work, apart from some negative results with Ti screens.
- It is difficult to assess the project this early. Understanding cooling issues with this new type of construction should be an early focus to evaluate practicality.
- The challenges in making these systems work seem very daunting. They have scaffolding, but the biggest challenge by far seems to be building an MEA and diffusion media inside of it. The PI’s insistence that he will lay it down “very carefully” is not reassuring.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.1** for its collaboration and coordination.

- The team consists of good partners, GrafTech and General Motors (GM), who can address the GDL and the testing and validation of the corrugated MEA concept. GM and GrafTech can help develop a workable stack using such corrugated MEAs.
- The team of GM, GrafTech, and Dexmet seem to work well with Ion Power.
- A good team has been assembled for this project. GM should help provide a realistic evaluation of this new construction.
- This project features good cooperation with industry.
- It appears that the PI has the right team.
- The rating of “good” is only if GM eventually comes on as a collaborator and modeler, as it must be aware of the issues in these systems.
- GM, GrafTech, and Dexmet are all subcontractors, but GrafTech and Dexmet are clearly more vendors than collaborators.

Question 5: Proposed future work

This project was rated **2.4** for its proposed future work.

- This reviewer is looking forward to seeing the progress next year.
- The proposed future work is aligned with the proposed work of the project. The team needs to study the effect of stack compression force on the performance and durability of corrugated MEA under temperature and relative humidity cycling.
- The proposed work is reasonable, but it lacks benchmarking in terms of volume and weight to conventional designs. Benchmarking is needed to see if there is real merit to the “corrugated approach.”
- Conducting testing in an actual cell as soon as possible is critical.
- The investigators will jump right into testing these systems, but some simple qualifications can be done. It remains to be seen whether a small enough radius of curvature can be achieved with an MEA/GDL combination to build a system with a small enough pitch. Others questions are whether heat can be removed fast enough through a small amount of attachment welds, and how resistance can be lowered. The project team showed that the plate resistance was higher than typical MEA resistance. These hurdles need to be completed before the investigators attempt to conduct tests.
- The work discussed involves fabrication of the “corrugated” stack, followed by evaluation of the performance of that design. For now, it seemed that much of the details of the design are “open.”
- The project team needs to be more aggressive and quantitative in presenting proposed work. It was unclear if there is any proposed work with GrafTech or GM.

Project strengths:

- The team is well organized and capable of evaluating such a different idea.
- The innovation in trying to use engineered structures to raise power density is an area of strength for this project.
- A good team has been assembled for this project. GM should help provide a realistic evaluation of this new construction.
- This work proposes a novel stack design, which may have utility for one of the DOE fuel cell applications. Having other stack designs might prove more useful.
- This project is testing something a bit off of the beaten track. DOE should foster this type of work—it will provide the breeding ground for needed innovation.

Project weaknesses:

- The team assumes that a corrugated MEA system would not incur any additional stress around the corrugation during temperature and relative humidity cycling. The team also needs to consider the heat dissipation mechanism along the wall of the corrugated surface of the GDL/MEA, which is not in contact with the bipolar plate. Therefore, the coolant will have very little effect along the walls of the corrugated surface and can give rise to heat-spots resulting in the formation of pin-holes in the membrane. The heat management and cooling of the MEA in a large stack is always a challenge, and the team should seriously consider heat management challenges for corrugated MEA.
- This project has poor benchmarking, so it is not clear if there have been any improvements.
- This appears to be an ill-conceived idea. It is a hardware program, but the PI is an MEA expert. If GM does not come onboard to guide the PI to the relevant issues, this project should be dropped.
- The premise of the proposed work is highly questionable. The corrugation of the electrochemical package (i.e., current collectors, diffusion media, electrodes, and membrane) enables a different packing ratio. The projected stack area is less than the active area because the active area is the full surface of the corrugated sheet, not the projected area of that sheet (such a corrugated design has been commonly used in solid oxide fuel cell hardware). The result is that the projected area is smaller than the active area. However, the corrugated structures are necessarily (if the active area is actually larger) thicker than a planar assembly, so the stack length is increased. The final volume most likely will be larger, but the active area will clearly be larger. The PI claims that the stack will operate with higher efficiency. However, the improvement results from operating at a lower current density, the actual mA/cm² (milliamps/centimeter squared), but the principal investigator claims that the current density is measured using the projected area. This is rather obvious. With low-loaded electrodes, the costs for membrane and current collectors add up. The cost issues are complicated and were not convincingly presented. Certainly a stack operating with decreased current density will yield a higher efficiency. However, other issues—such as weight, volume, cost, reactant and product management, and thermal management—remain important. The corrugated MEA and its advantages must be considered with the rest of the stack design issues and their advantages. The investigators did not present any convincing design results that would suggest inherent advantages. It remains to be seen if a planar stack that is designed to operate with lower current density will produce the same result. Such a stack would be larger, but perhaps far easier to engineer, especially for the details of current flow, heat, and mass transport.
- The project team needs to be more focused and organized in defining future work and goals.

Recommendations for additions/deletions to project scope:

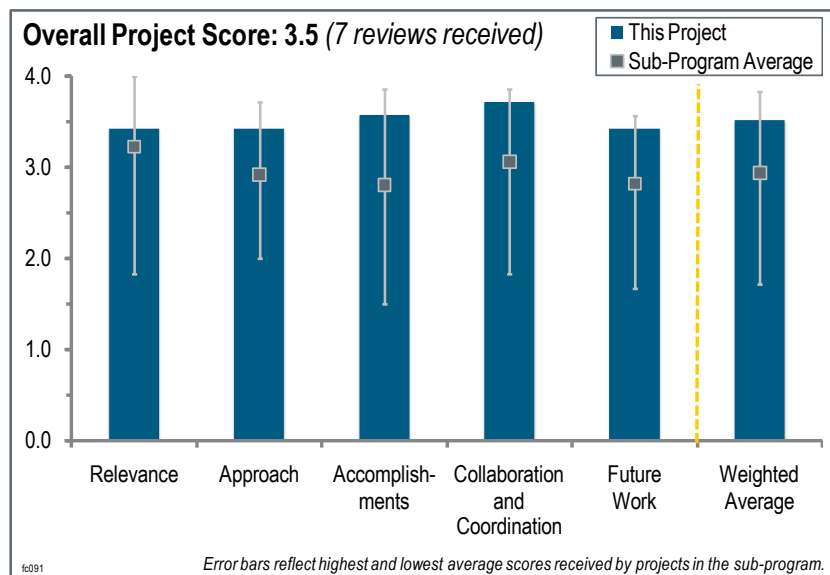
- The project should conduct benchmarking in terms of volume and weight to conventional designs.
- This construction could be useful for air-cooled, lower temperature applications, such as backup power, and should be investigated for such applications.
- Investigators should add a go/no-go decision to the program schedule. A detailed engineering design for the fabrication of this device should be performed, perhaps at the end of the first year, and evaluated as a go/no-go decision point. That design review needs to include a full understanding of all transport issues, as well as proposed fabrication processes. At that time, some significant “single cell” performance data should be presented to justify additional development.

Project # FC-091: Advanced Materials and Concepts for Portable Power Fuel Cells

Piotr Zelenay; Los Alamos National Laboratory

Brief Summary of Project:

The overall objectives of this project are to develop advanced materials (e.g., catalysts, membranes, electrode structures, and membrane electrode assemblies) and fuel cell operating concepts capable of fulfilling cost, performance, and durability requirements established by the U.S. Department of Energy (DOE) for portable fuel cell systems, and to ensure a path to large-scale fabrication of successful materials. Project technical targets are: (1) a system cost target of \$3/W (watt); and (2) a performance target for an overall fuel conversion efficiency of 2.0–2.5 kWh/L (kilowatt-hours/liter).



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.4** for its relevance to DOE objectives.

- This project directly addresses DOE durability, cost, and performance challenges for non-hydrogen fueled portable fuel cells. The investigators understand that the technical targets must be based on DOE goals, and have proposed reasonable solutions to achieve them.
- This project is working toward the key, system-level goals of performance, durability, and cost for portable power.
- This project addresses barriers for fuel cells for consumer electronics and portable fuel cell systems.
- The project is directly relevant to DOE objectives because it addresses three important limitations of state-of-the-art (SOA) direct methanol fuel cells (DMFCs): (1) low catalytic activity of C-PtRu for the methanol (MeOH) oxidation reaction; (2) high costs, depending on the high loading of expensive Pt- and PtRu-catalyst used; and (3) a low MeOH utilization rate, depending on the high MeOH permeation rate of Nafion® membranes.
- The project targets are well-aligned with the DOE Office of Energy Efficiency and Renewable Energy, Fuel Cell Technologies Program's *Multi-Year Research, Development, and Demonstration Plan* and the activities are aligned to the overall DOE Hydrogen and Fuel Cells Program goals.
- This project is relevant to DOE objectives. Quantitative technical milestones are provided that map to the DOE goals. The investigators clearly state what efficiency (cell voltage) is required to meet the DOE goals.
- This project fully supports all three critical Program objectives: durability, cost, and performance.

Question 2: Approach to performing the work

This project was rated **3.4** for its approach.

- The approach achieves an excellent balance of focus on improving the existing state-of-the art DMFC technology through significantly improved understanding of the catalyst and membrane with exploratory (but designed with go/no-go decisions in mind) work on other fuels. The project has a strong balance between fundamental research and practical catalyst, membrane, cell, and stack demonstrations. The project has a very

broad initial scope of studies (especially with respect to the various classes of catalysts), but appears to have clear go/no-go decision points and metrics to identify the most promising systems.

- It is unclear if Pt nanotubes can be made thin enough to reach the mass activity targets. Calculations based on currently achieved activity and thickness to determine how much thinner the nanotubes would need to be would be beneficial. The use of Rh and Ir is not beneficial on a cost basis. Rh is currently more expensive than Pt. Iridium is only slightly less than Pt, but it is scarcer.
- The project features well defined and challenging milestones. Using a multiblock copolymer with biphenol based polysulfone is a good approach to designing membranes with lower methanol crossover than Nafion®. However, at least for methanol as fuel, the ultimate goal of 96% fuel utilization appears out of reach with this type of membrane material.
- The approach is well-rounded and addresses many of the major durability and performance issues with portable power fuel cells, such as anode catalysts, membrane, and membrane electrode assembly (MEA) integration and testing.
- This project does not have a singular approach, but instead comprises many activities that contribute to meeting the DOE goals. These individual activities are generally strong, but given the size of the project there should be a critical path with intermediate milestones for each activity. Such a path would increase the chances of meeting the final project goals.
- The technical approach used in this project is adequate, systematic, and well planned.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.6** for its accomplishments and progress.

- Investigators have made excellent progress during the short time the project has been active. The only glitch has been the delay with the subcontract to Smart Fuel Cell, which is not surprising given the challenges of government sponsorship of foreign entities. However, it will be worth having Smart Fuel Cell's expertise once the details worked out. Overall, the investigators have been really conscientious about conducting the necessary benchmarking with SOA/standard catalysts or other appropriate baseline systems to assess the impacts of the new catalysts and polymeric materials accurately. The project featured early achievements and significantly surpassed the mass activity milestone for the thrifed (i.e., lower cost) Pt catalyst. The researchers have also adequately acknowledged the need to demonstrate durability. The initial nanotube results have been promising, but the project would benefit from a discussion of the likelihood of decreasing the tube thickness using the current or other synthetic methods. Multiblock polymer results have also been impressive, particularly the three-fold reduction in thickness. A direct dimethyl ether fuel cell (DDMEFC) has shown excellent preliminary results, particularly better performance at lower pressures than demonstrated in previous literature reports. The project team is really targeting key experiments to determine viability.
- The membranes show better conductivity and lower MeOH crossover than Nafion®. Investigators have developed a direct ethanol fuel cell (DEFC) catalyst with good conversion to CO₂, as well as a DMFC catalyst with improved Pt mass activity.
- The project team has made a very good effort in anode catalyst development. For the catalyst stability measurements, the cathode should also be measured with MeOH stripping to see if any Ru from the anode crosses the membrane. The partial fluorination of the hydrophobic block was successful in improving bonding with electrodes. The investigators have already achieved the milestone for membrane synthesis, which was targeted for April 2011. The novel membranes show lower methanol permeability than Nafion®-117, and the MEAs have better cell performance than Nafion®-based MEAs. However, the methanol crossover measured in limiting current experiments is higher than in Nafion®-117 based cells and not lower, as researchers hoped. The project team should reconsider selection of the membrane specifications.
- The project has made good progress and met most of its milestones, and is on track for the upcoming milestones.
- All of the activities are showing progress; some more than others. The accomplishments are expressed in quantitative terms so progress can be measured against SOA and past results.
- This project has made great progress and generated excellent results since it started.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.7** for its collaboration and coordination.

- This project features a strong team with complementary expertise that covers the project scope. In particular, Smart Fuel Cell is a rare industrial partner with commercialization experience in the portable power area. The role of each team member is clearly identified. In addition to the project team members, several strong international collaborations increase the likelihood of success and impact.
- A wide range of collaborators with appropriate sets of skills and contributions are involved in the project.
- This project features several collaborators, including a DMFC fuel cell company and a catalyst provider.
- There is good cooperation with the external project partners.
- The combination of collaborators on the project (universities, national laboratories, and industrial partners) is quite good. The contributions from each partner are logical and well-designed.
- The approach is multifaceted, with seven institutions participating. The investigators did not present any details regarding how this is managed, which should perhaps be discussed in future reviews.
- This project features an excellent team that includes renowned experts in the area. The project is well-coordinated and has full participation from each team member.

Question 5: Proposed future work

This project was rated **3.4** for its proposed future work.

- The proposed future work addresses challenges and questions that resulted from the initial results, and provides adequate tasking to enable key go/no-go decisions on Sn-based catalysts and DDMEFC viability during fiscal year (FY) 2012.
- Testing in MEAs under practical operating conditions is important to understanding the real performance and durability of the components.
- The proposed future work represents a logical progression to meet the targets.
- The work packages are well defined.
- The future work plan is reasonable. There is emphasis on the ternary catalysts and alternate fuels. These efforts should have clear targets and go/no-go decision points to conserve the Program's resources. This reviewer realizes that the proposed work highlights the short term, but some discussion is needed about the MEA and stack development tasks, particularly regarding metrics.
- The proposed future work is planned and phased in a logical manner with appropriately incorporated critical decision points.

Project strengths:

- The project slides were excellent, and clearly presented information regarding the project's rationale, approach, milestones, and status. The slides also included a data and key scientific/technical finding summary related to the data presented on each technical slide.
- This project features good teams and a sound approach to materials development and evaluation.
- This project's strengths include its strong team and good initial results.
- All of the project partners have great experience and expertise in their respective fields.
- The team that is assembled for the project is top-notch. Partners are well-integrated into the objectives.
- This is a powerful team of researchers. The metrics for technical performance are for the most part clearly defined and if achieved will meet the DOE goals.
- Excellent team.

Project weaknesses:

- It would be preferable to understand nanotube fabrication processes better to assess the potential for making thinner nanotubes. Maybe this is sensitive/propriety information based on the early stage of the project.

- More testing is needed in MEAs as well as testing at more practical temperatures and in multicell stacks. The MEA operating conditions of 80°C seem too high. Long-term testing of the catalyst and membrane under MEA operating conditions, including off spec conditions, is needed.
- A number of the estimated costs for the new anodic catalysts will be helpful comparing this with SOA catalysts.
- The cost metric has been largely ignored thus far. Some consideration should be given to this and approaches should be terminated that do not look like they could eventually meet cost goals.

Recommendations for additions/deletions to project scope:

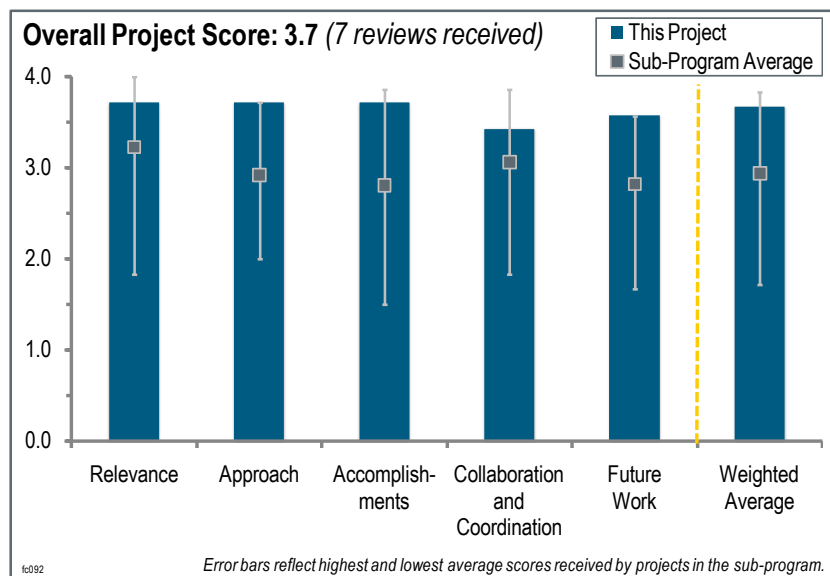
- It is too early to assess the need for additions or deletions. The ability to make key go/no-go decisions in several task areas in FY 2012 as well as the need at some point to down-select between the various MeOH catalyst approaches may require additional focus at a future date if project remains as broad as initial scope.
- Dimethyl ether fuel cell work is less developed and perhaps draws funds and resources from DMFC and DEFC work.
- The block copolymer membranes still have a relatively high methanol permeation compared to other hydrocarbon materials, leading to a loss of efficiency of fuel. The reviewer notes that this can be adjusted through material synthesis, and asks if there are plans to do this.
- The ternary catalyst and alternative fuels tasks should be monitored closely and deleted, if deemed necessary, so that resources can be focused on the core goals of the program.
- The project team should incorporate the study of the electrochemical stability of PtRhSnO₂.

Project # FC-092: Investigation of Micro- and Macro-Scale Transport Processes for Improved Fuel Cell Performance

Jon Owejan; General Motors

Brief Summary of Project:

The basis of this project is employing new and existing characterization techniques to measure transport phenomena and fundamentally understand physics at the micro-scale. Additionally, a comprehensive down-the-channel validation data set is being populated to evaluate the integrated transport resistances. This work will consider baseline and next-generation materials sets. The project is standardized by materials and operating space. Baseline and auto-competitive material sets are chosen based on parametric variations that consider degradation and cost versus performance tradeoffs. A database for data dissemination and modeling is available at www.PEMFCdata.org (development will continue throughout the project).



Question 1: Relevance to overall U.S. Department of Energy objectives

This project was rated **3.7** for its relevance to U.S. Department of Energy (DOE) objectives.

- The project addresses a range of DOE targets and challenges at a fundamental research level. The development and maintenance of a website provides ready access to methods and results for the broader community and could accelerate and increase the impact, particularly with respect to the objective of creating a modeling tool that the community can use.
- This project's relevance to DOE objectives is high. This project will enable the fuel cell community to use tools to identify rate-limiting steps, compare different sets of materials, and give recommendations for fuel cell stack improvements.
- The study of transport phenomena is critical to optimizing fuel cell performance.
- This project should result in advancements in cost and durability, with an improved understanding of key parameters and phenomena.
- This is the most comprehensive investigation of micro-and macro-scale transport processes ever attempted, particularly in regard to water distribution. The investigators have both a baseline set, which is higher in Pt loading and has a thicker membrane, and an auto-competitive set. The plan is to validate their pseudo two-dimensional model or update to a full two-dimensional model if required. The flow field is channel-land for both cases, and a materials list and operating conditions are presented for both cases on slide seven. The plan is to post data on a public forum database for at least the baseline condition. The investigators envision the data posted on their database to be a point of consensus within the DOE Transport Working Group. They are publishing the materials list for their auto-competitive polymer electrolyte membrane (PEM) fuel cell—therefore, even baseline unit data is enough to enable the fuel cell community to build its own PEM fuel cell that meets most of the goals for DOE's 2015 targets.

Question 2: Approach to performing the work

This project was rated **3.7** for its approach.

- The approach is excellent. The project team is connecting characterization with validation for a down-the-channel model. In separate experiments, a comprehensive macro-scale validation database is generated with fully integrated material sets and local down-the-channel resolution. The investigators list their fiscal year (FY) 2010–2011 deliverables as well as FY 2011–2012 and FY 2012–2013 expected work. The fact that they list material sets for both the baseline and current auto-competitive cases is unheard of—the work on the baseline case is expected to take the bulk of FY 2010–2011. The public database from this work is expected to be of immense help to the fuel cell community.
- It is hard to see how this could be done better. General Motors (GM) described an integrated performance evaluation and performance modeling activities. This is the necessary, highly interrelated approach. Critical parameters will be discovered in both activities: Measurements will show processes that are not in the models, but need to be, and models will disclose processes that improved (i.e., next generation) models need to include.
- This project uses a well defined baseline materials set and well defined excursions from the target data set to achieve a fundamental understanding of the impact of materials changes on performance and cost. The project is a good combination of theory and experiment. It will provide significant new insight into water transport mechanisms that is relevant to practical systems without sacrificing strong fundamental science.
- The approach to combine micro- and macro-level phenomena both experimentally and by modeling with industry standard validation and a front-end database that is accessible to the community is outstanding.
- The approach includes measuring a lot of important parameters and using a model to help maximize the understanding of the experimental results.
- The approach involves developing a simplified fuel cell diagnostic system that allows for the monitoring of key materials and operating parameters. Such a system will greatly aid in obtaining a fundamental understanding of the multiple processes that are occurring.
- The approach is in good accordance with the project objectives.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.7** for its accomplishments and progress.

- The results presented were very good. The different experimental tools are in place and working, and the different announced topics have been addressed. The project team has carried out much testing, enabling it to upload an impressive amount of data to the already existing website.
- The validation experiments have been excellent—the team has an appreciation for mimicking realistic temperature distributions. The database seems to be up and running and functional. The team conducted beautiful experiments on transport in Nafion using film thickness as a variable. The modeling seems appropriate. The investigators have used a nice set of advanced diagnostics throughout the project.
- The project has produced lots of good results to date. This reviewer looks forward to some more significant insights in the future.
- Although this project is only 20% complete, significant progress has been made.
- Accomplishments to date include acquiring, analyzing, and uploading to the public database one entire baseline data set using the standard protocol (117 test points).
- The most important activity to date is building the talented crew who are on the GM team. Even excellent people will take time to become useful and an excellent, useful team is essential. One hopes that these GM people are given the time, because it appears they do have the talent. It takes both time and talent.
- The project got off to a slow start due to subcontracting delays, which are now resolved. The initial results highlight the need for a reliable, simplified, one-dimensional water transport model. The project has a good mix of practical and model materials systems to develop and validate the modeling capability. The initial results have been promising in many of the materials characterization tasking areas, as well as modeling. This reviewer anticipates significant progress by next year's Annual Merit Review.

Question 4: Collaboration and coordination with other institutions

This project was rated **3.4** for its collaboration and coordination.

- This is a good team with complementary expertise.
- The collaboration and coordination between the partners must be very good in light of the number and quality of the obtained results.
- The project features good collaboration between an original equipment manufacturer (OEM), a membrane electrode assembly manufacturer, and universities. It seems that the project should have stronger interactions with a DOE national laboratory that is doing similar work to avoid duplication in the DOE Hydrogen and Fuel Cells Program.
- This project has a good team and good coordination by the principal investigator, and the website is outstanding.
- There are a number of institutions involved in the project and the coordination appears to be very good.
- GM is collaborating with three universities, DOE, the National Institute of Standards and Technology, W.L. Gore and Associates, and Freudenberg. This reviewer was unable to find the work of the University of Tennessee, unless it is going by the logo of “FCDDL.” It was unclear who this logo represents. The reviewer is sure that as the project unfolds, the work of all participants will be revealed.
- The concern is that this sort of activity will lead to new understanding that could have large economic benefit. For example, the modeling may show another thermal management approach. Commercialization certainly requires trade secrets. This reviewer thinks that the GM approach—making codes available and consuming the data of others—is right on. However, it is necessary to protect proprietary design information, so sharing can only go so far. It is also important to share data with other quality laboratories.

Question 5: Proposed future work

This project was rated **3.6** for its proposed future work.

- The future is really the rest of the entire project, as the work presented was really “getting ready.” The pathway described will probably need to be modified as new information unfolds, but for now, the plan is sound.
- The proposed future work builds on initial results and the tasking clearly advances the work toward achieving the overall project goals.
- The proposed work is in accordance with the expected project objectives. Sensitivity analysis may be added as the extension to the stack level.
- The project will finish diagnostic development, measurement for model validation, and selection of auto competitive components.
- The project team should keep up the transparency.
- The future work builds very well on what has been done thus far, and is clearly focused on key aspects.
- The investigators plan on completing the component characterization method development with an emphasis on diffusion as a function of saturation. Then they plan on defining the remaining auto-competitive components and applying the characterization methods to them, and completing down-the-channel validation and populating the database with full baseline and auto-competitive data sets. This latter public dissemination of future auto-competitive data sets is unheard of. The investigators are then going to identify and integrate component model divergent transport resistances into the pseudo two-dimensional model.

Project strengths:

- Strengths of this project include the availability of many experimental tools to characterize the different components and the good balance with modeling. The project also features a well adapted partnership to integrate all of the needed competences to achieve the project targets, and fast dissemination of the project results through the website.
- Investigators are making a considerable effort to measure a lot of important parameters. They are also evaluating novel materials that potentially offer lower cost. Other strengths include the transparency with results, including a website to share them, and a good team with complementary skill sets and capabilities.

- The detailed experimental monitoring of the fundamental processes that occur in a proton exchange membrane fuel cell is an area of strength for this project.
- One of this project's strengths is the public dissemination of both the baseline and future auto-competitive component data sets.
- It looks like the team is excellent, well organized, and moving. This will be a "people thing" in the end, because the team needs to win, not an individual. It appears, from the information shown, that the team is competent and on target.

Project weaknesses:

- There are no major weaknesses to date.
- This reviewer is somewhat worried about the stability of the metal coated gas diffusion layer. It is well known that uncoated metal bipolar plates fabricated from inexpensive metals passivate and corrode. It is unclear what the stability and long-term performance of these metal-coated glass fibers is expected to be.
- It is not clear what new and significant understandings have resulted from this project to date. The investigators have shown lots of results, and some are interesting and useful, but there have not been any real surprises yet. There has been nothing really innovative in this project, but this type of fundamental and rigorous work is indeed needed.
- Keeping sufficiently high levels of project coordination may not be easy.

Recommendations for additions/deletions to project scope:

- The project scope is excellent as is.
- The project has a good plan, and should continue as planned. The project team should keep up the transparency.
- Investigators should bring in more industry and laboratory partners.
- Fuel cell performance depends upon some number of interrelated parameters. So does chaos. The problem will be that the variables change depending upon the value of other variables. It is also obvious that many transient events happen on the local scale during fuel cell operation. The project team should think about other ways of describing overall stack performance that account for the fact that fuel cells are very non-uniform, just like all other heterogeneous catalytic reactors. One critical issue is heat transfer; another is stress and the implications of stress on transport. One suspects that this team is on top of all of this.

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