2010

Fuel Cells

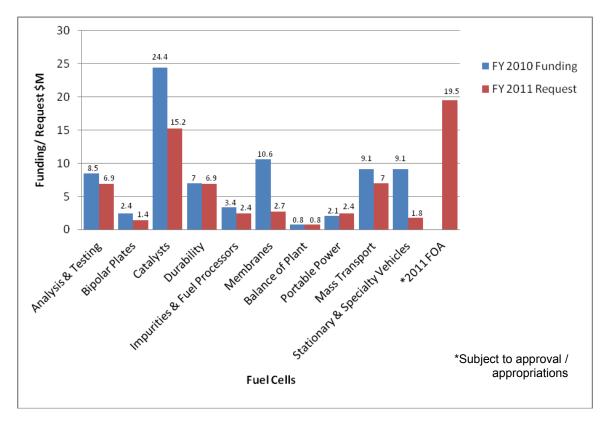
Summary of Annual Merit Review of the Fuel Cells Sub-program

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

Reviewers consider fuel cell and fuel cell system development to be a critical enabling technology for the success of the DOE Hydrogen Program. The strength of the Fuel Cells subprogram was evidenced by reviewer comments pertaining to the world class scientists working in strong, well-managed teams across industry, laboratory, and university settings. Teams continue to enhance understanding and research, develop, and demonstrate cutting edge technologies that will further enable fuel cell commercialization by allowing competitiveness with incumbent technologies.

Fuel Cells Funding by Technology:

The Fuel Cells sub-program continues to concentrate on the critical path technology of stack components—including membranes, catalysts, bipolar plates, mass transport, and analysis/characterization—as well as system balance of plant components and fuel cell systems for transportation, portable, and stationary applications. Additional projects address cross-cutting technologies, innovative concepts, and the effects of impurities on fuel cell performance. The primary focus of the sub-program is reducing cost and improving durability.



Majority of Reviewer Comments and Recommendations:

At this year's review, 77 projects funded by the Fuel Cells sub-program were presented and 55 were reviewed. All 48 presentations given during the oral segments were reviewed as were seven of the 29 poster presentations. Reviewer scores for the fuel cell projects ranged from 3.6 to 1.9, with an average score of 3.0. This year's highest score of 3.6 was equal to last year's highest. Both the average score of 3.0 and the lowest score of 1.9 for 2010 were higher than 2009's average score of 2.9 and lowest score of 1.6.

	Highest Score	Average Score	Lowest Score
2009	3.6	2.9	1.6
Presentations			
2010	3.6	3.0	1.9
Presentations			

Forty eight projects were reviewed during the oral sessions by between five and eight reviewers. On average, six experts reviewed each project. Either four or five experts reviewed the seven posters, with an average of just under five reviewers per poster.

Analysis/Characterization: Of the eight projects presented in this category, three were ranked in the top six of all projects in the sub-program. Overall, the average score of 3.3 is well above the sub-program average of 3.0. The analysis projects scored highly in their relevance to DOE objectives and goals. Reviewers encouraged the PIs to collaborate with more OEMs to deepen the understanding of the components that are available and also to allow the OEMs to better understand the models being developed. The cost of an 80-kW automotive PEM fuel cell system operating on direct hydrogen and projected to high manufacturing volume (500,000 units per year) continues to decline, and the cost studies being conducted help elucidate opportunities for greater cost improvement.

Water Transport: Two water transport projects were presented, with an average score of 2.9. A considerable volume of data was produced and a wide range of experiments were performed for model validation. Reviewers were interested in understanding details of the model, such as the accuracy of the parameters.

Impurities: This year the average of the four impurities projects presented earned just over the sub-program average, with an average score of 3.05. Projects were commended for having the unique ability to synthesize and characterize different ceramic supports and for identifying, modeling, and cataloging system contaminants and then disseminating this knowledge to the fuel cell community. Reviewers consider the impurities studies on both the fuel and air sides of the fuel cell to be very important to the success of the Program.

Membranes: Ten membrane projects were presented and reviewed, with an average score of 2.8. The highest ranked membrane project received a score of 3.6 and tied for the highest scoring project in the sub-program while the lowest ranked project scored a 1.9 and tied for the lowest scoring project overall. Projects were lauded for building on proven perfluorosulfonic acid chemistry and improving conductivity and durability while maintaining manufacturability and for addressing the need to provide conductivity in the absence of water. The portfolio of ten projects is especially relevant to the automotive applications the Program pursues, as the development of

robust, high-temperature, low-RH membranes is vital to the successful commercialization of fuel cells.

Catalysts: The average score for the 11 catalyst projects was equal to the sub-program average of 3.0. The highest score given to a catalyst project was 3.6, which tied for the highest scoring project in the sub-program and four of the nine highest scoring projects were catalyst projects. The lowest ranked catalyst project scored a 1.9 and tied for the lowest scoring project overall. Reviewers noted the advancements in cathode catalysts and supports and the progress that has been made in thin film electrolyte technology. 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.

Transport Studies: Four transport studies projects were evaluated and given an average score of 2.75, which is less than the sub-program average. The highest scoring transport studies project, with a 3.3, uses multiple approaches to model a single cell and stack to understand key performance cost drivers. The lowest scoring transport studies project also tied for the lowest scoring project in the sub-program's portfolio. The reviewers did not believe that the approach was satisfactory and questioned whether the project was producing transferable value to the research community. The two remaining projects scored near the average, and the reviewers offered many suggestions for how the projects could be improved in approach and future planning.

Degradation Studies: The average score for the five projects concerning degradation studies was 3.1. The projects in the degradation studies portfolio were regarded as having commendable approaches and excellent scientists with the capacity to potentially answer some key fundamental questions about fuel cell degradation mechanisms.

Bipolar Plates: This year three bipolar plates projects were evaluated, with an average score of 3.0, equal to the sub-program overall average. Reviewers were impressed with the new and innovative approach of using highly conductive and ductile aluminum to coat bipolar plates. Reviewers recommended including additional tests to demonstrate performance and corrosion resistance under real-world conditions and for greater periods of time for hardware projects.

Distributed Generation:

The two distributed generation projects received an average score of 2.75. The reviewers praised the project's development of a tubular solid oxide fuel cell and the project's success in leveraging niche markets. However, reviewers were disappointed that a high efficiency PEM fuel cell system had demonstrated lifetime, durability, and efficiency metrics well below DOE targets.

Portable Power:

One portable power project was presented this year and earned a score of 2.7. This project focused on improving the catalytic activity and durability of platinum ruthenium for direct methanol fuel cells. Reviewers noted that the most relevant finding of this project was demonstrating the first stable, high activity catalyst for methanol electro-oxidation.

Innovative Concepts:

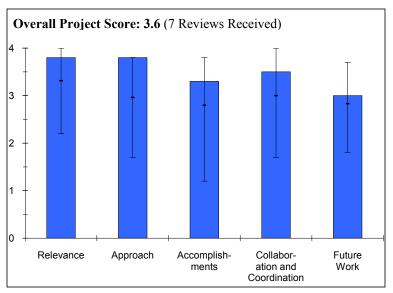
Three projects presented this year fall under the category of innovative concepts. The average score of the projects was 2.9. The highest scoring project earned a 3.1 and studies nano-scale ceramic supports as alternatives to carbon supported catalysts, which may result in a more durable cell. A 2.9 was given to a reversible solid oxide fuel cell project that addresses energy storage while pursuing high capital utilization. The third project in this category earned a 2.6 for

an anion exchange polymer electrolyte project for alkaline fuel cell applications, which was believed to have the potential to drastically reduce precious metal usage in fuel cells. The reviewers commended this diverse portfolio of projects for their strong teams and the potential of their innovative ideas. However, they noted that more research should be conducted to better leverage lessons learned from projects conducted previously by other teams.

Project # FC-01: 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 with gas diffusion media, fabricated by high volume capable processes, and is able to meet or exceed the 2015 DOE targets. Focus topics for past year were: 1) water management improvements for cool/wet operation through materials, electrode structure, and boundary condition optimization and understanding; 2) continued multiple strategies for increasing nanostructured thin film (NSTF) support surface area, catalyst activity and durability, with total loadings of $<0.25 \text{ mg}_{Pt}/\text{cm}^2$ per membrane electrode



assembly (MEA); 3) continued fundamental studies of the NSTF catalyst activity for oxygen reduction reaction in general and methods for achieving the entitlement activity for NSTF catalysts; 4) more severe accelerated tests to benchmark the NSTF/MEA durability; 5) development of faster, easier MEA break-in conditioning protocols; and 6) working with system integrators to validate NSTF functional properties or issues in short stacks.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.8** for its relevance to DOE objectives.

- 3M's work directly addresses one of the major DOE technical areas--catalyst performance and durability.
- They always keep DOE targets in sight and work to achieve them.
- This program remains a lighthouse for direction and accomplishment toward DOE automotive goals of reducing platinum group metal (PGM) content, increasing power, and increasing durability under automotive conditions.
- This is a large, complex project that involves many activities that are focused on the DOE performance goals. The impressive record of meeting and exceeding the DOE goals demonstrates the focus and relevance of the project.
- Efficiency is the key to fuel cell commercialization, and that thrust demands improvements in electrocatalysts and MEAs, which are the main focus of this large-scale DOE project.
- This project is perhaps the most relevant of all the DOE fuel cell projects. Improving cathode catalyst activity and durability is the most crucial task toward enabling fuel cell vehicles to overcome the cost and durability barriers that prevent commercialization. This project has been addressing both activity and durability in a manner that is both consistent with atomic-scale fundamental research and with a material that is amenable to large-scale mass production. If the DOE only funded one fuel cell project, this project would be it.

Question 2: Approach to performing the research and development

This project was rated **3.8** on its approach.

- 3M uses DOE accelerated test protocols, internal accelerated stress tests (ASTs), and automotive-systemrelevant cycles designed to stress 3M's NSTF materials and structures to evaluate progress against DOE technical targets.
- It is not clear whether or not the materials will transition to fuel cell systems.
- Accepted characterization techniques are a strong part of the program. This includes *ex situ* and *in situ* materials and components' performance and durability testing, as well as extensive use of characterization techniques such as transmission electron microscopy (TEM).
- The original design concept has continued to bear fruit and progress. It is good to see that the project has processed 37,000 linear feet of material and also the move to pre-production processes.
- 3M's approach to dealing with such a complex project is exemplary. Breaking the project down into manageable pieces not only facilitates performance, but also helps to explain what is going on to the reviewers. The technical approach is extremely good, although the solution to the flooding problem may have a weakness (see below).
- 3M is a good project lead because they have the ability to move quickly from the small to the pilot scale for evaluation.
- They still have a lot of uncertainty in their methods. This issue seems to be more programmatic and the project would benefit from a more detailed development method. The project is confusing as presented because the methods keep changing.
- The project combines detailed modeling, advanced manufacturing, materials science, and testing to develop new products for contemporary fuel cell systems. Dr. Debe has created a fuel cell culture that continues to evolve and adapt technical advances in real time. An example of this evolution is the inclusion of newer coreshell catalysts.
- This project is the benchmark for all other DOE fuel cell projects in terms of approach. For years, oxygen reduction activity on platinum has shown to be greater per unit area for platinum in bulk-like phases, particularly with preferred surface orientations. NSTF provides for this and, by doing so, has performed excellently with low catalyst loading in mass-produced MEAs.
- The approach of this project has been criticized in recent years for not paying enough attention to the major issues that have prevented commercialization, namely water management and conditioning. However, this year the project has addressed these issues, delivering interesting phenomenological results that may lead to practical solutions.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.3** based on accomplishments.

- 3M has met nearly all of the DOE 2010 targets for single cells and some of the targets for short stacks tested by General Motors (GM). However, not all of the targets have been met simultaneously by a single set of materials and structures.
- 3M continues to make steady progress, and are reaching the DOE goals.
- 3M has met some of the goals only through the use of non-conventional methods, such as requiring sub atmospheric pressures on the anode to assist in water management.
- There are still some concerns over water management as this still appears to be the last hurdle. 3M has suggested a clever approach to use the anode as the water removal element, along with thin membrane and differential pressure. However, the question of what the impact on the automotive system would be if the anode is run at lower pressure compared to the cathode needs to be addressed.
- There has been excellent progress in all areas. The response in regard to the flooding is innovative, but it may have a flaw. Would the flow of water over to the anode not encourage oxygen to reach the anode, thereby leading to increased chemical stress on the membrane? Perhaps the membrane stability is sufficient to stand up to this, but it would be good to demonstrate this.
- The team has made very good progress toward meeting DOE goals.

- 3M doesn't explain why its specific activities are so high (2,500 uA/cm², or about 8 times the standard platinum on carbon), but the mass activity has only a 2.5x increase. Where did the activity go? It is not clear whether this a measurement issue.
- The Pt₃Ni₇ seems very interesting, but it's disconcerting that 3M talked up its other manganese-based catalyst so much last year and now 3M is talking up its new catalyst. 3M was not particularly clear about how stable the catalyst was with cycling as nickel is lost or what the loss of nickel into the fuel cell might mean on a system level.
- There is much to admire in the results of this activity. 3M has an excellent, integrated team that has the ability to do many things well and thus retain a leading position. 3M has a long history of making large volumes of coated polymers, and it is obvious that the fuel cell products are part of that technology flow.
- In regard to the major issue--water management--the project has not yet delivered a practical solution. It is very difficult to see how sub atmospheric pressures can be targeted for particular operating modes without adding parasitic loads to the system, which would create more costs for needed components. However, it can be said that the sub atmospheric pressures used were good for demonstrating the phenomenon needed to enhance water management: removal of water from the cathode catalyst layer, particularly towards the anode. From a phenomenological standpoint, 3M reported some good work.
- The development of pure platinum NSTF is very welcome from a processing standpoint. It would be interesting to further develop apples-to-apples comparisons with PtCoMn mass activity and cycled durability.
- Major progress has been reported with respect to dryer conditioning. More progress is still needed to reduce dryer conditioning time from six hours.
- Pt₃Ni₇ is interesting, but it would appear that just a small change in nickel weight percentage would result in a drop in activity.
- Cycling tests (0.6-1.2 V) show major loss in performance over 4,000 cycles, but these cycles are not necessarily equivalent to realistic cycling. This underlines the need for more realistic cycling to be done.
- Open circuit voltages (OCVs) appear consistently low (near 0.9 V or below). It would be interesting to understand more about this, or whether OCV is actually being measured.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated 3.5 for technology transfer and collaboration.

- 3M's program involves collaboration with several universities as well as the U.S. auto original equipment manufacturers and some stack integrators.
- 3M is willing to provide NSTF samples to any credible fuel cell R&D entity.
- It is not clear how strong a link there is with the fuel cell manufacturers.
- There is a wide array of partnerships, especially with GM and Nuvera Fuel Cells.
- The presentation showed some of the collaborations and how they were integrated, but did not show all of them, which was probably due to a lack of time.
- It would have been helpful if they could have talked more about their relationship with fuel cell manufacturers, and try to present more non-proprietary results.
- It is obvious that 3M has effective collaborations and is teaming with quality institutions. It is also obvious that the principal investigator has highly effective procedures that have resulted in effective group productivity.
- The Dalhousie collaboration has been particularly valuable toward developing more active alloys, and perhaps even more valuable toward screening out less active alloys.
- Assistance from Argonne National Laboratory (ANL) with activity measurements has also been worthwhile in providing insight into expected activity measurements.
- The National Aeronautics and Space Administration's (NASA) role was not explicitly stated in this year's slides.
- Most other collaborations' results are either mostly unreported or will be expected to deliver more progress in the future. This is especially true in the case of the interactions with the water transport projects (e.g., the Lawrence Berkeley National Laboratory), and in the case of stack testing at Nuvera.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- The focus of future work will benefit from further evaluation of catalyst layer structures and compositions.
- Plans include optimizing catalyst compositions, structures, and fabrication processes to meet and exceed DOE catalyst targets on a single materials set.
- There is not much time left. Considering the extensive accomplishments in the past, a focus on stack testing and water management may be more than enough for future work.
- The plan for proposed future work is good in most respects, except for water management. Future work in this area should focus on whether the method of dealing with flooding affects membrane life.
- 3M keeps moving along making incremental, but steady, progress.
- The next act is obviously optimization. There appear to be concerns about water management ("flooding"), break-in details, and perhaps durability. Concerns will happen; the important attribute is to address them, and this is happening.
- It would be good to see some of the details within the categories of future work described. It is unclear whatthe strategy on water management is. They could assume atmospheric anode pressures are acceptable or to attempt to find other routes toward shifting water away from the cathode catalyst layer.
- There was nothing described in terms of the direction for durability cycling. There is still not a realistic drive cycle in the project, and it would be interesting to see if certain collaborations (e.g., GM) would be able to provide results. The results from 0.6 to 1.2 V cycling do not look good, but the cycle is more aggressive than needed.

Strengths and weaknesses

Strengths

- The project is simultaneously addressing all aspects of catalyst development.
- 3M is making outstanding progress on an innovative concept that is fraught with difficulty and requires new understanding of what happens with ultra-low PGM MEAs.
- 3M has a very strong team.
- Their mindfulness of the transition to manufacturing processes is also a strength.
- Even though this is a large complex program, it is being managed well. Breaking the component activities down into manageable parts brings order to what could easily be an out-of-control complexity.
- 3M has the capability to evaluate catalysts in pilot-scale MEAs.
- The project team is making slow but steady progress toward DOE goals
- The introduction of a new Pt₃Ni₇ catalyst with very high activity as demonstrated in MEAs is a strength.
- As with all successful technology, the 3M team is the key to this project's success, and is the most significant strength.
- This project holds the greatest promise toward developing a catalyst that will enable fuel cell vehicle commercialization. The project's approach is entirely consistent with the scientific fundamentals of improving oxygen reduction platinum-based catalyst activity and durability. At the same time, industrial processes for mass production exist.
- The project is well-organized and disciplined. It is able to deliver results each year toward improving mass activity and surface area, and quantifying durability. We can now say the project is on its way to understanding the reasons for water management deficiencies.

Weaknesses

- In one reviewer's opinion, the project has no weaknesses.
- The project has a clever approach to water, but there still seems to be a compromise to force an operation on the automotive system.
- As discovered in the program, water management on the cathode becomes the largest issue and ultimately may deserve the greatest effort to overcome.
- The methods being used are inconsistent.

- 3M is still battling water management.
- It is not clear if the use of 3M membranes in automotive fuel cells requires extensive reengineering to help with water management.
- The viability of Pt₃Ni₇ is not clear due to the impact of nickel loss.
- The project seems to be dominated by the fuel cell technical team, and its stringent focus on just one technology—light duty vehicles. Today, high purity hydrogen is the fuel, and thus lightly loaded anode catalysts perform well. One suspects that fuel specifications are unreasonable, and "technical grade" hydrogen will change things. The issues of water flooding, durability, shutdown/startup, and freeze protection involve stack design issues. 3M probably needs to be involved with stack technology, because fuel cell commercialization always requires optimization of the catalyst with the reactor.
- Practical solutions are still needed for water management.
- The conditioning time still needs to decrease.
- While NSTF is more durable than platinum on carbon, a proper cycle is still missing that would help toward understanding how performance would respond to vehicle operation.

Specific recommendations and additions or deletions to the work scope

- There needs to be an agreement on the last period of stack testing at potential customer sites and on the hardware.
- There should be more focus on water management.
- Membrane stability measurements under flooding conditions need to be tested, particularly under shut-down conditions where the voltage goes to over one V.
- In order to support 3M commercialization plans, the project should involve a large-scale product-testing activity, shipping products to both customers and developers, as part of a significant market-testing endeavor. This could be accomplished with a near-term commercial project, using, for example, a small-scale combined heat and power (CHP) system. However, the next phase needs to involve thousands of systems under test, and that is probably best done in early market activities.
- There is some "rumor" that the 3M products do not always perform as advertised. This needs to be thoroughly investigated, probably by a "round-robin" or some other focused testing activity.
- The project should remain focused on delivering water management solutions in at least atmospheric pressures.
- The addition of a realistic drive cycle (most likely through collaboration) would be useful to help understand the true durability of NSTF.
- The project should provide some reporting on OCVs. Some polarization curves pique curiosity by at least appearing as if OCV is a bit lower than 0.9 V.
- It would be interesting to know more about how grain sizes of PtCoMn and Pt₃Ni₇ compare (in a fashion similar to what was reported for pure platinum versus PtCoMn).

Project # FC-02: 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 DOE activity, durability and platinum group metal loading targets in a structurally optimized membrane electrode assembly (MEA) capable of performing at 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 DOE objectives

Overall Project Score: 3.1 (6 Reviews Received)

This project earned a score of **3.7** for its relevance to DOE objectives.

- This project addresses key issues of stability and activity.
- Durable alloy catalysts are essential to meeting 2015 cost and durability targets.
- The project is aligned with DOE goals and targets. It addresses critical issues of cost and durability.
- This is an excellent project that advances state-of-the-art catalyst technology.
- The DOE targets A, B, and C are addressed.
- The project aligns well with the hydrogen program objectives. It is focused on improving catalyst durability and also support durability, with more emphasis on catalyst durability. Catalysts investigated are ternary and coreshell-type alloys.

<u>Ouestion 2: Approach to performing the research and development</u>

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

- It is not clear whether this approach will get to the low platinum group metal (PGM) loadings required by DOE.
- Ther project has a good blend of theory and practice for a difficult core-shell target. As discovered by the team, more time and/or tasks to ink development should have been considered earlier.
- With limited time remaining, they need to combine the highest activity catalyst with the best support and try to optimize MEA activity. It's not clear the value added by modeling work.
- UTC is addressing technical barriers of cost and durability.
- The approach is sound and is yielding good results.
- With the three tasks being dispersed alloy catalyst development, core shell catalyst development, and carbon support investigation, the most prominent approaches are covered. The first two tasks are supported by modeling in addition to experimental investigations.
- The approach is broad, as far as catalysts are concerned, and there is still insecurity in manufacturing of the catalysts.
- The overall approach is good. The project spreads itself thin by focusing on two families of catalysts ternary alloys and skin/core-shell type. The core-shell approach still has unresolved issues in regard to cobalt or chromium leaching, due to the difficulty of coating the shell completely with platinum. It is not clear why the Pt₈IrCo₂ alloy was selected for carbon support durability tests, even though Pt₂IrCr was found to be more durable. The overall score approach on slide eight (fiscal year 2009) is an excellent way to grade the various catalysts. The improved MEA results for the Pt₂IrCr catalyst are encouraging.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.8 based on accomplishments.

- UTC was able to identify a possible failure mode for core and shell, which is the loss of structural integrity. Is this the link to loss of performance with MEA making?
- Modified carbon (C₄) has shown to experience the lowest loss, but there may not be enough surface area to support the activity goal. One wonders whether the DOE targets can be met. Ketjen black has far higher surface area and thus more corrosion compared to the other standard Vulcan XC72. It's surprising this group did not use the carbon employed for its 40,000-hour life phosphoric acid fuel cell--certainly that has to be a benchmark grounded in actual field testing and results.
- Some activity gains were made on the rotating disk electrode (RDE), but it is unlikely that they will meet MEA activity targets before project completion. MEA durability was not tested.
- UTC has decreased platinum group metal (PGM) loading from 0.8 g/kW to 0.5 g/kW, which is not at the 2010 target of 0.3 g/kW, but is still a significant improvement.
- UTC has downselected to one carbon support, and has demonstrated improved durability on this support.
- It would be nice to know why the MEA activities are so much lower than RDE activities (for the core shells especially, but also for the PtIr alloys).
- Tests with the fuel cell technical team recommended cycles would be useful for comparison to other projects (0.6V-1.0 V triangle wave at 50 mV/s).
- Excellent progress has been made and should be encouraged to continue.
- The 2010 DOE targets are not fully met for topic 1. In particular, cycling durability investigations are still to be performed. A corrosion-stable carbon material has been downselected; yet corrosion tests are to be extended to 5,000 hours.
- The project has made significant progress toward DOE goals. However, for the MEA tests, there are significant gaps for RDE results. While the team is focusing on MEA formulation optimizations to improve MEA results, it is not clear if this can be achieved in the time left for the project. The core-shell approach leads to leaching of palladium and cobalt; hence this is a high risk approach.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **2.8** for technology transfer and collaboration.

- It appears the collaborations do not extend beyond the immediate team. Since the core-shell approach is novel, new methods for the verification of structure may be needed and obtained through additional collaborations.
- It's not clear how Brookhaven National Laboratory (BNL) and Texas A&M University (TAMU) partnerships have provided value.
- The good collaborations within the team.
- UTC has very good collaborations in place, including top people at Johnson Matthey Fuel Cells, Inc. (JMFC), BNL, and TAMU.
- The collaboration structure was not shown clearly in terms of concrete results. The synergy of the collaboration is not clear, although the basic work share is outlined in chart 5. It is not clear to the reviewer whether modeling leads to a better understanding of the catalyst systems, and whether that information was taken up by the catalyst manufacturer.
- The team is very strong, with JMFC, TAMU and BNL. The project has very good collaboration, with each team role well-defined, as seen in slide five. JMFC provides the catalyst, while UTC Power tests MEAs (not clear who fabricates the MEAs). BNL looks at core-shell fundamentals. UTC Power also does testing to determine the corrosion resistance of alternate supports. TAMU does computational calculations.

Question 5: Approach to and relevance of proposed future research

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

- The core-shell approach may not have the potential to meet the program's targets. Author cites some stability issues in acid and leaching of transition metals. It seems this would make MEA production inherently difficult. It is unclear how the team would address a final catalyst that either loses activity with leaching or obtains a certain activity with a certain degree of (uncontrolled?) activity.
- This reviewer agrees with the author that, in the final phase, efforts should be dedicated to ink-making/electrode structure to show the potential to meet DOE goals.
- There is no MEA durability planned. With limited time remaining, they need to combine the highest activity catalyst with the best support and try to optimize MEA activity. The performance model is not providing value at this point.
- The project ends October 2010; the proposed work is logical to wrap up the project and accomplish deliverables.
- The project is going well and should continue as proposed by the PIs.
- The proposed work is described at a general level, which does not refer to the status achieved and does not show that the issues discovered will be tackled in a focused way.
- Since the project ends October 2010, the amount of future work proposed appears excessive compared to the time available for completion. The team did not state if the future work will be done by the end of this program, or if some of the proposed future work would be done as part of additional funding.

Strengths and weaknesses

Strengths

- This is a good team with depth--should the core-shell succeed, a commercialization partner is ready.
- They have screened many concepts for alloys, supports, and core shell concepts.
- Others can learn from their alloy work.
- Two strong and renowned industry partners are working together in this program.
- There is good collaboration among team members.
- UTC showed promising results for PtIrM alloy with chromium.
- UTC's ability to scale-up catalysts and generate scale-up data is a strength.

Weaknesses

- There was insufficient time reserved to develop the electrode structures necessary to support these new catalysts and/or carbons.
- UTC has not been able to make an MEA with good activity. There is no MEA durability. Others cannot learn from support work because materials are not disclosed.
- The project leaves the impression that the activities are still somewhat tentative.
- This project is overly focused on its core-shell approach.
- UTC has displayed an inability to get MEA activity comparable to rotating disk electrode (RDE) activity.
- There is a lack of criteria for catalyst selection for support.

Specific recommendations and additions or deletions to the work scope

- Focus on the ink/electrode structure.
- The project ends soon. With time left, try to combine the highest activity catalyst with the best support and try to optimize MEA activity, test MEA durability, and forget everything else.
- Demonstrate durability of the MEAs.
- There is a need for an academic partner to provide deeper understanding of the catalysts, or a more focused approach of the existing project partners.
- The team should focus on catalysts that work (especially for support durability tests), which is PtIrCr based. It should focus less on the core-shell approach, since leaching issues remain unresolved. The team should test the most stable catalyst with the most stable support before the project is over.

Project # FC-03: Development of Alternative and Durable High Performance Cathode Supports for PEM Fuel Cells

Yong Wang; Pacific Northwest National Laboratory

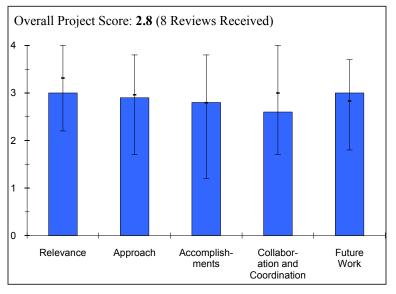
Brief Summary of Project

The overall project objective is to develop and evaluate new classes of alternative and durable high-performance cathode supports. The objective for 2010 is to demonstrate durability under accelerated test protocols that meets DOE lifetime criteria.

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of **3.0** for its relevance to DOE objectives.

- Some of the supports investigated consist of two or more components that may be difficult to use in real fuel cells.
- The relevance is obvious. A major decay mechanism is the oxidation or



- decay mechanism is the oxidation of the carbon support of the cathode catalyst.
- This project's relevance is somewhat limited in that it only addresses alternative catalyst supports, and therefore one major DOE barrier, i.e., fuel cell durability. Also, the overall objective of just two times higher stability than conventional carbon supports is a very modest goal and is subject to the test used to demonstrate it. Also, it depends on the type of carbon chosen as "conventional." Suppliers already sell much more stable carbon supports than Vulcan XC72, for example.
- They have a good focus area which is much needed in terms of catalyst development. Factors in development include understanding and development of the support as it relates to durability.
- The goal is to develop durable supports, which is critical to the DOE goals. It is difficult to determine whether their chosen supports are providing any real value, because of their inadequate electrochemical and physical characterization of the samples before and after durability testing.
- This project has been well focused on the critical issue of catalyst support durability which is necessary to meet DOE objectives and for commercial success of fuel cells.
- The DOE Hydrogen Program seeks to develop fuel cells that can last over 5,000 hours in real world automotive cycling. Historically, one of the barriers to durability has been the corrosion of the catalyst support, which is typically a carbon black (Vulcan, Ketjen black, or a more state-of-the-art graphitized carbon). This project is relevant because it is studying how to replace these carbons with a more stable oxide, or how to protect the high surface area carbon with a stable oxide or carbide.
- The use of carbon in a lot of the project threatens to reduce relevance, but, at the moment, the community lacks definitive public data that would dismiss all carbons as candidate catalyst supports.
- Elimination of carbon substrate corrosion is an important objective for the polymer electrolyte membrane (PEM) fuel cell system to decrease the degradation of the fuel cell performance.

Question 2: Approach to performing the research and development

This project was rated 2.9 on its approach.

- Although the role of support in a catalyst is secondary, selecting a good support can improve the catalyst's stability and activity. The selection of the systems seems primarily based on considering stability, rather than optimizing catalyst-support interaction.
- The approach is very good. Care should be taken on material selection. A prohibitively expensive catalyst support, while interesting, may be of little value.

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- The choice of new classes of alternative supports is very much in line with the literature and there are no surprises. A lot has been studied on these systems already. The innovative concept appears to be to use these inorganic oxides, nitrides, and carbides to protect a graphitized carbon surface.
- The project has a good approach covering major areas of supports--carbon and non-carbon. However, I wonder how relevant the benchmark Vulcan XC72 is as I thought the automotive companies have moved to more graphitic acetylene black standards.
- The impact of support on the structure of the electrode layer in the membrane electrode assembly (MEA), such as porosity and hydrophobicity, is not clear from the approach as described.
- The approach is systematic and the choice of supports is adequate; however, the lack of characterization is a serious deficiency. The synthetic and heating procedures should be adequately described, as they significantly impact catalyst stability. The use of theory is good, but is perhaps premature.
- The PIs overlook the effect of metal-support interactions and carbon wettability in their results.
- There is a good partnership with the right balance of theory and analysis.
- Given the instability of carbon, it can be said this project is looking for incremental gains instead of breakthroughs by focusing so much of the work on carbons. Thankfully, there is some work on oxides.
- While activity and electrochemical surface area (ECSA) results are reported, there has not been enough supporting data from microscopy to explain the results. Reviewers are left to wonder what the particle-size influence on reported activities might be.
- The stability calculations are interesting, but it remains to be seen whether pH and voltage were accounted for, similar to what would be expected in developing a Pourbaix diagram.
- The reporting of mass activity and ECSA is done for Pt/GMC (graphitic mesoporous carbon), but this needs to continue for other catalysts.
- The durability cycle seeks to eliminate convolution with the Pt dissolution/oxidation mechanism, which is fair. However, the stability of the catalysts at lower potentials, which do arise in realistic automotive operation, is still necessary to observe. The FreedomCAR (Cooperative Automotive Research) and Fuel Partnership's Fuel Cell Technical Team (FCTT) has generally avoided specifying accelerated stress tests beyond those that are obviously imperative for Pt and C. However, with alternative chemistries that involve other metals and poor metals (e.g., Ti, In, and Sn), that might have corrosive potential regions at low pH in between immunity and passivation regions, the investigators should take it upon themselves to learn the limits of stability.
- The approach is reasonable, but the comparison to Vulcan XC72 is questionable for a stable substrate. The comparison should be to graphitized XC72 (Vulcite), which is more stable than Vulcan XC72, and Vulcite has been used commercially in fuel cells.
- Testing at 1.3 V or 1.4 V is a severe test. I did not see a correlation of this "accelerated test" with actual test results.
- I did not see mass activity or specific activity in plots for Pt/WC. Is the platinum coverage is the same for Pt/C and Pt/WC-C?

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.8 based on accomplishments.

- The stability of Pt-based catalysts is apparently improved with several substrates investigated, but, occasionally, only marginal improvements were found.
- The activity of new catalysts is quite modest and Pt mass activity for the best samples is lower than that of Pt/C by E-TEK.
- The accomplishments to date are very promising.
- This project has achieved progress toward its milestones, although they were not too difficult to start with.
- The mass activities at 900 mV on slide 15 seem to be quite small compared to state-of-the-art materials (45 A/g_{Pt} or 0.045 A/mg versus the target of 0.44 A/mg).
- The percentage of retained ECSA should be compared to state-of-the-art Pt/C catalysts sold by Tanaka Kikinzoku Kogyo (TKK) for example, not just the old Pt/VC.
- The concept of stabilizing graphene with indium tin oxide (ITO) modification is very interesting and potentially useful. There were good results and work done to optimize it. Fundamental studies of why it works were also very good.

- ITO-mediated graphene is a good facilitator of catalytic site dispersion on the support, and there is good analysis to show why.
- The project has shown an increase in performance (activity and durability), but how does this improvement hold up to DOE final goals?
- The project is difficult to evaluate given its poor characterization.
- The density functional theory (DFT) model has predicted stable Pt/ITO/graphene with low activity. It is unclear whether the team has independently determined the conductivity obtained in their method of ITO deposition and optimization. There may be room for much improvement here.
- It is not clear why the team reported rotating disk electrode (RDE) with sulfuric acid. Perchloric acid would be better used alone (slide 12 and supplemental slides).
- The team could attempt going below 0.85 V to see how the various classes of supports fare.
- For the cycle used, a 2x durability improvement over Vulcan was demonstrated for GMC and ITO/Vulcan. However, it is unlikely that a 2x durability improvement will be sufficient for commercialized fuel cell vehicles.
- The investigators make a good point during their discussion of the Pt/carbon nanotube (CNT) work when they mention that MEA durability is different than RDE durability. Because RDE durability can be convoluted with adhesion issues, MEA durability should be used for reported measurements.
- No reporting was given for catalyst layer thicknesses.
- Microscopy data are needed.
- Good progress has been made providing 3x stability with ITO at an initial ECSA of $40 \text{ m}^2/\text{g}$.
- All RDE measurements reported were in sulfuric acid. Supplemental slides show that perchloric acid RDE was done. Because of sulfuric acid poisoning of Pt(111), sulfuric acid data should be thrown out, and the perchloric acid data should be those that are reported.
- Oxygen reduction reaction (ORR) mass activities are all below those of conventional Pt/C. Of course, the objective of this project is durability, not activity, but it is fair to comment that the supports studied have not contributed a catalyst-support interaction effect on activity.
- Good progress was made, but some questions remain. The PIs claim no degradation of TiO₂, but the V-I curve shows a loss in performance due to the addition of TiO₂ and it is not clear if there is an advantage. The PIs claim the CNT is "very stable," and this does not appear to be correct. CNT is more stable than XC72, which is a proper claim.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **2.6** for technology transfer and collaboration.

- There are a number of experts in the group of collaborators.
- The collaboration with industry partners is lacking. Collaboration in this area might yield additional materials to consider, political and technical support, and a path to commercialization.
- There were no specific slides on the coordination or functionality of the collaborators beyond the overview slide. Based on that, it appears most of the work was done by the Pacific Northwest National Laboratory.
- More industrial collaborators are needed. The Automotive Fuel Cell Cooperation (AFCC) is only a consultant, while a catalyst company or experienced partner such as General Motors may make a difference in the program.
- The PIs could do a better job getting input from industry on relevant supports by reaching out to the DOE community for help with characterization.
- Good guidance of experimental work by theory.
- DFT predicts good ORR and stability for Pt-VC(111), yet this is not borne out in testing. No reason was given for why.
- The project has been light on reporting fuel cell testing this year (with the exception of some ECSA for Pt/CNT and Pt/ITO/graphene catalysts), so the guidance from AFCC has probably not been extensively used.
- In general, the PI needs to take better advantage of collaboration or it needs to provide more explicit reporting of collaborations. Perhaps ORNL could be leveraged for more microscopy results.
- The partner list looks good; however, it is unclear which contributions came from each of the partners.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- The focus of future work should be selecting the most active, simple, and stable systems.
- The future work plans are very good.
- Focusing on the ITO-treated graphene system would seem to be warranted given its novelty and use as a model for a new type of alternative support.
- More work on how the ITO contributes to stability is needed. It is unclear whether ITO is a facilitator or a direct contributor to durability.
- The PI should plan for ink development, with respect to compatibility, to make key porosity and hydrophobicity targets for the electrode layer in the MEA.
- I think more work with conductive metal oxide-modified CNTs would be important.
- Given the greater durability of CNT and GMC, it is surprising that ITO and WC have not yet been supported on these carbons. Thankfully, this is identified in the future work.
- MEA tests on Pt/GMC and Pt/ITO/graphene are worthwhile to see if the durability is actually greater than two times. MEA tests, of course, are better than RDE for this measurement.
- Further work to improve non-carbon materials (such as Pt/ITO) is good.
- More emphasis needed on microscopy and catalyst-layer thickness. Future work should also include cycling at lower potentials. So long as MEA mass activities are reported, there will be expectations that hydrogen crossover will be verified to be low.
- "Study carbon degradation under more severe and representative conditions" is stated in future work plans, but it is not clear why these conditions are not representative. The exact solubility of metal oxides in water was never touched upon, and neither was the effect it would have on the stability of the membrane. It is hard to understand why 1.4 V was so representative. There don't seem to be many instances of a fuel cell reaching 1.4 V. It is also unclear whether the mechanism for carbon corrosion is the same at 1.4 V as it is at 0.7 V.

Strengths and weaknesses

Strengths

- The project has a good team and good facilities.
- The project has well-defined concept and material selections.
- The project seems to have a good rate of productivity.
- There is a good connection between theory and practice.
- The project team has shown great productivity and advances.
- High durability catalysts used in this project are of importance to DOE and the fuel cell community.
- There is good synergism between theory and experiment. CNTs have shown good stability compared to the control.
- The project has made progress with a more stable, higher surface area ITO.
- The PI has been one of the more responsive PIs in regard to technical team and reviewer feedback, as evidenced by doing RDE in perchloric acid, adding non-carbon materials to their plan, and removing some nitrides suspected to be unstable.
- The project includes many needed experiments in its future work plan.
- The program is evaluating important substrate materials such as CNT and graphene.

Weaknesses

- The activity measurements are not easily compared with the data in the literature. In similar research, 0.85 V is generally not used for comparison; it indicates a low activity of the catalysts.
- There is a lack of a path to commercialization.
- The homemade fuel cell testing, questionable ORR activity values, and questionable (low) MEA performances shown may be compromising the ability to accurately compare the effectiveness of the different supports.
- The PIs are underplanning for ink/electrode structure development using these novel materials.
- Electrochemistry is the only characterization tool, and it is used in a non-standardized format, using H_2SO_4 as the electrolyte. Thus, all of the results are unstandardized and difficult to understand.
- No physical characterization tools are used.

- There is no insight to determine whether there are any metal-support interactions which might affect the catalysis, or how the wetting of the different catalysts might affect their performance.
- I would like to see more work done with CNT, which by far showed the best stability results.
- The project approach was conservative at the outset. Instead of looking at non-carbon supports, the project has sought to stabilize carbon-based supports.
- More microscopy is needed to understand whether trends in activity and stability are affected by particle size, surface orientations, and other nanoscale phenomena.
- Beyond Pt/ITO, the project may not have delivered a catalyst material that would be of interest to the end customers of the research. This is partially dependent on how MEA testing proceeds with some of the materials that have been identified through RDE as two times more stable.
- The results were not reported as mass activity or specific activity. This does not permit adequate comparison from sample to sample or with the work of other researchers. There did not appear to be any collaboration with industry.

Specific recommendations and additions or deletions to the work scope

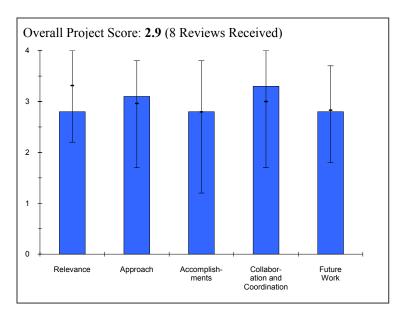
- The PIs should select one to two systems and optimize them.
- Find an industry partner.
- Evaluate the materials for other technologies, like phosphoric acid. PEM fuel cells use a sulfur-based transfer ion at temperatures up to 120°C. Phosphoric acid fuel cells use a phosphorus-based transfer ion at temperatures up to 240°C. Look at the other technologies opens up the potential commercial possibilities.
- Have Los Alamos National Laboratory cross-test the activity and durability of their best candidate materials.
- Obtain state-of-the-art dispersed catalysts from TKK and compare them to that rather than Pt on Vulcan.
- Use remaining time on ink/electrode structure development.
- The project should not continue without redirection to include extensive physical characterization of the catalysts with techniques such as transmission electron microscopy, scanning transmission electron microscopy, Brunauer-Emmett-Teller surface area analysis, chemical analysis (ICP), and/or point of zero charge.
- Researchers should do their electrocatalyst characterization using standardized methods, e.g., 0.1 M HClO₄ References exist in literature and there is significant help within the DOE laboratories to support.
- Test optimized ITO on CNT both in DOE RDE and MEA stability tests.
- Microscopy needs to be added to understand the relationship of nanoscale properties to activity and stability.
- Durability measurements should rely on MEA testing. Reporting of catalyst layer thickness is needed. There should be some reporting that hydrogen crossover was verified to be low.
- Modeling calculations for stability should provide some assurance that results are valid for low pH, high voltage operation.
- Durability cycling should be performed at lower potentials, in addition to cycles being used.
- Reporting procedures should be updated to include mass activity.

Project # FC-04: Non-Platinum Bimetallic Cathode Electrocatalysts

Debbie Myers; Argonne National Laboratory

Brief Summary of Project

The overall project objective is to develop non-platinum cathode electrocatalysts for polymer electrolyte fuel cells to meet DOE targets that: 1) promote the direct fourelectron oxygen reduction reaction with high electrocatalytic activity, 2) are chemically compatible with the acidic electrolyte and are resistant to dissolution, and 3) are low cost (5/kW), 0.3 mg_{PGM}/cm²)). This year's specific objective is to prepare and characterize model systems (bulk alloys) and nanoparticles of Pd-Mo, Pd-Re, Pd-Ta, and Pd-W binaries and Pd-Cu-Mo, Pd-Cu-W, Pd-Cu-Re, Pd-Cu-Ta, and Pd-Cu-Ni ternaries.



<u>**Ouestion 1: Relevance to overall DOE**</u> <u>objectives</u>

This project earned a score of 2.8 for its relevance to DOE objectives.

- While I appreciate the need for alternatives to Pt as a catalyst, I am concerned about the perception that Pd is a low cost material. The precious metals market has shown in the past that the Pd price is volatile at best. This means that, given the poor stability and performance of Pd as a catalyst, this approach may not meet the objectives of the RD&D.
- Searching for more cost-effective Pt alternatives has merit; success could be a game changer in light of performance and durability issues with Pt at low loadings.
- My only concern is that the entire presentation was deep into the science without any obvious connection to practical targets being made. The project addresses cost barriers, so a simple parametric study of material/cost/performance tradeoffs would work wonders to frame the work and the talk, and place the candidates discussed in proper context.
- This project directly supports, in theory the major barriers of electrode performance, durability and cost. It focuses on reducing cost by replacing Pt while achieving the same Pt mass activity targets. The widely known negative impact of excess transition metals dissolved into membranes that reduce fuel cell limiting currents is a counterweight to this objective that does not appear to be factored in. Lower high current density performance can have a greater effect on increasing stack costs (by requiring more cells) than would be lowered by replacing Pt with lower cost noble metals.
- The development of low cost catalysts does address the objectives of the Hydrogen Program. A more detailed analysis of the likelihood of this project meeting the DOE cost targets, if it meets technical targets, should be provided.
- Cost and durability are the most critical gaps for fuel cell commercialization and the use of a platinum group metal (PGM) catalyst is a large fraction of the cost. A non-PGM catalyst approach is expected to lower the catalyst cost. However, current performance of non-PGM catalysts is far from a performance target that could replace the PGM catalyst.
- DOE shows a catalyst target and requires measurement of its performance in the membrane electrode assembly (MEA), regardless of whether it is PGM or non-PGM. This project should follow this target and metric.
- The team needs to enact relevant, basic research to determine whether d-band effects with alloying of Pt are admissible for other metals, such as Pd.
- This project is focused on the DOE R&D plan leading to fuel cell commercialization, focusing on these twin thrusts: reduced dependence on foreign oil (due to reducing overall energy demand by creating more efficient

technology), and environmental concerns. It is safe to say that newer DOE plans will dictate fuel cell technology competitiveness in the global marketplace, and the activity also addresses that!

Question 2: Approach to performing the research and development

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

- The combined theory and analysis is an excellent approach.
- I appreciate the approach to use bulk alloys, as this has been missing for a long time in working with alloys. The progression from this to dispersed catalysts with complimentary calculations is well thought out.
- Strong team competencies were revealed in a sophisticated approach from molecular theoretical calculations to innovative and informed material production methods.
- This project's fundamentals approach is very good for generating basic information on the likelihood of ever engineering multi-material compositions that can effectively simulate a Pt surface.
- The PIs have a very strong team with all the basic elements represented: theory, synthesis, characterization, model system study, and functional testing.
- New Pd alloy materials are being developed. Computational studies, nanoparticle synthesis, and characterization are well focused. Model system work is also a plus. More characterization focused on stability toward oxide formation and dissolution in acid is needed. MEA testing should also be done at this point, as this can help understanding of stability, as well as activity and performance.
- The use of quantum mechanical molecular dynamics is a good idea to help understand oxygen reduction reaction (ORR) catalyst mechanism and materials screening.
- The project team uses a very solid approach to make alloys and characterizes them well.
- The degree of alloying might be better assessed with Reitveld analysis (neutron or high energy X-ray diffraction), although this might be irrelevant as the surface structure is what is most important.
- There does not seem to be any evidence that the bulk alloys will stay alloyed at the nanoscale, and the formation of oxides (for example of molybdenum or tantalum) seems inevitable. If the PIs want to retail alloys at the nanoscale, they will have to introduce a rigorous synthetic approach, and perhaps explore capping the nanoparticles.
- The ANL project appears conventional and is designed to explore the "latest" catalyst, the core-shell, and concept. This direction is necessary and potentially useful.
- The project should continue to focus on bi- and tri-metallic systems that have both the highest ORR activity and show the lowest tendency for surface oxide formation.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.8 based on accomplishments.

- I think that the project as stated is on track. The only concern is that the MEA fabrication and fuel cell testing task is not on track for 2010, yet it does appear to be a deliverable. One wonders if this has been abandoned. It would be the ultimate demonstration and culmination of the project. Perhaps this task should be a focus of the last six months, instead of further fundamental materials research. It would be preferable to be able to do both.
- Extensive calculations, material preparations, and activity tests are good. With that being said, it was not clear why Pd was the pre-eminent specie of choice around which the majority of the work was conducted. Even if this case has been made before, it is critical to educate the audience each time the work is presented, otherwise it begs many questions. This choice would not impress most as a cost-reducing alternative to Pt.
- There has been an excellent amount of fundamental work completed. However, because of its fundamental nature, it is difficult to conclude whether the main DOE barriers will be overcome by this approach. The progress on the Pd₃Mo bulk alloy is promising, but when compared to PtCo or PtNi state-of-the-art catalysts, it shows there is a long way to go yet to equal Pt. The nanoparticle systems based on Pd also indicate the very large parameter space associated with synthesis and the low ORR activity of these best-to-date prepared systems.

- It would be useful to know if any of the Pd-TM alloys studied were perhaps explored decades ago for ORR by more empirical approaches, and if there was any evidence of any activity approaching Pt. These do not seem to be unusual materials to combine.
- The ultimate stability of a Pd skin on a non-noble metal core is also a critical assumption in this approach and could perhaps be validated by alternative types of tests.
- New alloys show that progress can be made. Computational models are providing insight into the structure and composition activity characteristics.
- It is interesting to see an optimized d-band center for ORR activity. It seems to be similar to Pt.
- Progress toward DOE targets for fuel cell catalysts is only fair because this is really a basic research project and the system cannot meet DOE targets. If this project had been an Office of Basic Energy Sciences (BES) project, their progress would have been outstanding.
- Progress is evident and the project (80% completed) is moving forward. The PI should explore techniques for reducing surface oxides on bi- and tri-metallic nanoparticles. This approach will improve ORR results, which should be reported at 0.9 V.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.3** for technology transfer and collaboration.

- There are no industrial partners and this means that the project is research-based, rather than delivering to the program. Los Alamos National Laboratory (LANL) has had no input at all, as no MEA fabrication has taken place, nor is it planned before the end of the project.
- The impression is that a lot of work was done with the California Institute of Technology and ANL. A limited contribution from the University of Nevada, Las Vegas and the University of California, Irvine is also evidenced. The role of the Oak Ridge National Laboratory and LANL may be reflected in the content, but it is not apparent, given their central role in studies of this kind. Their contributions and perspectives should have been better highlighted.
- These are very strong team members and their organizational contributions are well utilized.
- The project has an excellent team for computational, electrochemical and in situ (MEA) evaluation.
- The collaboration is working. It is too early to include industry partners.
- There is good work being done with theorists.
- It is unclear why the project has not been collaborating with another ANL catalyst group (Markovic, et al.).
- ANL has teamed with competent and supportive organizations. The tasks seem well partitioned.
- They have excellent collaboration with their partners, who appear fully engaged.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- The proposed future work seems to be to do more and more research without ever demonstrating that it is worth doing. The demonstration in an MEA is not proposed before the termination of the project. A key deliverable is being dropped. The assumption seems to be that this research may continue indefinitely without a hard deliverable, and perhaps that is the case, but this project must deliver.
- The project should focus on model systems. The evolution of certain analytical and preparation techniques is enduring; however, it is not clear the specific extent to which the team anticipates progress toward technical targets -- this should be quantified.
- In general, the proposed future work extends the most promising routes identified to date. However, it focuses on the surface compositions and less on the surface structure. They may not have identified the key factor for effective ORR by assuming it is related to just d-band center or valence band width or segregation energies. Consideration of surface defects or ideal surface structure seems to be less appreciated.
- There doesn't appear to be much reason to expect that ternary systems will be better than the binary systems previously studied.
- Testing in MEAs should begin.
- Materials stability is not well defined.

- There is no clear path to forming alloyed nanoparticles. It will be a significant accomplishment if they can make alloyed nanoparticles with Ta, Mo, etc., with no oxides.
- The project is about completed and has produced new literature on structured alloy catalysts. These results support the thesis that this direction is worthwhile.
- Since only Pd₃Ir and Pd₃Os are segregating in the presence of absorbed oxygen, possibly add iridium and/or oxygen to your tri-metallic future work with your alloys showing the best ORR.
- A large increase in ORR is seen in PdRe with only a small amount (95:5) of rhenium added. I wonder if more could be done with this system.

Strengths and weaknesses

Strengths

- Very good fundamental science is used. The approach is sound and the project is well organized.
- The PI and certain team members have experience with the spectrum of work, bridging theory and experimentation.
- The expertise of the respective team members and access to facilities is good.
- The new alloys show that progress can be made. Computational models are providing insight into structure and composition activity characteristics.
- The project is well coordinated between theoretical modeling approaches and empirical validations.
- The project builds upon basic research and understanding of how d-band effects affect ORR.
- The PI is competent and an excellent manager. The integration of theory and fabrication is well done.
- Great computational work has been done. The concept of the project is sound and true advances in ORR over palladium have been reported.

Weaknesses

- The project does not seem to be reaching any conclusion in its closing stages.
- There is little connection to practical technical targets. The project seems to be on the verge of becoming a BES project.
- One weakness is associated with the basic premise for this project, and a lack of a larger appreciation for the economics of the proposed approach. It should be possible to state for given Pt/Pd cost ratios, for example, how much performance decrease with a Pt-free system can be tolerated to compensate for the gain in cost reduction brought by the Pd. That is, from the Directed Technologies, Inc. or TIAX system cost breakdowns and estimates of stack cost per cell, how many additional cells would it take to eat up the cost savings by replacing the Pt with Pd? This would give a minimum performance target on the one hand, that may be less aggressive than with Pt, at least to indicate how far the materials are from a practical standpoint.
- A more detailed analysis of the likelihood of this program meeting the DOE cost targets, if it meets technical targets, should be provided.
- This should not really be an EERE project.
- The transition from bulk alloy studies to nanoparticles was not made clear.
- The core-shell concept has been around for decades: alloy surface modifications have been observed and described. The real issue is durability addressing the utility of these formulations. The project needs to be focused on significant testing of corrosion on these catalysts, which necessarily involves a highly focused testing activity. ANL has shown good ability to do this and should do this, because the tests need to be inclusive. The source of all materials must be known, and the fabrication steps must be done carefully and reproducibly. Then tests need to be completed with sufficient replication to get reliable results. Critical postmortem analyses, including "tear-down" investigations on samples that are still performing well, need to be done, with continuous sampling throughout the testing period. The effects of the testing regimen, say the voltage scan, on the rate and mechanisms of degradation must be documented.
- I see no obvious weaknesses other than possibly becoming spread too thin and evaluating too many tri-metallics in the time remaining.

Specific recommendations and additions or deletions to the work scope

• I recommend that the project take the best that it has in regard to a dispersed catalyst and demonstrate the performance and durability in an MEA before the project ends.

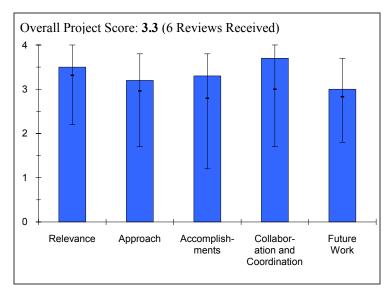
- They need simple analyses framing the targets and progress toward them. These analyses should include materials, costs, performance, and tradeoff calculations.
- The project should justify why they expect ternary materials to perform better than binary materials.
- The project should address the stability of Pd-transition metal alloys in acid media.
- The project should add considerations for how surface structure might affect ORR activity of the materials, not just composition.
- As mentioned, the end target of this project should be consistent with DOE metrics.
- This project should now focus on durability testing.
- I would suggest focusing on techniques that could reduce formation of the surface oxides from the base metals and then adding a few that show promise for ORR.

Project # FC-05: Advanced Cathode Catalysts

Piotr Zelenay; Los Alamos National Laboratory

Brief Summary of Project

The overall project objective is to develop oxygen reduction reaction (ORR) catalysts as alternatives to pure platinum and electrode structures suitable for new catalysts that together are capable of fulfilling cost, performance, and durability requirements established by DOE for the polymer electrolyte fuel cell cathode and to assure a path to large-scale fabrication of successful compositions. Project impact in past year has been to: 1) significantly advance the knowledge of factors affecting ORR activity of the platinum monolayer (facet, shape, particle size), and to achieve specific activity and mass activity targets for catalysts with ultra-low platinum content, 2) develop a non-platinum group



metal (non-PGM) catalyst with combined activity and durability better than shown in any prior reports, and, after mass-transport correction, meet the DOE 2010 target, 3) demonstrate an active non-PGM catalyst with high durability to voltage cycling, and 4) scale-up the synthesis of selected core-shell catalyst to 20 g per batch.

Question 1: Relevance to overall DOE objectives

This project earned a score of 3.5 for its relevance to DOE objectives.

- The objectives of this project align well with the objectives of the DOE research, development and demonstration (RD&D).
- This program is very relevant to DOE R&D objectives.
- This project is of high relevance for the development of PGM and non-PGM catalysts for polymer electrolyte membrane (PEM) fuel cell systems and fully supports the goals and objectives of the DOE Hydrogen Program. The PIs developed a number of core-shell PGM catalysts that significantly reduced Pt content, while not sacrificing the required catalytic activity and stability. The PIs also accomplished industrial scale-up of the first generation of core-shell catalysts. Even more importantly, the PIs demonstrated that activity and stability of non-PGM catalysts are approaching the DOE targets.
- This is a highly relevant project. The electrodes studied were all very interesting and showed promise.
- This project fully supports DOE RD&D objectives.
- The project is very relevant to the Hydrogen Program objectives, targeting catalyst durability, cost, and performance.

Question 2: Approach to performing the research and development

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

- This project is massive (\$10 million) and is made up of two seemingly unrelated parts--one with Pt core-shell catalysts and the other with non-PGM catalysts. It is unclear to me why these two catalysts are included together in one project, especially when the research takes place in National Laboratories that are not close.
- This project has an excellent approach, targeting both non-PGM and low PGM catalysts. This approach is very ambitious and increases the probability of success.
- The project's approach is unique, combining many important synthetic and characterization methods for unraveling the most active and stable sites for the ORR. By utilizing this approach, many milestones are fully or

partially completed. Go/no-go decisions are carefully chosen, in particular for the non-PGM catalysts and the methods for peroxide detection in real systems.

- The approach was very good and well thought-out. The PIs were effective at meeting their milestones and goals, as well as leveraging the technical strengths of the various group members.
- This team has been sharply focused on the technical barriers and has done an excellent job in overcoming them to meet the DOE milestones.
- The project has obtained very good results for low Pt loading and non-PGM based catalysts. The agreement between rotating disk electrode (RDE) and membrane electrode assembly (MEA) results is quite good. The hollow nanoparticles approach appears to be quite novel. The increase in high current density results for the PANI-Fe-C catalyst, while quite encouraging, needs further analysis to demonstrate what is going on.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.3** based on accomplishments.

- Good progress has been made and some elegant science has taken place on this project. I am concerned that the use of Ir and Pd in the core-shell catalysts is not low-cost as the precious metals market is so volatile. The current price of these metals is not a good guide as to the ultimate cost. Therefore, claims to have met the DOE cost targets, while accurate to the letter, are perhaps not on a path to enable the commercialization of fuel cells in the 2015 timeframe.
- The project has good, interesting experimental results.
- The project has good durability results.
- An understanding of how these alloys work is being developed.
- Technical accomplishments for both PGM and non-PGM catalyst developments are relatively high. For the former systems, the highest activity is observed for the Pt shell-Pd/Ir core, with an improvement that exceeds almost three times the DOE target of 0.44 A/mg_{Pt}. Although the mass activity of such catalysts is significant, the stability issues are not addressed adequately. The PIs should focus more on how to stabilize the Pt shell. The same issue applies for the non-PGM catalysts. It is very important that industrial scale-up of the core-shell catalysts is accomplished.
- The group as a whole showed excellent progress toward their goals, specifically by improving the mass activity of the core-shell catalysts and the stability of the PGM nanoparticles.
- I am a little surprised at the stability of the gold in the gold displacement method, as I wonder whether some of the gold would ultimately end up on the surface.
- The exact surface area of the hollow platinum spheres and whether or not one can access the center of the sphere were questions that did not get answered.
- We have also seen no performance loss in non-PGM iron-based catalysts, even with the loss of significant amounts of the iron. It has been speculated that the iron is necessary to form the site but is not needed after the site is formed. It is conventional wisdom that the lost iron would be detrimental to the integrity of the membrane, but I have not seen it.
- It has not been shown that the hydrophobicity changes as a function of voltage, and whether or not that is the reason for the variation of flooding with voltage for the PANI-Fe/EDA-Co-C. To me, it seems that the flooding should have been more severe at 0.4 V.
- It is not clear why sulfuric acid was used as an electrolyte in the PANI-Fe/EDA-Co-C RDE, instead of perchloric acid.
- The project has made good progress toward achieving high ORR activity for core-shell, hollow nanoparticles and non-PGM catalysts. Scale-up has been achieved for core-shell based catalysts. Non-PGM catalyst results look very good. The Br-Br adsorption/desorption approach to improve performance appears to work well. The significant improvement in mass activity for Pt₈₀Ir₃₀/Pd/C is clearly evident from the zoomed plot in slide 7. The progress shown in slide 23 for ORR activity of non-PGM catalysts is very impressive.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.7** for technology transfer and collaboration.

- This project seeks to provide funding for a wide swath of national laboratories and achieves this end. The involvement of industrial partners is minimal, and this may affect the scope of the project.
- There is excellent collaboration with universities and other national laboratories. Collaboration with Cabot should also be very important.
- This project is multifaceted, and as such requires a close collaboration between the PIs. Eight partner organizations with highly complementary skills are put together in catalyst development, electrode-structure design, materials characterization, and catalyst fabrication. The degree of collaboration is indeed outstanding and certainly a big part of the success of the entire project.
- The group did an excellent job leveraging the individual strengths of the various group members.
- There has been excellent collaboration with the partners over the duration of the project.
- The project is well coordinated, with the team members' roles clearly indicated. The assembled team is very strong, with each team member a leader in PEM fuel cell development.

Question 5: Approach to and relevance of proposed future research

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

- No details are given as to how the proposed work is to be carried out. For example, there is a proposal to "reduce mass-transport resistance in non-PGM cathode" with no indication of how this is to be carried out. This task is not trivial and I would like to know the approach.
- There is a good plan for future research. Understanding stability and/or durability aspects of non-PGM catalysts is important.
- Future research focusing on scale-up of low PGM is important.
- The project clearly builds on past progress and is sharply focused on barriers.
- The project seems to have met most of its goals and is starting to wind down. It is difficult to address future work.
- Future work could include modeling using density functional theory to help demonstrate Pt-O interactions on Pt(ML)/PdIr/C and give direction to other possible metals to evaluate.
- There is a very good outline for future work. Durability issues of non-PGM catalysts have been addressed. Scale-up of these catalysts has also been proposed. The durability of non-PGM catalysts would be a point of high interest.

Strengths and weaknesses

Strengths

- The science is very good at a fundamental level and great progress has been made. This is a very large project and appears to have been managed well.
- An understanding of how these alloys work is being developed.
- The major success of this project is based on the fact that the PIs are developing real systems based on fundamental knowledge of the ORR and enacting novel synthesizing approaches for creating the most active sites.
- Overall, the group had a very good plan and did an excellent job executing it. The PIs worked on very interesting and relevant materials and made significant progress with the materials.
- Excellent results were obtained with the non-PGM catalysts, especially considering the fact that the actual reaction site is still not known.
- The PIs had very good collaboration with their partners.
- There were some very good performance results, especially for non-PGM catalysts.
- The durability results look promising.

Weaknesses

• There needs to be more focus on the *in situ* durability of these catalyst approaches. It is not helpful to make a high performing catalyst that cannot deliver that high performance for the lifetime of the product.

- The weak point of the project is that stability of the catalysts is not fully addressed, as in the case of activity. The PIs should develop a new strategy for handling stability of active sites. This is the key requirement for the development of non-PGM catalysts as well as for preserving high activity of a monolayer Pt shell.
- I see no weaknesses other than possibly optimizing the non-PGM catalyst to an optimized support.
- The project lacks durability information for non-PGM catalysts. It is not clear how PANI-Fe-C catalysts would hold up at 1.2 V.

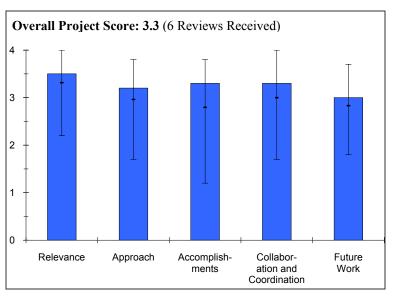
Specific recommendations and additions or deletions to the work scope

- I recommend that this project be split into two separate projects: one PGM-based and the other non-PGM based. The smaller projects would be easier to manage.
- A cycle between 0.6-1.2 V vs. NHE to determine the durability of catalysts using a MWNT substrate (which is known to be stable) should be used (3M-FC-006 adopted this approach). For ultra-low Pt content catalysts, slide 12 shows a 0.05 mg/cm² anode. It would be preferable to use higher anode loading so that the anode is not limiting performance.

Project # FC-06: 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 (>1.2 V) during the transient periods of startup/shutdown and fuel starvation, by favoring the oxidation of water over the dissolution of platinum and carbon. Such catalysts are required to make it possible for the fuel stacks to satisfy the current 2010 and 2015 DOE targets for performance and durability.



<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of 3.5 for its relevance to DOE objectives.

- Fuel starvation is a real concern in automotive fuel cell systems. However, there are other known ways to mitigate fuel starvation via system controls and corrosion resistant supports.
- The project is aligned with DOE Hydrogen Program goals and objectives. It addresses key issues of fuel cell durability.
- This is a very good project that addresses a key area in terms of durability.
- This project has well -performing fuel cells that address a technology relevant to DOE objectives. This program is well aligned with those goals. Obviously, catalyst durability is a key issue, and this work addresses those concerns.
- This project aligns with DOE R&D objectives. It focuses on durable catalysts for fuel cells during transient conditions.
- This project is well aligned with DOE objectives, addressing major cathode durability issue. While the odds for success are not high, a possible payout is significant.

Question 2: Approach to performing the research and development

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

- The project has a fundamental approach of modifying both the anode and the cathode catalysts to favor the oxidation of water over carbon corrosion without compromising performance that is solid. I'd like to see the scope expanded to materials that would most benefit from this type of mitigation, such as dispersed carbon catalysts.
- The project is sharply focused on improving durability during start/stop cycles, which is one of the key issues concerning fuel cells for automotive use.
- The project takes a robust approach by looking at two independent methods to alleviate degradation from fuelstarved regions during startup and shutdown (enhance oxygen evolution reaction (OER) at the cathode; decrease/inhibit oxygen reduction reaction (ORR) activity at the anode). Both methods will reduce corrosion.
- The project's approach is excellent and the hypothesis is sound.
- The philosophy here is obtuse. The question now should be "what is necessary to assure contemporary fuel cells survive with acceptable durability," rather than "what is necessary to make contemporary fuel cells 'bullet proof', no matter how they are abused." It is probably worth thinking along these lines. However, the early

markets are being established now, and other developers have solid, durable products. Indeed, there may be a variety of modern membrane electrode assembly (MEA) designs that prove useful.

- The idea of using an OER catalyst at the cathode to compete with and inhibit carbon corrosion is novel. The idea to incorporate an ORR inhibitor at the anode is also novel. This approach addressed the technical barrier of catalyst durability under startup and shutdown conditions. The scale-up is being done by 3M.
- The success of the proposed cathode approach appears contingent upon solving the conundrum of catalysts for "reversible" fuel cell/electrolyzer systems. It is not obvious what makes this effort more likely to succeed than previous attempts aimed at developing dual-function ORR/OER catalysts for such systems.
- While a "discrete-nanoparticle" approach is, in principle, attractive, ruthenium migration from the OER component to the ORR one, with ensuing formation of a platinum-ruthenium alloy with poor ORR activity, is very likely. It is not clear how such a migration can be prevented, especially over long operating times of the fuel cell.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.3 based on accomplishments.

- In a short time they have already identified several candidates for both anode and cathode catalysts.
- The project has been active for less than a year.
- The project has identified trends in ruthenium-iridium catalysts for OER, and has achieved an initial milestone of 1mA/cm₂ at 1.45 V and 10 mA/cm₂ at 1.5V.
- Results were found that platinum tantalum decreases ORR on the anode by an order of magnitude with no losses in anode performance.
- There has been a limited project performance period thus far, but the work that was reported is very good.
- The approach is thoughtful and novel. The challenge is to reformulate the anode catalysts without impacting hydrogen oxidation. Certainly the amended anodes will react differently to anode impurities, and will complicate fuel quality issues. This all seems to be leading away from commercialization, no matter how interesting it will be.
- Significant progress has been made toward identification and testing of an ORR-suppressing catalyst anode (tantalum based). They have also identified a ruthenium-based catalyst for OER increase at the cathode. Cycling to 1.25V showed little changes; however, at 1.45V, ruthenium starts to disappear, giving rise to platinum behavior. Since the cathode may be exposed to voltages as high as 1.45V during startup and shutdown, this may be a concern. The MEA results for OER catalysts were given, but no MEA results for ORR inhibition catalysts were made available.
- Good progress has been demonstrated to date in simple voltammetric experiments.
- Ruthenium is a questionable choice due to its many stability issues. A promising replacement for ruthenium at the cathode needs to be identified, and both of these points should be raised by the PI at the next AMR.
- Most performance targets appear to have been reached (but only in cells operating for a limited time).

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.3** for technology transfer and collaboration.

- There is already evidence of valuable contributions from both the Oak Ridge National Laboratory and Dalhousie.
- There is close collaboration between partners. These collaborations worked well in the previous program and appear to be working well here.
- Collaboration with electrolyzer developers may be useful in screening and/or development of OER catalysts.
- The project has a very good team in place.
- 3M has people and facilities as good as any.
- The tasks among the team members are well defined.
- The project seems well integrated among the participants. No collaboration with other institutions has been demonstrated to date.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- Future work should include proof that mitigation strategy works under real start/stop conditions and with dispersed carbon catalysts.
- Using 3M nanostructured thin film (NSTF) will allow the researchers to isolate the reactions of interest, while minimizing carbon corrosion. This approach should be even more applicable to improving durability of dispersed catalysts on carbon. Some tests with platinum or carbon would be useful to show how much this approach could help there. Measurements of carbon corrosion in both instances (by carbon dioxide evolution measurements) would be beneficial (microporous layer (MPL) or gas diffusion layer corrosion in NSTF case).
- Future work should include investigating whether there is iridium, ruthenium, or titanium plating on the anode.
- Future collaboration with electrolyzer developers may be useful in screening OER catalysts.
- The work proposed is excellent; however, it would be great if focused efforts were set up to ensure that the added catalyst materials were stable under all operating potentials.
- There needs to be a clear back-up plan, because the described attack seems to involve high risk. Because others seem to be achieving useful durability with their MEAs, 3M needs to sort through its procedures. It is possible that the durability testing protocols are too severe, and that maintaining the MEAs within same potential limits might greatly increase durability. Achieving durability now is very important.
- The future work is well planned and laid out. More details are needed for MEA testing of ORR reduction catalysts at the anode.
- More advanced kinetic studies beyond cyclic voltammetry are needed.
- Catalyst stability is missing in future research.

Strengths and weaknesses

Strengths

- They have exhibited a sound fundamental approach. The targets being set are based on electrochemical fundamentals. There is a nice combination of *in situ* and *ex situ* experimental techniques.
- 3M's experience with durable MEAs and novel approach to limiting start/stop degradation, are good contributions to the project
- The project has a sound hypothesis, an excellent team, and very good preliminary results.
- The people on the project team are impressive.
- The project uses novel ideas for OER enhancement at the cathode and for ORR inhibition at the anode.
- The MEA results have been good for OER catalysts at the cathode.
- Solving a system challenge through electrocatalysis is very attractive and represents a major strength of this project.

Weaknesses

- A weak point involves the fact that this team's strategy is only being applied to electrode design that is already robust to potential excursions. Not exactly a weakness; however, it did not come across that stability of added materials will be examined under all potential ranges (if the PIs intend to do this, then this comment can be ignored).
- The problem is to kill the anode without killing the anode, so to speak. Certainly the explanation was acceptable, and may be correct. However, there is danger along this path: if you make the anode even more susceptible to low level contaminants, the progress may turn to be negative.
- No MEA performance for ORR inhibitor at anode has been provided. (the project is young).
- RuO₂ almost disappears at 1.45V.
- This project is not fundamental enough in its approach.

Specific recommendations and additions or deletions to the work scope

- There needs to be validation on dispersed carbon electrodes.
- Hydrogen peroxide formation results should be reported for these catalysts.

- The DOE accelerated test protocol (full voltage cycle test) needs to be performed on materials that have been shown to be useful, but also include lower voltages (e.g., 0.1-0.65 V) to test stability of the added metals.
- Consider measuring carbon dioxide on at least some of the experiments; e.g., during high potential and simulated start/stop testing (there is carbon in the MPL).
- Look at applying concepts to dispersed catalysts on carbon supports.
- There seems to be various reactor engineering solutions that would solve this concern in rather simple ways. Certainly, 3M can figure ways to keep oxygen out of the fuel cell anodes. It is possible that durability will turn out to be a fuel cell stack issue, not really addressed by the MEA alone. 3M is talented, and needs to use its chemical engineering resources to address "durability" broadly, including reactor design; i.e., stack design. Like any chemical fixed bed reactor, the catalyst and the reactor engineering are necessarily merged.
- The team should continue this direction.
- In principle, this project represents an effort in fundamental electrocatalysis and, as such, it would benefit from more thorough characterization using both electrochemical (other than voltammetry) and non-electrochemical tools.
- Poor durability of several proposed anode and cathode electrocatalysts needs to be addressed as soon as possible and alternative solutions should be proposed.
- More challenging second-year milestones should be proposed. They ought to combine desired OER (cathode) and ORR (anode) activity with stability.

Project # FC-07: Extended, Continuous Pt Nanostructures in Thick, Dispersed Electrodes *Brvan 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. 3M's (and others) demonstrated improvements 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.

<u>Ouestion 1: Relevance to overall DOE</u> <u>objectives</u>

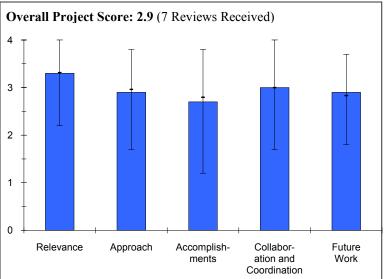
This project earned a score of **3.3** for its relevance to DOE objectives.

- The National Renewable Energy
 Laboratory (NREL) is directly addressing a key DOE target--catalyst/electrode structure and performance.
- The project is well aligned with the DOE research, development and demonstration objectives.
- Alternative pathways for cost reduction are imperative.
- The project proposes to address two critical barriers--activity and cost of electrocatalysts. It will probably be forced to address durability as well.
- It is not clear how the alternative approaches considered will reduce cost as there is no consideration at this early stage for manufacturing capabilities for many of the alternative, extended-surface catalyst supports.
- Cost and durability are the most critical gaps for fuel cell commercialization and the catalyst is important for both attributes. It is good to look at bulk characteristics, which would be a breakthrough from the current state-of-the-art dispersed platinum on high surface area carbon technology.
- It is worthwhile to look at the impact on morphology and "novel" structures on catalyst activity within the platinum/carbon system.
- The project includes using different methods for making the catalysts.
- The cost of implementing the new catalysts is not clear, as the synthetic methods could be expensive, but perhaps this problem could be addressed later if they find a particular structure with exceptional attributes.

Question 2: Approach to performing the research and development

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

- The project's several different approaches seem to be modifications and/or enhancements of approaches taken previously by other researchers. The emphasis seems to be on alternative ways to prepare catalyst layers or electrodes.
- There is a strong modeling component to the project.
- The project has a good approach. I am a little concerned that there are too many avenues for exploration, but it is early in the project and I expect that some downselection will take place during the course of the project.
- Working with known material, and interrogating roles of substructures and morphology, is an exciting innovation. The team has a bold approach with regard to fabrication methods that is applauded.
- Some of the assumptions inherent in the approaches cited may need to be revised based on acquiring a deeper understanding of the 3M nano-structured thin film (NSTF) catalysts used as a model and baseline.
- It is not clear exactly what the electrode studies will entail, that is different from much of what is already in the literature, in order to pursue extended-surface catalysts; e.g. nanowires, carbon nanotubes (CNTs), and vertically aligned carbon nanotubes (VACNTs).



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- A clear strategy is not seen and activities are kind of exploratory if CNTs and the like will work.
- It is necessary to dig in bulk characteristics. This project is looking at atomic layer deposition. Additionally, crystallite orientation and interaction with substrate materials should be considered.
- Materials stability is important for durability and it is necessary to develop its metric for this oxygen reduction reaction (ORR) catalyst concept.
- As for the modeling approach, it would be good to add quantum mechanism molecular dynamics to understand the ORR activity enhancement mechanism for this catalyst concept.
- This project includes using a variety of synthetic tools to make new platinum structures.
- The overall characterization of the project is good.
- There are questions about whether the catalysts can be heated to the high temperatures needed to order platinum and impart stability.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.7 based on accomplishments.

- The project is less than a year old, so progress is limited but meaningful.
- Some approaches have already been downplayed.
- The initial results are encouraging.
- These are early days for the project, but the team is off to an excellent start. Clearly, much work has been done to climb the learning curve of substrate materials and novel material production methods.
- This project is very early in its beginning. It is too early to judge, but there has definitely been some progress to date.
- The relevance of their achievements is hard to judge.
- The project has produced a good atomic layer deposition result.
- Their accomplishments are adequate, considering that it is still early in the program.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.0** for technology transfer and collaboration.

- Collaborations are plentiful and varied and include an automobile original equipment manufacturer (the Nissan Technology Center of North America).
- . I would like to see more industrial involvement from the end users of the technology to be developed.
- This is a "dream team", and Pivovar is an apt maestro. Coordinating this number of contributors is a tall order; however, the level of interaction and focus at the start seems excellent.
- The PI has enlisted a wide variety of collaborators with clearly defined functions. This is good for a start.
- The processes for applying catalysts may be limited by just relying on academia and NREL. It is critical that, whatever approaches are considered, that consideration be given to ultimate scalability to the high rates and low costs needed for automotive production.
- More cell manufacturers on the team is desirable.
- The project incorporates an effort to bring university accomplishments (e.g., at Riverside) to a DOE laboratory.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.9 for proposed future work.

- Much of the future work is aimed at fabrication of structures and not on new catalyst materials.
- Milestones and/or go/no-go points in the next year are not clearly defined.
- The plan is clear and well thought out. I hope that some downselection takes place as time passes to reach a preferred approach to deliver or demonstrate a significant breakthrough.
- The presentation offered a fantastic vision of what could be possible. It will be exciting to see the results of successful fabrications and the performances of the synthesized structures.

- The currently proposed work is focused on the material structures that appear to look like the 3M NSTF as a model of the extended surface catalysts; however, many other properties must be considered besides one structural aspect in order to make the whole electrode and its catalytic properties and manufacturability feasible. These aspects still do not seem to be appreciated or understood.
- It looks more like "trying out", rather than pursuing a clear strategy.
- As mentioned above, the crystal orientation of the atomic layer deposition should be investigated.

Strengths and weaknesses

Strengths

- NREL employed a clear plan, and has displayed good organization starting out with the project.
- The team has unprecedented bandwidth and capability, from theory to testing. The research builds on platinum attributes which lend high confidence for a positive outcome.
- The project has many collaborators with good capabilities and a strong PI.
- It is important to consider how novel structures can be used to affect catalyst activity and durability for polymer electrolyte membrane fuel cells.
- There is good collaboration with the University of California, Riverside.

Weaknesses

- It is too early to see where the cracks are.
- Team size and coordination challenges may interrupt quick progress at times.
- This project is trying to duplicate the 3M catalyst project without really narrowing in on the one aspect that makes it so unique, the crystalline organic whisker support. This technology area could benefit from high level fundamental work befitting a national laboratory consortium. The organic pigments represent an extremely broad and novel approach for high volume manufacturable, high durability catalyst support concepts. Materials and structural options are far broader than for any type of CNT or VACNT type system. These options should be looked at far more broadly. This suggestion has been communicated to the PI in private, with the encouragement to look at this class of materials in more depth. 3M cannot do the fundamentals alone or at the level potentially possible at national laboratories. They should not assume they cannot bring a great amount of further fundamental understanding to the 3M catalyst system, and therefore have to spend resources studying less promising, and, in many cases, already evaluated catalyst systems.
- The team needs more fuel cell manufacturers.
- The odeling approach used is relatively weak. It would be good to consider inclusion of Professor Goddard, at the California Institute of Technology, as well as an Argonne National Laboratory non-platinum bimetallic cathode electrocatalyst specialist.
- Some of the results for the catalysts look poor. The project team should enlist more help from Shyam Kocha to aid in the analysis.
- It is not clear if the catalysts can be annealed to higher temperatures needed to impart stability on the platinum.
- It is not clear how NREL is working with 3M. Because the 3M catalyst system doesn't seem to be workable in practical fuel cell systems, it seems like a waste of effort to try to improve this catalyst system and/or format.
- It's not clear how the alternative deposition methods would be used at a large scale and what the cost would be. If using the atomic layer deposition method and no nucleation takes place until after 100 cycles, is platinum lost? If so, this factor should be taken into account for the overall cost of the catalyst.

Specific recommendations and additions or deletions to the work scope

- Eliminate the focus on CNTs and VACNTs, which will always have the issues of cost, speed of manufacture, limited currents from thick dispersed electrodes, and carbon corrosion.
- Add a focus on the entire new class of organic heterocyclic pigments of which the 3M perylene red is the classic and first example.
- The project should include two or more fuel cell companies.

Project # FC-08: 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 PtM_1M_2 ternary systems that would lead to the development of highly efficient and durable real-world nanosegregated Pt-skin catalysts with low Pt content. ANL's materials-by-design approach will be utilized to design, characterize, understand, synthesize or fabricate, and test nanosegregated multi-metallic nanoparticles and nanostructured thin metal films.

<u>**Ouestion 1: Relevance to overall DOE**</u> <u>**objectives**</u>

Overall Project Score: **3.5** (6 Reviews Received)

This project earned a score of **3.7** for its relevance to DOE objectives.

- This project team is probably the only group to set targets beyond DOE-specified goals, although the outcome is more theoretical. This setting of a target beyond current goals is probably necessary as one moves to the membrane electrode assembly (MEA) and stack stage where the inevitable loss due to scale-up is found.
- The relevance is clear and well defined. DOE targets are somewhat outdated and ANL has taken a forward-looking step of promising to achieve even higher catalyst activity targets.
- We agree that a new DOE target for catalyst total content target should be 0.1 g/kW or 10 g_{PGM} per 100 kW stack to keep up with the improvements in catalyst activity as well as to reflect the increased cost of Pt (\$50/g). It is not necessary to define a specific activity. A mass activity of ten times today's Pt/C is also appropriate (rather than four times).
- This is a really relevant project.
- The project addresses the top stack component R&D need: decreasing the loading and increasing the performance of oxygen catalysts.
- This is an excellent project that addresses an area of need (high activity electrocatalysts).
- The project, with its focus on ultra-low PGM loading, is well aligned with DOE R&D objectives.

Question 2: Approach to performing the research and development

This project was rated **3.7** on its approach.

- The experiments correlate well with the theory. The theory has guided these experiments well.
- The approach has a sound scientific basis that demonstrates improved activity in half-cells followed by implementation in fuel cells.
- This was a really interesting project, very well thought-out and planned. The team made significant progress in a relatively short period of time. It was an outstanding approach, starting with fundamental mechanistic studies to optimize the electrodes, and then moving to both small scale and short stack fuel cell testing. It will be interesting to see the results following the fuel cell experiments.
- The project seeks to exploit lessons learned in the study of extended surfaces to the realm of nanoparticles, contributing directly to addressing technical barriers.
- This was a great approach from a strong team. The PIs have an outstanding prior publication record in this field.
- The technical targets are three times the DOE activity targets and focus on alloys and skin/core-shell type catalysts. Activity has been as high as 2,100 mA/mg.

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Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.3** based on accomplishments.

- The project is very good, but it is still unknown as to why or how the effect could be translated into final systems.
- Task 1.2 is related to the reaction mechanism of ORR and is supposed to be 90% accomplished. If this is simply based on the data showing all Tafel plots are similar, then the task has not been accomplished. The ORR mechanism is complex and has not been resolved for the last 40 years. There is no evidence of any results or data showing any further clarity on the ORR mechanism in the presentation. Deeper analysis of the rotating disk electrode (RDE) data, reaction orders, and Arrhenius effects have not been shown. An explanation of why nanosegregated particles have higher specific activity compared to extended films or bulk Pt and Pt alloys has not been touched upon.
- It is not clear from the presentations as to which materials are supported on carbon and which are not. It is unclear whether or not unsupported materials are considered films or nanoparticles. All the other accomplishments are outstanding!
- The group really made significant technical progress in a relatively short period of time. They did an excellent job optimizing the activity of the electrodes through modeling and experimental activities.
- The project has shown high performance and durability of novel catalysts in many *ex situ* tests.
- By using well-ordered and defined systems, the project has demonstrated improvements in electrochemical activity and durability.
- The only weakness is that these results have not yet been verified in fuel cell tests leaving some concern that the promising results shown will not translate into in-system gains.
- It is not clear when carbon supports have been used in testing and if the results presented are ever for unsupported catalysts. The role of the catalyst-support interaction is critical for some of the conclusions presented and it is unclear to what level this has been considered.
- Great progress has been reported in terms of enhancement in catalytic activity.
- The project determined the effects of particle size, preparation temperature, and annealing temperature on specific and mass activity. The platinum skin protects the Pt₃Ni core. 30,000 cycles at 0.5-1 V. Mass activity for Pt₃Co of 1,500 mA/mg was obtained, while ternary PtNiCo has an activity of 2,100 mA/mg and 5 mA/cm². The MEA results are needed.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.2** for technology transfer and collaboration.

- The addition of GM as the MEA fabricator is critical to take this discovery to the next step.
- Determining the lifetime under real conditions may give additional insight.
- Collaboration is sufficient at this stage of the project.
- The group contains highly technical and accomplished individuals who work together seamlessly.
- The project consists of several collaborators who are appropriate for the project.
- It is not clear from the presentation what role, if any, the collaborators played in the data presented. While the high-resolution transmission electron microscopy (HRTEM) is likely from ORNL, the roles (if any) of JPL, Brown and Indiana University-Purdue University Indianapolis for the data presented are unclear.
- The team in place is very strong and has very good interaction.
- The tasks for team members are well defined and well laid out.

Question 5: Approach to and relevance of proposed future research

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

- This reviewer agrees that more work on stability will be done.
- There is concern that there is not enough time left in the project to address MEA making and testing.

- The PIs need to explore the mechanism of ORR for the new materials in depth and explain the reasons for the high activity of their materials in half-cell studies. The first steps in incorporating these materials into MEAs need to be attempted sooner.
- The group has a clear plan going forward. It will be very interesting to see the performance of the MEAs and short stacks they plan to make.
- Future work, particularly for fiscal year (FY) 2010, focuses on a continuation of prior work. It is not until FY 2011 that MEA testing is incorporated and the exceptional results shown in *ex situ* testing should be validated sooner in fuel cells.
- The project started in Fall 2009. The future work is well laid out, with focus on MEA testing in 2011.

Strengths and weaknesses

Strengths

- There is an outstanding combination of theory and practice to identify a "super" class of catalysts on this project.
- New materials that exhibit high catalytic activity have been demonstrated in half-cell experiments. DOE targets are somewhat outdated and ANL has taken a forward-looking step of promising to achieve even higher catalyst activity targets. The project has implications both on the fundamental level as well as potential for application at a practical level.
- This is overall a very solid project that has already produced interesting results in a relatively short period of time. The work achieved has been very nice.
- The project has demonstrated durability and performance in *ex situ* testing.
- There is a strong fundamental basis for developing next generation materials.
- The project has a strong team, excellent prior publication record, and a very good and rational hypothesis.
- There is a very high specific activity for all types of catalysts.

<u>Weaknesses</u>

- There is a delay in bringing this advance to the MEA and short stack testing stage. This reviewer is not sure if enough time is left for this critical final task.
- - There is a lack of demonstration of materials in application: for example, testing in fuel cells or studies investigating the carbon support and/or catalyst support interactions.
- The MEA tests are not done yet; however, this is not a weakness because the project is young.

Specific recommendations and additions or deletions to the work scope

- The PIs need to explore the mechanism of ORR for the new materials in depth and explain the reasons for high activity of their materials in half-cell studies. The PIs need to describe the materials more clearly.
- The PIs should focus on MEA fabrication with partner GM.
- The PIs should accelerate tests involving fuel cells.
- The PIs should perform studies investigating the influence of carbon support, and the processing and characterization of electrode structure using novel catalysts.

Project # FC-09: Contiguous Platinum Monolayer Oxygen Reduction Electrocatalysts on High-Stability-Low-Cost Supports

Radoslav Adzic; Brookhaven National Laboratory

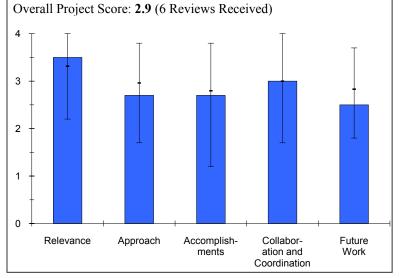
Brief Summary of Project

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, carbon nanotubes, scale-up syntheses of selected catalysts, and membrane electrode assembly (MEA) and stack testing.

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of **3.5** for its relevance to DOE objectives.

This project addresses three barriers



- associated with electrocatalysts, but only for the electrocatalysts. The scope is not broad enough to address catalyst/membrane or catalyst/ionomer interface effects or MEA operating condition effects, which would be outstanding.
- BNL uses one basic catalyst approach that is quite innovative in trying to overcome the barriers; i.e., developing contiguous Pt monolayers on various supports.
- The project is aligned with the DOE Hydrogen Program targets and goals. The project addresses the critical issue of fuel cell cost.
- The project addresses the top stack component R&D need: decreasing the loading and increasing the performance of oxygen catalysts.
- The team is trying to develop new, active catalysts and apply new synthetic methods to do so.
- Justification of catalyst costs with Pd and Ru seems unrealistic in that there is no information given on how these costs were derived and whether it was for massive production quantities.
- Clearly, improved fuel cell catalysts address DOE goals pertaining to cost and performance.
- The development of new ORR catalysts is perhaps the most important activity to address the commercialization barriers of cost and durability. This project is working on this topic exclusively.
- Two arguments can be made against the relevance of this project: 1) that Pd is used to replace Pt and therefore cost will remain high, and 2) that the catalysts will be unstable. In regard to the first argument, there are catalysts being made that have shown 0.44 A/mg_{PGM} (mg of platinum group metal) for both Pt and Pd) from the rotating disk electrode (RDE), so relevance is not lost. In regard to the second argument, there is a contention that Pt will be stable on Pd, which remains to be seen.

Question 2: Approach to performing the research and development

This project was rated **2.7** on its approach.

- The basis of the research plan appears to be one factor affecting ORR activity that is directly related to surface fundamentals of the nanoparticles, e.g., how to obtain properly compressed Pt(111) facets. This is good as well as specific to the basic approach of trying to develop stable contiguous Pt monolayers. There may be other fundamental characteristics of the supported monolayer that will be important for finding the best ORR to use. It would be useful to see a consideration of these.
- The approaches with BNL focusing on more standard Pd nanoparticles and with MIT and Johnson Matthey on high aspect ratio support particles is a good start to cover a wide variety of support types.

- The project is directed at increasing the activity of ORR catalysts through manipulation of structure and use of extended surfaces.
- The durability of Pd cores (and Pd nanorods) and the redeposition of Pd on Pt is a concern. There is also a concern about stability in the cycling of Pd materials to low potentials (~ 0.1 V), where hydrides could potentially form.
- For Pt monolayer on Pd nanowires, the PIs should be able to extrapolate activity to see if it is feasible to make Pd nanowires thin enough to reach the target mass activity (per gram PGM).
- The project presents a range of materials and approaches to addressing the issue of decreasing Pt use in fuel cells. While each of the individual approaches has merit, the project lacks clear organization and direction for advancing and evaluating specific materials.
- The project seems like several independent projects that do not benefit specifically from synergies between the different approaches.
- This project should be synthesis driven. For Cu underpotential deposition (UPD), pulsed wave, etc., it is not clear how compatible the synthetic approaches are with MEA manufacturing.
- A better approach would be to understand what the catalyst manufacturers need, and then work backwards toward practical approaches.
- It was not clear which catalysts were carbon-supported.
- The PI assumes that spreading the platinum to a maximum extent will improve performance. However, fuel cells are influenced by a wide variety of factors which result in non-homogeneous three-dimensional space. So, this approach might be useful, but not necessarily so. This process focuses on carbon supports, which is a very well-studied area. Carbon-containing supports might not be the best choice.
- For the most part, the PI seeks to layer Pt on Pd, Ru, or Pd alloys (with the exception of some samples such as Pt hollow spheres). Pd and Ru are both precious metals and the price of both will rise with high volume automotive commercialization. While the PI has been able to show that high PGM-based mass activity can be achieved on an *ex situ* basis while using Pd or Ru, the approach of doing so implies that the PI is making it tough on himself. Layering Pt on a cheaper base metal would be more robust to precious metal prices.
- The durability of the samples generated is still in question. With the exception of work at 3M (for samples where mass activity was not reported in these slides), cycling reported here has been in RDE with no potential below 0.6 V as the cathodic limit. Durability needs to be shown *in situ* with lower potentials.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.7 based on accomplishments.

- Each of the approaches suggests pathways to overcome the barriers of nanoparticle ORR activity, PGM cost, and stability. This project is very good in considering a wide variety of support particle approaches.
- The amount of data shown is very limited; however, it suggests an opportunity for much more testing and evaluation to statistically support the conclusions. Also, the testing conditions were very sparse or not reported at all, which limits conclusions as to the effectiveness of the tests.
- Testing stability under voltage cycling should be done under more severe conditions (higher temperature, wider voltage range, step function cycling) to determine the limits of stability.
- It is not clear how the Pd nanorods were made. The STEM (scanning transmission electron microscopy) photo is very unclear, with no size information given.
- The fuel cell testing demonstrates one potential caveat on the ultimate utility of any of these approaches, and that is the intrinsic limitation of dispersed electrodes to enable high current densities, ~2 A/cm², which is the fastest route to reducing stack costs besides reducing PGM loadings. The DOE targets for mass activity do not dir^ectly correlate with peak power and therefore are not sufficient metrics for predicting catalyst utility.
- The project is slightly less than one year from its start. The team has made a good start.
- The project team should make sure all mass activity measurements are reported in terms of total PGM. Adjusting activities for today's PGM price is not useful because the price of Pd (or other PGMs) will undoubtedly increase if demand increases with use in fuel cell applications.
- Hollow Pt spheres show good mass activity, exceeding DOE target.
- For a Pt monolayer on Pd nanowires, the PIs should be able to extrapolate activity to see if it is feasible to make Pd nanowires thin enough to reach target mass activity (per gram PGM). Pt/Pd on CNTs may be more likely to get to the target, and use of Pd seems to provide more stability than Pt on CNTs from cycling test at BNL.

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- There needs to be an explanation for the large electrochemical area loss for Pt/Pd/C MEA testing at 3M.
- Significant progress has been made in the area of bromine treatment to remove low coordinated sites and hollow nanospheres. These advances move toward overcoming technical barriers associated with electrocatalysis.
- Studies involving multiple Cu UPD steps and displacement with Pt and Pd may be difficult to scale to meaningful quantities. The potential cycling to only 0.9 V may limit some of the potential aging effects.
- The work to date on CNTs has not shown increases in performance that are particularly compelling.
- Pd nanorods and nanowires are interesting from an academic point of view, but may not offer meaningful cost benefits when used as supports for Pt monolayers.
- 3M's fuel cell results showed meaningful, although modest, losses from potential cycling after just 1,000 cycles on durability.
- The team has made some progress, although it seems to be a repeat of progress made in prior years, and the PIs do not appear to have any big breakthroughs this year.
- The results, to date, appear modest.
- The project has done a nice job of quantifying activity for a number of samples. However, the value that counts is mass activity on a PGM basis. This value is highest for Pt hollow spheres (1.1 A/mg_{PGM}), Pt/Pd/Ru/Pd particles (0.44 A/mg_{PGM}) and Pt/Pd nanorods (0.42 A/mg_{PGM}). PGM-based mass activity was not reported for Pt/Pd nanoparticles and Pt nanowires, but should be.
- Durability needs to be shown using *in situ* data.
- Microscopy on some samples (e.g., Pt nanowires) would help toward explaining whether Pt coordination or crystal orientations contribute toward higher activity. Microscopy would also help toward predicting whether coordination or orientation would remain stable after cycling. Because fuel cell cathode half-cell potentials will inevitably rise high enough to create surface oxides, it would be interesting to see if such oxides would cause restructuring or piercing of Pt monolayers.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.0** for technology transfer and collaboration.

- BNL has strong team members with MIT and JMFC, but it was not brought out how they are mutually collaborating to make any one approach succeed.
- There appears to be good collaboration between the partners. Collaborations outside of project partners with key MEA manufacturers and stack developers are present.
- BNL has a history of collaboration in the area of catalyst development.
- The project team has limited collaborations, but project members each bring specific strengths.
- Johnson Matthey and 3M provide significant support with catalyst scale-up and fuel cell characterization.
- MIT's role seems largely disconnected from the primary focus and most promising results to date of the project.
- The project has a good team of DOE laboratory, university and commercial partners; however, the activity level of the commercial partner was not very clear.
- BNL teams with excellent partners.
- Most collaborators listed (MIT, 3M, Johnson Matthey) have a clear role in assisting the PI (BNL), and their efforts are well represented in the project. The exceptions are United Technologies Corporation (UTC) and the University of Wisconsin. It is clear in the future work slide that UTC will be expected to fabricate MEAs and test them. University of Wisconsin appears to do calculations on onion catalysts, but it is unclear from the slides if the calculations pointed in the direction of Pt/Pd/Ru/Pd formulations, or if the modeling was done following the synthesis.
- Johnson Matthey has already been incorporated into catalyst scale-up work, and 3M has been used in cell testing. The specific activity data are interesting, but hopefully mass activity can be shared from *in situ* tests in the future as well.

Question 5: Approach to and relevance of proposed future research

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

- The breadth of work in any one of the three approaches is enormous, so it would be expected that the rate of progress might be small.
- For Pd-Nb, future work needs to reduce the total noble metal content in the core.
- Future plans are primarily a continuation of the different thrusts presented to date. The future work is quite disconnected with little synergy or overlap between UPD synthesis of Pt/Pd, the synthesis of Pd particles and metallization of CNTs, oxides, and nitrides.
- Scale-up of catalysts seems premature without some sort of performance metric associated with the decision.
- Future work shows no clear path to scale-up to 20 g of catalyst.
- Certainly, detailed testing and stack testing will take more resources, and would depend on the preliminary results to be achieved successfully here.
- The future work should focus more on those samples that have shown the highest A/mg_{PGM} values. It would be interesting to see Johnson Matthey scale-up these catalysts and have 3M or UTC test them *in situ* for activity and durability.
- Improved or initiated synthesis of Pd-W and Pd-Nb core materials is worthwhile, although low limiting current for Pd-W may point to two-electron reduction. Stability of Pd-V seems dubious. Calculations from University of Wisconsin may be helpful in this regard.
- At some point, a go/no-go decision may need to be made as to whether CNTs are too rough for layering of extended catalyst structures. If roughness causes more PGM to be laid down, perhaps a base metal could be used to "coat" the CNT. If not, then perhaps this aspect of the project needs to be cut.
- Islands of Pt on Ni, Fe or Co would not be expected to be stable.

Strengths and weaknesses

Strengths

- The project has good fundamental concepts and strong team members.
- There is a good understanding of ORR activity dependence on Pt structure.
- Collaborations in place with major companies in the area of MEA and stack development are worthwhile and productive.
- BNL uses an excellent materials set and has demonstrated activity improvements using specific systems, namely bromine-treated nanoparticles and hollow nanoparticles.
- There should be a focus on scale-up of some of BNL's outstanding catalyst systems.
- Presentation slides gave a lot of information on synthesis methods.
- The team is admirable, and appears to have adequate resources.
- The project demonstrates consistent reporting of RDE measurements, as evidenced by limiting currents shown. RDE is performed cleanly and accurately.
- The PI has sought to incorporate novel catalyst synthesis methods and morphologies.
- Collaborators provide useful content, whether through novel fabrication (MIT), catalyst scale-up (JMFC), or testing (3M).
- Some materials, such as Pt hollow spheres, have demonstrated very high PGM-based mass activities. In future years, this could prove to be an interesting discovery.

Weaknesses

- The team uses too many approaches to pursue the basic concept of developing contiguous Pt monolayers on a stable support. There are many questions to be answered that are common to all the various supports being considered. Faster progress on the fundamentals, as well as practical utility, might be made if the team chose one approach and focused on it.
- There are too many disconnected materials and approaches without a clear sense of direction or information regarding the decision-making process for each of the materials presented.
- The role of carbon and support interactions is largely unclear for the systems presented.
- There is a lack of hard focus on synthesis, and it is not clear why BNL cannot get a serious synthetic partner. JMFC seems to be playing a minor role, and no progress was reported from them beyond mentioning that JMFC is going to scale-up.
- The basis of the cost analysis was not explained.

- In general, the testing environment appears weak. There needs to be a real focus on experiment replication, and on highly specified testing conditions. Tests need to follow device changes during the test period, for instance, migration of catalyst materials. Fuel cell testing involves complicated fabrication, and there needs to be considerable interaction between the catalyst people (generally this team) and the MEA people (probably JMFC).
- More emphasis needs to be on PGM-based mass activities, not Pt-based ones. Decisions about which catalyst to scale-up need to be based upon PGM-based mass activities.
- More emphasis is needed on microscopy, and how atomic-level features influence observed activity.
- Better reporting of the role of modeling is needed.
- Some of the future work points toward catalysts whose instability may be obvious before testing. Perhaps calculations could help screen unstable catalysts.

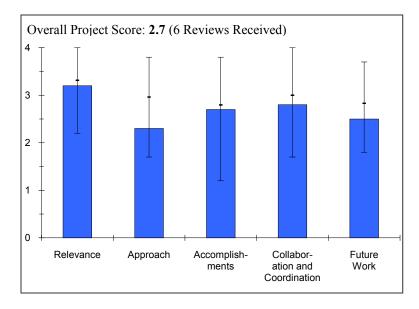
Specific recommendations and additions or deletions to the work scope

- The work scope should include trying to downselect one of the approaches for determining the entitlement potential of the "continuous Pt monolayer" concept, and focus the whole team on it.
- The high activity of hollow Pt spheres suggests that work on these should be emphasized in the near future durability studies on MEAs, etc.
- There should be a more narrow focus on the material set to include only a few of the most promising materials. Downselect MWCNTs or quickly establish ability to form two-dimensional islands as a go-no go decision for this material.
- Reconsider quantity of catalysts synthesized, based on some combination of performance metrics.
- The team should have stronger synthetic chemistry. The team at MIT is not known for synthesis, and might be replaced with a better team.
- The testing plans need to be crafted and published. Testing protocols need to avoid, at least initially, moving the fuel cell specimens into destructive conditions, such as fire and high voltage. After adequate performance has been thoroughly and successfully demonstrated, more vigorous testing may make sense. Those types of tests might be called "crash testing."
- The work scope should include probing whether PGM loading of CNT-based catalysts can be reduced using base metals. Rough surfaces may necessitate high loading of PGMs if only PGMs are used.
- The project must stay focused on PGM-based mass activity.
- A scale-up of Pt hollow spheres is worth trying.
- BNL should not include unstable catalysts in future work by using calculations to discern likely instabilities.
- BNL should make greater use of microscopy for understanding the fundamental reasons for activity increases.

Project # FC-10: The Science and Engineering of Durable Ultralow PGM 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 to 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 to 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 high activity PGM catalysts to validate advanced concepts.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.2** for its relevance to DOE objectives.

- The project focus is on catalyst materials and structure, which directly address critical DOE technical objectives. Performance, durability, and cost are considered.
- Potential cost reduction deriving from novel supports exploiting known catalytic materials have compelling prospects.
- This project had particularly good relevance for understanding. Final goal should be realized at larger scale membrane electrode assemblies (MEAs) or short-stacks.
- Durable ultra-low PGM catalysts are needed to meet DOE cost and performance targets.
- Although the relevance appears reasonable, work in this area of catalyst nanoparticles, low loadings, etc., has been carried out for 40 years. Fundamentally, smaller particles with high surface area to volume will always be less stable than larger particles or extended films. There is no scope or basis for improvement in this direction. Spheres have the highest area-to-volume ratio.
- Automotive companies have already succeeded in preparing low-loaded electrodes; as low as 0.05 mg_{PGM}/cm², and employ these in their stacks. Other pathways to low-loaded thin catalyst layers have been heavily funded through the work of 3M in nanostructured thin film (NSTF) for the last decade.
- The project's focus on ultra-low loading is well aligned with DOE R&D objectives.

Question 2: Approach to performing the research and development

This project was rated 2.3 on its approach.

- The approach includes depositing low PGM loadings on novel support structures and materials. The novel support structures may make uniform PGM deposition and ionomer infusion into the catalyst layer difficult.
- I am not sure why the project flow slide in the supplemental slides was not shown-this would have added needed clarity to the vision of the project. Also, theoretical approaches to catalyst design are mentioned, but there is no explanation of how this will be done-the framework needs to be explained.
- The project seems well rounded with multiple approaches to achieve the objectives.
- It would be interesting to know whether sputter deposition on nanotubes will be implemented in a manufacturing process, and if so, how it would be implemented. Including a schematic could be helpful.

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- This project clearly builds on the premise of the successful NSTF project, which has brought positive results. It is not clear why we would expect anything superior would come from this project. Electrodes tested to date are too thick. Thickness optimization will be needed.
- The approach lacks any clear pathway in that it basically claims to employ modeling to come up with an improved catalyst layer. It is unlikely that a significant improvement will result from work in this direction, considering that the lab does not have an in-house catalyst layer that meets currently attained performance and durability standards reported in the literature by automotive fuel cell companies.
- Different shaped catalysts were synthesized. Novel supported catalysts such as conductive polymers and Pyrograf nanowires were used. Various methods such as PVD and RF sputtering were used to deposit low PGM on supports.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.7 based on accomplishments.

- The project is less than a year old, but meaningful progress has been reported. Test articles have been fabricated and tested.
- All of the major areas of research have been initiated, including rotating disk electrode (RDE), modeling, and characterization.
- LANL has very impressive micrographs that show interesting structures and morphologies. It seems that some of the enabling methods for materials preparation have been developed successfully.
- LANL has good results demonstrating low loadings of Pt distributed over the various supports (sputter deposition), but operation in an actual MEA appears to be inhibited, with the exception of the use of ionomer. The fact that ionomer is necessary to extend the three-phase interface implies that these are not really thin layers of catalyst, or that the innate hydrophobicity is higher than typical materials and the ionomer assists in making them more hydrophilic.
- Electropolymerization, with the use of a heparin template to keep discrete tube architecture instead of aggregates, provided very controllable results. This is an exciting development that may be important later on for developing a controllable manufacturing process.
- Limited fuel cell (fuel cell) data is very poor. No RDE or fuel cell activity measurements were available. The use of ionomer in the cathode, not surprisingly, slightly improved performance.
- The preparation of Pt on polypyrrole nanowires is interesting work.
- A baseline characterization of standard materials demonstrating the benchmarks established by DOE for Pt/C has not been done. Without the basic electrochemical characterization of currently available electrocatalysts and meeting benchmarks already achieved by the fuel cell community, it does not make sense to proceed forward with new materials.
- The fuel cell H₂|Air curves show, along with the conclusions, very poor performance with no analysis of electrochemical surface area and oxygen reduction reaction (ORR) activity. There is no scientific basis for claiming poor utilization without all the requisite and obvious electrochemical diagnostics.
- Current density of 0.65-0.9 A/cm² was achieved at 0.05 mg_{Pl}/cm². The project is only one year old, but the team has done a lot of synthesis on various types of carbon supports and different catalyst types. The team has shown rapid progress in testing MEAs from these synthesized catalysts.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **2.8** for technology transfer and collaboration.

- The team includes a fuel cell stack/system integrator, universities, and National Labs with relevant expertise. Input from an automobile original equipment manufacturer (OEM) and a catalyst supplier would be beneficial.
- The collaboration slide explains the roles of the partners clearly, but the extent of actual coordination and contributions to the results is not as clear.
- The project has a well-rounded team comprised of National Laboratories, universities, and industry partners.
- So far the subcontracts have not contributed much to the project, but this is due to delays in getting contracts in place. Also, LANL should be careful of overlap with the Pivovar's NREL project.

- LANL should partner with fuel cell companies who can show how to prepare a good state-of-the-art electrode that is currently used in automotive fuel cells and has high performance and durability. This would be a starting point for further improvement (rather than show improvements over poorly functioning in-house MEAs).
- The tasks among the collaborators are well defined.

Question 5: Approach to and relevance of proposed future research

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

- Most of the elements to understand and advance the concept are included.
- Catalyst/support interactions should be understood for the new support materials/structures.
- The work so far has produced some interesting materials, and has developed in the audience an appetite for seeing additional experimental results.
- The project has operated dry to minimize flooding. It will be interesting to see how this limitation (dry operation) is addressed in future plans. Similarly, it is unclear what will be done to reduce the use of high cathode flow to get around the flooding problem.
- As has been pointed out with 3M's "whiskers," the use of ultra-low precious metal electrodes brings with it a new challenge in developing alternative ways to remove water on the cathode. This topic should be addressed in future plans.
- More effort on developing techniques for introducing Pt is likely required.
- While carbons may still be of interest to automobile OEMs for supporting high activity nanoparticles, it is difficult to see where a study that focuses on carbon sites for nanoparticle nucleation is relevant. Even with conventional Pt/C fabrication, nanoparticles are not necessarily nucleated as the LANL project described.
- A modeling effort on large particles (which are more likely to be stable) requires an extraordinary amount of computational throughput. It is doubtful that much will be gained from the computational efforts described.
- Significant changes in future work based on baselining and benchmarking, as well as achieving the performance and durability of currently available electrodes and catalysts, needs to be undertaken. Any improvements should be on top of currently achieved performance and durability by the fuel cell industry.
- The future plan includes ORR testing, optimization of catalyst synthesis, and MEA durability performance. This plan is well laid out.

Strengths and weaknesses

Strengths

- The project team has an understanding of fundamental issues.
- The team has selected a great portfolio of approaches, which increases the chance of success in achieving DOE goals.
- There is a very good balance of theory and practice.
- The project team is well selected, whereby the strengths of the various team members complement each other.
- A wide range of conductive supports will be evaluated for continuous thin platinum surfaces.
- Some of the new substrate materials are interesting.
- The project has produced good results at low Pt loading.

Weaknesses

- There is a lack of articulation of the project structure and a clearly explained connection between development efforts and ultimate technical targets.
- Interfacial contact, as mentioned by the author, is a current weakness that should be addressed in the future plan.
- There are no plans that anticipate issues with water balance in the cathode ultra-low PGM structures.
- There is no clear fundamental basis why these concepts will be an improvement over existing materials. The project plan is too quick to jump to fuel cell testing while significant materials development is still required.
- A baseline characterization of standard materials demonstrating the benchmarks established by DOE for Pt/C has not been done. Without the basic electrochemical characterization of currently available electrocatalysts and

meeting benchmarks already achieved by the fuel cell community, it does not make sense to proceed forward with new materials.

• The approach lacks any clear pathway in that it claims to employ modeling to come up with an improved catalyst layer.

Specific recommendations and additions or deletions to the work scope

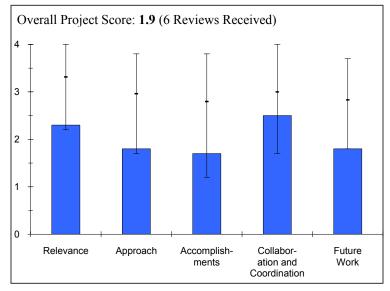
- The project needs to include durability tests.
- Some of the supports are hydrophilic. It will be worth seeing how this will impact mass transport at high current densities.
- Remove work on studying nucleation sites on carbons, as this is not seen as value added. Focus on RDE work before scaling up to MEAs. Be sure to include Pt alloys.
- It is questionable to continue funding this project based on the work and approach so far.
- The team should proceed in the same direction. The team may want to consult with LANL regarding their microporous layer development to mitigate flooding issues for hydrophilic supports.

Project # FC-11: 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

Project objectives are to: 1) demonstrate that non-platinum group metal (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 this is an effective matrix for the testing of new catalysts; 3) demonstrate that the three-dimensional structure of polymer-coated electrocatalyst layers can offset slower kinetics of the catalyst centers when compared with twodimensional platinum or non-platinum catalysts; 4) demonstrate that significant stability of the matrix is possible; 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 DOE objectives

This project earned a score of 2.3 for its relevance to DOE objectives.

- The project does address and support DOE objectives by considering non-PGM catalysts.
- Cost and durability are the most critical gaps for fuel cell commercialization and the electrocatalyst is the most important factor for these attributes. A non-PGM catalyst has the potential to make a breakthrough.
- It is unlikely that this project will contribute to furthering fuel cell objectives of 2015. This is a fundamental research project that will take 20 years to demonstrate useful results and should correctly be funded through other sources.
- The development of improved non-precious metal catalysts offers the potential to overcome the largest stack component cost in fuel cell systems. The decrease in turnover frequency and packing density of such materials limits their potential impact.
- This is a project on non-Pt catalysts in polyelectrolyte membranes. It has no apparent benefit to DOE. While non-Pt catalysts are of interest, this project is just a jumble of ideas that seek to replicate successful projects on non-Pt catalysts by making them in a different format. A significant effort was made in the presentation to justify the concept, but there was no quantitative path. This project might be a good Office of Basic Energy Sciences project for \$300,000 a year, but it is not ready to be an EERE project.
- Achieving cost, performance, and durability is essential for fuel cell commercialization. This project is projected to make progress in all three areas. Clearly, the focus is correct.

Question 2: Approach to performing the research and development

This project was rated **1.8** on its approach.

- The approach is novel, but it is very high risk with insufficient rationale for expectations of success.
- The expected conductivity, activity (turnover frequency), and stability is in the polymer electrolyte membrane (PEM) environment should be included with reasons why the teams expect those values.
- It was not clear what the technical basis was for considering this approach.

- It will be interesting to see how materials will be characterized for structure and activity.
- Homogeneous catalysis in a fuel cell would likely add ohmic losses.
- The go/no-go criteria at the 24-month milestone are not well defined.
- Basically, it is too early to discuss *in situ* level testing. Prior to discussions on how to make the catalyst layer with ionomer, it would be good to focus more on catalyst mechanism understanding to see whether or not the non-PGM catalyst can meet the end-game target.
- The approach is not well described and does not take into account the work done in the literature.
- The implementation of non-precious metal catalysts in fuel cells has significant challenges associated with the performance and durability of the catalytic centers.
- The project focuses on biomimetic complexes and oxygen separation systems that are unlikely to be suitable for commercial applications. Biomimetic systems operate near neutral pH and have significant lifetime limitations, perhaps due to the fact that biological systems do not prefer having long-lived species that can accumulate inside living systems.
- The approach to using a three-dimensional catalyst layer (<500 nm) does not make sense. The turnover frequency of these systems will be slower, the packing density of active sites much lower, and the traditional catalyst layers are much thicker (~10 microns). Add to this the short lifetimes of materials and the ion and electron transport issues, and it seems almost certain this project will not contribute to overcoming DOE barriers involving performance and durability.
- The speaker gave no path toward meeting the DOE targets. The PI does not quantify in any way how they can meet the DOE targets. The team is blending a lot of old ideas, and it is not clear how they will now be reformulated or leveraged into a catalyst breakthrough.
- Much of the "new" stuff is old and well tried. "Enzymes" clearly work as they are abundant in nature. However, such catalysts are short-lived and readily rebuilt, as needed. Fuel cells are invariably not homogeneous with flow starvation, varying reactant conditions, and a large number of factors that result in variable current densities across the current collecting surface. A "differential reactor" approach (essentially high stoichiometry) could sort some of these factors. Of course, one key issue is that the water concentrations are not uniform. Indeed, the same cell may be experiencing flooding and dehydration at the same time.

<u>Ouestion 3: Technical accomplishments and progress toward project and DOE goals</u></u>

This project was rated 1.7 based on accomplishments.

- The project is less than a year old. There is little real progress that is evident.
- It is difficult to distinguish between actual new results and summaries of previous work by the PI and others.
- It is difficult to evaluate this project with limited results shown.
- Results to date are essentially nonexistent. A handful of voltammograms based on either commercial catalysts, "Rh" polymer, or di-nuclear copper complexes show no promise for the catalysts to enhance the oxygen reduction reaction (ORR) in a meaningful way.
- Data on the effects of polymer chain mobility using traditional acidic polymers with traditional catalysts are not insightful for the systems proposed.
- The rest of the "technical accomplishments" section is essentially future work.
- The team is nine months into a four-year program (~20%) and no progress has been made. Slow progress was blamed on graduate students. The PI did not explain where approximately \$2 million allocated to date has gone at the National Labs and other university laboratories.
- Not much new was described, although the classical electrochemistry was well presented and fun to listen to.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **2.5** for technology transfer and collaboration.

- National Laboratories and industry partners are appropriate and will benefit the program.
- A little collaboration exists, but it is primarily a two-National Laboratory project where the synergies between the two National Laboratories is not well defined. The role of 3M as an in-kind contributor is potentially useful, but also poorly defined.

- The PI has made a good effort to bring together an expansive and knowledgeable team, however they have not produced any new ideas or results, so the effort does not appear to be well managed or coordinated.
- It appears as if the teaming is well considered, and tasks are well assigned. One wonders about the capacity of the National Laboratories to do successful polymer synthesis, especially these polymers. They probably need to find additional talent to pull off the polymer engineering tasks, with the understanding that synthesis of polymers is just the beginning of the process required to make useful materials.

Question 5: Approach to and relevance of proposed future research

This project was rated 1.8 for proposed future work.

- Next steps are logical and include modeling and synthesis followed by characterization and testing.
- If Milestone 1, ORR catalyst demonstrated with polymer coated electrodes, is not accomplished, then Milestone 2 cannot be met, and serious consideration to ending this project must be given.
- The future work has little chance of eliminating barriers based on the significant challenges of the system presented.
- No specific plans to take on limitations with packing density, turnover frequency, performance, or durability are presented. Similar systems have been studied in detail without addressing these limitations.
- There is no quantifiable approach going forward. Most of the talk was spent as a Gordon Conference format giving the general physical and chemical background to their ideas.
- There is much to do, and the targets are set high.

Strengths and weaknesses

Strengths

- It is a fundamental research project of scientific curiosity and interest.
- The role of polymer catalyst interaction and factors controlling their interaction are worth further study.
- The project has a large, talented team that is schooled in the classic ideas of polymers, and have been brought together to make a breakthrough in catalysis.
- The people are excellent, and they have access to quality tools and appear to have adequate financing.

Weaknesses

- Project management seems to be relatively weak.
- Minimal results have been produced and the approach is unlikely to succeed and be useful to the fuel cell industry for the next 20 years.
- The systems presented operate biologically under very different conditions than those for fuel cell systems. It is unlikely that much beyond increased fundamental understanding can be gained.
- The project is very expensive, unproductive, and poorly organized.
- The technical progress is non-existent.
- The testing regimes are not appropriate. So, a key weakness is that, as crafted, this project will not gain much new knowledge.

Specific recommendations and additions or deletions to the work scope

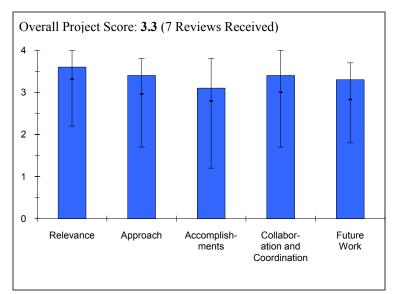
• Testing has to come first to achieve performance; probably most specimens are not worth testing (discard junk quickly). Then there needs to be experiments conducted that carefully expose the test specimens to highly controlled testing conditions, protecting the specimens from harmful (say, high voltage or toxic) events. A significant number of parallel tests should commence to assure reproducibility. After specific testing times, some fractions of the specimens should be extracted and thoroughly examined, so that an understanding of time-dependent changes is garnered. That is, the experiments need to determine normal aging processes, including those that might, for example, result in time-dependent improved performance. A thorough investigation of totally dead test articles might be useful, but usually the "bullet hole" is pretty apparent, and not much is learned by doing that (The totally dead cathode shown by ORNL should invoke only the question, "who killed this fuel cell?").

- Very early go/no-go decisions should be made on materials for those that demonstrate theoretical feasibility, based on packing density and turnover rates and some sort of durability under relevant conditions.
- The project should be cancelled or scaled back to ~\$300,000/year either immediately or after their review next year.
- It is not too late to refocus the testing plans that should be done. There could be some interesting homogeneous (flow battery) type systems that could work in alkaline conditions, and those might be considered in the longer term.

Project # FC-12: Polymer Electrolyte Fuel Cell Lifetime Limitations: The Role of Electrocatalyst Degradation Deborah Myers; Argonne National Laboratory

Brief Summary of Project

Project objectives are to: 1) understand the role of cathode electrocatalyst degradation in the long-term loss of polymer electrolyte membrane fuel cell (PEMFC) 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 type of electrocatalyst on the performance degradation; and 5) determine operating conditions and catalyst types or structures that will mitigate performance loss and allow PEMFC systems to achieve the DOE lifetime targets.



Question 1: Relevance to overall DOE objectives

This project earned a score of 3.6 for its relevance to DOE objectives.

- This project is critical to the DOE R, D&D objectives. There needs to be a deeper understanding of the failure mode associated with catalyst degradation in PEMFCs.
- The project objectives of understanding and mitigating performance losses defined by catalysts properties are directly relevant to the DOE catalyst durability targets.
- This is a very relevant project, specifically because the group is looking at the durability of electrodes that have not really been durability tested before.
- The barriers "Durability, Cost, and Electrode performance" are well addressed.
- The project is highly relevant to further increase the lifetime of PEMFCs. Catalyst and catalyst support degradation are the major life-time limiting factors for fuel cells. Enhanced lifetime contributes to cost reduction as well.
- The project is we focused, has good partnerships, and theory plays an important part.
- The project focus on catalyst degradation, cost and performance is well aligned with DOE objectives.
- It is an important effort to develop an understanding of Pt/C degradation and performance loss. The efforts should address the Pt alloys used for fuel cell systems.

Question 2: Approach to performing the research and development

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

- The approach is clear, logical, and well thought out. I am concerned about the scale of the work fitting into the timeline. It may be necessary to focus on some key tasks.
- There is a strong combination of experiment and modeling work in this effort. The systematic approach is well thought out and looks very good.
- This is a nice, simple, straight-forward approach with a number of critical variables planned; however, in studying the various electrodes, I think it will quickly become very complex. I would advise the group to look at both overall effects on the electrodes, as well as the morphological changes to the actual material. In a true fuel cell environment, the changes in activity associated with morphological changes may be overshadowed by membrane-electrode interfaces or water management, e.g., "whisker" penetrating the membrane over time in nanostructured thin film (NSTF). Additionally, I would advise the group to monitor the state of the ionomer in

the electrode ink, as this may also affect electrode durability. Again, these comments may fall under the other durability projects, but hopefully the groups will work together.

- The approach encompasses a broad range of well-selected methods. Cell degradation tests are well combined with *in situ* and *ex situ* analytics, as well as fundamental materials properties studies. Modeling exceptionally complements the approach. For the *ab initio* modeling, it may take longer than the project duration to devise optimization strategies for the catalysts. Yet, from this method, the most fundamental impact on catalyst optimization can be expected if pursued persistently.
- The milestones and decision points are appropriate.
- It might be rather ambitious to develop a cyclic voltammetry model and oxygen reduction reaction (ORR) catalyst layer performance model parameters by September, 2010, but with the basic code developed and studies under way to determine the relevant input parameters, this may be accomplished.
- ANL describes novel catalyst synthesis methods. It identifies degradation modes. *In situ* and *ex situ* studies have been carried out. Modeling work supplements the testing very nicely.
- The approach appears to be systematic.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.1** based on accomplishments.

- The initial work presented is excellent and is a very good start.
- Good technical progress appears to have been made already. ANL has produced a lot of experimental data early on, as well as a sophisticated kinetic Monte Carlo (KMC) model to predict behavior in a fuel cell environment.
- The project is just getting started, so there is not too much to discuss at this point. The experiments performed to date have generated interesting initial data, but much more work needs to be done.
- The project results are impressive for the short time the project has been in place, underlining the pre-existing skills of the project partners.
- It was not clear what the pressure was on slide 12.
- On slide 11, it appears there is something different going on with the 7.1 nm particle size sample. There is no initial decay and no long-term decay approaching the mass activity of the 12.7 nm sample such as the smaller initial particle sizes exhibit. This behavior was never explained. It could be that there is a critical size where the initial decay occurs, and if that were the case, I would also like to hear why that is, as well.
- It will be interesting to see what you find out for Pt₃Sc and Pt₃Y, as we have tried Pt₃Y without any real improvement.
- The KMC model will be interesting if you can obtain fuel cell parameters to incorporate into it that represent all the degradation modes.
- ANL determined the effect of Pt particle size on degradation rate. Smaller particles led to a faster degradation rate.
- Anomalous Small-angle X-ray scattering (ASAXS) and transmission electron microscopy have been carried out before and after degradation. The effect of relative humidity on degradation was determined. Low relative humidity led to low degradation. Rapid progress had been made in less than a year.
- The explanation of data on page 11 misses an important point. While the surface area of the small initial particles degrades to that of the particle with larger (7.1 nm) surface area, the mass activity of the degraded smaller catalyst is much less than the mass activity of the 7.1 nm catalyst. An explanation would be welcome. It appears there must be a specific crystallite orientation for the 7.1 nm material that is not reached by the degrading 1.9 nm or 3.2 nm catalyst, suggesting crystallite size effect. An explanation for that would also be welcome.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.4** for technology transfer and collaboration.

- This project is relatively small and does not seek to involve as many National Laboratories as possible. There are two key industrial stakeholders involved and that is a preferable position.
- There is an extremely talented group of collaborators.
- The activities of all partners look well integrated at this point in time; e.g., the *ab initio* results shown are definitely relevant for the industry.
- There is a well-integrated team.
- The tasks for various team members are well laid out. The team assembled is very strong.
- The program has brought in a good mix of industry, academic, and National Laboratory contributors.

Question 5: Approach to and relevance of proposed future research

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

- The proposed plan of work is very good. I am concerned that it may be too much to complete in the allotted time. This issue may become clear in the future, in which case I would recommend a focus on task three.
- Future work builds on the current experimental and modeling progress and extends the effort toward platinum alloy catalysts.
- The proposed plan for future work was in general good; however, it did lack specifics. This lack of specifics was most likely because the project was just starting and the durability groups were still distinguishing themselves.
- The tasks are structured along the skills of the partners. The longer this complex project will be going on, the greater the risk will be that the tasks get or stay less integrated. A strict project management is advised to keep the project on track.
- I agree with the first critical assumption. There will be process variables in the membrane electrode assembly (MEA) studies that will not be applicable in the aqueous studies such as diffusion/current collector issues, compression, crossover and shorts, etc., and verifying the relation between the two by modeling is prudent.
- Proposed future work is well laid out, with detailed description of synthesis, modeling, and MEA test plans.
- It does not appear that this team is going to answer the question about decreased surface area and mass activity for *in situ* decrease in surface area versus as-prepared larger surface area.

Strengths and weaknesses

Strengths

- A clear plan to deliver has been established that incorporates a manageable project scale and a good approach.
- There is a talented group of collaborators.
- The project is focused on the understanding of fundamental fuel cell durability barriers.
- The approach is planned and very systematic.
- The team working on the project has highly skilled individuals and they know, in general, what they want to accomplish.
- The project incorporates a broad approach and skilled project partners.
- This is an important project that could answer some very key fundamental questions about fuel cell degradation mechanisms. It appears to me to be a very sound approach to solving this long-term decay problem.
- The effect of particle size on degradation rate (RDE and MEA) is backed up by ASAXS results.
- The team has access to electrochemical and analytical methods, which are necessary to resolve the research problems.

Weaknesses

- The scope is perhaps a little too ambitious to be able to complete all planned tasks within the timeframe.
- The plan is missing some details.
- There is a high level of complexity.

• Viable modeling will be very important, which means valid parameters for the major processes are needed soon.

Specific recommendations and additions or deletions to the work scope

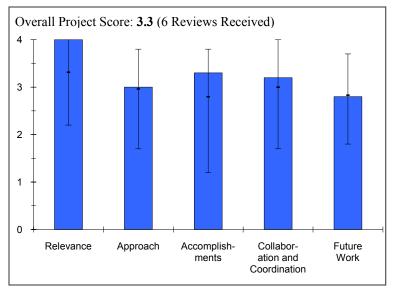
- As far as determining the effects of contamination, using HClO₄ is a good starting point; however, I would also include sulfates. There have been a couple of papers looking at the effects of sulfates on Pt/Vulcan electrodes with high loading. It would be interesting to study the effects on low loaded novel electrode materials.
- It was not clear how the KMC code would predict Pt nanoparticle evolution at fuel cell conditions. It is not clear if the model is independent of experimental data and is just predicated on first principles. It would be worthwhile to figure out how this model would predict catalytic activity as a function of surface area and/or particle size. If the model is not able to predict catalytic activity as a function of surface area or particle size, then it should not be a part of the program, unless it serves some other purpose.

Project # FC-13: Durability Improvements through Degradation Mechanism Studies

Rod Borup; Los Alamos National Laboratory

Brief Summary of Project

The objectives of this project are: 1) identification and delineation of individual component degradation mechanisms; 2) development of advanced in situ and ex situ characterization techniques for analysis of fuel cell component degradation; 3) quantifying the influence of the operating environment on different fuel cell components; 4) degradation measurements of components and component interfaces; 5) elucidation of component interactions, interfaces, and operating conditions leading to cell degradation; 6) individual degradation models of all fuel cell components; 7) development and public dissemination of an integrated comprehensive model of cell degradation;



and 8) identifying methods to mitigate degradation of components.

Question 1: Relevance to overall DOE objectives

This project earned a score of 4.0 for its relevance to DOE objectives.

- Understanding the fundamental degradation mechanism is vital for the successful development of highly robust and price competitive materials, which in turn are vital for the successful commercialization of the fuel cells.
- Degradation due to interactions among components is an important area of study, ultimately leading to advancing DOE's goals. This topic is missing from many other studies. The final product is a model, not physical deliverables *per se*.
- The program is relevant and addresses the barriers: "Durability, Cost, and Electrode performance". It includes durability with cycling.
- This project is clearly relevant.
- Cost and durability are the two vehicle-related barriers to polymer electrolyte membrane fuel cell (PEMFC). commercialization. This project is scoped to investigate nearly every mode of PEMFC performance degradation. For this reason, the project is entirely relevant to DOE's objectives.
- LANL seeks to study durability on a fundamental component basis. In other words, LANL is not seeking to derive empirical relationships for durability (or "acceleration factors") based on one particular technology that may be irrelevant to future commercialization.

Question 2: Approach to performing the research and development

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

- Obviously, with the technical competence of the PI and all of the various subcontractors, the approach is top notch. Doing the fundamental characterization from scanning transmission electron microscopy (STEM) and transmission electron microscopy (TEM) all the way to *in situ* fuel cell testing is vital for success. My only major concern is the shotgun approach of trying to do everything, which could lead to a dilution of heads, dollars, and effort. Perhaps a more focused approach may be necessary. Perhaps to help clarify this, a Global Gantt Chart should be created, clearly stating who is doing what and when.
- The project has a good general approach to performing the R&D, but, with a team this large, a flow chart showing how information and materials is supposed to flow would be beneficial.

- It appears electrodes and membranes are the prime focus.
- There is a clear and systematic approach that is well oriented at real-world conditions.
- The work is very comprehensive, if not too comprehensive, for one single project. The different degradation mechanisms addressed are not necessarily interrelated and could be addressed in single projects. The objective to "quantify the influence of inter-relational operating environment between different fuel cell components" needed more specific substantiation. The overall objective of this project is hard to identify, even more so as it includes electrode production methods in the applied science tasks.
- My key question is the integration with the other durability projects. I was very glad to hear that a working group has been formed and met during the review meeting. I wouldn't mind some overlap between the various durability groups--multiple teams performing similar experiments and reaching the same conclusions will give us a lot of confidence in the outcome. At the same time, complete duplication of effort is not good, and if each of the durability efforts can have their own area of focus we will get the most out of the DOE investment. It is too early to tell--I hope next year we will clearly see that the different groups are effectively coordinating their activities.
- At this stage, the approach still suffers by the size of the scope. Some prioritization should be given to the different activities involved. For example, after studying catalysts, membranes, gas diffusion layers (GDLs), and plates, it remains to be seen how relevant a study of seal and seal degradation will be.
- The project will benefit from including a modeling effort, which will help to limit the experimental scope.
- The project has a considerable amount of characterization techniques at its disposal. It would be good to see a preliminary plan as to what the work flow will be.
- The use of low catalyst loaded membrane electrode assemblies (MEAs) is excellent.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.3** based on accomplishments.

- The amount of results and progress (some from before the project began) in all of the various subprojects is clearly stated and at a very high level. Especially of interest is the work being done to understand the inconsistency between electrochemical area (ECA) and fuel cell performance and how using different solvents in the catalyst-coated membrane (CCM) fabrication leads to very different fuel cell fuel cell polarization curves.
- LANL has produced significant findings for solvent-dependence on catalyst ink and MEA durability.
- It is hard to really judge the project because it is only six months old; however, results so far promise that the project is headed in the right direction.
- The progress is hard to assess since the project start date is not provided in the charts. The results in catalyst degradation owing to freeze cycling and the carbon corrosion results, for instance, do not seem to be interrelated, though each single result may be good. This leads to too superficial a description of the single effects throughout the presentation.
- I particularly liked the "applied science" task on solvents used to fabricate electrode structures. I think it's relevant, and should certainly be continued with other high boiling point solvents and with input from groups that directly apply catalyst structures to membranes, as opposed to the LANL decal method. I would be particularly interested in seeing what fraction of these high boiling solvents like glycerol are left behind in the electrode structure (not necessarily intact as glycerol--there are a number of acid-catalyzed decomposition/polymerization possibilities). The freeze start and carbon corrosion results also look promising, although it is far too early to know that these investigations will lead to effective mitigation of these problems.
- The project reporting would benefit from further notations of experimental details. For example, when comparing two different GDLs identified as "cloth" and "paper," it would be good to know if they had the same microporous layers (MPLs).
- The systematic evaluation of how half-cell potentials affect carbon corrosion is very well designed, but more useful knowledge would be obtained if the corrosion rates were also linked to properties of the carbon; e.g., graphitization level, agglomerate size, etc.
- The project should aim to extract further meaning from the data. Performance losses that trail electrochemical area (ECSA) losses are known. Investigating whether lower ECSA loss would affect kinetics due to underutilization of the catalyst, yet still affect mass transport due to change in MPL surface energy, would be beneficial. Phenomena like these need to be explored.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.2** for technology transfer and collaboration.

- Collaboration between the partners exists and is ongoing; however, it is not readily transparent on who is doing what and where. It is my recommendation that the partners' responsibilities are clearly labeled in the presentation to make it more evident.
- In this particular presentation, it is hard to gauge what was done before the project and what will be done during the project and by whom.
- This is a broad group covering National Laboratories, one university, and two industrial concerns, with advanced modeling and characterization.
- Collaboration and dissemination look excellent.
- Engaging relevant industry is good.
- Though each of the activities looks worthwhile, the whole project leaves the impression of too little overall coordination.
- It looks good on paper—next year's review will tell the real story.
- The collaboration planned is extensive, but it primarily involves the National Laboratory community. Industry is represented by a stationary / material handling vehicle application original equipment manufacturer (OEM), Ballard Power Systems, and by the MEA supplier, Ion Power. Collaboration with automotive stack OEMs may be helpful.
- It may be useful to compare supplier or in-house catalyst layer properties versus those from other suppliers to check how representative catalyst layer properties are to other commercial catalyst layers.
- Collaboration has created an almost unmatched set of characterization techniques at the project's disposal.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.8 for proposed future work.

- Future work focus is largely on individual components. I am hoping the future will show more on interaction effects. I applaud the future topic of studying the impact of mechanical compression on the MEA and durability.
- The future work is no logical continuation of the results presented, yet it introduces a very important but also very different topic, the impact of mechanical loads to degradation.
- The parametric aging studies are needed, but I am very concerned about whether they will ever lead to a model that can predict durability for a new material or operating condition. It will be easy to look at the data and say what does not work, but it will be harder to predict what may work. Without a predictive model, I do not see this effort shortening the development cycle significantly. The presentation mentions "science-based degradation models," but we heard no details about this effort.
- The scope needs to be further refined and prioritized. At this moment, the project seeks to investigate every type of performance degradation.
- For prioritized studies, it would be good to see a deeper plan. At the very least, the initial stages of some parts of the research (applied science for electrodes excluded, since this has begun in earnest) should be defined so as to provide a starting point. Experimental designs could be presented showing expected control factors (operating conditions), response variables (voltage, material properties), etc.

Strengths and weaknesses

Strengths

- World class scientists are on this project doing the fundamental characterization from STEM/TEM all the way to *in situ* fuel cell testing, which is vital for success.
- LANL has assembled an outstanding team that was recruited for their complimentary specialties.
- Novel approach of looking for interaction effects and interface degradation.
- LANL has already identified the impact of electrode ink solvent and durability, which is a surprising and significant finding, along with the analysis of why this may have an effect.
- There is ample relevant background knowledge.
- The project has very relevant research topics and renowned partners.

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- There is a good team with the capabilities to get a host of useful data.
- There is an immense amount of experimental capability.
- The project intends to use low catalyst loaded MEAs where possible, which reflects the direction of the technology.
- A modeling effort should help to contain the experimental needs.
- The project has considerable capability for developing systematic studies. The study of carbon corrosion, both with Pt/C and Pt black is a great example. The studies of electrodes prepared with different ink solvents, in concert with SANS characterization, is another great example.

Weaknesses

- The shotgun approach of trying to do everything which could lead to a dilution of heads, dollars, and effort is a weakness, and perhaps a more focused approach could be beneficial.
- There is a lack of clarity as to who is doing what with the various subcontractors.
- There needs to be a flow chart of how materials and information flow between the numerous groups.
- There could be a "one size fits all" degradation model that would benefit a highly diverse fuel cell community.
- Too many research topics seem to be patched together. The future work is inconsistent to the previous work.
- LANL must show effective coordination with other durability projects.
- The synthesis of results into a predictive model is uncertain. No details about the modeling tasks were presented, so it is impossible to know what is planned.
- The project scope is large. There is a need for prioritization and for further definition of experimental designs.
- Reports of experiments should contain all details, including operating conditions, material sets, and cell assembly parameters, if needed.
- The collaboration is National Laboratory-heavy. More extensive industry collaboration may be useful. One specific example would be in terms of defining what GDL degradation occurs from light-duty automotive drive cycling, in terms of material properties.

Specific recommendations and additions or deletions to the work scope

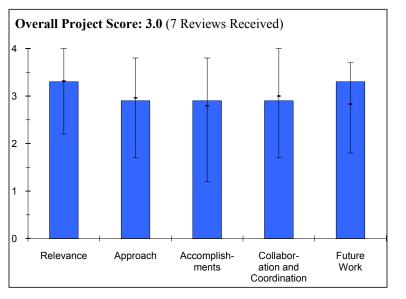
- Consider streamlining and consolidating some of the projects.
- Make the work of the collaborators more transparent. Perhaps to help clarify this, a Global Gantt Chart should be created, clearly stating who is doing what and when.
- The reviewer is looking forward to the output from this prestigious group.
- The scope is fine, but overlap with other durability projects must be considered.
- It may be best to truncate a project after more progress is made to see where efforts can be most effective.
- LANL needs to maintain focus on going beyond cell-level characteristics and making sure to drill down to relationships with material characteristics.
- A water transport model (from such a project) would help in the event that realistic degradation of GDL material properties is known. Then perhaps a model would be able to predict whether performance losses could be expected from the observed material changes.

Project # FC-14: Durability of Low Platinum Fuel Cells Operating at High Power Density *Scott Blanchet: Nuvera Fuel Cells*

Brief Summary of Project

The objective of the SPIRE program is to study decay mechanisms and identify strategies to assure the durability of fuel cells capable of achieving DOE's 2015 cost target. The most significant enablers for achieving stack cost are increased power density and reduced platinum loading. The technical approach of the SPIRE program is to elucidate the critical durability mechanisms for a stack operating at a power density and platinum loading that can achieve DOE's 2015 cost target.

<u>**Ouestion 1: Relevance to overall DOE**</u> <u>**objectives**</u>



This project earned a score of **3.3** for its relevance to DOE objectives.

- Cost and durability are among three of DOE's most important targets.
- The project is truly relevant for achievement of durability and cost goals.
- The project is focused on meeting goals and supports the overall modeling effort.
- Understanding durability at high current density operation is important to enable the achievement of DOE cost and durability targets. Nuvera must be careful that projects provide universal value and not just value for their flow field design and materials set. I was glad to see that the plans included testing of more conventional plate designs.
- The project is looking at the durability of fuel cells using current technology, but operating at conditions necessary to meet the DOE 2015 cost target.
- The project is very relevant to the DOE objectives to reduce the cost of fuel cells to meet the DOE target of \$30/kW.
- Achieving durability and performance at the DOE cost target (2015) are two factors of critical importance to moving the technology to commercialization. This is a good program, but the presentation was difficult to follow and the single cell approach, with a claim that it is not an isothermal cell, was hard to rationalize.

Question 2: Approach to performing the research and development

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

- Reducing the cost by reducing platinum loading is planned. There is, however, no proposal for reducing the platinum loading.
- The project's approach to performing the R&D is pragmatic, straightforward, and well structured: good!
- I have serious questions concerning correlating sub-single cell testing to stack testing (heat rejection, flow concentration, etc.). The Principal Investigator (PI) presented neither data that indicates testing will be valid nor any information on the number of samples to be tested or statistical sampling (although some information was provided in supplemental slides).
- The proposed durability cycles cover reasonable ranges of current density and relative humidity (RH). I was pleased to see Nuvera take advice from the FreedomCAR (Cooperative Automotive Research) and Fuel Partnership's Fuel Cell Technical Team (FCTT) and run more testing at 2 A)/cm², rather than focus on 3A/cm². I'd like to see testing at higher temperatures included, but I am concerned that the available material set cannot

handle it. It is imperative that postmortem analysis of the membrane electrode assemblies (MEAs) used in these studies be shared, or results will be of little value to the community. W.L. Gore & Associates (Gore) does not have a great track record of sharing such analyses. The degradation model developed must also be universal to a range of materials sets to be valuable.

- Interesting approach to optimize: 1) power density and 2) lower platinum loadings to achieve the 2015 cost target and then analyze durability under those conditions.
- The project's use of multiple cell designs is good and extends the relevance of the project.
- The PI has a planned, integrative approach to selecting relevant model parameters phenomenologically, and it seems like a good approach.
- The approach seems systematic and well planned.
- Nuvera is taking a bold approach to reduce cost by operating at higher current densities and low platinum loading. This project is investigating the durability of low loaded platinum electrodes at the higher current densities. The work is balanced between modeling and experimentation. The project is developing a decay model that will attempt to determine the impact of current and local conditions on the catalyst. Single-cell experiments will be conducted with different material sets over different test cycles to accelerate degradation.
- Nuvera's interpretation of low loading appears to be different from most interpretations: low loading at a targeted power density. Nuvera may be onto something in using higher platinum loading per cm² while achieving sufficient power density to achieve the total cost target.
- The ideas of using a single cell area will be very difficult to provide the corresponding thermal distribution in the cell.
- The independence on compression (slide seven) was demonstrated by Ballard Power Systems five to ten years ago.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.9 based on accomplishments.

- The initial activities seem adequate.
- The project is only half a year old, but achievements and arrangements among the partners look convincing.
- The project is reasonably on schedule. One major accomplishment is the development of a sub-single-cell test jig.
- It's too early to judge Nuvera's progress. To date, Nuvera has shown the ability to run some of the durability protocols. Until durability results and analyses come in, there's not much to base a ranking on.
- Nuvera developed a test fuel cell to simulate a realistic stack cell to share with partners.
- Nuvera is developing a phenomenological model to describe observed fuel cell durability.
- Progress has been good for the project which began in October 2009. A single-cell test fixture has been fabricated and tested. With this test fixture, Nuvera expects to be able to preserve local gradients seen in a full area stack. The performance model has been verified against Nuvera data. The test matrix has been defined.
- It was not explained if *in situ* performance mapping was a model or actual experimental data (page 11). If it was actual data, Nuvera should explain how it was generated. If it was a model, Nuvera should explain how it validated the model. The single cell does not have segmented areas, so it would appear the data is a model. The problem was they put data-point markers in current density and MEA RH plots rather than smooth lines. I took the data point markers to represent data.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **2.9** for technology transfer and collaboration.

- A strong, collaborative effort is planned.
- The small group of competent partners promises to be very efficient.
- Nuvera has strong national laboratory partners. As mentioned earlier, it is unclear if their results will be meaningful.
- Dependence on Gore limits material sets to expanded polytetrafluoroethylene (ePTFE) supported membranes and dispersed carbon catalysts. The interaction with FCTT is encouraging. There is concern that this project could become a mechanism to fund Nuvera's Orion Stack development.

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- The collaboration with national laboratories is good.
- Collaboration partners are very strong and there is good evidence of effective interchange among the partners. Nuvera has incorporated suggestions from the FCTT into their future plans.
- The collaboration team has several strengths. It appeared that the only efforts reported are for Nuvera. It is unclear what the Los Alamos National Laboratory (LANL) did during this seven month period. I saw supplemental slides referring to LANL, but nothing in the main body of the presentation. Similarly Argonne National Laboratory (ANL) work was not presented, although supplemental slide 21 indicates thermal modeling was done, but the results were not presented.

Question 5: Approach to and relevance of proposed future research

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

- A call for a "durability task force" seems appropriate.
- The project will produce a large quantity of test data; however, a better definition of how test data will be analyzed is needed.
- I'd like to see temperature explored more explicitly.
- The inclusion of the impact of different materials (catalysts, membrane, electrode) properties should be included to test the universality of the models.
- Future work will build on what is learned from accelerated stress tests (ASTs). It is a good plan to focus the project on the most important parameters to optimize for iteration back into the final planned testing and development.
- Future work plans are well laid out. If successful, the project output (validated model and data set) should be useful to the community, if the impact of different cell architectures can be elucidated.
- The future work is going to be the most interesting part of this program. The plan looks ambitious, but possible. There was little discussion about catalyst degradation or membrane degradation.

Strengths and weaknesses

Strengths

- The project has a good team with good funding.
- Nuvera has strong national laboratory partners.
- The ability to get accelerated durability data using DOE recommended protocols on stack and small scale platforms with different flow filed designs is a strength. Nuvera is considering heat rejection constraints in the test matrix. Nuvera's tie-in with LANL for their AST program is also a strength.
- Multiple cell platforms for testing extends the relevance of the project to non-Nuvera cells. Nuvera has good national laboratory partners.
- MEAs are being provided by Gore; commercial and pre-commercial materials (down to 0.2mg total platinum loading) are being provided.
- Both land/channel and open flow field configurations will be tested.
- Single cell testing will be done at LANL and stack testing at Nuvera.
- ANL will develop a durability model.
- The cell stack developed by Nuvera appears to be very robust and the concept of operating at much higher CD and lower voltages is very interesting.

Weaknesses

- Nuvera has no plan for platinum loading reduction.
- The correlation of sub-single cell to actual stack performance.
- Without knowing the specific MEA materials sets used, the value to the fuel cell community will be limited. The depth and details of material characterization before and after testing is not clearly defined. The approach to develop a predictive life model is not clearly described and is difficult to evaluate.
- Durability findings may be specific to the cell platform and may not apply as broadly as hoped.

- There is some uncertainty regarding the ability of the decay model to account for and capture the relevant decay mechanisms. Not sure how the temperature effects on durability will be captured. Operation at higher power densities will tax the ability to reject waste heat, at least in conventional vehicles.
- It may be difficult to maintain materials characteristics (carbon support, ionomer, platinum particle size, carbon/ionomer ratio, etc.) while varying catalyst loading.
- The single cell has a low probability of yielding a true temperature distribution. The cell will most likely be isothermal and need to have the temperature varied in a series of experiments to map the performance profile.
- The lower voltage of the high current density stack means more current and bigger buss bars and a lower voltage to the power conditioner; both of which are considered negative. I would like to see an explanation of why Nuvera does not consider the lower voltage to the power conditioner and the larger bus a negative.

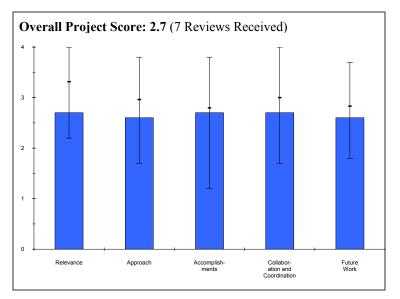
Specific recommendations and additions or deletions to the work scope

- Need to show, with data, that approach of comparing sub-single cells operating at high current density within the test jig to stack data has any validity.
- I'd like to see temperature and MEA material type explored more explicitly.
- Experimental results and detailed model should be published in a peer reviewed journal.
- The thermal issue will be the biggest for original equipment manufacturers. Even at 2 A/cm², heat rejection will be an issue. Nuvera should plan to determine sensitivity to current density coupled to temperature.
- Consider how to relate the thermal behavior of the different architectures (land/channel and open flow field).
- Near the end of the project, Nuvera should be encouraged to look at materials (alloy catalysts and/or membranes) that can enable higher temperature operation.
- The team should discuss with ANL (Myers) the electrochemical surface area and mass activity data that indicates little correlation.

Project # FC-15: Improved Accelerated Stress Tests (ASTs) Based on Real World FCV Data *Tom Madden; UTC Power*

Brief Summary of Project

The objectives of this program are to: 1) compare conditions and materials in bus field operation vs. DOE 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 metals (PGM) decay, carbon corrosion, membrane mechanical and membrane chemical.



Question 1: Relevance to overall DOE objectives

This project earned a score of 2.7 for its relevance to DOE objectives.

- The need for such a test is outstanding.
- The "real world" data used here to assess and potentially modify the ASTs for automotive applications of fuel cells are limited to high loaded membrane electrode assemblies (MEAs) in heavy duty bus duty cycles. The DOE automotive objectives are for light duty vehicles at much lower loadings. The PI is aware of the risks in missing key stressors and failure modes using this data. Also, stress tests for the other fuel cell applications are not addressed in this project. There may be "real world" data from these other applications that may be more relevant to where that technology is expected to go.
- It is important to attempt to compare on-road testing to accelerated stress tests.
- The project plans to correlate ASTs with observed real-world degradation and to look for gaps in the currently used ASTs. This work is relevant to the DOE's durability targets and could provide an important connection between real-world and laboratory-world testing.
- Certainly, these ASTs should be part of membrane/catalyst/MEA development projects, but just having United Technologies Corporation (UTC) running these tests on commercial materials is of limited use. I wouldn't expect accurate acceleration factors to be derived from them, especially given the variability of the real-world cycles run on them (not to mention busses vs. automotive). These programs should run various cycles to attempt to gauge the influence of temperature, relative humidity (RH), and catalyst loading on durability, as well as the impact on the acceleration factor.
- This UTC project will determine if the DOE accelerated stress protocols can account for the relevant decay mechanisms as seen in bus fleet field data. If so, the accelerated tests can reduce the need for costly durability testing in the field by a factor of three or four. This project is very relevant to the DOE program objectives.
- Product durability is important in commercialization. Clearly, engineering studies that result in enhanced durability are an important part of the commercialization activity, but the results of this study might support only a specific commercial technology.

Question 2: Approach to performing the research and development

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

- A very comprehensive plan is proposed with the real-world fuel cell vehicle (FCV) data to come to the test.
- The reliance upon field data from busses that are made with technology that is old and does not meet the DOE targets is a huge effort. I would prefer that the brass board system they are building be used to test more relevant MEAs or stacks under conditions that more closely resemble light duty.
- It's not clear that one stack is a good representation (good or bad) of typical results. UTC needs to have more data presented on the stack (number of faults, time at idle, etc.) to better judge. No data was presented on the number of cells to be tested and whether there will be any correlation to cell performance; i.e., how do weak cells show degradation as defined by stress tests.
- Significant effort might be required to validate short stack results to full-scale results.
- The project includes comparing real-world bus stack failures to degradation in the laboratory, based on ASTs. If a correlation exists, then the project will continue. Presumably, numerous gas diffusion layer (GDL) and MEAs will be tested, but only one, the MEA by W.L. Gore & Associates (Gore), was mentioned. The stack being used to test is proprietary, so it is not clear how the information obtained will be considered generally useful, as the stack conditions will not be published.
- It was not made clear how the testing will allow for isolating the four different proposed (and "other") mechanisms from each other to enable developing acceleration factors for each of the four DOE test procedures.
- Plans regarding lower catalyst loading and correlating results versus loading should be put into place, if they haven't been already. This will be key for automotive commercial viability.
- The operating conditions of bus stacks (air and hydrogen stoics) was never made clear, as well as the likelihood of anode starvation.
- The modeling goals were ambiguous.
- The approach is good. UTC is gathering real-world degradation data from their bus fleet and is attempting to match the conditions and failure modes in a laboratory environment in a short stack running the accelerated tests.
- The PGM loading in the bus fleet data is higher than the DOE targets. UTC will consider using different materials in the stack to better characterize the performance of newer state-of-the-art materials.
- The real world data may highlight several failure modes, but it may not reflect all modes. UTC needs to guard against introducing new failure mechanisms that are not seen in the real world.
- The project has a go/no-go decision point to determine if UTC can correlate the observed degradation with process conditions.
- The procedures seem obvious and expected.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.7 based on accomplishments.

- The beginning of the work seems good.
- Progress against the plan and milestones is fair, but I fear the project is falling a little behind.
- The project appears to be on schedule.
- Go/no-go decision point will be reached in July 2010, after which more solid progress assessments can be made.
- The cell voltage range in the bus operation was not revealed. The plot on slide five doesn't show values on the y-axis, therefore, the severity the real world bus voltage cycles was not presented.
- Regarding slide eight –explain the discrepancy in particle size and electrochemical area (ECA) results between X-ray diffraction (XRD) and ECA tests. How they correlate the measured 15 mV loss at 200 mA/cm² is an important question that was never answered. It seems like an ECA loss would result in greater mV loss, whereas an ECA calculated from XRD is more in line with mV loss.
- Referring to slide 11 an ECA loss of 70%, but only an 18mV loss @ 0.8 a/cm is surprising. Can these results be explained further with fundamentals?
- UTC appears to be making good progress in the short time the project has been funded. Characterization of the PGM decay in the bus fleet has been completed. ECA loss on the cathode is greater than the DOE targets, but the performance loss is less than the DOE targets. More analysis is needed to understand these results. UTC should subtract out the contribution to decay for startup and shutdown.
- The short stack has been fabricated and is installed on the UTC test stand.

- Catalyst decay seen in the accelerated testing at Los Alamos National Laboratory (LANL) agrees with that observed in the bus stacks after 2850 hours.
- The project is in its early phases. The results shown appear typical, similar to many earlier data pertaining to fuel cell performance loss.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated 2.7 for technology transfer and collaboration.

- Several experts are listed as collaborators.
- Apart from failure analysis at national laboratories, there is no collaboration outside of UTC and the relevance of their analysis of UTC field heavy duty data is limited outside of UTC.
- The project has a strong team; however, more information concerning LANL test capability (comparison to inhouse UTC data) is needed.
- Their collaboration with national laboratories is good. It is not clear from the presentation what role LANL will play in this project. Presumably, some of the AST testing will be done at LANL.
- Collaboration is evident and it seems the primary expertise is well leveraged.
- The collaborations are strong. LANL is performing the accelerated tests and the Oak Ridge National Laboratory (ORNL) is characterizing the degraded materials.
- Collaboration with the National Renewable Energy Laboratory (NREL) to compare bus fleet durability with light-duty vehicle (LDV) durability should be encouraged to the extent possible.
- This is an "internal" investigation (internal to UTC), and the collaborators seem more like contractors.

Question 5: Approach to and relevance of proposed future research

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

- The future plan reflects the good initial plan.
- It seems to me that the future work planned here is the implementation, within UTC, of an AST to enhance their capability for development of their heavy-duty product.
- It would be very valuable if additional stacks can be included in study. UTC needs better analysis of individual cell (within the stack) performance versus degradation.
- If the project is successful to make a correlation between the laboratory ASTs and the real world, then extensive ASTs will be performed or developed on the a UTC laboratory stack. It is unclear if there will be benchmarking of test data with other cell designs.
- It is not obvious how various experiments are going to be linked together.
- Future work is intended to lead to the development of a system-based test protocol that accelerates all relevant decay mechanisms in bus stacks.
- It is possible that improved understanding might result in identification of "danger signs," or other metrics that result from undue stress on working systems. Accelerated testing can be useful.

Strengths and weaknesses

Strengths

- UTC has a good team, with a good source of real world FCV data.
- The approach for reconciling and validating ASTs with real field data is commendable.
- UTC has a strong team using on-road tested hardware.
- The focus and approach of the team using real world degradation issues from bus fleet is a strength. Connecting the real world to the laboratory world is extremely important.
- Overall, they've done a nice job developing stack materials for their bus application, and this project is indirectly describing the key factors involved.
- The project is focused on providing a method to expedite material qualification for use in bus stacks that can meet the UTC target of 15,000-hour durability.

- The project will determine the applicability of the DOE accelerated test protocols for LDVs to bus operating conditions.
- To minimize the risk that the accelerated tests do not introduce a new mechanism that is not seen in the real world, UTC is trying to make the potential, temperature, RH, and time as realistic as possible. UTC also plans to screen the test results for a "reality check." The FreedomCAR (Cooperative Automotive Research) and Fuel Partnership (FreedomCAR) Fuel Cell Technical Team (FCTT) indicated a willingness to validate the UTC results.
- The go/no-go decision on task 1 will be made on the basis of whether UTC can correlate the degradation mechanisms to process conditions.
- UTC probably knows its stack well, and UTC has some excellent technical support.

Weaknesses

- The use of high loaded heavy-duty data for this means that the value is internal and does not filter down to meet the DOE research, development and demonstration targets.
- The project has produced questionable test results given only one stack. UTC indicated next-generation stack lifetime approximately double that of stack used in this analysis. If these stacks were tested, it would be interesting to know if whether or not the results would change substantially.
- The real-world stack is proprietary and little of the testing information will be shared. The project would be stronger if a connection to an open cell or stack design could be made.
- It is not clear to me that this is at all viable for light-duty automotive: high catalyst loadings, minimal RH and temperature cycling, no information on the number of starts or stops, air starts, freeze, etc. And it is not clear what the maximum voltage was on the voltage cycles. It is likely a very benign environment compared to what one would expect for passenger vehicles.
- The sample size is limited, will results be statistically significant?
- It is not clear how much of this work will apply to LDVs.
- Developing accelerated tests to project lifetimes for old material sets may not be totally applicable to newer materials and lower PGM loadings. Newer materials may have different failure modes.
- Existing engineering of similar fuel cell systems frequently showed that stack failure (or performance loss) can be the result of component failure, and a failed component stresses the stack in unanticipated ways. Consequently, there needs to be through system-wide system analysis, component by component. Components with short durability need to be exchanged, as required. It is also well understood that degradation processes proceed at a rate that vary with the log of voltage; i.e., a few minutes of poor system control can result in the equivalent of years of degradation, problems that would be totally avoided if appropriate conditions had been maintained. Therefore, accelerated testing indeed might work, but only after there is a considerable knowledge of chemical dynamics at the stack level, and the anticipated durability of all system components.

Specific recommendations and additions or deletions to the work scope

- To be relevant to the DOE and not just UTC, this project should be rescoped to light duty data acquired on itsbrass board with light duty relevant cycling and low loaded MEAs. UTC should partner with their catalyst-coated membrane supplier to carry out this work.
- UTC needs to add more stacks, if possible, and UTC needs to compare cell performance within overall stack and clearly identify stack history (number of faults, percentage of time at idle, etc.).
- Since the bus stacks are proprietary (including the planned laboratory stack for AST), there still needs to be a clear connection made to cells used for laboratory testing using standard hardware. The project's members should consider other cell/stack designs as well.
- Incorporate FCTT durability protocol (load and RH cycling).
- UTC should be encouraged to work with NREL to compare the durability results from the bus fleet with the durability for the LDV fleets in the Technology Validation Program.
- UTC should consider working with lower catalyst loadings. The higher loadings in the current fleet may mask the effect of airborne contaminants. However, the absence of recovery cycles in the test protocols will be a problem with lower platinum loadings. UTC should reconsider including performance recovery cycles, since this can help keep low platinum materials from being unnecessarily eliminated from consideration.
- UTC may need to run multiple accelerated tests with different stresses and failure modes in order to really get a fundamental cumulative damage model, in order to predict lifetime.

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• There needs to be some standard testing done to back up the proposed tests. Certainly, one year of quality testing takes the entire year, and that involves costs. However, without such a baseline, results might be of limited use. Other developers have learned this. Clearly, a company as well-endowed as UTC has necessary resources for product testing, especially because potential customers will be eager to review such results.

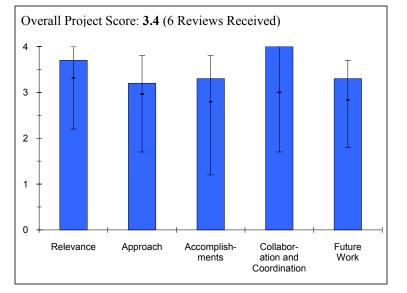
Project # FC-16: Accelerated Testing Validation

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 (GDLs), bipolar plates, and interfaces.

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>



This project earned a score of **3.7** for its relevance to DOE objectives.

- The objectives of the project are very important to be able to better predict the degradation of the fuel cell (fuel cell) and to propose improved materials.
- The relevance to DOE objectives is well defined. If the project is successful in validating ASTs by comparison with real-world data and understanding mechanisms, then there will be a good foundation for developing mitigation strategies which will help to achieve DOE's durability targets.
- This project is utilizing relevant materials. However, it is not clear yet how degradation in buses correlates to automotive applications.
- Having standardized AST protocols is very useful for the development of advanced materials, and it is important that these ASTs be validated with real-world data.
- This project, while new (August 2009), combined with Tom Madden's project at United Technologies Corporation (UTC) and Rod Borup's project at LANL (many of which have the same collaborators, particularly LANL), is critical to the Hydrogen Program as it evaluates AST for membrane electrode assemblies (MEAs) from Ballard Power Systems pressurized gas bus systems. Further, since only bus systems are available, the program is working with W.L. Gore & Associates (Gore) and Ion Power to develop automotive MEAs and subject them to an LANL-developed accelerated drive schedule to determine if current AST standards are sufficient. ORNL is providing metal stacks and LBNL is performing the modeling. Madden's program at UTC looks at ASTs developed from buses operated at atmospheres of pressure. Therefore, the DOE R,D&D objectives are met with all currently commercially operating fuel cell vehicles, other than forklifts.
- This project is critically important for the fuel cell developer community.
- Validating ASTs with real-world data and correlation will enable future testing to provide more confidence to developers that they are emulating the major degradation mechanisms observed in the real world.

Question 2: Approach to performing the research and development

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

- The approach is in accordance with the project targets.
- The parallel approach between "real-world" data and laboratory testing is adequate.
- Real-world fuel cell data is expensive to obtain, so it is understandable that this project cannot rely on a fleet of cars to get real-world durability. However, a bus system may have significantly different stressors compared to a light-duty vehicle (LDV). For this reason, it will be important to thoroughly compare the operating protocols

of the real-world bus with the target operating protocols of light-duty vehicles for which the current ASTs were developed.

- The overall approach is logical and the scope seems reasonable. The use of low catalyst loadings is critical to the development of ASTs. The justification for developing GDL ASTs is not clear, especially if one looks beyond LANL results. Also, the bus data seem limited. It will be interesting to note if the results are statistically significant.
- This project appears to have a good plan to start to validate the ASTs. This will be challenging due to the many differences in real-world applications; e.g., stack and system designs, operating conditions, material sets, etc. Therefore, it is probably good that a single laboratory, LANL, is involved in both AST-validation projects.
- The project is sharply focused on technical barriers, as degradation mechanisms within operating MEAs are not well understood. LANL is trying to develop mitigation strategies, while simultaneously meeting cost and durability targets.
- The approach seems solid, although there is some question of whether applying ASTs developed for LDVs and validating them with fuel cell bus data is the best approach.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.3 based on accomplishments.

- The obtained results are numerous and of quality.
- It will have to be checked that the correlation between electrochemical surface area (ECSA) and performance loss is not only true for the tested material as other presentations showed much worse correlation.
- The project is in its first year and the electrochemical-diagnostics data presented demonstrate that the project is off to a good start.
- The team has made good progress for less than a year of work. The relative humidity (RH) cycling results have good correlation with experiments by other groups.
- Excellent amount of results presented for a project that recently got underway. Results are also well presented.
- The program has just started and progress to date has been concentrated on the bus data. Some AST data have been verified, while others, like catalyst cycling, impedance and carbon corrosion, impedance and RH cycling metrics, and characterization need further work to verify or develop new ASTs.
- Despite the recent start on this project (August 2009), it appears as though a significant amount of the laboratory work has already been completed at LANL (unless results presented were from a previous project).

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **4.0** for technology transfer and collaboration.

- The collaboration between the partners seems to work well.
- PI should maintain close communication with Ford and General Motors (GM) to make sure that progress remains focused on DOE goals for LDVs. Start/stop durability can be highly dependent on stack design and system protocol in addition to MEA materials. LANL should solicit input from industry and consider implications of different operating strategies while developing a start/stop AST.
- The project appears to be well coordinated, with a good balance of work for the various partners.
- It is excellent that the project includes a MEA supplier, Ion Power, that allows full analysis of this key component. It is unfortunate that there are restrictions on analysis of some key components that may be the actual materials used in fielded power plants.
- The collaborations are excellent; with great partners and other LANL investigators such as Rod Borup, Ion Power, Gore, LBNL for modeling, Ballard, ORNL for metal stacks, and GM provided protocols for cell life testing, etc.
- The PI has assembled an outstanding team, including Ballard, LBNL, Ion Power, ORNL, and Gore.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- The proposed work is in accordance with the project objectives.
- The work plan is well laid out and focuses resources on the appropriate activities.
- The impact of temperature and RH on the acceleration factor should be looked at, if it has not been already.
- It is not clear how dropping freeze investigations and substituting forklift data improves the scope of the project. Adding forklift data will most likely add more uncertainty.
- This reviewer feels that if the team stays away from freeze data points while sticking to AST testing by initiating combined mechanical and chemical testing of membranes, catalyst cycling, and GDLs, it will have a successful project. LANL should continue cell-life testing (accelerated drive cycles), correlate ASTs to life data using LBNL modeling (already underway), and develop new ASTs where appropriate, keeping in mind the goal is to develop mitigation strategies while simultaneously meeting cost and durability targets.
- As this project is still ramping up, there is much future work to be done.
- Details of future work were light (only listed at task level), but based on the clear approach of the project they appear to be appropriate.

Strengths and weaknesses

Strengths

- The project is well organized with a good partnership.
- There is a strong team with coordinated experiments that is focused (in part) on relevant materials.
- There is a good plan and a good team, and it is good that additional components (metal plates, Ion Power MEAs) are included.
- Excellent collaborations, goals, and objectives.
- Good partnering with industry.
- The project is focused on a key enabler for fuel cell developers to validate accelerated testing of new materials in the laboratory and proceed to the marketplace more quickly.

Weaknesses

- Activity on bipolar plates should be more precise.
- Real-world data should deal with a bus application. As the lifetime of systems in buses is longer than that in LDVs, how will it be ensured that the results will also be applicable to automotive cars?
- It is not clear if bus data is relevant to automotive ASTs and this project partly depends on this.
- It is unfortunate that some of the future "field data" will be just from running simulated bus cycles, not real buses.
- It is unfortunate that the catalyst loadings are even higher (1 mg/cm²) than the other AST-validation project. No real-world data will be obtained on low loaded MEAs.
- •
- Using bus on-road data to validate ASTs designed for cars is a weakness.

Specific recommendations and additions or deletions to the work scope

- Stay away from freeze data.
- As freeze data are no more concerned regarding the tech team proposal, analyzing fork lift data is fine.
- As many stationary systems are commercialized today, comparable data should also be obtained under stationary conditions dealing with either neat or reformed hydrogen.
- There should be less emphasis on GDL degradation.
- Forklift data may complicate an already difficult task.
- The project should include additional diagnostics (such as IV curves with different oxygen partial pressures, as suggested by an audience member) to obtain additional insight into source of performance losses, e.g., oxygen transport vs. ohmic losses in cathode.
- Follow the recommendations of the fuel cell TT.
- Recommend partnering with a light-duty fuel cell manufacturer (Automotive Fuel Cell Cooperation, GM, etc.) to obtain related on-road data from cars to do similar validation of the ASTs.

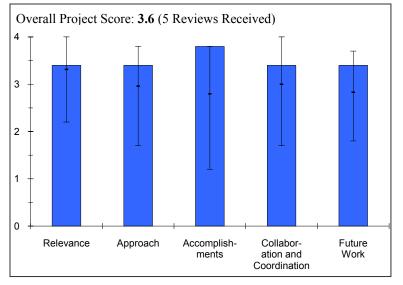
Project # FC-17: Fuel Cells Systems Analysis

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 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 highvolume manufacturing cost estimation.

<u>**Ouestion 1: Relevance to overall DOE**</u> <u>objectives</u>



This project earned a score of **3.4** for its relevance to DOE objectives.

- This is perhaps the best fuel cell model developed. The 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. This project is critical to the Hydrogen Program and DOE RD&D objectives as it addresses the key targets for performance, system thermal and water management, start-up and shut-down time, transient operation, and cost.
- The relevance is rated "good," not "excellent," for the reason that, unlike many other projects in the program, the results of this project will not be directly useful to end developers of fuel cells. Such end developers already have their own system analyses. However, this project does fulfill an informational role that benefits DOE, and, to that extent, it is relevant. The project feeds information to the cost analyses, and therefore has a "second order relevance" to end developers.
- The PI has been responsive to reviewer feedback and has revised its component selection for this year. The PI's willingness to do so has enhanced its relevance.
- The project has made very good progress over the last few years and offers an integrated-systems look at the fuel cell and importantly contributes to the overall understanding of the fuel cell system problem. This project provides a base for comparing most of the DOE program goals and objectives.
- This project probably has the most value in how it establishes a detailed fuel cell that can be used by Directed Technologies, Inc. (DTI) and TIAX, LLC for their annual fuel cell cost projection updates.
- This project may not be very relevant to the industry partners, as it relates to a generic fuel cell that does not represent (in detail) the system of any particular company.

Question 2: Approach to performing the research and development

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

• The technical barriers are cost, performance, system thermal and water management, air management, startup and shutdown times, and energy/transient operation. The approach to solving these barriers are to develop, document, and make available versatile system design and analysis tools existing at ANL, to validate the models against data obtained in laboratories in collaboration with external organizations, and to apply the model to issues of current interest by working with the Fuel Cell Technical Team and with DOE-requested contractors. While validation of system models still needs to be done, this project's approach has made a considerable improvement in the past year. First of all, the system concepts are much more up-to-date, as evidenced by the

removal of anode humidification and the option to study lower stack inlet pressure. Temperature differences across the stack are now recognized as well. Second, the models now recognize that low power conditions play a role in determining system components and sizes (which applies to air compressors, recirculation blowers, and humidifiers), which has made for a considerable and long-awaiting improvement in the methodology. The investigators are to be applauded for these changes.

- The approach is solid and attempts to bring the best data available and approaches to understand the system issues. The validation of the impurity models and data is very important. The breakout of the different operating conditions for pressure, Pt loading, water management, etc., is informative. I think that the approaches being used are very good and demonstrate a systematic, stepwise evaluation of the system.
- The approach of validating GCTool with in-house testing data is good.
- Interactions with Fuel Cell TT and DOE contractors are necessary to keep this project focused on relevant fuel cells and issues of interest.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.8 based on accomplishments.

- The model has been successfully utilized to operate a stack at 1.5 atm without a cathode humidifier, but with hydrogen recirculation ejectors and a hydrogen blower using 3M's new 20 µm membrane with 0.05 g/cm²(anode)/0.10 g/cm²(cathode) PtCoMn catalyst. The membrane results agreed well with 3M's single cell testing. Also, the model predicted the water and air management results with turndown, the size of the radiator resulting in an increase in stack operating temperature at maximum power, except for the S-3 stack. The model also predicted that lowering the full load efficiency to 40% decreases the peak efficiency by only 1% and increases the battery charging mode fuel economy by 4%. There was a general consensus in the room that the project met all of the concerns expressed last year.
- Even though validation still remains to be carried out, excellent progress has been made. Compressor turndown is now defined in the context of surge limitations. The model provides the flexibility of numerous fuel subsystem configurations with dual ejectors and variable geometry ejectors included. Furthermore, the model attempts to provide detail on which power densities ejectors and/or blowers are needed. With proper assumptions, this will be useful to DOE to be able to figure out whether fuel cell developers are using blowers or ejectors or eliminating certain components.
- There is still progress to be made in validating component assumptions, extending the model to other operating regimes, e.g., low temperature, and reviewing battery charging mode assumptions, but it can still be said that a large improvement has been made in this project.
- It was good to see the enthalpy wheel finally removed from the system. The data are defining the compressor expander motor (CEM) module and providing a system view of the effect of catalyst loading. Good progress appears to be made on all topics; however, the level of activity is so high that I am having difficulty assimilating all of the progress.
- Significant updates to the system model were made this year, enabling the system to be more representative of a typical fuel cell in industry.
- Updates were key for TIAX and DTI to have improved fuel cell cost projections.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.4** for technology transfer and collaboration.

- Collaborators have been expanded to include 3M, LANL, and DTI. The collaboration is appropriate and well coordinated and the partners are full participants. Vairex International need not be consulted regarding the CEM unit.
- The collaboration with two particular partners--3M and Honeywell--is very good, but the PI must be careful not to become too dependent on these organizations. In principle, there should still be inputs from alternative component suppliers (although given the uniqueness of nanostructured thin film (NSTF), alternative suppliers are not necessary for now to understand the catalyst layer).
- The project could benefit from seeking alternative inputs on humidifier membranes. The focus should be on finding performance for lower cost membranes, so this could be done collaboratively with DTI or TIAX.

- The experience of ANL results in recognition of the top groups to team with.
- There is a high level of interaction and collaboration with Honeywell.
- It would be nice to see more direct and/or ongoing interaction with the original equipment manufacturers (OEMs) outside of the FreedomCAR (Cooperative Automotive Research) and Fuel Partnership's Fuel Cell Technical Team (FCTT) forum. The possibility that ANL could model GM's actual next generation system and add value into the process for the subsequent generation is intriguing.

Question 5: Approach to and relevance of proposed future research

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

- The future work, of course, builds on prior results. The system analysis supports the DOE/FreedomCAR development efforts. The model has been used to verify the performance of 3M's membrane/catalyst at low loadings and pressure, but collaborations continue with 3M regarding durability of their electrode structures. The future work remains focused on the barriers listed for the project. The project remains critical to the Hydrogen Program and the relevant DOE RD&D objectives, as this is the relevant systems model that others are adapting. It is also the model that DTI and TIAX use for future costing of fuel cell systems.
- ANL has the ambition to take on drive cycle and durability modeling, but further validation and optimization of the present model (most likely at component level) should come first.
- A drive cycle or durability model will require much deeper knowledge of the stack that has been presented here, but might be possible in collaboration with some of the new durability projects. For this, a work plan is in order.
- It is understood that stack performance mapping with 3M single cell data has been done. A prerequisite to some of the future work should be that this performance map should be presented and reviewed to see whether the map can extend to operating regions that might be experienced during transients.
- The future work is consistent with the objectives and there was no information presented that indicates some of the efforts should be terminated.
- Of all the future work, the most important is the drive cycle simulations for durability enhancement. Unless the real-world cyclic nature of fuel cell operation is included, changes to the model could be making dramatic (and negative) impacts on the durability of the system. OEMs will already know this, but it will lead to a modeled projection of cost that is cheaper than what is actually possible.

Strengths and weaknesses

Strengths

- The system model is very powerful and applicable to the work of other collaborators and non-collaborators. Stack and system designers have been brought on board.
- ANL has made the system concepts much more contemporary.
- ANL has begun to model components throughout the range of power density required during operation. As a result, the PI has developed a much better understanding of system component selection based on phenomena that occur at low power operation.
- The PI's experience and the progress this project has made over the last several years demonstrate the continued growth in expertise and fuel cell leadership.
- Backbone of defining a reasonable and detailed fuel cell to enable parallel cost projections to be made by TIAX and DTI.
- Some interesting insights can be learned by exercising the model in various different ways.

Weaknesses

- Perhaps a little more modeling on cathode air-borne contaminants should be included.
- The model still requires validation in many areas and some system components should be studied further to see whether the representations in the model are the most competitive versions possible.
- Use of 3M NSTF provides a low loading catalyst, but it also implies some limitations in terms of operating conditions, particularly at low temperatures.
- The future work section indicates that the project will attempt to address durability and drive cycles. However, some restraint is needed to be sure the present model is validated and that collaboration on such matters will occur with the new durability projects.

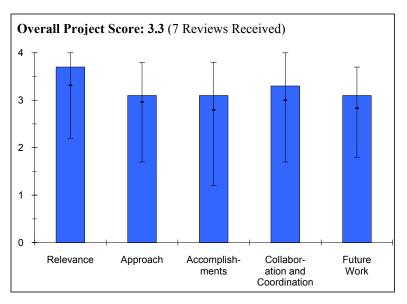
- The model does not represent an actual fuel cell operating anywhere, so complete model validation is not possible.
- ANL needs to make sure model queries are focused on the key issues facing fuel cell developers within the OEM's programs.

- No deletions but perhaps a little more emphasis on modeling the effects of cathode air-borne contaminants.
- Other collaborators on system and stack should be added beyond Honeywell and 3M. The project should be able to gain a competitive understanding of what components could be available.
- Durability and drive cycle modeling should be restrained until after performance model is validated.
- Some analysis should be made of the stack performance map to see whether it agrees with a more phenomenologically-based performance map (with science-based relationships with partial pressures, relative humidity, temperature, etc.).
- Some analysis may be of interest to see whether the stoich / voltage relationship from 3M single-cell testing would be applicable for a stack. Single-cell stoich sensitivity is usually different (less sensitive) than that for a full stack module.
- The complexity and maturity of this project makes it hard to evaluate using the AMR format. This project has progressed so far that a separate review is recommended (a review that could take several hours and would include a preliminary report to assist the reviewers' understanding of the project progress).
- Closer collaboration with OEM programs on actual system designs and tradeoffs within the model that will affect future product design would be helpful.
- Make a version of the model to enable analysis of fuel cell forklifts, as this is an area where the fuel cell developers could probably use some help with the issues they are finding from real-world product placement.

Project # FC-18: Mass-Production Cost Estimation for Automotive Fuel Cell Systems *Brian James; Directed Technologies, Inc.*

Brief Summary of Project

The overall objectives of this project are to: 1) identify the lowest cost system design and manufacturing methods for an 80-kW direct-hydrogen automotive polymer electroylte membrane fuel cell system based on two technology levels, current technology and 2015 projected technology; 2) determine costs for these tech level systems at varying production rates from 1,000 to 500,000 vehicles per year; and 3) analyze, quantify and document impact of system performance on cost, using cost results to guide future component development.



Question 1: Relevance to overall DOE objectives

This project earned a score of 3.7 for its relevance to DOE objectives.

- It is the only publically available source for discussing costs. DOE needs these programs to discuss future R&D directions and policies.
- The goals of this project are in line with the DOE objectives. This project provides valuable insight into the cost projections of current and future technology. These costs would otherwise be very difficult to estimate based on the limited amount of information that is disclosed by industry.
- This is one of two projects that seek to realistically assess the mass production cost for a prospective automotive fuel cell systems. The highly detailed research of this project is essential for informing overall policy on the realistic prospects that fuel cells will be an affordable alternative to other power plants; e.g., batteries and internal combustion engines. This project integrates the best available engineering and design knowledge it has developed over the years, particularly with respect to the membrane assembly and the balance of plant (BOP), to provide a reference fuel cell system. This focuses planning and elucidates what additional research might be done to further reduce costs.
- The cost model is useful for cost-benefit analysis.
- This attacks key problems for implementation and does so in a well-aligned way.
- This is a continuing program. Directed Technologies, Inc. (DTI), unlike TIAX LLC, uses extensive interactions with industry and researchers to solicit design and manufacturing metrics as their input to their cost analysis. TIAX, on the other hand, uses the latest model from Argonne National Laboratory (ANL) for their cost analysis. Thus, the Hydrogen Program and the DOE research, development and demonstration (RD&D) objectives have two independent cost studies to draw upon. DTI uses the cost study to guide future component development.

Question 2: Approach to performing the research and development

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

• Both cost studies are converging on more automotive representative systems. Generally, DTI does a great job of aggregating all the known manufacturing techniques and suppliers. They do a good job of using the limited and highly contrasting information provided to them. If improvement is possible, PI could offer suggestions for advanced, new, or upcoming techniques. The PI should not invest much effort into this analysis or include it in the cost summary since it could be highly speculative, but offering thought provokers would be helpful.

- It is helpful that they switched to 3M nanostructured thin film (NSTF) design in 2009 instead of an average system.
- Assumptions regarding the practicality of "current (2010) status technology" should be more clearly stated in the presentation.
- The approach seems to be good, but it is not transparent. It would be beneficial to know what came out of the talks with industry and what those talks were about.
- The PIs laboriously developed a reference design in consultation with the national laboratories; e.g., ANL, and leading firms developing fuel cells (fuel cells) and their components.
- Well-established methodology (e.g., DFMA) is used to estimate materials and manufacturing costs.
- They are taking a reasonable approach. There are some differences between ANL mechanization and the mechanization used for this study.
- There is a good level of detail in price estimation. It could be better if at least an order of magnitude costing were provided for the excluded cost items.
- They use suitable assumptions where they must.
- They did not look into improvements in supplying industries and assembling techniques, which means estimates of true steady state cost may still be high. This is a place for continuous improvement.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.1** based on accomplishments.

- The model is now entering into the refinement stages. They continue to converge on realistic automotive designs.
- The model has improved the accuracy of its system and performance assumptions since last year.
- For this project, accomplishment is an accurate assessment of the state of the technology, regardless of the DOE goals. They seem to be doing a credible job of it. Their findings are that the DOE goals are "nearly" met for 2010, but 2015 goals are overly optimistic (slide 23). Their continued refinement of the model (slide seven) demonstrates excellent progress towards their charge of pricing the lowest cost design consistent with good engineering practice.
- The major components of cost model are complete and have been refined.
- Their progress is quite suitable, and it is in useful areas.
- The effect of scale is useful in avoiding work on areas that will not matter at scale.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.3** for technology transfer and collaboration.

- PI regularly seeks input from vehicle and stack OEMs. They seek information from the appropriate suppliers and manufacturing experts
- DTI could benefit from collaboration.
- DTI has done well with the challenge of working with industry partners and publicly available process information to build accurate cost models.
- By its nature, this work is highly collaborative. The PIs seem to be doing an excellent job of seeking out information from laboratories; e.g., ANL, and relevant firms (fuel cell producers and their suppliers; e.g., Nuvera, Ballard Power Systems, 3M, and Honeywell) and, in turn, vetting their results with them before publication.
- This work could have much more impact if the workshops were held where the original equipment manufacturers (OEMs) interact with the model on their own and use the model to improve their system tradeoff studies. This should be done not at a gross-cost number \$70/kW, but at a detailed component and subsystem level.
- They chose good institutions to work with and worked with them closely.
- The work with ANL is critical, too.

Question 5: Approach to and relevance of proposed future research

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

- Lifecycle cost analysis would be a useful addition to the project. Particularly, results from the recycling studies could be integrated into understanding true total cost of ownership of these systems.
- It is not clear that the 2010 technology would work as predicted. Practical demonstration of robust performance and degradation rates; i.e., freeze-start, and voltage cycling stability of alloy catalysts, will be important to validate the assumptions needed for a meaningful lifecycle cost analysis.
- Investigation of recycling cost and issues might be added.
- The PI's attention to detail extends to their future plans, which are laid out on slide 25. They intend to explore design tradeoffs such as operating pressure vs. catalyst cost, and the effect on the system price of quality control in more depth.
- One would like to see a workshop where OEMs interact directly with the model.
- The approach and relevance of proposed future work is suitable.

Strengths and weaknesses

Strengths

- 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 technical approach and use of OEM input are both strengths.
- Working with manufacturers ties the program to the real world.
- Have one of the best DFMA analyses of all the contractors. They are sharply focused on costs and barriers.

Weaknesses

- One may consider the reference design separate from the pricing of it. Further validation of the pricing can be had by encouraging others to price the reference model.
- There appear to be no apparent provisions for detailed use of the model by OEMs for developing lower cost system architectures.
- They are not looking far enough toward possible improvements in component and raw material costs, and so they are not getting as good an estimate of the long-term high-volume cost as possible, which is what is really needed.

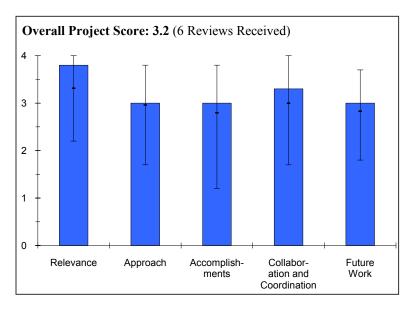
- A collaboration with a national laboratory could provide the data needed to validate the 2010 technology assumptions.
- A sensitivity analysis of membrane thickness is also recommended. Inefficiencies due to gas crossover should be balanced with the benefits of increased water back diffusion and reduction in the total ionomer cost.
- Strongly recommend adding workshops where OEMs can directly interact with the model to better understand internal subsystem and component cost trades.

Project # FC-19: Direct Hydrogen PEM fuel cell Manufacturing Cost Estimation for Automotive Applications

Jayanti Sinha; TIAX, LLC

Brief Summary of Project

The overall objective of this project is to conduct bottom-up manufacturing cost assessment of an 80-kW direct-hydrogen polymer electrolyte membrane fuel cell system for automotive applications. Objectives for 2009 include: 1) a high volume (500,000 units/year) cost projection of a current (2009) fuel cell system configuration, 2) independent peer review of cost analysis methodology and results, and 3) comprehensive report on the 2008 fuel cell cost analysis. Objectives for 2010 include a high-volume cost projection of a 2010 fuel cell system configuration.



<u>Ouestion 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of 3.8 for its relevance to DOE objectives.

- It is the only publically available source for discussing costs. DOE needs these programs to discuss future R&D directions and policies.
- This project provides valuable insight into the cost projections of current and future technology. These costs would otherwise be very difficult to estimate based on the limited amount of information that is disclosed by industry.
- This is one of two projects that seek to realistically assess the mass production cost for a prospective automotive fuel cell system. The highly detailed research of this project is essential for informing overall policy on the realistic prospects that fuel cells will be an affordable alternative to other power plants; e.g., batteries and internal combustion engines. This project integrates the best available engineering and design knowledge as it has developed over the years, particularly with respect to the membrane assembly, the bipolar plates (stamped and nitrated, as per ORNL), and the balance of plant (BOP), to provide a reference fuel cell. This focuses planning and elucidates what additional research might be done to further reduce costs.
- The cost model is useful for cost-benefit analysis.
- TIAX LLC works closely with ANL; in fact they collaborate with ANL on the system configuration and modeling and they receive feedback from the FreedomCAR (Cooperative Automotive Research) and Fuel Partnership (FreedomCAR) Technology Team (FCTT), Developers and Vendors. TIAX uses a bottom-up DFMA® approach, using a high volume scenario. This reviewer believes that it is necessary to have two independent companies working on the cost as they now use approximately the same system and the work is highly relevant to the DOE RD&D objectives and the Hydrogen Program.

Question 2: Approach to performing the research and development

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

• Both cost studies are converging on more automotive representative systems. Generally, TIAX does a great job of aggregating all the known manufacturing techniques and suppliers. They do a good job of using the limited and highly contrasting information provided to them. If improvement is possible, PI could offer suggestions for advanced, new, or upcoming techniques. PI should not invest much effort into this analysis or include it in the cost summary since it could be highly speculative, but offering thought provokers would be helpful.

- Collaboration with ANL to validate the stack performance assumptions is a strength.
- The PIs laboriously develop a reference design in close consultation with the national laboratories; e.g., ANL, ORNL, and leading firms developing fuel cells and their components. Solid methodology is used to estimate materials and manufacturing costs.
- The costing method used for estimating motor costs (catalog cost of dissimilar technology; e.g., single phase ac motor versus three phase dc brushless motor) is deficient and unlikely to produce meaningful, accurate results. This raises concern that: 1) other cost numbers may suffer from a similar inappropriate approach, 2) the cost report was not reviewed by suitable experts, and 3) the internal review process in the project team is insufficient.
- The utilization of the ANL system, which is approved by most of the OEMs and their vendors, lends credence to the TIAX selection and sizing of components. It was stated that the ANL system is thermodynamically correct and thus TIAX is costing an actual working system. TIAX also uses a sensitivity analysis now in response to one of the reviewer's remarks from last year. The ANL stack uses industry and vendor input and attempts to design a system using the minimum and most efficient components available. TIAX then costs this stack.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.0** based on accomplishments.

- The model is now entering into the refinement stages. They continue to converge on realistic automotive designs.
- The model has improved the accuracy of its system and performance assumptions since last year.
- For this project, accomplishment is an accurate assessment of the state of the technology, regardless of DOE goals. They seem to be doing a credible job of it. This study emphasizes multiple "scenarios" and variability due to uncertainties.
- It is surprising to have such a significant approach error this late in the project.
- TIAX has made excellent progress toward meeting DOE objectives; using the ANL system as a costing
 mechanism suggests that the cost barriers are close to being met. Both the stack and the BOP components have
 been updated since the last review. This costing is not supposed to move the technology, but state what the best
 technology costs are. Good collaboration with ANL, OEMs, other developers, and vendors; keeps the costing
 accurate. Based on the 2009 ANL model, TIAX's cost was \$55.2/kW. This cost is within the error bound of
 DTI's cost of \$51.30/kW for essentially the same system. DTI has perhaps more experience with DFMA than
 TIAX does.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.3** for technology transfer and collaboration.

- PI regularly seeks input from vehicle and stack OEMs. They seek information from the appropriate suppliers and manufacturing experts.
- Partnership with TIAX is particularly useful since it provides better understanding of system sensitivities TIAX is working with several well-informed consultants: Ballard Power Systems (Ballard), Ford, the Oak Ridge National Laboratory (ORNL), and ANL. Collaboration with ANL is crucial to achieve good results—well done!
- By its nature, this work is highly collaborative. The PIs seem to be doing an excellent job of seeking out information from laboratories; e.g., ANL, ORNL, and relevant firms regarding fuel cell production and supply (such as Ford and Ballard), and vetting their results with them before publication.
- The project results do not appear to have been vetted by an expert review panel prior to presentation at DOE AMR.
- This work could have more impact if workshops were held where the OEMs interact with the model on their own and use the model to improve their system tradeoff studies.
- There appears to have been no communication between this cost study team and the other nearly identical cost study team. In the future, such interactions could benefit DOE by uncovering gaps and inconsistencies as noted, as well as lead to better cost models for the customer (DOE).

• TIAX has a close relationship with the ANL system and costs it. They also receive feedback from DOE and the FreedomCAR Fuel Cell Technical Team, developers, and vendors.

<u>Question 5: Approach to and relevance of proposed future research</u>

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

- Their approach and it's relevance is appropriate. I believe that increased focus will be needed on BOP components as they are increasingly representing a larger percentage of the system cost. This may prove challenging, as BOP components are increasingly dependent on proprietary system designs.
- They should focus on completing the BOP cost analysis.
- Should we be considering different configurations for different degrees of configuration–ultimately yes, but for now it is important to stick to the targets until the baseline configuration changes (partnership working on targets currently).
- Bottom-up cost projection for stack conditioning was the only major task proposed in the future work. Quality control analysis and lifecycle cost analysis could be valuable future tasks.
- Investigation of recycling issues and cost might need to be added to the project.
- These cost estimates are a continually evolving thing as reference designs improve. The next steps (slide 32,33) indicate that future work will integrate recent information about reinforced membranes, non-woven gas diffusion layers, and stamped metal bipolar plates. In addition, cost projection for stack conditioning is a priority item.
- There appears to be no discussion in the future work of where said work will be published, what reports are forthcoming, and there are no presented plans for having OEMs interact with or utilize this model. This is a significant deficiency of the stated future work.
- TIAX will use the newest available ANL stack and BOP system and cost it using DFMA and input from DOE, the FreedomCAR Fuel Cell Technical Team, developers, and vendors.

Strengths and weaknesses

Strengths

- 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 a current state-of-the-art (SOA) program. Further, the detailed analysis highlights what areas continue to need attention to further reduce costs.
- The development of bottom-up cost for stack components is a strength.
- TIAX uses the latest ANL stack and BOP system (which is thermodynamically correct) and performs a DFMA cost study on this stack. This DFMA is reviewed by DOE, the Fuel Cell Technical Team, developers, and vendors.

Weaknesses

- The format of the data presentation makes it difficult to see the yearly trend in price predictions.
- There were inappropriate/insufficient costing methods for some components.
- I think that TIAX is not as strong in DFMA as is DTI.

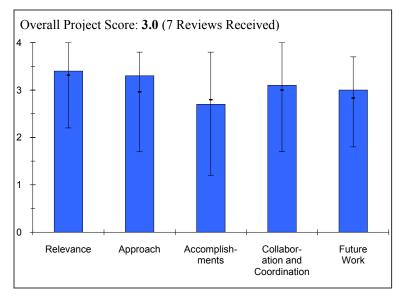
- A sensitivity analysis of membrane thickness is recommended. Inefficiencies due to gas crossover should be balanced with the benefits of increased water back-diffusion and reduction in the total ionomer cost.
- Add workshops where OEMs can interact with the model and use it to improve system and subsystem architecture tradeoffs.
- TIAX could perhaps bring on personnel who have a strong background in DFMA.

Project # FC-20: Microstructural Characterization of PEM 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, and unique specimen preparation methodologies, for the µ-Åscale characterization of the material constituents comprising fuel cells (catalyst, support, membrane); 2) apply advanced analytical and imaging techniques for the evaluation of microstructural and microchemical changes that correlate with fuel cell performance; 3) elucidate microstructure-related degradation mechanisms contributing to fuel cell performance loss; and 4) make techniques and expertise available to fuel cell researchers outside of the ORNL.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.4** for its relevance to DOE objectives.

- This project provides a vital service to the DOE projects in developing and refining high-resolution transmission electron microscopy (HRTEM) and scanning transmission electron microscopy (STEM) methods for characterization of fuel cell materials.
- This project is very relevant to DOE's RD&D plan objectives. Most of the work in this project is focused towards DOE's goal. However, the relevancy of statistical data analysis methods for nanoparticle analysis is a bit farfetched for this project. The analysis of carbon corrosion and Pt migration through the membrane to form Pt bands is a known phenomenon. Analytical method development to understand these behaviors is necessary to understand MEA decay/durability effects and is relevant to DOE's goal.
- Novel imaging techniques accelerate our learning of fuel cell degradation mechanisms and novel materials characterization.
- Clearly interesting—but I still don't think I have heard anyone say, "because of what we learned in the TEM, we made XYZ change to our materials/processes/operations and now our performance/durability is X% better than it was." The new batch of durability programs is clearly providing an opportunity to change this.
- The characterization of fuel cell materials is very useful for materials' durability and performance analysis. It provides guidance to the catalyst and support synthesis, as well as membrane development.
- The comprehensive techniques really help to correlate fuel cell performance with material structures.
- For the core technology of fuel cells, i.e., MEAs, it is extremely important to explore the catalyst active sites, to maintain the active structure of catalysts, and to know how to avoid the degradation of fuel cell components.
- The fuel cell community will benefit from the achievements of the project.

Question 2: Approach to performing the research and development

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

• I feel that the focus on carbon supports is lagging behind a considerable effort in alternative, non-carbon supports. This would require the project to embark upon leading research on its own rather than wait for a client to ask for something that would be a significant paradigm shift.

- The use of electron microscopy techniques for the analysis of MEA components is a good approach. Investigation of carbon corrosion mechanism and Pt migration is necessary to understand the durability behavior of MEAs. However, the reason for the approach of developing a method for statistical data analysis to locate elements within the nanoparticles is not evident from the presentation. Further explanation of the future use of this statistical data analysis method would aid the team in understanding the benefit of this approach.
- The PI has many characterization methods, which is good.
- ORNL develops tools and conducts measurements to meet the needs of the various projects it supports.
- This user facility provides needed analysis for the fuel cell community.
- I strongly suggest dropping the *in situ* TEM effort. From my point of view, I see two years of method development with no result, and even if they are successful in the future, I highly doubt the relevance of the results. It will not be *in situ*. You may be able to apply a particular potential to a sample prepared for TEM, but that sample will not be the original intact material in the real three-dimensional fuel cell environment. Also, since you really cannot see the ionomer in the TEM, I think you are missing any information that might be gained by an *in situ* study. I think we are better off accepting that post-mortem analysis is the best we can do and focus on ways to make those studies yield the maximum information that they can.
- The development of the techniques allows us to collect valuable information about the fuel cell components. However, it is also critical to recognize industry needs and apply the techniques to address the issues in industry.
- The application of statistical analysis is extremely important for the nanoscale characterization. The case study of failure mechanism is strongly recommended to be high priority.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.7 based on accomplishments.

- The images and data from this project are globally state-of-the-art (SOA). No other group is currently producing such high quality data, although several other groups now have the same technical capability.
- The team consists of a good mix of industry, academia, and National Laboratory partners, and good interaction between these partners is expected in this project. However, not much has been mentioned or discussed about the joint activities that had been done last year. A brief explanation of the contributions from and to the other partners will help in understanding the nature of partnership and the extent of collaboration between the partners in the team. Any opportunity for technology transfer between ORNL and other partners should be envisioned and explored.
- There were many results, but not many conclusions.
- No novel tools were developed this year, but ORNL's support has been critical for several projects, particularly those focusing on understanding carbon corrosion and Pt distribution in the ionomer.
- The images are excellent and they identify the physical mechanisms occurring during the carbon corrosion on the cathode.
- Based on the data presented, the PI should be able to begin at least speculating on the chemical mechanism occurring for Pt migration. ORNL limited it to the physical process.
- The mechanism for carbon corrosion was proposed.
- It would be interesting for the activity's PIs to hypothesize on how carbon corrodes and Pt migrates.
- While the results presented are interesting, looking at one Pt alloy catalyst or one carbon support isn't good enough to say you have gained real understanding. Without comparison to either the same materials after different treatments, or comparisons to different, but similarly treated materials, what have you really learned? I know this team wants to do those studies, and instead of trying to do a little bit of work with everyone, they may need to focus their efforts instead.
- Liquid STEM technique development has encountered the understandable obstacles.
- The project has really moved in the right direction; i.e., using statistical data analysis methods for nanoparticle analysis. The sample selection should also use a statistical method.
- The carbon corrosion mechanism study has been carried out using electrochemically-aged MEAs. It would be beneficial to continue creating new protocols to mimic the cases of MEAs in the cells, in particular, the non-uniform temperature distribution or current distribution.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.1** for technology transfer and collaboration.

- As what is essentially a service project to the industry, this project meets its objectives in providing that service to all.
- Opportunities for more cutting edge, exploratory research should be explored and encouraged.
- The team consists of a good mix of industry, academic, and National Laboratory partners, and good interaction between these partners is expected in this project. However, not much has been mentioned or discussed about the joint activities that had been done last year. A brief explanation of the contributions from and to the other partners will help in understanding the nature of partnership and extent of collaboration between the partners in the team. Any opportunity of technology transfer between ORNL and other partners should be envisioned and explored.
- The project has a very well-balanced team.
- ORNL supports many other fuel cell projects with valuable images and analysis.
- How the project collaborates is unclear. There are a large number of partners and it is unclear what are they doing. The presentation suggests that all the work is being done at ORNL. If this is the case, there doesn't seem to be a need for the long list of collaborators. If it is not the case, then the PI needs to give credit to what the partners are doing. I would have liked to rate it higher, but there is not enough information on what the partners are doing.
- They have a good number of publications.
- This team clearly needs to be more closely integrated with the new efforts on durability headed by LANL and ANL. Dr. More and her team have the tools to greatly assist those efforts, and from them, one can receive the range of materials needed to truly gain some insight.
- No doubt, both industries and academic institutions have been involved in the project. The owner of the facility may want to provide training to users so that the innovative users can easily use the facility or work with ORNL to facilitate their progress.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- It would be helpful to see more studies of metal/support interactions on non-carbon, e.g., oxide supports. The interface between Pt and conductive oxides is a region of interest in catalysis that could be aided greatly by the expertise at ORNL.
- The plans for future work address some of the gaps present in the project. Planning for further development of an *in situ* liquid holder suggests that the team has overcome past obstacles or has new ideas to overcome the obstacles. A brief explanation of past learning and new ideas will be helpful in justifying the continuation of liquid holder development activities. More emphasis in technical collaboration between all partners should be continued and opportunities for technology transfer between partners should be explored.
- Adding capabilities to the *in situ* liquid holder for nm-scale microscopy of PEMFC material constituents operated under relevant operating conditions (liquid electrolytes, temperature, potential cycling) should be extremely beneficial.
- The project has the information and capability between the members such that they should be able to identify the chemical mechanisms for corrosion, as well as the physical mechanisms, and they should be able to propose pathways to mediate the problem.
- The future work seems to be "continue what we're doing." With the facilities and the team, they should consider offering some recommendations on how to improve fuel cell performance.
- Bullet points 1, 2 and 5 on the future work slide are fine, but to do them justice, omit bullet points 3 (*in situ* TEM) and 4 (more new collaborations).
- Developing appropriate protocols for sample preparation or selection is important in parallel to the characterization techniques' development.
- The PI has proposed the right directions and topics.

Strengths and weaknesses

Strengths

- SOA equipment is driven by some of the best in the business.
- The project is being carried out by a great team comprised of a good mix of academic, industry, and National Laboratory partners. Due to the diverse mix of partners, there is an immense opportunity for obtaining MEA samples aged under many different kinds of fuel cell operational conditions. ORNL has a great set of analytical tools and a relevant knowledge base for conducting MEA component analysis.
- ORNL provides unique microscopic tools that support numerous EERE fuel cell projects.
- ORNL has excellent equipment and facilities.
- ORNL has already developed a suite of analytical techniques to aid fuel cell developers.
- The project has good tools and good people who are trying hard to be useful to a wide range of customers and/or collaborators.
- The equipment and technical skills of the team are exceptional to explore the microstructural information from the MEAs.

Weaknesses

- More interactive collaboration between the partners is needed. More emphasis should be given to the analysis of a variety of aged MEA samples using existing analytical methods. MEA samples aged under different, such as, automotive, stationary, and portable, fuel cell operational conditions, should be studied to determine the impact of fuel cell operational conditions to MEA degradation.
- Speculations not based on direct evidence, i.e., Pt particles lead to mechanical membrane failure, where water flows in carbon particles, impurities accelerate graphitization, can mislead other researchers and should be avoided.
- The focus has been solely on understanding what is happening. There seems to be little effort directed towards how to improve the materials.
- The progress has been slow. *In situ* TEM efforts have lasted at least two years without result. The carbon corrosion study has been mentioned as a milestone in 2008, 2009 and 2010 presentations, and is now complete for XC72 only.
- The team needs to discuss with their colleagues in the fuel cell community defining the analysis metrics.

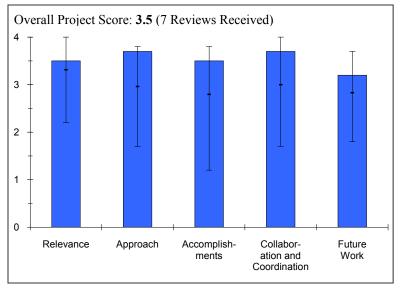
- The project focuses upon providing a service to others, rather than pursuing its own research. I would like to see this approach change so that the expertise is used to undertake more independent exploratory research in collaboration with other groups.
- Look for ionomer in corroded electrodes.
- The PIs need to be clearer on what their partners' contributions to the work are.
- The PIs need to speculate on "why" things are occurring, not just report what has happened. For example, why does the Pt nucleate preferentially on the surfaces of graphitic domains?
- It would be nice to see the progression of the carbon corrosion. So far there seems to be fresh and completely spent studies. Intermediate studies would add a lot of information on the mechanisms.
- Drop work on *in situ* TEM. Make assisting the LANL-led durability study the number one priority.
- It will be extremely important to characterize the carbon corrosion and catalyst particle changes around proton exchange ionomers in the MEA catalyst layers.

Project # FC-21: Neutron Imaging Study of the Water Transport in Operating Fuel Cells

David Jacobson; National Institute of Standards and Technology

Brief Summary of Project

This National Institute of Standards and Technology (NIST) project aims to develop and employ an effective neutron imagingbased, non-destructive diagnostics tool to characterize water transport in proton exchange membrane fuel cells (PEMFCs). Objectives are to: 1) form collaborations with industry, National Laboratory, and academic researchers; 2) provide research and testing infrastructure to enable the fuel cell (fuel cell) industry to design, test and optimize a prototype to commercial grade fuel cell 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 and academia needs.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.5** for its relevance to DOE objectives.

- It is highly relevant that NIST provides the mechanism to study water transport in operating fuel cells. This can be an important tool for optimizing fuel cell performance and durability.
- *In situ* water imaging gives valuable insight which can be used to validate models, improve membrane electrode assembly (MEA) robustness, and design better flow fields, all of which are relevant to DOE performance objectives.
- This project is very relevant to DOE's RD&D plan objectives. Direct visualization of water in various parts of fuel cell hardware is of great help in understanding the water distribution phenomenon and in developing a water-management strategy for fuel cells. Understanding the effect of gas diffusion layer (GDL) properties on water distribution within the cell is very relevant to developing good water-management strategy, and hence developing durable stack technology.
- This is an outstanding project that really gives insight into water management within the fuel cell system (fuel cells). It is outstandingly relevant.
- This is excellent work. The mix of collaborators from laboratories, industry, and academia clearly shows that this project is relevant and useful. This is a tremendous bargain for DOE at the cost.
- Monitoring the water transport in operating fuel cells is extremely important. Fuel cell durability and polarization performance at high power density are determined by catalyst activity, component stability/conductivity, and water transport properties, in particular water transport, is more important for the fuel cell stack and system design.

Question 2: Approach to performing the research and development

This project was rated **3.7** on its approach.

• The NIST approach of close collaboration with users allows each partner to contribute in their specific area of expertise. This is a very effective and efficient approach for fuel cell investigative studies.

- The approach of mapping the high frequency resistance (HFR) with the amount of water present in GDL is interesting. This approach is good as it provides a direct correlation between the amount of water vs. HFR in a fuel cell. The approach of using graded anode GDL and flow field channels with different hydrophobicity is also unique. Overall, all the approaches taken in this project to study water transport within fuel cell components are valid, and their technical feasibility has been addressed in this work.
- The approach was outstanding in addressing the technical barriers of neutron scattering and two-dimensional imaging.
- The enhanced capability for freeze/thaw studies is an excellent addition.
- Neutron imaging is an excellent technique for investigating water transport. The NIST team has shown the capability to utilize the technique to address the water transport issues in fuel cells. Better understanding of the techniques' resolution and improving the resolution allow the team to have deeper insight into water transport in fuel cell components.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.5 based on accomplishments.

- The large number of users (fuel cell researchers) that NIST has worked with in the past year is truly impressive. At the same time, NIST has made significant improvements to its diagnostic tools.
- 13-µ spatial resolution is a major accomplishment, which enables imaging water content in distinct GDL, membrane, and most electrode layers.
- It is encouraging to see that the presence of 20 µ of water layer in GDL has significant influence in the HFR of the cell. The study should be extended to see what level of cathode inlet humidity is required to maintain such water level in the GDL. This information will help system engineers to develop an adequate system for retaining such level of water in the GDL. The study of the thermal conductivity of the GDL and its effect on the water saturation point in fuel cells is very informative. It is interesting to see that higher thermal conductivity induces more water deposition into the GDL. The effect of hydrophobic flow field channels on water accumulation in GDL is very convincing, and is observed in the neutron radiograph. The radiographic evidence of temperature-gradient-mediated phase-change-induced flux and capillary action is very interesting.
- This presentation was excellent. Now that we are building up a substantial body of results with different materials and flow field designs, I think we are seeing some real understanding grow, especially if we look more at comparing these results with computational fluid dynamic (CFD) model predictions and see where the CFD models are lacking.
- High resolution and high scan rates help to reveal the instant and dynamic water transport information. Individual cell diagnosis for water transport in stacks is really helpful for the stack design engineers.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.7** for technology transfer and collaboration.

- The number of fuel cell collaborators that NIST has worked with is outstanding and includes academia, National Laboratories, and the private sector. It is important to note that proprietary services can be provided to industrial partners.
- The long list of influential collaborators demonstrates that this project is valuable to many fuel cell researchers.
- Since this is a unique facility, there seems to be lots of users of the instruments, where NIST is providing an analytical service. However, not much description has been given about the collaborators. The improvement of imaging techniques and instrument operation was done solely by NIST. More direct collaboration will be beneficial for the research community.
- Publishing fifteen papers in refereed journals this last year is excellent. In addition, I do not hear anyone complaining about the beam time allocation process. The team has worked extensively with industry and academic groups in universities and National Laboratories.

Question 5: Approach to and relevance of proposed future research

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

- The plans to continue the study of water transport in partnership with fuel cell experts is excellent. In addition, the plans for continued improvement in the NIST diagnostic tools are important.
- Water attenuation and other causes of beam hardening are very important to understand so that researchers can have confidence in neutron imaging results. All common fuel cell materials should be checked for beam hardening interference.
- The future research plan is in line with the development needed to explore the limits of the neutron radiography technique for *in situ* fuel cell evaluation. It's encouraging to see the plan of building a new facility.
- Bringing the larger field of view detectors on line next year can only help. Continuing to look into systematic errors will also be beneficial.
- Further development of advanced imaging methods will be appreciated. However, it is also very important to work with industry to address the issues in industry and define the fundamental challenges for future research.

Strengths and weaknesses

Strengths

- They employ unique diagnostic tools.
- It is an effective approach to work in partnership with fuel cell researchers.
- Neutron radiograph is a unique method for visualizing the water transport through various components of the fuel cell. Involvement of a large number of research groups, from academia, industry and National Laboratories, makes this program very active and effective. This is a very effective, non-destructive method for visualizing inside the operational fuel cell.
- This is a nice project that should be a model for DOE. This group has really expanded the boundaries of fuel cell understanding by providing unparalleled insight into water management within a fuel cell. They have an excellent approach to operating the instrument and using it to probe fuel cells. They work seamlessly with various groups on other DOE projects.
- This is relevant, useful, and a good balance between industry and academia.
- The excellent facility, equipment and technical skills make the team very competitive.

Weaknesses

- The experiments can be carried out in small foot print only. It will be advantageous if this method can be implemented to visualize cells with larger dimensions.
- Further improving the resolution and scan rate has technical challenges. The team needs to consider how to use the current equipment and methods with the realistic resolution to address reasonable issues in the fuel cell stack and system development.

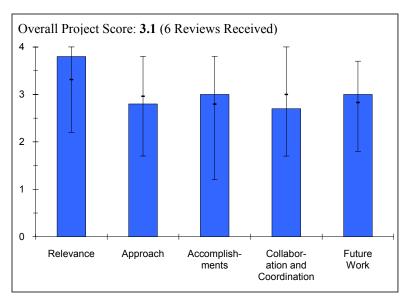
- Further improvements in the spatial resolution would be useful to study water management in MEAs with thin $(<10 \mu)$ electrodes.
- I think that detectors with modest spatial resolution but higher temporal resolution may be of interest to the community in the future.
- The team may want to help address the water transport and distribution in the fuel cell stacks, in particular, to help diagnose the failure mechanism of the individual cells in the stacks.

Project # FC-22: Nitrided Metallic Bipolar Plates

Peter Tortorelli; Oak Ridge National Laboratory

Brief Summary of Project

The objective of this project is to demonstrate nitridation to protect stamped metallic bipolar plates (BPP). The overall goal is to demonstrate potential for metallic BPPs to meet automotive durability goals at cost of < \$5/kW. Milestones include: 1) no significant warping or embrittlement of stamped, 15-cm² active area plates by nitriding; 2) single-cell fuel cell test behavior for 15-cm² stamped and nitrided metallic BPPs equivalent to that of graphite; and 3) complete automotive original equipment manufacturer (OEM) BP manufacturing assessment and single-cell, drive-cycle test protocol.



<u>**Ouestion 1: Relevance to overall DOE**</u> <u>objectives</u>

This project earned a score of 3.8 for its relevance to DOE objectives.

- One of the keys to achieve the cost, power density and volumetric density targets for the successful commercialization of fuel cell vehicles (FCVs) is the ability to make cheap, chemically robust, ultra-thin bipolar plates. This project attempts to do just that.
- The development of low cost bipolar plates is of great interest considering the stack durability and cost. Therefore, it is critical to achieve the proposed objectives.
- Bipolar plate improvement and cost reduction is critical for the Fuel Cell Technologies Program to meet its cost and durability targets.
- Metallic bipolar plates offer vital advantages over graphite bipolar plates. Corrosion protection for metals as bipolar plates proved to be mandatory.
- Metal bipolar plates can contribute to overcoming the cost and durability barriers that prevent commercialization of polymer electrolyte membrane fuel cells (PEMFCs) for many applications. For this reason, the study of metal bipolar plates is relevant to DOE's objectives.
- This project has not avoided the targets that are relevant to the study of bipolar plates. It focuses on cost, corrosion resistance, and conductivity. In principle, the other targets (mechanical strength, formability, weight, and low gas permeability) could possibly be met with the candidate materials.

Question 2: Approach to performing the research and development

This project was rated 2.8 on its approach.

- From a materials perspective, the approach of this project is very strong.
- When looking at the size/area of the manufactured plates, this reviewer wonders if the ability to scale up to full-scale stack-sized bipolar plates is feasible.
- It would be worthwhile for the PI to demonstrate the ability of producing plates larger than 400 cm², which are uniform in thickness, are flat, etc., to demonstrate the viability of this project.
- This project contributes to overcoming some barriers such as formability, performance, and to some extent, cost.
- The approach is correct, but it could have been improved by considering bipolar plates with representative active areas (on the order of 200-300 cm² instead of 15 cm²) and considering stacks instead only single cells.

- What about the sealing? The gaskets can be put either in the membrane electrode assemblies (MEAs) or in the bipolar plate. This option has not been integrated into the project.
- This is a somewhat academic report. It does not address low cost manufacturability.
- A comparison with competing technologies should be made to check viability.
- There has been a lot of work in nitrided metals.
- The use of cycle, as opposed to steady state conditions, to evaluate performance is good, but the test cycle used does not seem to represent the drive cycle of a car, which would be what the bipolar plate would experience for this application.
- The plates were fresh (no welds). The plates should have had a part welded to be able to evaluate the impact of the welding.
- The simple flow field used does not seem representative of what is expected. For the stamping test, the team should have included more details and tried more complex geometries.
- With the new material being developed, will there be joining problems? This question should be addressed.
- Though this approach has been pursued over a couple of years, new challenges, like using stainless steel instead of more expensive Ni-containing metals, were taken up recently, and the requirements for the metals could be lowered substantially, leading to further cost reduction.
- In terms of the targets that DOE has identified for bipolar plates, this nitrided stainless steel project has generally had a decent concept for achieving interfacial contact resistance requirements. However, cost, corrosion resistance, formability, and joining have always represented the primary doubts.
- Cost estimation should have preceded the material development. Now that the project is in its final stages, cost has been shown to be within range of targets, but the sequence should have been reversed.
- Corrosion still remains a question mark. The cycle used in *in situ* testing does not represent startup/shutdown cycling. Officially, the tests prescribed by DOE, including the *ex situ* tests, will never be able to address high voltage corrosion. While the materials in this project passed the tests, there is still some desire to know what would happen under realistic cycling.
- Formability and joining were addressed late. Formability necessarily followed the change in base metal, while joining still has not been evaluated.
- There has never been an interest in investigating corrosion through coolant channels.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.0** based on accomplishments.

- The PI has obviously done a lot of work optimizing the materials used, the chemistry of the treatments, and the optimization of the surface treatment. The technical accomplishments and progress are high and commendable.
- The ability to produce large-scale, high quality, thin, uniform, bipolar plates still remains to be demonstrated. Also, the cell potentials, which the PI uses to investigate the chemical robustness of these cells, are a bit modest. Spiking the cell potential to 1.3-1.5 V as experienced during startup and shutdown, will eventually be needed to validate the chemical robustness of these cells.
- The different characterizations performed in this project (nitridation process, polarization curves, membrane electrode assembly (MEA) contamination) are interesting and indicate that the proposed nitridation solution might be interesting.
- Questions are still open, like the formability of this ferritic stainless steel regarding real GM bipolar plates with small channels (in particular in the curves), the welding of the plates, and the real cost.
- The cause of the $35-\mu$ MEA failure was not made clear.
- The 1,000-hour test is a significant accomplishment.
- For the size of the team and the amount of funding, one would expect more accomplishments.
- The stamping of the simple flow field design was good, but more complicated and larger designs would be more representative of the industry.
- The nitridation process for stainless steel has been developed well. The process seems to be adequate for mass manufacturing.
- The material exhibits good longevity under normal operating conditions. Failure-mode conditions should be introduced later in the project or reserved for a follow-up project.

- Despite concerns about the approach, the PI has managed to consistently respond to concerns throughout the project's life through testing.
- Low interfacial contact resistance has been demonstrated.
- Formability has now been partially demonstrated, although it would still be interesting to see if depths beyond approximately 250 µ could be achieved with a channel span of approximately one micron. At present, the formability shown is likely not sufficient to enable robustness to a wide variety of cell components, particularly gas diffusion layers (GDLs).
- Corrosion remains a mystery. Before and after cycling, *in situ* high frequency resistance (HFR) and *ex situ* through-plane resistance measurements need to be done. One slide showed evidence of some discoloration and some explanation is needed. The X-ray fluorescence measurements could still have produced the same data with corrosion, especially with high cell humidification (inlet RH not noted).
- Cost analyses performed in other projects. Although the cost does not meet the official DOE target, the cost is within striking distance. Some decrease is still preferred.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated 2.7 for technology transfer and collaboration.

- Research collaborations with all of the various entities are at a high level and are transparent. Adding GM certainly makes this program stronger. If the technology proves to be feasible and practical, it will be interesting to see who would scale it up and commercialize it. ATI Allegheny and AGNI GenCell are two companies that come to mind.
- The collaboration between the partners seems correct.
- More stack manufacturing expertise should be included in the project
- The addition of GM to the team was good.
- The PIs have a large team. They defined the roles and responsibilities of each partner well. Communication between the partners is unclear.
- With GM and other industrial partners, it was surprising to see that intellectual property was available for license.
- With GM, an OEM is part of the project team. Though industry input is generally appreciated, they should be sure that other companies still have sufficient access to the technology.
- Collaboration has been done with many partners and has been improved with the incorporation of an automotive OEM (GM).
- The roles of Allegheny Ludlum, GenCell, NREL, and LANL are clear.
- The role of Arizona State University was not made entirely clear from the presented slides, but one could guess that they played a role in the cycle tests.

Question 5: Approach to and relevance of proposed future research

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

- Future work is very strong, especially now that GM has been added to the program. GM's most robust drive cycle test protocols will do a more thorough investigation of the bipolar plates' chemical robustness under a more extreme variety of cell potentials. It is good to see the area of the single cell be increased to 50 cm², but ideally it would be even larger.
- The proposed further work is in accordance with the main objectives of the project. One might just ask if all the jobs will be done by the end date of the project (September 2010).
- This project is ending, so future work is unclear.
- Completing GM's manufacturing study is very important.
- As the project is ending soon, the proposed work is appropriate. The activities basically focus on cost reduction.
- The project is near completion and is now directed towards those activities that should be its final focus. These activities include laser welding, realistic drive cycle evaluations, and further efforts to reduce cost through quartz lamp nitriding.

• It is hoped that the GM single cell drive cycle will include startup / shutdown cycling. It is also hoped that further stamping activities will test formability limitations and investigate the draw depth that can be achieved given a narrow channel span.

Strengths and weaknesses

Strengths

- A lot of work optimizing the materials used, the chemistry of the treatments, and the optimization of the surface treatment has taken place. The technical accomplishments and progress is high and commendable.
- The project strengths are its partnership covering the whole need of production, and characterization.
- This is a large team with sufficient funds.
- Adding GM strengthened the project.
- Good materials approach, which has been pursued with perseverance. The focus on normal operating modes other than failure operating modes is positive, since convolution of too many criteria was avoided. Good materials have been identified. Further optimization to harsher conditions should be reserved for future projects.
- The PI has been able to respond well over the years to considerable feedback from reviewers and OEMs. The project has added activities related to formability, joining, and cost estimation, and has changed base metals to meet targets.
- The project has made use of an intense amount of collaboration amongst approximately a half-dozen partners.
- The future work is well focused on remaining activities.

Weaknesses

- The ability to produce large-scale, high quality, thin, uniform, etc., bipolar plates still remains to be demonstrated, and, as of now, no plans are included in this activity. Also, until now, the cell potentials that the PI uses to investigate the chemical robustness of these cells are a bit modest. Spiking the cell potential to 1.3-1.5 V, as experienced during startup and shutdown, will eventually be needed to validate the chemical robustness of these cells. This activity probably will be investigated by GM.
- The main weakness is that they only worked on small-size, single cells. Even with the larger 50-cm² GM bipolar plate may not be really representative with nitridation process of real-size bipolar plates for automotive applications. Warping remains a potential issue.
- Stack testing would have been appreciated to evaluate the performance and durability of these bipolar plates.
- The cost analysis could have been more detailed, to be more convincing in achieving the cost target.
- Stamping is much more difficult than people realize. The designs and plate size are extremely important. They needed to examine more complicated flow fields over a larger plate (at least 25 cm²; preferably 50 cm²) for stamping size matters.
- The cycle testing seemed very tame compared to what the fuel cell will experience in a vehicle.
- Doubts about corrosion under realistic drive cycles remain, as well as doubts about the effectiveness of the cycle that was generated by the project to test the plates. Hopefully, the GM collaboration will help to address this issue.
- More *in situ* diagnostics needed, e.g., HFR.
- Formability and cost evaluations have improved considerably, but the limits of each are still unknown.
- Throughout the project, development carried forward without first answering critical questions about cost, corrosion, formability, and welding. For the most part, the project has serendipitously succeeded, but there was perhaps more risk than necessary in getting there.

- Large scale $(400 + \text{ cm}^2)$ plate making would help the work scope.
- More extreme cell potentials need to be included in the work scope.
- The PIs needed to examine more complicated flow fields over a larger plate (at least 25 cm²; preferably 50 cm²) for stamping size matters.
- The cycle testing seemed very tame compared to what the fuel cell will experience in a vehicle.
- Needed additions, e.g., realistic drive cycling, cost estimations, welding, *in situ* diagnostics, are being addressed in either ongoing work or future work.

- For apples-to-apples comparisons, it would have been good to have seen cycling on an untreated $Fe_{20}Cr_4V$.
- Unless better evaluations are to follow, it would be good to understand what the discoloration on slide 13 represents.
- Some information about coolant channel corrosion would be beneficial.
- *Ex situ* evaluations before and after cycling are needed.

Project # FC-23: Low Cost PEM Fuel Cell Metal Bipolar Plates

Conghua Wang; Treadstone

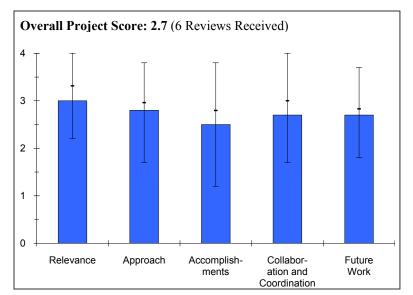
Brief Summary of Project

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

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of **3.0** for its relevance to DOE objectives.

• The development of low cost BPs is of great interest, considering the stack



- durability and cost. Therefore, it is critical to achieve the proposed objectives.
- They have a strong focus on reducing cost, which is a critical step for fuel cell implementation.
- The project seeks to produce a low cost BP base material to meet the DOE 2015 cost target.
- Innovative concepts for low cost BP development are critical. However, the cost basis for this concept wasn't clearly established in the presentation. It also wasn't clear how the coolant layer or gas manifolds are integrated.
- The TreadStone Project addresses the BP durability and cost, and is very relevant to the DOE Fuel Cells Technologies program.

Question 2: Approach to performing the research and development

This project was rated 2.8 on its approach.

- The approach to insert some conductive vias in a non-conductive coating is original. The characterization means are classical.
- Task two is referring to carbon steel and aluminum based plates, but only carbon steel plates are detailed.
- No details are given regarding the gaskets. As gaskets may be put on the BP, it should have been addressed.
- The cost objective is mentioned, but no corresponding milestone is presented.
- They employed an interesting approach to utilize only a portion of the surface as conductive material. Fabrication cost analysis will be critical information.
- Unique approach using non-corrosive, electrically conductive vias to support conduction through a nonconductive surface treated plate. Such an approach allows the use of cheaper material for the bulk of the plate and utilizing a small amount of expensive material to support conduction through corrosion resistant layers.
- The via concept is not clear for stainless plates.
- This technology should be compared with gold-plated, stamped stainless steel that has the thickness of gold minimized. This could be considered a baseline and enable direct comparison. If the via technology is capable of five nm equivalent thickness, the demonstration metrics should be compared.
- The approach is unique to TreadStone—vias that represent a small fraction of the surface of the plate that conduct electrons through the plate while the rest of the plate is covered by a non-corrosive, non-conducting coating. TreadStone is investigating lower cost materials for use as conductive vias, as well as carbon steel and aluminum base materials.
- There have been other attempts to coat low cost metals with corrosion-resistant coatings. Some sort of conductive additive/particle is needed to ensure electrical conductivity. TreadStone indicated that the expensive

via materials need cover only 1.4% of the surface area for acceptable conductivity and thus contribute a small part of the material cost.

• Their efforts are not high focused. The work on carbon steel should be eliminated.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.5 based on accomplishments.

- The presented results lack precision to be evaluated in a relevant way. For instance, on slide four contact resistance values are presented, but without précising the clamping force, or if the measurements have been performed on stamped or flat samples.
- Data have been presented neither on carbon steel nor on aluminum based plates, but only on stainless steel (SS)316L! In the discussion, we learned that corrosion rate on carbon-steel was between 5 and 10 μ A/cm²; i.e., far over the target.
- Costs are given on SS, but no indication of the assumption of the number of plates is considered.
- In the presentation, it is not précised if the coating can be stamped or if it has to be processed after stamping and welding. And if it is before stamping, how will be the welding be done?
- No active peaks with the coating is underlined, but even on bare SS 316L, there is no active peak.
- For the 10-cell stack, 2.5 kW for 3,000 cm² is less than the target. As the stack is running for 800 hours, polarization curves or performance graphs would have been appreciated.
- Not clear how many samples have been tested for performance repeatability.
- The experimental data looks promising, and the cost per kilowatt is close to the 2015 target. More extensive testing is necessary to provide convincing evidence that this approach can be successful.
- The stack demonstration should include a resistance measurement (high frequency resistance/internal resistance drop). Moreover, it would be interesting to monitor changes in this resistance over more time.
- TreadStone earlier proved the concept of their metal plate technology by achieving 8,200 hours in a long-term stability test. They have recently completed initial performance testing of a 1-kW stack for stationary fuel cells and Ford is testing TreadStone coated SS plates that contain gold dots. The testing includes realistic driving cycles. The stack has achieved 800 operating hours.
- A manufacturing cost analysis indicates that the current technology can meet the 2010 DOE targets, but not the 2015 targets.
- There were different via materials developed. Stack current-voltage performances were tested. A similar curve should be given with graphite BPs. Authors should give contact resistance curves (instead of the number). Moreover, corrosion testing for other via materials should be given. The influence of defects on the corrosive behavior should be monitored.
- As an add-on coating, the thermal coefficients of different materials and their effect on the corrosion of the substrate material should be addressed. This is very important since proton exchange membrane fuel cells are expected to operate at a wide temperature range.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated 2.7 for technology transfer and collaboration.

- The collaboration inside the project seems correct.
- It is not clear from the presentation level of interaction between partners; as they simply list tasks each will complete.
- For a project like this, there should be even more automotive original equipment manufacturers (OEMs) involved as partners. The project should look into collaborating with other National Laboratory groups.
- This reviewer recommends reviewing the cost analysis with OEM.
- The collaborators are all experienced and are experts in specific areas. The collaboration appears to be very good and covers the needed skill sets to provide TreadStone with an independent assessment of their technology. Ford brings automotive expertise to the project.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

This project was rated 2.7 for proposed future work.

- The proposed future work is in accordance with the project targets. Fabrication cost using aluminum based plates should be inserted.
- They will not develop aluminum plates until 2011. If possible, this task should be moved earlier.
- Other via materials are slated to be studied. Utilizing carbon nanotubes (CNTs) might be a way to further reduce cost. Corrosion studies that are ongoing or planned for the future are critical to provide convincing proof of the applicability of this approach.
- Future work seems ambitious based on the current rate of progress.
- TreadStone is focusing on developing lower cost conductors such as CNTs, palladium dots, and metallic carbides. They are also looking at carbon steel and aluminum plate materials, upon which their coating will be applied.
- The work plan appears reasonable but lacks decision or down-selection points.
- The project should be focused on a stainless steel substrate.

Strengths and weaknesses

Strengths

- The main strength of the project is the partnership.
- They are focused on cost.
- This is an innovative approach to bipolar plate design. The material and fabrication costs are quite attractive.
- The concept is unique and warrants evaluation.
- The project has demonstrated feasibility and the ability to meet the DOE 2010 cost target. Lower cost approaches are being investigated, which give some hope of meeting the 2015 targets.
- Ford is subjecting one design iteration of the plates under rigorous automotive test conditions.
- The project is target oriented, includes stack testing, and exemplifies team work.

Weaknesses

- The main weakness of this project is that no performance and cost comparisons are done with alternative BP materials or coatings, like composite or carbon based coatings on metallic plates. For the latter, the coating cost should be comparable, but with a lower-cost material.
- As the number of contact points is reduced, operation at high current densities may be an issue, due to thermal hot spots where the current is passing.
- They were not clear on the number of samples tested or test repeatability.
- They were not clear how robust the plates will be.
- It is not clear what this technology is being compared to, and a baseline should be established. It isn't clear how this will be accomplished.
- It is not clear if the paths being followed will be able to meet the DOE 2015 cost target. The ultimate target may need to be even lower to enable widespread commercialization.
- The presentation lacked clarity on how the conducting vias are formed.
- The efforts are not very focused.

- Performance and cost comparisons with alternative BP materials or coating, in particular carbon based coatings on metallic plates, should be added.
- Cost evaluation should take into account the real stack performance and not the assumption of 1000 mW/cm², which artificially lowers the number of plates.
- Move aluminum plate development forward to ensure adequate time for testing.
- Bring in more automotive OEMs into the collaborators. The project should demonstrate performance and corrosion resistance under real world conditions and for greater time periods. Perhaps accelerated stress testing should also be performed.

- Add down-selection points and appropriate metrics for the decision.
- Work with stationary and portable fuel cell developers to qualify the plates for those applications.
- Focus on stainless steels. Testing with some 200 steels may be helpful.

Project # FC-24: 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 (BPP) that meets the DOE performance and durability targets for BPPs that are thinner and more durable than machined graphite BPPs 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.

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>

Overall Project Score: 3.2 (6 Reviews Received)

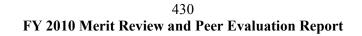
This project earned a score of **3.8** for its relevance to DOE objectives.

- One of the keys to achieve the cost, power density, and volumetric density targets for the successful commercialization of fuel cell vehicles (FCVs) is the ability to make cheap, chemically robust, ultra-thin bipolar plates. This project attempts to do just that.
- The development of low cost bipolar plates is of great interest, considering the stack durability and cost. Therefore, it is critical to achieve the proposed objectives.
- The project has the potential to develop low cost, lightweight bipolar plates.
- The project addresses the DOE barriers of performance, cost, and water transport within the stack by forming a hydrophobic surface of the bipolar plate.
- The project is in alignment with the intention of the DOE program. It seeks to lower bipolar plate weight through use of aluminum, while keeping costs lower through the expectation of stamping. Corrosion and conductivity are also accounted for in the coating selection.
- Bipolar plate work is still relevant within the DOE program since plates can contribute to the cost and durability barriers that most applications experience. Within the context of DOE work, these barriers have not yet been cleared with regards to bipolar plates.

Question 2: Approach to performing the research and development

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

- The approach of using aluminum as the base material instead of stainless steel is a valid approach thanks to its low cost and weight savings. Also, the concept of using fluoropolymers as a coating makes sense from a chemical robustness perspective.
- The weakness of this approach, however, is the poor selection of electrically conductive dopants, TiB₂ and CaB₆. While both chemicals are reported to have higher conductivities than graphite, their incredibly weak tolerance to acid and fuel cell (fuel cell) potentials is a huge detriment to this project and approach.
- The approach to develop composite coating is correct. The characterization means are classical.
- No details are given regarding the gaskets. As gaskets may sometimes be put on the bipolar plate, it should have been addressed.
- The cost objective is mentioned, and, even if the low cost methods are already accepted by the original equipment manufacturers (OEMs), a corresponding milestone should have been presented.
- The approach is sharply focused on the technical barriers.
- The possibility that, if the coating fails, the aluminum can react with water to form hydrogen and aluminum hydroxide should be explored.



- The approach is smart and the production cost for such a painted surface looks reasonably low.
- While the team acknowledges that the acid stability of the borides needed to be checked, a more thorough understanding of both boride and polymer stability in acidic oxidizing/reducing conditions should have been investigated prior to the beginning of the project.
- In general, corrosion tests are not following protocols that have recently been specified by the Fuel Cell Technical Team. Potential sweeps were not used at low voltage, and NaF was used in place of HF. The temperature was not reported.
- For both the boride rotating disk electrode (RDE) work and the plate studies, no cyclic voltammetry was reported that could be used to show whether currents were faradaic or related to other charge transfer processes. It would be interesting to see this.
- Given the instabilities that have been seen in fuel cells for aliphatic C-H bonds, it will be interesting to see ethylene tetrafluoroethylene copolymer (ETFE) stability in the presence of an operating fuel cell where Pt presence can account for higher levels of peroxide. Polychlorotrifluoroethylene (PCTFE) stability is questionable as well, and any loss of chloride might affect Pt.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.8 based on accomplishments.

- The amount of high level, thorough work at this point in the project and for the dollars spent is quite impressive.
- While TiB₂ and CaB₆ have both been shown to not be viable for this application, the synthesis and characterization of these materials clearly demonstrates top-level research.
- The apparent quality of the uncoated aluminum plates, which were stamped and tested for hydrogen permeation, is very high.
- Presented results are not in total agreement with the milestone table presented on slide 6. Milestone: "Fabricate a composite coated aluminum plate": okay for the achievement, but 24-hour testing is too short for making any conclusions. Milestone: "Synthesize TiB₂ and CaB₆ using low cost process": okay for the achievement but no indication on the real cost of the powder fabrication and spraying. Milestone: "Regarding CaB₆ and TiB₂ low conductivity and corrosion resistance": there may be no interest in using them in the future.
- There was a significant amount of progress made in a short amount of time.
- The conductivity of TiB₂ and CaB₆ is significantly larger than that of carbon, but the carbon composites have yielded conductivities that are comparable to what has been seen with the TiB₂ and CaB₆.
- The conductivity of the coatings is much lower than might be expected from the conductivity data of the fillers. The conductivity of the layers is not out of range, though. A conductivity of 5 S/cm of the layer is about 50 times higher than that of the electrolyte. As a 100 µm coating is needed on either side and the electrolyte thickness may be between 50 and 100 µm, the resistance of the layers would contribute approximately 4% to 8% to the overall resistance, provided all other resistances were negligible compared to these two. If other resistances are to be considered, the effect would even be lower. Nonetheless, a higher volume fraction of the filler, up to 90 %, should be tried to achieve a better conductivity.
- Many of the results obtained so far are as expected. For example, uncoated aluminum plates have been shown to be corrosive and lack conductivity. These data may serve as a good baseline. Furthermore, the borides were not suitable for the coatings.
- Despite the recent start of the project, more results should be expected when considering the project's short timeline. The project needs to move more quickly towards coating materials that are more promising, and particularly, towards more challenging tests. Only two slides show positive results on composite coatings, and the two coatings represented are different (hydrophilicity on graphite/PCTFE and corrosion resistance on carbon fiber (CF)/ETFE).
- Electrochemical techniques should be validated versus a standard. The currents shown could be affected by infrared compensation if not addressed.
- During the presentation, the thickness of the coatings was mentioned to be about as thick as the web thickness ($\sim 100 \ \mu m$). The coatings need to be thinner!!
- Room temperature corrosion testing cannot represent the results at PEMFC operating temperature. There seems to be some technical challenge on measuring the contact resistance of the coatings.
- A stack testing with graphite bipolar plates should be used for comparison.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.3** for technology transfer and collaboration.

- Collaborations with key partners are clearly stated, reported, and easy to understand. It would be good to know whether Orion Industries (Orion) will begin their development for coating the aluminum sheets when the electrically conductive dopant is finalized, or will start before that.
- Collaboration between the partners seems correct.
- The collaborations appear to be working well.
- The work share and expertise of the partners is clearly defined and the partners supplement each other's skills.
- The collaboration partners appear to be providing input to the project. The design and stamping of aluminum substrates through the Gas Technology Institute (GTI) have been shown. Coatings have been provided through Orion and borides were provided by Southern Illinois University. No partner has been uninvolved.
- The project also deserves merit for appearing to justifiably let go of the efforts from one partner, which is difficult to manage.
- If coatings are too thick, further work needs to be done with Orion to produce thinner coatings. If this cannot be done, an alternative coating partner may be needed.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.8 for proposed future work.

- Plans for future work with their various collaborators are clearly stated and are logical.
- The lack of having a viable electrically conductive dopant for their protective film is clearly a major weakness which could eventually derail this project. At this time, it is unclear if a viable candidate will be discovered, and, if it does, how will it affect the pending cost analysis?
- The proposed future work is in agreement with the project targets.
- Preliminary cost analysis should start from now and not in fiscal year 2011.
- Stack testing should be inserted in the work program.
- Future work is targeted at the key issues.
- Investigation of alternative filling powders is strongly supported. The borides might also be cross checked to see whether the low film conductivity might be a property of the specific powders used and not the chemistry in general, or even depend on pretreatment of these powders. Systematic check of one of the boride powders is advised to clarify, for instance, whether they form thin oxide layers reducing the conductivity.
- Some of the future plans are exactly what is needed: corrosion testing at 80°C, through-plane resistance, *in situ* testing, and cost analysis.
- Investigators should be able to provide some estimation of plate formability, including the present plate status (channel span, channel depth, radius, land width, etc.), as well as some effort to understand the limits of local elongation. Finite element analysis modeling can help in this regard.
- More information about alternative filler powders would be beneficial. The project does not have much time to waste with materials that are high risk. It would be interesting to know whether alternative fillers could be other carbons or metal carbides.
- In situ test plans need to be discussed.

Strengths and weaknesses

Strengths

- Management of this project appears to be very high with good coordination between the various groups. Stamped aluminum plates also appear to be very high quality.
- The project strength is its partnership, which covers production and characterization needs.
- The project has the potential for a high payoff.
- The project uses a new and innovative approach, introducing aluminum as a new material which is highly conductive and very ductile as a bipolar plate.
- The project is using a low weight material for a substrate (aluminum).

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- The PI has managed to use collaboration opportunities well to deliver the results from each partner.
- The PI was able to move nimbly from the boride concept to carbon-based fillers once the boride materials showed their deficiencies.
- Good effort to provide baseline data for uncoated aluminum, materials synthesis, and stack testing with untreated aluminum plates.
- Team work is a definite strength.

Weaknesses

- The lack of having a viable, electrically conductive dopant for their protective film is clearly a major weakness that could eventually derail this project. At this time, it is unclear if a viable candidate will be discovered, and, if it is, how it will affect the pending cost analysis.
- No stack testing is foreseen. Referring to the total project budget, there might be no financial issues.
- The benefit of TiB₂ and CaB₆ materials for composites as compared to carbon has not yet been demonstrated.
- Project needs to eliminate materials faster if they are not meeting requirements, and do so with the philosophy of using a reasonably aggressive test first. Hopefully the carbon fiber/ETFE composite coating will be tested in a cell soon.
- The *in situ* test plan should have already been formulated.

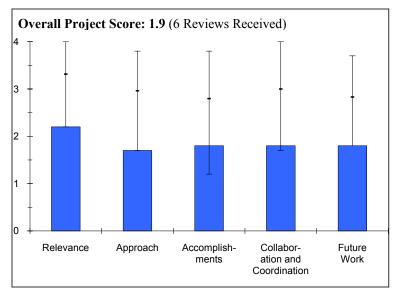
- Corrosion testing at high temperature and contact-resistance testing should be performed.
- Cost analysis is needed.
- Find a dopant as soon as possible.
- Performance comparison with usual bipolar plate (graphite, composite or metallic gold coated) using the same design and operating condition should be added.
- Evolution of the contact resistance at long term may be an issue in particular on the cathode side with oxidation of the aluminum plate. Coating permeability could be measured before single cell testing.
- This PI may want to examine issues associated with reactions of aluminum with water in areas where the coating is not protective.
- A systematic analysis of the reason for the low conductivity of the borides needs to be carried out. The team could also embark on a development of paints with a higher degree of filler.
- Focus on aluminum and use lower risk coatings.
- Formability estimates for the plate should be provided, as well as detailed cost estimates.
- Material status should be reported consistently for all candidate coating composites.
- Some test protocols (particularly with respect to corrosion) need to be updated to fit DOE guidelines.
- Electrochemical testing should be reviewed to be sure it is validated.
- Borides appeared to have been removed and should remain so.
- More collaboration is needed.

Project # FC-25: Air-Cooled Stack Freeze Tolerance

Dave Hancock; Plug Power, Inc.

Brief Summary of Project

Project objectives are to: 1) advance the state-of-the-art for air-cooled stack technology by determining stack failure modes and root causes, developing baseline understanding for freeze tolerance, validating mitigation strategies for failure mode root causes and design improvements to improve freeze tolerance; 2) test and evaluate air-cooled stacks and components developed for increased freeze tolerance and durability; 3) evaluate failure mechanism mitigation in stack and/or system design; 4) perform life-cycle cost analyses for freeze tolerance strategies; and 5) document and publish summary of stack freeze failure analyses.



Question 1: Relevance to overall DOE objectives

This project earned a score of 2.2 for its relevance to DOE objectives.

- The key topics concentrate on advancing and understanding major issues and barriers of fuel cell (FC) technologies. They address technology development aspects, cost analysis, and increasing understanding of real-world operating conditions; i.e., improving freeze start behavior of stacks and system components. The overall project, however, appears to be tailored too much to one particular application. Instead of concentrating on air-cooled stacks alone, the inclusion of stack systems with alternative cooling technology would widen the scope of the project and increase its value to the community.
- The development of freeze tolerance and air cooled stacks can help DOE with early market penetration in some markets, but the vast majority of higher power applications will require liquid cooling. The DOE program cannot be justified by low power applications.
- This project is aimed at the forklift market-in/out of freezers.
- Freeze tolerance and operation at sub-freezing temperatures, due to forklifts operating in refrigerated areas, seems like an incredibly small niche market. There doesn't seem to be a substantial market for fork lifts that operate at low temperatures. As the market is very small, the technology for air cooling and freeze tolerance, does not apply to other higher power applications where removal of waste heat is critical.
- Increasing FC tolerance to freezing conditions is important.
- It is not clear how an air cooled stack will fit with DOE targets. The volumetric power density needs to be identified. The market niche proposed seems very narrow.
- Although this is a Topic 4B (Transport within the PEM Stack-- Freeze Effects) project, and is 40% complete, this project is not addressing freeze effects. Only two slides (10 and 11) had any sub-freezing data, and these did not provide any insight or anything new.
- This project addresses very important aspects that are limiting FC commercialization and holds relevance to the DOE Hydrogen Program goals. More specifically, the relevant DOE objectives addressed are sub-freeze conditions, durability, and cost.

Question 2: Approach to performing the research and development

This project was rated 1.7 on its approach.

- The approach was presented almost like a trial and error development of a stack system. According to the presenter, much iteration is needed to improve the system, which seems very insufficient and incomplete regarding the limited amount of two iterations proposed for the course of the project. This approach concentrates on a single FC design only, which offers very little potential for improving the freeze tolerance of FC systems in general. Instead, a scientific approach that helps to develop a general understanding of the processes involved in freeze start is missing completely.
- Approach uses multiple design, build, and test (DBT) cycles. However, two DBT cycles cannot be considered multiple.
- The approach is broken into three phases:
 - PHASE 1: Stack and Module Baseline Testing and Failure Modes Identification,
 - PHASE 2: Freeze Effect Design Mitigation Strategy, and
 - PHASE 3: Freeze Effect Failure Mode Mitigation Testing.
- In terms of freeze tolerance design and air cooling, there is little to no information to understand what the design iteration is.
- One would need more information of how the project will be focused, not simply DBT.
- They need to present much more information on test procedures, such as how the system is "frozen."
- There was no discussion of cost or how to achieve a 25% reduction metric.
- The DBT approach is effective, but expensive. The project team should plan to do modeling to determine the limits of the technology. It would be interesting to see whether the same air-cooled design that works and starts in a sub-frozen environment could work in a 40°C ambient temperature. Go/no-go decision point 1 requires a 25% cost reduction, but it is not clear how this can be evaluated, as the cost analysis was not presented.
- This project is not addressing freeze, to date it has primarily focused on start/stop strategies. They have not focused on freeze start/stop strategies, just normal start/stop decay. This is not supposed to be the focus of this project. Additionally, it does not appear that much freeze work will be done based on the slides. Their goal does not even include developing cold-start capability! It is, therefore, not clear why this project addresses DOE's Topic 4B goals.
- The project approach includes multiple DBT iterations to address the key issues relevant to freeze tolerant air cooled stacks. However, the lack of information regarding test and design plans raises concern on whether the project objectives will be accomplished within the project timeline.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 1.8 based on accomplishments.

- The presented data only described the problem, but it did not show any advances in developing a more freezetolerant system. While defining the subject matter of such a project is very important, a thorough scientific study of the subject matter was entirely missing, and merely a compilation of data describing the topic was presented. No information was shared with respect to failure modes. No information was given with respect to the improvement of the design iterations. Very little, if any, data was presented that would help the FC community to understand and develop a startup procedure from freezing temperatures. If any useful information was gathered during the project, it was not shared with the audience. The impression created was that all useful information that may have been created during the project was company owned intellectual property. This is a big conflict, and thus I believe the work should not be funded with DOE funds.
- Significant progress was made in the one year of the project to date.
- There is voltage degradation as a function of stack hours from 2,500 hours to >4,500 hours (still running). There is stable performance with >1,000 start/stops (still running).
- The durability cycle shows membrane leak at 1,200 cycles.
- Testing at sub-freezing conditions shows that the system cannot maintain operation at low power levels (<65A) below -20°C, but only have a 2% power de-rate to -20°C.
- Limited accomplishments were produced for one year of effort.
- The data does not back up the statement that stack performance is 100% recoverable.
- Given that this project recently began, the accomplishments are difficult to evaluate. However, the baseline data set that is forming seems to be favorable. It would be interesting to note what metrics will be improved in the next generation design.

- It appears the focus to date has been primarily on system mitigation strategies to normal (above freezing) start/stop decay, which are known (and have been published in both the open and patent literature). The broader FC community has gained nothing from this project to date.
- There is insufficient evidence of project advancement. There is only one graph that shows the test results generated during this project. In addition, there is limited information given on the test conditions, and that makes it extremely difficult to evaluate the merit of the technical accomplishments.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **1.8** for technology transfer and collaboration.

- It seems collaboration consists of two companies only. Neither a national laboratory, nor a university, nor any other research institute contributes to the work. This may explain the lack of scientific observations and may have led to an engineering approach that is based on a multitude of iterations to develop a product.
- To shift this focus to an approach that would create results that are of value for the entire FC community, the collaborations would have to be extended, the scope would have to be expanded to a less product-oriented objective.
- Partners include Plug Power (Plug), Ballard Power Systems (Ballard), and the National Institute of Standards and Technology (NIST).
- NIST was never mentioned other than on a partners slide.
- It appears that most of the design and development work is being done by Ballard with testing arranged by Plug.
- There does not appear to be close coordination between collaborators. The presenter did not answer any stack questions.
- The collaboration between Ballard and Plug seems strong. However, in the interest of the broader community, and since there is no collaboration outside of industry, it is uncertain how the findings will be disseminated.
- Their collaboration has been limited to Ballard to date. It is not apparent what is intended with their NIST partner, since NIST is never mentioned, and there does not appear to be any fundamental freeze-cell testing planned.
- The team includes Ballard, Plug Power, and NIST as described in slide 2. The individual companies' roles and their collaboration are clearly defined. However, the NIST role is not mentioned, as well as either when or how it will participate in the project. In addition, it is not shown what data NIST will generate, and who will be the primary data beneficiary, Ballard or Plug.

<u>Question 5: Approach to and relevance of proposed future research</u>

This project was rated 1.8 for proposed future work.

- The project pretty much proposes to continue with the general approach. Since this approach does not create any benefits to the FC community, the presented future work does not offer any improvements. Although some points such as "develop mitigation strategies based on failure analysis" seem valid things to do, no failure analysis was presented. If the failure mechanisms are not shared, not even in a common way, and are only used for product development of the two collaborators, again, no benefit will be given to the FC community and thus, I personally think that such a project should not be funded by DOE.
- Future plans include operable air cooled stack subsystem and commence freeze testing using real motive power application loads and environmental conditions, and then performing failure analysis. Future work should include completing failure analysis of baseline and advanced stack concepts for durability assessment and developing mitigation strategies based on failure analysis, and then building stack systems with mitigation design changes and performing mitigation verification testing.
- Considerable effort will be required in the final year of the project.
- The future work description is very broad, but all elements will contribute to project goals.
- It does not appear that any fundamental freeze testing is planned. Although there will apparently be some additional testing of stacks and a subsystem in a sub-freezing environment, it is not apparent how this will result in improvements in understanding or mitigations, much less how any findings will actually be shared with the community.

• Given the lack of specific test plan details, insufficient experimental data related to durability, and no data on failure analysis, it is not possible to evaluate the relevance of the proposed future work. Furthermore, it is not clear whether the team will be able to accomplish proposed future work within the project timeline.

Strengths and weaknesses

Strengths

- Use of real FC stacks is a strength.
- They are experienced partners.
- They have a clear understanding of market needs, system requirements, and system limitations. Although it was not presented, I expect that the cost benefit of an air cooled system is driving the design. These elements are often lacking in projects not tied to industry.
- None. It has contributed nothing relevant to the topic of interest, and it does not appear to intend to do so either.
- The project has a good mix of team strengths.

Weaknesses

- No sharing of any information that would be of real value to the FC community.
- No science aspect was presented.
- Product development oriented project with only one to two companies as beneficiaries.
- This project has limited ability to advance overall FC technology beyond a small niche market.
- The project does not appear to be focused; and adopts a very general approach.
- It seems like the pendulum between academia and industry swung too far toward industry on this project. It is not clear how this project will benefit the FC research community. This project seems as if DOE is funding the development of a product with no clear mechanism.
- Many. One of the main partners is not a U.S.-based company, which seems particularly egregious when the project appears to provide minimal impact on the broader FC community.
- Results presented to date are insufficient, and there is a concern that the proposed future work will not lead to significant improvements or accomplish the project objectives.

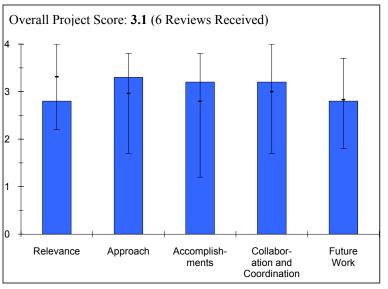
- Add a scientific aspect to the project.
- Redirect work so that it increases general understanding of freeze start phenomena.
- Share findings with FC community.
- Do not waste DOE money for company-owned product development.
- The addition of more DBT cycles would make this a stronger project.
- The project presented is very general. A better focus on the project goals is required.
- Please consider the value of this project to general FC research and adjust such that findings are transferable, not exclusive.
- Stop this project. At a minimum, insist that the balance of the funding be dedicated to freeze-relevant work. However, even this re-direct option would probably not result in a very satisfactory result, since the fundamental issue is that the application which Plug is focused on, does not require developing cold-start capability.
- The team needs to intensify the work in order to be able to accomplish the project objectives within the given timeline.

Project # FC-26: Fuel-Cell Fundamentals at Low and Subzero Temperatures

Adam Weber; Lawrence Berkeley National Laboratory

Brief Summary of Project

Project objectives are to: 1) obtain a fundamental understanding of transport phenomena and water and thermal management at low and subzero temperatures using state-of-the-art materials by examining water (liquid and ice) management with thin-film catalyst layers, and enabling optimization strategies to be developed to overcome observed operational and material bottlenecks; and 2) elucidate the associated degradation mechanisms due to subzero operation to enable mitigation strategies to be developed. Improved understanding will allow for the DOE targets to be met with regard to cold start, survivability, performance, and cost.



Question 1: Relevance to overall DOE objectives

This project earned a score of 2.8 for its relevance to DOE objectives.

- Fuel cell start-up and operation in sub-zero temperatures is vital for the automotive sector, but perhaps of less relevance to other market sectors. Perhaps this is a legacy of the automotive focus of the program in prior years.
- The understanding gained may only be relevant to the one company involved.
- Start up, operation, shutdown, and storage at low (freeze) temperatures are important to understand and address for devices that use or produce water, like fuel cells. In the fuel cell, many components might freeze. Evaluation of the effects of freezing wet graphite components and developing possible mitigation strategies is an important and timely investigation.
- The freeze topic is important for automotive applications. I only hope the same level of excellence in execution of an R&D project can be applied to the more urgent water transport issues in operating stacks.
- Understanding transport fundamentals at low and sub-zero temperatures is important for achieving fuel cell performance and durability targets. Understanding physical properties that limit thin electrode performance is most important, as it is difficult to link gas diffusion layer (GDL) water to performance.
- The project addresses the need for more theoretical understanding of phenomena in fuel cells at low temperatures.

Question 2: Approach to performing the research and development

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

- Well integrated and planned across the various partners involved.
- The approach is suitable and detailed. The delays in the timeline are unfortunate.
- PI and team have an impressive command of theoretical principles and experimental methods needed to understand the salient phenomena; this project has a very high likelihood of success.
- Stack temperature gradients at freeze shutdown and end cell ice are design-specific and should not be the area of focus.
- It is not clear how all the various experiments link together.
- In general, this project should focus more on "what parameters does the model need" rather than "what can we measure."
- The approach, in principle, is a valid and reasonable one.

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Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.2** based on accomplishments.

- In the short timeframe, much has been done; however, it is unclear how much presented is previously existing data and how much is new. A lot of the data shown is from the industrial partner's own work.
- The accomplishments are in line with the expected completion points within the schedule.
- Very good progress for a project less than one year underway.
- The design map is an excellent graphic and informative tool.
- It is unclear why ice affects electrochemical surface area (ECSA) and the conclusions are debatable.
- Imposed temperature gradients show valid fundamental phenomena, but may not be relevant in real systems, where stacks are insulated.
- Conclusions of the neutron-scattering water distribution experiment upon shutdown were complicated by purging.
- Isothermal start measurements have already been done. Common membranes are required in testing to isolate catalyst effects.
- Several milestones have been significantly delayed and are incomplete, such as the baselining of cells.
- No cyclic voltammograms showing the underpotential deposition of hydrogen peaks, used to determine the ECSA loss, have been shown. Considering the extremely low ECSA of 3M catalysts (10 m²/g), it is questionable as to how accurately one can measure the ECSA loss due to ice in the catalyst layer. A 10% change in ECSA is about 1 m²/g, and is within the error of typical ECSA measurements. More details should be presented to demonstrate the validity of this method.
- Progress is a bit behind due to the delays with getting partners under contract, but nothing serious. Progress on the water movement during shutdown and the diagnostics on ice formation in the GDL and the catalyst layer is interesting.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.2** for technology transfer and collaboration.

- The partners are well coordinated, but there is only one industrial partner. This is a weakness.
- The level of collaboration is appropriate to the topic.
- The roles of the partners are stated, but it is not entirely clear what organizations have conducted which pieces of the work to date.
- This is a very strong team; it is important to show the contributions of all involved.
- Collaboration with LANL and 3M seem to be going well. It is too early to evaluate other partners.
- Catalyst layers from UTC should be used for this project as they may represent a more general, valid, and useful system that deserves detailed study.
- Good balance of partners and resource distribution in the project. This is key to the success of the project, as the partners are providing the input data for the models.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.8 for proposed future work.

- I am concerned that the desire appears to be to optimize and understand the impact of all other membrane electrode assembly components on the thin-film catalyst layers from the industrial partner without focusing on the source of the problem, which is the ultra-thin catalyst layer itself, with its own unique structure and composition. I feel that the focus should be the catalyst layer, as the project seeks to engage the industrial partner that supplies the thin film catalyst layer and who stands to benefit most from a successful project.
- The proposed future work appears to be appropriate for this topic.
- The vision of the ultimate testing needs to be more fully explained.
- Much work on modeling has already been reported on start-up/shutdown and cold starts. It is not clear what is new in the future work proposed in this project besides the fact that the catalyst layer is atypical.

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Strengths and weaknesses

Strengths

- Focus is on a critical issue for the automotive fuel cell sector.
- Good team with input from an industrial stakeholder.
- The strength of this project is the approach to the problem.
- The PI has the team and member capabilities to successfully execute.
- Well resourced, strong team with strong collaborations. Their focus on water transport issues of NSTF is encouraging.
- The general approach is good.
- The team put together for the project is well rounded.

Weaknesses

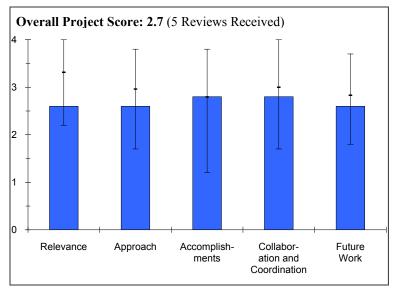
- The focus is on every component but the one with the problem, the catalyst layer.
- The project has limited value to any other party except the single industrial partner. It is possible that this DOE project will give a competitive advantage to a single company, which is undesirable.
- A key weakness may be the lack of re-evaluation of the pore structure of the components after repeated freezethaw cycles. Answering the question of whether forces generated by the expansion of water while freezing increase the pore sizes with time, changing the water distribution within and between components after repeated freeze thaw cycles, would help to clear this up.
- The test plan is weak.
- Scattered results from a wide variety of experiments may overcomplicate the modeling effort. Not enough input from OEMs regarding real-world shutdown and startup freeze parameters is a definite weakness.
- The choice of 3M NSTF catalyst layer is highly questionable in terms of usefulness to the fuel cell industry.

- Many complicated experiments will require complicated models. Reduce the number of methods and improve rigor with statistics.
- The focus should include the understanding of water and freezing within the thin catalyst layer structure.
- Identifying whether the hydrophobicity of the components change with time in an oxidizing or reducing atmosphere could be useful.
- Clarification of issues at extreme low temperatures (<-35°C) is needed.
- Provide more clarity around the link between experiments and models, and how it all fits in the big picture.
- The project should be changed to evaluate a standard Pt/C catalyst layer currently used in the fuel cell industry rather than the 3M NSTF.
- This reviewer would like to see a bit more work put in the scope for non-NSTF catalysts.

Project # FC-27: Development and Validation of a Two-phase, Three-dimensional Model for PEM Fuel Cells *Ken Chen; Sandia National Laboratories*

Brief Summary of Project

Project objectives are to: 1) develop and validate a two-phase, three-dimensional transport model for simulating polymer electrolyte membrane fuel cell performance under a wide range of operating conditions; 2) apply the validated fuel cell model to improve fundamental understanding of key phenomena involved and to identify ratelimiting steps and develop recommendations for improvements so as to accelerate the commercialization of fuel cell technology; and 3) employ the validated fuel cell model to improve and optimize fuel cell operation. Consequently, the project helps to address the technical barriers on performance, cost, and durability, and to achieve DOE's near-term



technical targets on performance, cost, and durability in automotive and stationary applications.

Question 1: Relevance to overall DOE objectives

This project earned a score of **2.6** for its relevance to DOE objectives.

- A two-phase, three-dimension transport model backed by cell/stack data will help elucidate transport phenomena in the fuel cell, including the microporous layer (MPL) and interface discontinuities.
- It is intended to provide an executable model to the industry.
- The project addresses the modeling of the performance so far in a one-phase model with future plans to move to a two-phase model. It is not clear how the model will address the objectives of improving cost and durability at this point, except as related to some possible improvement in performance which will depend on the choice of material parameters used in the model.
- The attempts to address performance, cost, and durability are all relevant.
- It is not clear how this project will support DOE's objectives. The PI claims that it will address "performance, cost, and durability," but not a single specific example of how this will be done is provided in this presentation, not even on the future work slide. Perhaps something useful will eventually result from this project, but the PI does not appear to have any clear objectives. If he has any, they are not stated or shown.

Question 2: Approach to performing the research and development

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

- Continuity equations serve as the governing equations. Presumably, these equations will be simplified to make the model manageable.
- It is not clear what is different from other previous modeling efforts.
- Experimental information from Ford and Ballard Power Systems (Ballard) will support the model.
- It is not clear what diagnostic tools will be used to verify the three-dimensional aspects of the model.
- Transient capability (or the possibility thereof) is not evident.
- The modeling approach seems to be a sound development of the parameters for the model moving in a stage wise approach to the two-phase model. The team seems to be seeking good input parameters to develop a model that will be applicable to real systems.
- Development and validation of model progress has been acceptable to date.

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• It appears that the major contribution of the PI is to simply incorporate sub-models provided by others (The Pennsylvania State University and Lawrence Berkeley National Laboratory (LBNL)) into a big threedimensional model. However, it is not clear what additional value this three-dimensional model potentially offers.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.8 based on accomplishments.

- The project is less than a year old.
- A single-phase, three-dimensional, single-cell model has already been developed.
- The model correctly predicts trends, but absolute values are not always in line with experimental data.
- Progress on the model seems to be good with a good fit to the performance parameters. While progress in modeling has been accomplished, there is no mention of error or error magnitudes, and there is essentially no verification discussion or line-on-line comparison between the model and direct measurement.
- Although a lot of pretty slides were presented, it is not obvious what work done by the subcontractors was actually supported by this project (vs. results obtained on other projects and simply incorporated here). More importantly, it is not clear what anyone was learned (or, for that matter, is going to be learned) from the project.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated 2.8 for technology transfer and collaboration.

- Use of Ballard's experience to set model inputs and boundary conditions, as well as material properties, is helpful.
- The participation of Ford to provide real-world operating conditions is beneficial.
- The program has good collaboration efforts, bringing together the modeling effort with good experimental measurements of materials properties such as the gas diffusion layer (GDL) and MPL properties, fuel cell performance, and vehicle performance targets.
- Collaborative efforts could be a little stronger. Partners seem academia heavy.
- This project obviously requires a lot of collaboration, since all of the team members must provide substantial input to generate the complex model of the sort envisioned here, especially if the model is also going to be validated (instead of just being used to "predict general trends"). It is also good that the project has an original equipment manufacturer (OEM) like Ford participating with no funding from DOE.
- The PI also takes credit for what seems like a lot of dissemination to date; e.g., two journal publications, two proceeding papers, and three conference presentations were generated so far. A team member also served as co-editor of a book on fuel cells). However, it is not clear that any of this had anything to do with this project (all of the subcontractors have multiple other projects and funding sources), much less what was actually communicated; i.e., what have we learned from this to date that is worth publishing.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.6 for proposed future work.

- A logical progression of model development and validation is planned.
- Interim model validation will be undertaken.
- The proposed approach of continuing to the partial two-phase model with the validation of the current model seems to be sound. The continued incorporation of the DAKOTA approach to make the model predictive and allow for uncertainty is good. I think it is important to address the water flux as described in the future work.
- Future validation and data generation were specifically mentioned and noted as necessary.
- Not a single specific example of how this project will support the DOE objectives is given in this presentation, not even in the future work slide.

Strengths and weaknesses

Strengths

- The approach of modeling the behavior and trying to build in the uncertainty is an important step. The focus on generating good data for the model, under a range of conditions, as well as gathering fundamental data on the mass transport and the effect of materials properties, is a definite strength.
- It is very challenging to simulate fuel cells on such a detailed level. But it would be great if that works.
- The project has good subcontractors that can provide good sub-models and good validation data. It is not clear what the value of the lead is here.

Weaknesses

- Although the model development is at the early stages, it is not clear how the model's predictive capabilities will address the cost and durability goals. Some more information on how the cost and durability will be addressed is needed in the project work scope.
- Validation of the modeling to date is weak.
- The PI does not appear to be very familiar with what has been done in fuel cell modeling. For example, he states that MPL has not been treated by fuel cell models, which is not true (he appears to qualify this statement with a "multi-dimensional model," but it is not clear what has been, or can be, learned from a "multi-dimensional model" vs. the models that have already been published on MPLs).

- I recommend comparing the simulation results with other simulation groups based on a more commonly used flow field design.
- More validation with possible line on line comparisons or discussion regarding errors and causes is needed.
- Make sure that this project has specific deliverables that actually add value; i.e., milestones other than just model maturity goals. For example, predict trends that are not obvious, or predict decay mechanisms. Otherwise, if the project does not have any clear goals, the project should be stopped.

Project # FC-28: Transport Studies Enabling Efficiency Optimization of Cost-Competitive Fuel Cell Stacks James Cross; Nuvera Fuel Cells

4

Brief Summary of Project

The objective of this program is to investigate transport limitations at high current densities in order to optimize the efficiency of a stack technology meeting DOE 2015 cost targets. Goals for fiscal year 2010 are to demonstrate a 1 W/cm² stack with 0.2 mg_{Pt}/cm^2 and to implement a predictive model for a high power density version (version 1.0).

<u>**Ouestion 1: Relevance to overall DOE**</u> <u>**objectives**</u>

This project earned a score of **3.6** for its relevance to DOE objectives.

- This project addresses critical barriers to be overcome to achieve DOE research, development

Overall Project Score: 3.3 (7 Reviews Received)

- to be overcome to achieve DOE research, development and demonstration (RD&D) targets.
- The program is a good fit with DOE cost and performance goals. The program objectives are focused on the goals to address cost and performance, as well as water and thermal management.
- Operating at high current densities is essential to the practical application of fuel cells (FCs) in vehicles. Basic understanding of the cell and stack dynamics is necessary to improve operating performance.
- If heat rejection is carefully considered in the cost model, this project will remain relevant.
- The project is clearly focused on cost reduction and performance.
- The concepts of increasing stack efficiency and cost reduction are in the heart of the DOE program. The center of the program is a detailed predictive model, but this model appears to be only valid for Nuvera. If this is true, then the value of the program should be decreased.

Question 2: Approach to performing the research and development

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

- The overall approach is interesting as it studies high current densities in order to achieve the cost targets and does not take efficiency as the first aim.
- Starting the study on a stack level is appreciated as it should lead to shorter transfer time to a system level.
- The program team appears to be adopting a sound approach to a series of models of the single cell, stack, and the membrane electrode assembly (MEA) behavior. The models are being linked in the overall approach with good testing of the single cell and stack to validate the two independent models. Although the program is in the early stages, it looks like it shows some promise of contributing to overcoming some of the barriers.
- Well-designed, logical project plan. Good use of modeling and experimentation to achieve results.
- Common model development approach that will result in a model that is only as precise as the experimental database. Uncertainty analysis of experimental data is recommended.
- Maximizing power density, even at the expense of overall operating efficiency, is certainly worth pursuing, especially in transportation applications (where the time spent at maximum power is relatively low). The PI seems to have a good understanding of the objections may be to this approach; i.e., heat rejection and possible durability impacts.
- Chart 4 in the presentation is a series of loops that never go anywhere but around the development circle. The presentation did not indicate how the predictive model could be used by DOE. The model will be specific to Nuvera, which is of limited to value to the rest of the FC community.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.1** based on accomplishments.

- Results are correct regarding the start late in 2009.
- The comparison between the Nuvera model, which is based on experimental data fitting, and the new model, which is based on physics, is interesting.
- The difference between the tests in preparation and the first results indicates that the data will be useful in the model.
- The program is still in the early stages, but the initial modeling and data looks promising for contributing to cost improvements.
- Good progress has been made considering the early-stage of the project. Reviewer is looking forward to future presentations!
- Limited results thus far, but too early in the project to accurately score.
- The project has good progress for 15% complete.
- In chart 11, the slope of the Orion baseline has changed because of the thin membrane. The benefit of the Orion system, other than the membrane change, is not clear, since it starts at the JM Andromedia hardware.
- The Nuvera results in Chart 13 show 3M performance for an approximately 66% increase in catalyst content. It is not clear why this is beneficial.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.1** for technology transfer and collaboration.

- Collaboration between partners seems correct.
- The team seems to be well coordinated with good initial progress on the program in both the modeling and the testing of the materials properties of the MEA.
- Good collaboration with appropriately chosen partners.
- Collaboration and organization seems strong, but it is not clear how work from the Lawrence Berkeley National Laboratory (LBNL) doesn't overlap other projects.
- It is good that partners not officially included, like 3M, are being included as well.
- It was not clear if the partners were contributing to the program. The Pennsylvania State University (PSU) data was given, but LBNL effort is in future, and the program is 15% complete. Humidified cathode results appear to be new to Nuvera, but others have seen the influence of cathode humidification before. Not sure why this was such a revelation.

Question 5: Approach to and relevance of proposed future research

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

- Proposed future work is in accordance with the targets of the project.
- The proposed future works is a good approach to combine the MEA model into the model. The program also incorporates goals for improvements of the MEA materials
- Good plans. Will need strong management and leadership to keep project focused. Somewhat unclear what specifics will be done as far as material development; i.e., how to decrease platinum loading without reducing performance and how to increase membrane conductivity (reduced resistance) without sacrificing cycling durability.
- It is not clear what fundamental understandings may result from this project, or how they will be shared. For example, will non-cell design specific barriers to mass transport at high current density be addressed and shared here? Perhaps challenging the targets on system efficiency is sufficient.
- Future work is proposed as activities. The benefit of the future work is unclear.

Strengths and weaknesses

Strengths

- Nuvera has adequate partnership.
- The comparison between a model based on experimental data fitting and a model based on physics is a strength.
- Testing and modeling at a stack level is a strength.
- Good modeling approach using multiple approaches to the modeling of the single cell and stack to understand the key performance cost drivers. The modeling also includes modeling of MEA and plans to incorporate this into the overall model. The goals include improvements in the material properties for improvements based on the model.
- Nuvera has a good team and a good plan.
- Validation of developed model with other simulation from the very beginning. Liquid water visualization in the FC is great to support modeling results.
- Competitive material sets are being evaluated.
- Nuvera has a good objective and approach to achieving the goal.
- Nuvera has a high performance FC system that operates at high power densities with usually higher catalyst loadings. If the design of the Nuvera polymer electrolyte membrane fuel cell (PEMFC) can be shown to operate at low catalyst loadings and also at high power densities, then this concept should be beneficial to the DOE automotive goals. Nuvera has a creative and experienced team.

Weaknesses

- Stack morphology is not a common design. Therefore, the results of this project will be difficult to be applied by the other actors in the FC community.
- Apparently, there is no link with this PSU model and the PSU model developed in the project "Development and Validation of a Two-phase, Three-dimensional Model for PEMFCs" (FC027).
- Material development details are somewhat weak and may get flushed out as project proceeds.
- It is not clear if the high current density goal is applicable to a wide range of FC applications.
- It is not clear what fundamental findings will be derived from this project that can be shared and can help the wider FC community.
- The program will benefit Nuvera if successful. It is not clear how a successful program will help the rest of the FC community.

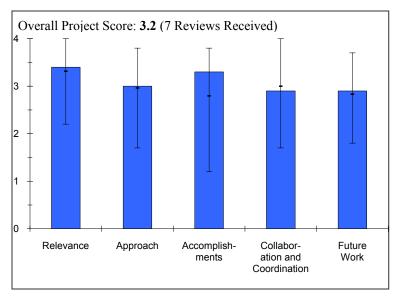
- The project covers single cell and stack level, but in order to fully validate the DOE stack targets, the system architecture and the system cost impact should be evaluated.
- Should have more emphasis on fundamentals of mass transport and sharing these findings with the community (via the "detailed predictive model" that is supposed to be central to this work).

Project # FC-29: Water Transport Exploratory Studies

Rod Borup; Los Alamos National Laboratory

Brief Summary of Project

The overall objective of this project is to develop understanding of water transport in polymer electrolyte membrane fuel cells (PEMFCs) (non-design-specific). The objectives are to: 1) evaluate structural and surface properties of materials affecting water transport and performance, 2) develop (enable) new components and operating methods, 3) accurately model water transport within the fuel cell, 4) develop a better understanding of the effects of freeze/thaw cycles and operation, 5) develop models that accurately predict cell water content and water distributions, 6) work with developers to better state-of-theart designs, and 7) present and publish results.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.4** for its relevance to DOE objectives.

- Water transport deserves and has received a lot of attention lately. This project directly addresses the issue and demonstrates some approaches to managing water transport in the gas diffusion layer (GDL).
- This study targeted some of the central issues in fuel cell water management that are essential to understanding the material impacts on performance and, by association, durability.
- Understanding of water management in cells and stacks will likely lead to enhanced performance in commercialized fuel cells. The ability to accurately predict water content at various points and under various conditions will lead to a better understanding of failure modes, particularly for freeze/thaw. This reviewer would have liked to see an automotive or other large stack manufacturer take a role in the project. W.L. Gore is listed only as a supplier.
- Water management is very specific to a given set of materials such as GDLs, catalyst layers, membrane thickness, flow field coatings, as well as operating conditions of flows, inlet relative humidity (RH), etc. It is not clear how the results of such a general non-design project with varied materials can be combined to produce a high performing stack. Ideally, the project should focus on a particular stack and show how the understanding has improved its performance and durability.
- The understanding of water management is critical for fuel cell performance and durability. This is why this project is highly relevant to the overall DOE objectives and fully supports the Hydrogen Program.
- Water management phenomena are critical to understand to develop fuel cells with thinner, lower-loaded catalyst layers that are less prone to flooding. In this sense, water management facilitates achieving cost targets.
- This project is directly relevant to understanding water management phenomena since it attempts to link membrane electrode assembly (MEA) material properties and operating conditions to observable catalyst layer flooding or, at the least, lower performance that is attributable to greater presence of condensed water within a catalyst layer.
- Water management projects would lose relevance if their results were specific to a particular cell design, plate material, GDL, or other cell component. This project has been designed to avoid this and to provide a general perspective.

Question 2: Approach to performing the research and development

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

- The approach effectively covers modeling, experimental validation, and successful application of techniques to improve water management and, consequently, cell performance by GDL design and materials selection.
- LANL is also attempting to design GDL properties to emulate the aged properties to enhance stability and constant behavior throughout the life of the cell or stack.
- The approach has been very clearly articulated. PI and team exhibit commendable focus and discipline in pursuit of their results.
- While process condition impacts are assessed, the role of compression, the overall mechanical state of electrochemical package, and the flow field structure appears to be absent. There was no analysis presented that suggested these could be neglected for the adopted experimental design.
- The PIs employ a good approach, with a combination of modeling with *in situ* experimentation.
- The use of an approach on a non-specific basis so that general conclusions may be spread across a broad segment of devices is significant.
- To study water management, the team uses advanced *in situ* and *ex situ* experimental techniques. The approach is methodical and scientifically sound. However, it lacks the explicit correlation between experimental results and the modeling effort.
- There are some very interesting experiments designed here, but they do not entirely connect with the fundamental water management "questions of angst" that a fuel cell developer asks, such as: "What material properties help me get out to higher current density?" or "How do I keep from drying out the membrane even though my operating conditions are dry?" For example, the studies of GDLs with different tetrafluoroethylenes are a good benchmarking study. However, a National Laboratory effort should probe deeper. If SGL 24DI is a good water-holding GDL, then why? Is it because the MPL has 10% TFE (hydrophilic relative to sample 25BC), or is it because of a lack of defects in the microporous layer (MPL) (slide 34 in the supplemental slides shows some hint of this work)? What is the water transport mechanism through the MPL and how do the MPL characteristics influence whether or not it happens? Are there contributions from GDL water vapor diffusivity and thermal conductivity?
- Regarding the Schroeder's Paradox experiments, more water is in the membrane when liquid water is outside, but the real question is whether water stays in the membrane when the operating conditions are dry. Does Schroeder's Paradox imply that it would be better to design electrodes with high thermal conductivity so that liquid water will more likely exist outside the membrane? Or will this be negated by the urgency to remove water from a thin catalyst layer to avoid flooding?
- Despite the need for experiments to be more focused on material properties, there are some nice overall trends shown in the work, exemplified by SGL 25BL versus 24BC contrast, where performance losses are clearly more associated with catalyst layer water, not GDL macroporous substrate water.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.3 based on accomplishments.

- Modeling and experimental data have been used to design a graded-property GDL that results in a meaningful increase in cell performance, especially in the mass transport, high-current-density region of the polarization curve.
- The results offer fresh insights into gas diffusion media behavior in operating stacks.
- Good, quality work as expected from a top-notch National Laboratory. It is unclear how this knowledge will translate to real-world applications. Seems more like an Office of Basic Energy Sciences (BES) project. It seems like there is already a large body of knowledge around water transport, states of water, hydration in cells, freeze/thaw, etc.
- Although a significant amount of work has been shown on GDL/MPL characterization *in situ* using electrochemical impedance spectroscopy (EIS) and water profiles, a characterization of the different pores, separation of bulk diffusion from Knudsen diffusion, etc. and a thorough study of the limiting currents using different carrier gases for oxygen does not seem to have been explored. Most of the results are qualitative and correlations and valid for arbitrary sets of materials and operating conditions.

- Controversial claims on the validity of Schroeder's Paradox are questionable. The key to the Paradox is the equilibration time, which is not discussed by the authors. There is likely no paradox for long equilibration times.
- The technical accomplishments are outstanding in terms of quality and quantity. The team demonstrated an excellent progress towards water transport imagining under sub-zero conditions.
- For what the project intends to do, the progress is voluminous and outstanding. Although people have talked about ideas like "distributed GDLs," where performance is increased by GDLs that have dry/wet-amenable properties in different regions of the active area, this is the first DOE-funded project to actually show it.
- In many experiments throughout the project, there are examples where *in situ* diagnostics, particularly AC impedance, is used to derive relevant information.
- Successful completion of research efforts to address hypotheses about Schroeder's Paradox and membrane humidification versus compression were shown. It may be interesting to see whether compression redistributes the water between channel and land.
- Higher MPL polytetrafluoroethylene (PTFE) loadings were associated with lower degradation in freeze/thaw, but it would be interesting to find whether it was the PTFE itself that allowed water to exit the cathode catalyst layer, or another property of the MPLs. Recent literature has a lot to say about MPL defects.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **2.9** for technology transfer and collaboration.

- The team includes National Laboratories, universities, GDL and membrane suppliers, and a stack/system integrator.
- Deep and broad technical competency in the team is excellent. Testing at the stack level with an industry partner could have strengthened the project even further.
- The level of "strong" collaboration is unclear. The team could likely use a stack manufacturer as a close partner.
- I do not see much input from industry on this project in regards to fuel cell and stacks. I have not seen any modeling results and their contribution to this project.
- Very broad and comprehensive list of collaborators.
- The efforts of the team members are well managed and integrated. Collaboration with National Laboratories, academia, and some industry members (Gore and SGL) is well demonstrated. However, it is not clear how Nuvera is involved.
- Collaboration is very instrumental in the execution of this project. The roles of the NIST, SGL, and Gore are fairly obvious in the work that has been done so far.
- The project is taking on new collaborators, as evidenced by assistance to be provided to SNL and LBNL transport projects. ORNL appears to be newly involved in investigations of water in the catalyst layer.
- From the slides provided, the roles of Nuvera and the University of Texas are not entirely clear.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.9 for proposed future work.

- The project is nearly finished.
- Model validation will be completed.
- Accelerated stress tests (ASTs) will be applied to the graded-property GDL to evaluate GDL property stability.
- Good considering the project is nearly complete.
- Quantitative characterization of mass-transport losses due to water management issues is needed. Input from industry should be solicited. Proof of the usefulness of this type of generalized work should be demonstrated by applying the understanding to a real fuel cell stack and showing overall improvement in performance with the recommended operating conditions and materials set.
- Future work planned completes tasks currently underway while launching appropriate new investigations based on observed data.

- The proposed future work seems logical and aims at generating data necessary to finalize the water profile and multi-phase transient models. However, there is no plan to evaluate different GDL materials and interaction of the GDL and bipolar plate configurations, which may affect the universality of the water transport models.
- The X-ray tomography is a welcome addition to investigate the water transport mechanisms that exist in GDLs. This technique is exactly what should be done to drive the project from near-benchmarking to a fundamental study.
- The intention is noted to share the models that were derived in this project. Some experts have talked about how DOE water transport projects have not yet delivered mathematical models that can be used by the fuel cell research community; hopefully, this project will break that trend.
- The segmented cell measurements are interesting, but they must be accompanied by material characterization and proper focus on water transport mechanisms.

Strengths and weaknesses

Strengths

- The use of a wide array of investigative tools, an understanding of water transport and correlations to performance are all strengths.
- The PIs perform good basic studies, and use powerful characterization techniques.
- Significant amount of work has been shown on GDL/MPL characterization in situ using EIS and water profiles.
- An excellent National Laboratory and university team with excellent analytical capabilities is complemented with experienced industrial partners.
- The project is able to deliver considerable volumes of data and experiments.
- The project contains a wide range of collaboration, in both industry and national laboratories.
- The project is able to formulate a hypothesis and deliver data to either prove or disprove the hypothesis. Unfortunately, there are not many DOE projects that are as adept at doing this as this project has proven to be.
- The investigators have considerable experience in the field and demonstrate cutting-edge knowledge of the issued involved.

Weaknesses

- The testing at larger active areas in stacks and lack of investigation of flow field/GDL interactions are both weaknesses.
- It is unclear how the findings will translate to real-world applications.
- Characterization of the different pores, separation of bulk diffusion from Knudsen diffusion, etc., and a thorough study of the limiting currents using different carrier gases for oxygen do not seem to have been explored. Most of the results are qualitative and correlative and only valid for arbitrary sets of materials and operating conditions.
- *Ex situ* measurements of water transport properties of porous materials need to be conducted and related to the improvements in the cell.
- The PIs need to demonstrate industry interest in this kind of work to improve stack performance, and conduct and demonstrate stack testing.
- The influence of the fuel cell flow field-GDL configurations is not considered in the project.
- The project has a large scope and more discipline is required to chase after the issues that are most meaningful to fuel cell stack developers.
- The cell-level experiments need to be better connected to water transport mechanisms and material properties. To be sure, there are examples within a project with as large a scope as this one where the connections are made, but on some important experiments, the connection is not made.
- Modeling could play a more significant role in explaining some of the data obtained and towards generating more useful experiments.

- More modeling results would be useful to see.
- The PI needs to summarize the relevance of experimental data to the overall project goals.

- The project could take the cell-level results that have been generated, and spend more time understanding component parameters and water transport mechanisms. This understanding might lend itself to improved water transport models.
- The small effort to understand how to measure water presence in a catalyst layer might benefit the research community tremendously, if successful.
- There should be some reporting on whether GDLs in segmented cells are all the same thickness or whether there might be unequal compression on the GDLs. It would be interesting to know whether this influences performance and impedance measurements.

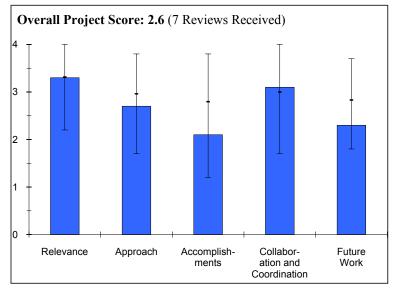
Project # FC-30: Water Transport in PEM Fuel Cells: Advanced Modeling, Material Selection, Testing, and Design Optimization

Vernon Cole; CFD Research Corp.

Brief Summary of Project

The overall objectives of this project are to: 1) improve understanding of the effect of various cell component properties and structure on the gas and water transport in a proton exchange membrane fuel cell (PEMFC), 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.

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>



This project earned a score of **3.3** for its relevance to DOE objectives.

- The control of water transport in fuel cells (FCs) is a major performance issue; this project intends to address and optimize the water transport issue and is certainly relevant.
- Optimized water balance and water transport is critical to FC performance, cost reduction and commercialization.
- Understanding water transport fundamentals in FCs is critical for achieving DOE performance and durability targets.
- The project addresses water transport, which is a major technical barrier for automotive FCs.
- The project objectives are overly generalized and not specific. It is not clear what is being measured, determined, or how the results integrate with the overall goals of EERE's FC efforts.
- The project addresses water transport in a FC—a critical technology barrier that affects FC performance, durability, and cost. The project is relevant to the DOE Hydrogen Program and fully supports DOE objectives and goals.

Question 2: Approach to performing the research and development

This project was rated **2.7** on its approach.

- The proposed approach is focused on using computational fluid dynamics (CFD) to model water transport in FCs with the emphasis this year on gathering experimental data and modeling water transport in membrane electrode assemblies (MEAs). This approach supports the project's goals and objectives of using CFD modeling to improve overall FC performance.
- The approach correctly develops the fundamental knowledge, models, and applications to improve water transport within PEMFC. This includes improved understanding of the effect of various cell component properties and structure, demonstrating improvements in water management in cells and short stacks, and encapsulating the developed understanding in models and simulation tools.
- The methodology to advance the science includes Lattice Boltzmann Modeling models, CFD models, *ex situ* characterization of components (gas diffusion layers (GDLs)), and *in situ* diagnostics including current and water distribution. All are excellent methods to advance the science.
- The relative permeability experiment is not clear. It is also unclear how liquid saturation is achieved and quantified.
- Water transport is best measured by doing direct water balance rather than pressure drop measurement.
- There was no mention on whether liquid water in the anode is considered.

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- Water accumulation in u-bends of serpentine channels is already well established.
- It's not clear how wet flow distribution in a four-channel array is relevant. It is not clear whether the final model includes inlet liquid water.
- They need to look at extreme operating conditions (low temperature/low stoic).
- While the overall approach for experiments and modeling is good, the mathematical approach for solving the transport equations does not appear to be the correct approach. Model convergence and numerical stability are a problem.
- The approach of combining modeling work with experimental characterization and the introduction of improved FC component concepts sounds good on paper, but the actual accomplishments of the research team were below expectations, and I did not see the integration of project tasks and project results.
- The team has a good approach in studying water management. They use the *ex situ* characterization of key materials properties to develop a model that is verified *in situ* diagnostic tests. They also incorporated the GDL-flow field that is critical for water management. However, the approach is missing the effects of compression that is of a significant importance for water management through GDL.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **2.1** based on accomplishments.

- Although progress has been made in the past year, the project is behind schedule. Validation problems were noted as the reason for the schedule slip; validation of the CFD models developed in this project is critical to the ultimate success of the project. Thus, it is important that model validation be confirmed before water transport improvements can be identified.
- The gas as permeability was found to increase significantly below a water saturation threshold (typically 20%) for SGL Group media; however, in operating FCs, GDL water saturation rarely is above 20%. This does not appear to be the limit issue in terms of performance related to mass transport limitations.
- Significant work on droplet emergence analysis, prediction, and measurement has been produced; however, other projects have shown that this portion of the water transport is not the rate-limiting area leading to mass transport losses.
- The model evaluation for the operational cell and comparison to experimental data leaves much to be desired. The single-phase model and two-phase models do not appear to agree. Neither model appears to agree with the experimental data, which is likely due to the fact that the experimental data has issues. Current distributions along a cell length do not typically look like the experimental data used in this project. Therefore, it is relatively impossible to match the data.
- Identified "intermediate" polytetrafluoroethylene (PTFE) loading as optimal. This is a trivial conclusion, not quantitative, and not a substantial improvement, as this was known more than a decade ago, with actual loading numbers presented.
- Wicking channels have been around since fuel cells and the Gemini program. Hydrophilic surface coating for channels as well were widely discussed, but difficult to implement in mass manufacturing.
- Some caution is recommended to avoid the model improvement being based too much on test data and less on first principles. The task is modeling and not adaptation of a first model until it fits.
- One cannot validate the two-phase model. All studies based on the model are questionable without validation.
- Wicking channels have been shown to reduce pressure drop. One wonders if that can be done cost effectively. Also, wicking channels may create other problems such as water accumulation in the smaller channel that will be harder to remove.
- Code convergence and stability problems are slowing progress.
- The models do not do a good job of matching experimental data. One-phase flow model provided better projections than an improved two-phase model. The project needs to validate the models with *in situ* data and verify that the models can predict observed behavior and are applicable to real systems.
- Obtain some water balance data to validate water crossover.
- The microporous layer (MPL) water transport needs to be validated.
- More details on options being pursued to improve transport would be helpful. For example, the wicking channel design needs principles behind it to be of use to people with different plate designs and materials.
- The water management in FCs is an ongoing concern. It is not clear whether this project has gotten us closer to solving or understanding the water management problem in PEMFCs. It was difficult to assess the modeling

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work because no details of the theory were presented. Plans and milestones listed numerous challenges such as difficulties, in the approach. How was water transport modeled in the membrane? How do the model results compare with experimental data? I do not see the value of flow visualization (droplet emergence) experiments. What important conclusions were drawn from such work? Also, the absence of water balance measurements was troubling.

• The team presented very good experimental results regarding water transport using screening tests at the component level. However, the results of the experimental model verification show discrepancy between the experiment and modeling results. The models developed require further refinement in order to achieve the project objectives.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.1** for technology transfer and collaboration.

- Good collaborations with industry and university partners. The partners appear to be playing an active role in the project and are making substantial contributions. Several publications in the technical literature were listed.
- The overall team and partners are an excellent list of collaborators. The team seems to be a good collaborative unit.
- Ballard Power Systems (Ballard) provides materials and data, but little details on materials or designs. The University of Victoria did poly(dimethyl siloxane) chip droplet dynamics studies for controlled experiment, mimicking water emergence at channel/GDL interface. The input of the other partners is not clear.
- The collaborations within the project partners are good. Collaboration outside the project partners is not evident.
- The project has numerous collaborative partners, but it appears that most of the work was carried out by Ballard and BCS Fuel Cells. The coordination of subproject tasks and the integration of subproject results are both questionable.
- The team involved is well balanced with credible members. However, it is not clear how the information exchange is managed within the team members.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.3 for proposed future work.

- Plans for future work are adequate—emphasis should be on completing model validation and then identifying water management improvements.
- There needs to be convergence between the experimental data and the different models.
- There was no mention or representation of the Lattice Boltzmann modeling. That was a portion that was novel to this project.
- They should not explore new channel designs and surface coatings unit the two-phase model is validated with the experimental data.
- The future work slide in the PI's presentation is far too generalized. The future work appears to be nothing more than a continuation of present project tasks. For example, it is highly doubtful that BCS, Techverse, and CFDRC will ever do a specific GDL modification.
- The proposed future work seems logical and in agreement with the project objective. However, the plan does not include the evaluation of different GDL materials and effects of stack compressive forces. They are not considering that these aspects can dramatically affect the model usefulness for predicting FC performance.

Strengths and weaknesses

Strengths

- They have experience and expertise in CFD modeling.
- Two-phase permeability measurement could be a useful test that others can employ.
- The overall project tasks (understanding and improving water management in an operating FC) are important and of relevance to the DOE. Some of the results are worthwhile.
- The team demonstrated excellent component screening tests.

Weaknesses

- Validation is lagging behind schedule—milestones have slipped.
- The experimental data used for model comparison does not appear accurate from other literature.
- The project examines a portion of water transport (droplet emergence from the outside of the GDL), which other developers have shown to not be the limiting issue with water transport and related to mass transport limitations.
- Model details were not provided, so there is little use for the community.
- Details of wicking geometries were not provided, so there is little use for the community.
- The studies focus on serpentine flow fields, which are not of interest to original equipment manufacturers.
- Overall, more experimental rigor is required.
- Model stability and convergence issues and the model have not done a good job of replicating data.
- Project accomplishments were not impressive.
- Model details were not presented and the accuracy of parameters in the model are not known.
- No specific recommendations were given for new flow channel designs.
- No preliminary data is provided, which material properties need to be modified to optimize water management and how they will be included into the model. On slide 16, they mentioned that intermediate PTFE loading is optimal for a particular GDL; however, the PTFE loading can affect not only the GDL wettability but also can change permeability, thickness, thermal and electrical conductivity, etc.

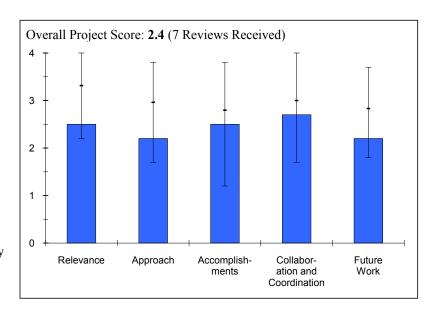
- Focus near-term on completing model validation.
- Perhaps a go/no-go decision is appropriate prior to beginning cell experimental activities.
- The experimental data for model comparison needs additional work, and is likely the cause of significant differences. The different model agreements should be better aligned.
- Revisit fundamental material parameters as part of two-phase model validation efforts. Perhaps do a sensitivity analysis of membrane water diffusivity or electro-osmotic drag coefficient, etc.
- In the future, the team plan must include the effects of compression on the GDL properties and the material anisotropy that can drastically offset experimental and modeling results.

Project # FC-31: 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 proton exchange membrane fuel cell (PEMFC) combined heat and power (CHP) system and to demonstrate it in an International Partnership for the Hydrogen Economy (IPHE) country (United Kingdom (UK)) outside of the U.S. Project objectives for 2009 - 2010 are to: 1) build, test and validate high efficiency (absorptionenhanced reformer (AER)) fuel processor bench-scale rig with 40% electrical efficiency, 2) build and test an integrated CHP system with multiple thermal recovery streams to achieve greater than 70% efficiency, and 3) perform 24-hour a day stack testing to demonstrate 40,000 hours durability.



Question 1: Relevance to overall DOE objectives

This project earned a score of 2.5 for its relevance to DOE objectives.

- The relevance is good: project to develop a high efficiency 10-kW PEMFC CHP System. However, it seems the Project Objectives for 2009 2010 to meet DOE targets should be updated to meet the new 2015 targets.
- The goal of this product is highly relevant. The published goals are aggressive and are approximate to the current, published accomplishments of commercial phosphoric acid fuel cell power plants. Electrical efficiencies should be achievable. Overall efficiencies and durability should be an interesting challenge, especially for direct hydrogen systems. It is unclear what the level of reclamation of heat is for the CHP. It could be envisioned as hot water or space heating.
- Should goals be met, the relevance would be good or better. There is not a compelling path to the stated goals in the content provided.
- This project does not offer any technology or system advancement to the program. The presenters did not demonstrate any reason why this technology integration would offer an advantage over existing projects if they were to be successful. The systems engineering was not present to drive component selection, cost management or market understanding.
- There isn't anything new in the project and there is virtually no R&D work, despite the starting point of the
 project severely underperforming relative to goals.
- The "AER" doesn't make much sense. It is very difficult to believe that carbon specie adsorption will exceed the current 95 % or so, and this isn't close to being sufficiently pure for a low temperature fuel cell (FC). They are stating that gas shift or pressure swing adsorber (PSA) is not needed and doesn't make sense. It is difficult to believe that ultra-high purity will be achieved without a PSA. Without shift, instead adsorbing carbon monoxide, which will always be in equilibrium with the carbon dioxide, it reduces the energy that can be extracted from the fuel.
- Project is in line with DOE 2011 targets:
 - Project objectives.
 - 2009 2010 objectives.
 - 40% electrical efficiency.
 - Build, test and validate high efficiency (AER) fuel processor bench-scale rig.

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Question 2: Approach to performing the research and development

This project was rated **2.2** on its approach.

- This project includes the phased development of an open architecture system with a pure hydrogen interface between the FC and the fuel processor.
- Assuming it is successful, there are advantages to this approach.
- Excess electricity is not going to the grid, but is being dissipated. This approach is not great for a high efficiency system.
- The approach appears sound. It is suggested that the design conform to EN62282-3-1 (the International Electrotechnical Commission (IEC) equivalent to ANSI FC1-2004).
- The approach appears to be a bit of a hodgepodge with some elements introduced to resolve issues (such as the AER).
- The project has selected steam methane reformer (SMR) + PSA + PEM for a heat and power application. This selection demonstrates a very poor understanding of the system application and integration.
- PSA is a poor choice for a low cost system, as it does not offer any advantages in such a system.—The presenter claimed that it would simplify the system and offer higher hydrogen utilization. However, a PSA unit is a very high cost component, especially at this scale, and even as established technology, it does not offer low cost or complexity. The presenter suggests that PSA would allow one to use all the product hydrogen for electricity production, while in reality, PSA units have a finite hydrogen recovery capability. High recovery ratio of ~85% may be practically recoverable with a PSA unit, which is similar to the innate recovery rate of a FC operating on reformate in the first place. The PSA thus does not provide any benefit in hydrogen recovery and overall electrical efficiency. The other point the presenter offered was that PSA units would block the carbon monoxide from getting to the stack. This would be true, but this function can be accomplished by significantly cheaper and simpler means; i.e., high temperature shift + selective partial oxidizer catalyst. Alternative solutions would have a small fraction of the cost of a PSA unit.
- The next question is the selection of PEM for a CHP system. Typically, hydronic systems operate in the 60°-80°C range. Therefore, coolant coming to the house water coming to the CHP unit is at 60°C, and the system has to heat up the coolant to 80°C before returning it to the house heating loop. When asked, the presenter offered that stack heat would be first applied to the coolant loop, followed by the higher quality heats from the reformate cooler and the system exhaust. However, in such a scenario, the heat management does not add up. Stack heat in a high efficiency system should account for a vast majority of the heat generated from the system and reformate heat and exhaust heat are a small fraction of that. However, the stack is operating typically in the 75°C to at most 85°C range. This means that transferring heat to a coolant loop may drop the heat quality to 65°-75°C, and then once more through a liquid heat exchanger to the house heating to 55°-65°C (house water cannot be run directly through the stack to gather heat since it has high impurities level and thus is electrically conductive and shorts the stack. Therefore, two temperature approaches have to be considered in transferring heat first to the stack to a coolant, and then from the coolant to the house water). The stack heat can at most heat the house water—best case from 60°C-70°C. With such a temperature rise consuming a majority of the heat available from the system, there is no longer sufficient heat available to accommodate the remainder of the temperature rise.
- Besides the system's useful heat rejection requirement, the system has another major issue. It requires that water be dropped before entering the PSA unit. PSA materials get damaged from moisture content. The presenter suggested that 40°C is required before the PSA unit. In a CHP application at this scale, such heat sink is not available unless they tap into a radiator heat sink or such. Commonly, however, this is not an option. At best, they may be able to sink heat into the water going to the house (which is intermittent). The system analysis is further double-tapping this resource, as they claim that they would use the heat for CHP applications!
- The presenters suggested that mass and energy balance modeling has been performed for this concept. However, they did not offer any of the analysis results and opted for a cartoon chart of arrows. This is very in appropriate for a technical review if adequate analysis was actually performed!
- There is virtually no development work in the project, so no real expectation that the system will perform better than the many others of this type that have been tested. The "AER" doesn't make sense from the start and appears to be the only development effort.
- A statement is made that the balance of plant (BOP) will use "fixed orifice devices," which are usually assumed to be eductors or something of that nature, of course it is difficult to tell, and is, for the review, a problem itself.

Typically, "fixed orifices" are a problem because of the narrow, if not "fixed", operating range. This usually causes problems for off-nominal operation, which will occur due to normal degradation, load changes, etc., and these types of systems typically require a blower or compressor for transient operation anyway, which increases the overall part count plus a motive system for the eductor. In almost all cases where an eductor is analyzed, it is rarely worth the negatives. If not an eductor then it's not clear what the reference to "fixed orifices" is, but that is the presenter's responsibility.

• Phased development of an open architecture system with a pure hydrogen interface between the FC and fuel processor is focused on overcoming the barriers of improved FC performance, increased FC lifetime, lower FC cost, smaller reformer, high fuel utilization, simplified integration, independent operation of the FC, and fuel processor plug and play."

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.5 based on accomplishments.

- The high efficiency promised by this project is lacking to date.
- The chemical efficiency of natural gas to hydrogen = (21574 + 2700)/35635 = 68.2%. The chemical efficiency of FC = (10,370 + 610 + 620)/21574 = 53.4%. Direct current power delivery efficiency = 10,370/(10,370 + 610 + 620) = 89.4%. End-to-end Efficiency = 32.6%. Combined CHP/electrical = 60.8%, which is not very close to any targets.
- The accomplishments to date are impressive.
- There has been some progress on life testing, but far less than is needed for practical application.
- The project has done some system integration, although in a misguided direction. This may be better suited to an education and technician training exercise than a systems development effort.
- It is claimed that a system was tested, but the data is inconsistent. The FC would not have survived more than hours at best on 95% pure hydrogen as indicated for the AES system. There are other data discrepancies. The polarization (IV) curve and area specific resistance (ASR) don't agree. Based on the IV curve, the ASR should be about 1.0 ohm-cm² versus 0.06 ohm-cm² or so.
- Lifetime, durability, and efficiency are all well below goals and, given the lack of development work, there is no expectation that this will improve. The data shown is typical for a low temperature FC system.
- Cost was not addressed other than saying that the unspecified cost would get lower with higher production numbers.
- A degradation rate is given for 7,000 hours but the data indicates that the stack (cells?, not sure what the system was) are failing catastrophically at that point, so a "degradation" rate isn't particularly meaningful with a 40,000 hour goal.
- Many accomplishments have been achieved, including detailed engineering completed June 2009, construction and shakedown tests completed October 2009, first power production achieved in November 2009, automated combustor startup and full system safety shutdown implemented in February 2010, system in continuous operation since March 2010 (350 hours) achieving 33% efficiency (end-to-end efficiency = 68.2% * 53.4% * 89.4% = 32.6%), and continuous hydrogen production at low temperatures with >95% purity achieved.
- However, the hydrogen generator efficiency is below expectation due to natural gas (rather than PSA off-gas) firing of the combustor.
- 4 of 8 metrics achieved during CHP test.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated 2.7 for technology transfer and collaboration.

- There is a list of partners, including the California Polytechnic University, the University of South Carolina, Sandia National Laboratories, Intelligent Energy, and IPHE partners Scottish and Southern Energy, Logan Energy, Element Energy, and Nedap.
- It is clear what Intelligent Energy is doing in the project, but it is unclear what the other partners have contributed to date.
- The collaboration partners are suitable. Leveraging the expertise of Logan Energy is important.
- There is a good mix of partners, though their roles are a bit unclear.

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- Collaborations were listed on the slides. If collaboration with these entities was effective, I would have expected much better results.
- Several collaborating institutions are mentioned, but the work plan doesn't indicate what, if anything, they are doing. There is no evidence of collaboration in the presentation. The building identified as the demonstration site in the UK that is under construction seems rather large to be built solely for a demonstration. No other details are given concerning the purpose of the building.
- Good technology transfer between a joint venture with Scottish and Southern Energy, system-installers subcontractor Logan Energy, site modeling and controls subcontractor Element Energy, and an inverter development in collaboration with Nedap.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.2 for proposed future work.

- System optimization (SMR+PSA+FC) and redesign to accommodate future plug-in AER fuel processor—this should be the priority, as the system does not currently have the efficiency required to be marketable. However, there was not a plan articulated to meet the DOE targets for efficiency.
- System optimization includes shorter PSA (so it fits through the door). Seems like a rather obvious development that is required.
- Demonstration of system with IPHE partners.
- The future work appears to be appropriate.
- The link between the future work and a quantitative estimate of potential progress to goals is unclear.
- It seems that a relatively standard PEM system that doesn't meet EERE goals will be operated in 2011. The AER, the only developmental aspect of the project, won't be operated and requires additional funding to complete and test, negating any actual advances associated with this project, assuming the AER is an advance. In 2012/13, all EERE goals will be achieved despite no actual R&D work indicated prior to that time and the current system well short of these goals.
- The hydrogen generator efficiency is below expectation due to natural gas (rather than PSA off-gas) firing of the combustor. Need 80% perhaps in future to develop a better AER. Intelligent Energy may need more funding.
- The six month field demonstration in UK is good.

Strengths and weaknesses

Strengths

- The strengths of this project appear to be the focus and the collaboration.
- The project has good focus on CHP at micro-scale.
- Overall, it is a good project.
- The project is interesting.

Weaknesses

- Overall system efficiency is not close to the targets.
- System is currently too big for intended applications.
- The potential weaknesses may include deodorizing (desulfurization) of the fuel gas and recovery of usable heat.
- There is an unclear link between tasks and gaps to current goals. Carbon monoxide content in AER exhaust is not specified and may require added clean up.
- See comments.
- It needs more funding to achieve 40% efficiency.

Specific recommendations and additions or deletions to the work scope

• System optimization (SMR+PSA+FC) and redesign to accommodate future plug-in AER fuel processor should be the priority, as the system does not currently have the efficiency required to be marketable.

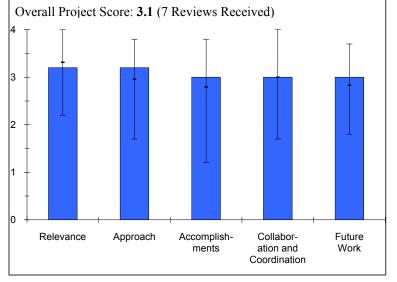
- This reviewer suggests a detailed review of the AER approach to ensure that it should stay in the program. Also, the project team needs to ensure that detailed benchmarking is conducted against the Japanese EneFarm activities.
- The group leading this project does not appear to be capable of systems development. This funding could be applied in other projects and provide actual technology development improvements.

Project # FC-32: Development of a Low Cost 3-10kW Tubular SOFC Power System *Norman Bessette; Acumentrics Corporation*

Brief Summary of Project

Acumentrics is developing SOFC systems for a variety of applications, including remote power, military and micro combined heat and power (micro-CHP). 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; 4) prototype test meeting system efficiency and stability goals; and 5) integrate to a micro-CHP platform.

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>



This project earned a score of **3.2** for its relevance to DOE objectives.

- This project is aligned well with DOE objectives for stationary power and CHP applications. The objectives are to: improve cell power and stability, to reduce cost for cell manufacturing, increase stack and system efficiency, and integrate to remote power and CHP platforms to allow short, medium, and longer term market penetrations.
- The project has good relevance to stationary technical targets.
- This system is definitely relevant to the DOE objectives, as it pursues high efficiency, durability, and economics of use for a potentially renewable fuel (biofuels). Currently, the system is focusing on feed stocks that are non-renewable, but is also leveraging niche and mass markets to develop the technology further.
- The work is consistent with a recent (Dec 2009) Request for Information and proposed goals for CHP and auxiliary power units (APUs).
- A solid oxide fuel cell (SOFC) is most useful as a fossil fuel power generator, although its advantages in reversible operation make it an excellent choice for energy storage. By using ultra-pure hydrogen as a fuel, the SOFC will, in general, not perform as well as a low temperature FC. However, an SOFC that meets goals, and an SOFC can certainly meet the EERE goals, is better than no FC at all and still should outperform reciprocating engines, and will certainly outperform a micro-turbine, particularly for distributed applications. A high performance state-of-the-art SOFC will also outperform the competition in large generation. In that sense, the relevance is good. The objectives of the project do address achieving the DOE goals.

Question 2: Approach to performing the research and development

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

- The project works to improve performance by improving the individual system components.
- The PI discussed cell manufacturing to improve processing yield and productivity, but these are not being improved by cell manufacturing in this project.
- The project could benefit from some better analytical methods, cost breakdown and/or efficiency estimates.
- This is one of the most complete system development companies I have seen. I was very impressed with the work done.
- To the extent that technical details were disclosed, the approach appears sound. There is appropriate focus on power density, stability and manufacturing process development.
- The project identified "barriers" well. Rather than barriers, this is a list of areas that could be improved, simultaneous improvement in all of them is not required to meet goals.
- The objectives are well stated and the focus is correct.

- The approach is very R&D focused and is focused on the right issues. This organization knows what needs improvement.
- The 1,500-hour durability test looked stable, but there may be a need to run a longer test. It has been demonstrated that there shouldn't be catastrophic failure in a well designed, manufactured, and operated SOFC (one of its advantages), so the short term tests are valid, but at some point, long-term performance needs to be verified. This stable long-term performance has been demonstrated by Siemens and Versa Power for 80,000 (tubular) and 27,000 hours planar—the planar was stable but stopped for analysis after showing stable performance and degradation around 1%/1,000 hours.
- The project's list of barriers include: cell power density, stack power density, cell cost reduction, system cost reduction, system efficiency, and lifetime.
- The approach is attacking barriers by improving cell power and stability, stack and system efficiency, the ability to operate on liquid fuels (funded through the Office of Naval Research (ONR) and the Department of Defense (DOD)), and integration to remote power, military, and micro-CHP platforms to allow short, medium, and longer term market penetrations.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.0 based on accomplishments.

- This project has shown a high degree of cell performance. However, this big percentage improvement is due to the cell performance in prior years being very poor. The polarization curves shown are still not state-of-art for SOFC performance at these temperatures (750°C and 800°C).
- The project states that the operating temperature has been reduced by 100°C and could be reduced by 200°C, which is an excellent achievement; however, this is not shown by the data presented. Other developers show 0.7 V at 1.0 A/cm² at 550°C, compared with the 0.6 V at 1.0 A/cm² and at 800°C in this project.
- The project exhibited good overall progress, but they need to recognize that the lifetime achieved is very far from where it needs to be.
- The project demonstrated improved system design, life, and efficiency, while having full consideration of system cost implications. This is all one could possibly ask for in driving to meet market needs.
- One percent per thousand hour degradation is a good status. Power density has improved; however, this needs to be presented in the context of operating cell voltage and fuel utilization, along with associated system efficiency. For example, what were the operating conditions associated with the demonstrated ~41% lower heating value (LHV) efficiency?
- The achievement of efficiency is easy at the expense of power density.
- The project showed cost targets based upon the CHP metrics, but the associated status of this technology was not. At the program level, it's by no means clear that residential FC CHP is a viable domestic market.
- Thermal/electricity demand profiles versus CHP system was a relevant accomplishment.
- The project demonstrated good BOP/integration progress.
- Good progress has been made in maintaining cell performance while reducing temperature. This is important to cell/stack life and provides operational flexibility. Power density has increased substantially, this is a major contributor to reducing cost.
- The project's present status reflects progress as follows:
 - Efficiency proven over 40% on stack in a demonstrated system in 2010/2011.
 - CHP efficiency of 85% proven on 1kWe wall hung systems.
 - Demonstrated start-up and load transients as part of ONR liquid fuels testing.
 - The latest generation systems operated with stacks over 5,000 hours and total system tests over 8,000 hours.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.0** for technology transfer and collaboration.

• Collaborators include EFESO, however, it is unclear if these collaborations are part of this project, as they appear to be a separate project. Other collaborators are listed, but there is no indication about what they are doing.

- The project demonstrated good leverage to other government programs.
- The company has worked with its customer base very closely to provide and supply products that are ultimately accepted by the market. The bottom line is sales.
- The project builds upon Solid State Energy Convergence Alliance (SECA) work, and Acumentrics leverages needed outside expertise appropriately.
- The project has demonstrated good collaboration through multiple government projects. Funds through ONR for fuel processing and small business innovation research (SBIR) for an important heat exchanger development are well coordinated with the work in this project. The activity in Italy is also impressive, although it is not part of this project. Technical collaborations do not appear to exist, although, given the funding level, it would be difficult to support them considering the financial requirements required for a commercial effort.
- The Italian government program granted to Ariston thermal group and 15 partners including Acumentrics is a key partnership.
- Acumentrics is the first foreign company to be issued an Italian government grant for a green energy program.
- Three year, \$1.1 million program culminating in a 1-kW_{electric} and 2.5-kW_{electric} CHP prototype.
- New ONR \$10 million project on logistics fuel.
- Acumentrics utilizes key technology contributors on inverters, balance of plant (BOP) components, testing laboratories/universities, and certifying bodies

Question 5: Approach to and relevance of proposed future research

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

- The project's future work appears straight forward, and is required for commercialization.
- Efficiency enhancement should be a key focus of future work, (as indicated, with cost as a constraint).
- Keep doing what you're doing!
- The project's future work is appropriate.
- There was no direct slide on future work. Clearly there will be future work, and the entire project is focused on the right tasks, but unfortunately this topic is not directly addressed.
- The project's future work, consistent with project objectives, should assure cell stability, correlate stability versus current density, demonstrate stability over thermal cycles, compare results of catalytic partial oxidation (CPOX) and steam reformed systems, resolve thermal issues in stack due to higher power density, test improved thermal management techniques, continue cost reductions on each product platform, continue cell manufacturing automation, and continue "make/buy" decisions on generator and BOP components.

Strengths and weaknesses

Strengths

- Acumentrics shows the ability to manufacture cells and SOFC stacks with developing technology.
- There is good focus on micro-CHP for SOFC.
- The functional demo systems are a strength of the project.
- The project is fully focused on accomplishing fuel cell improvements.

Weaknesses

- There is less focus on analytical methods (simple semi-empirical analytics are fine) than would otherwise be optimal.
- By nature of the open review, reviewers don't have access to material details. For example, what material changes permitted the operating temperature reduction?
- The project could put effort into assessing potential stack lifetime-limiting issues, given the lack of viable accelerated testing protocols. Engineering analysis and microanalysis can pinpoint areas of concern (e.g., materials volatility).
- It's not clear if the project will achieve a high enough power density to get to cost targets.

Specific recommendations and additions or deletions to the work scope

• The reliability of the multi-cell tube bundles should be assessed.

Project # FC-33: New Polyelectrolyte Materials for High Temperature Fuel Cells

John Kerr; Lawrence Berkeley National Laboratory

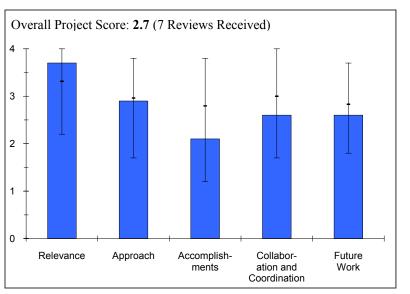
Brief Summary of Project

The project objectives are to: 1) investigate the feasibility of polyelectrolyte proton conductors that do not require additional water to achieve practical conductivities; 2) determine stability of these materials to oxidation and strong acids; and 3) fabricate and test membrane electrode assemblies (MEAs) to determine gas crossover.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.7** for its relevance to DOE objectives.

• The project is relevant to the requirements for high temperature



membrane development and it meets overall DOE objectives. The goals and objectives of the project are designed satisfactorily. The multi-year plan is in line with DOE RD&D objectives. The study of the model compounds and modeling work pursued by the team is aligned with the development of proton transport knowledge in the low relative humidity (RH) medium. The team should consider *in situ* testing of the material properties at low RH.

- Development of a membrane that is highly conductive and stable at 120°C would enable significant fuel cell system simplification and cost savings.
- The project is geared towards developing membranes and MEAs for hotter and drier operation of fuel cells.
- This certainly is a very relevant project and will help the design of simpler fuel cell systems for practical purposes. Being able to operate at a higher temperature with no or low humidification, while maintaining high performance (requires higher membrane conductivity) and efficiency, are key requirements. The project is aligned with DOE goals and targets. Recent change to "feasibility of polyelectrolyte proton conductors that do not require additional water" from "conductors that do not require water" aligns it more with practical systems.
- Improved membranes with greater operating range, higher performance and better durability are critical to the Fuel Cell Technologies Program. Membranes for efficient fuel cell operation without external humidification at operating temperatures between -40°C and 120°C will greatly benefit both automotive and stationary fuel cells.
- Although nearly all membrane projects have conceded the need for humidification to conduct protons, this project is one of two that in theory could still fabricate ionomers that do not require water. In that respect, the project is particularly relevant to the stated desire to simplify the balance-of-plant (BOP) system.
- The targets mentioned for the project, such as conductivity, gas crossover, and durability, are aligned well with the objectives of the DOE program.

Question 2: Approach to performing the research and development

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

• The project has been designed appropriately to understand the proton transport barriers under low RH. The approach of the project is good and its outcome is expected to address some of the key technical barriers. The technical feasibility of the synthetic route to an oxidatively stable ionometric polymer is doubtful. The oxidative stability of the new polymer should have been considered. The grafted imidazole polymer possesses benzylic hydrogen, which may affect the durability of the membrane. The team is studying polystyrene-based model compounds to understand the proton transport mechanism, but the instability of polystyrene under low RH fuel

cell conditions may not give the team an opportunity to test such materials in real-life fuel cell conditions. From slide 5, it is not clear whether 3M perfluorosulfonic acid (PFSA), which degrades similar to heterocyclic polymer-containing materials, contains peroxide inhibitor or not.

- It is still unclear if this approach is feasible. The base ionic liquids (IL) do not meet the DOE conductivity targets, so why would one expect a less mobile tethered imidazole to be any better. I do not see how the modeling work is helping to make a membrane that meets the DOE targets. The electrode work distracts from required membrane focus.
- There are good strategies to make new functionalized polymers via either modification or new monomers. There is also nice complimentary work going on in electrode development. It all seems a little disorganized though, as there is no clear path to achieving conductivity of 0.1 S/cm.
- It is a difficult project. But the PI was successful in narrowing down the choices for the backbone to polystyrene, polysulfone and PFSA. Now, these materials perhaps can be attached with imidazoles, blended with acid materials and then fabricated as membranes and MEAs. If these membranes achieve conductivity goals, even with some humidification (<25%), it would be a great accomplishment!
- Good idea and sound experimental approach. Very nebulous Go/No-go decision criteria "How close to 0.1 S/cm at 0 25% RH and the full range of temperature (-40-120°C) is possible without free solvents?" It is unclear what would constitute a No-go when judging the criteria of this approach. There needs to be more specific Go/No-go criteria.
- Discontinue electrode work and focus on membrane work.
- Goal of achieving 0.1 S/cm at 120°C and 25% RH is appropriate. Three backbones, liquid ionomers and blends cover a large area of investigation, commensurate with the funding level.
- The approach of tethering strong acid / weak base groups to a polymer backbone is appropriate. Given high conductivities observed for ionic liquids, a study of whether tethered analogues of these species could provide for a conductive membrane was needed.
- The project approach has addressed the need to understand the conductivity limits of the materials.
- In later stages, the focus upon conductivity limits has driven the project towards understanding the connectivity of conductive routes and has involved a modeling effort. These directions are more appropriate than continued fuel cell testing and electrode fabrication.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.1 based on accomplishments.

- Good progress has been made by the team towards the understanding of proton transport mechanisms in fully humidified and dry membrane conditions. The synthesis of various block co-polymer-based morphologies is commendable. The ionic liquid model work in slide 10 is very interesting. However, in slide 12 it would be nice to compare the conductivity of polymer blends and free solvents with same host PFSA polymer. Comparison of 1100 equivalent weight (EW) Nafion® PFSA (effective EW 2000) with 800 EW 3M PFSA with imidazole alone does not show the true difference between the effects of polymer blend and free solvents.
- The one encouraging result is the improved conductivity with λ of BPSH blended with PAES-COOH-Im, but the conductivities are still below targets and measurements were not done as a function of RH. The other materials tested showed poor conductivities. The other measurements (NMR, TGA, small-angle neutron scattering (SANS), DMA) have not proved helpful to understand conductivity results and trends.
- There were new polymer systems investigated, but conductivity was too low. Small-angle x-ray scattering/ SANS work and modeling is on-going, and the PIs have developed an impressive new electrode system.
- A lot of good work was done for this project. But the fact remains, we are still a long ways away and perhaps have identified the bare essentials to achieve membrane conductivity: the presence of mobile solvents, the high degree of connectivity through membrane (difficult to achieve and maintain), or the presence of water.
- From a system integrator point of view, water management for an automotive power plant with a minimum operating temperature of -40°C is almost a No-go situation. Even though it is suggested that <25% RH may improve conductivity significantly, I remain concerned due to the practical implications of this approach.
- A good amount of work has been done, but the results do not look promising. Conductivities have been low and well short of targets.
- There has been significant activity with some progress towards the program goals.
- Progress needs to be accelerated for the project to meet all program goals within the remaining time.

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- The progress has been slow in getting this project to explore fundamental limits of conductivity and then tethering ionic liquid-derived groups to less exotic backbones before progressing to more exotic chemistries. The 3M 850 mix with imidazole was a step in the right direction, but a tethered version has not yet been reported.
- At the moment, polysulfones appear to be the preferred class of backbones for the polymer (given low conductivity robustness with PSS-PMB), but the rationale is not entirely clear. The project still appears to be searching for a tethered sample that achieves reasonable conductivity, and has been characterized for things such as swelling, crossover, durability, etc.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **2.6** for technology transfer and collaboration.

- The team has very good collaborations with many different research groups with solid understandings of the field. The team also has access to good polymer characterization analytical tools to obtain relevant information in support of the research work.
- Electrode work at LANL is not well aligned to the rest of the project. It's not clear what 3M is doing. Most work seems to be done at LBNL.
- The collaborations with university, National Laboratory and MEA manufacturer need more input from the automotive industry and require input from the catalyst manufacturer.
- Appears to be good collaboration with LBNL and LANL, with topics ranging from various characterizations to polymer synthesis.
- Collaborations within the project are good.
- There is good interaction between partners and great access to analytical facilities.
- The collaboration appears to have improved over the past year, but could be improved more. Modeling calculations with Utah have been a step in the right direction, although this is still a collaboration within the primary contractor organization (LBNL).
- Collaboration could have been improved with 3M to tether imidazole or benzimidazole groups to PFSA materials. This would be a good baseline to serve as a link between the PFSA/IL mixes and the tethered materials from hydrocarbon backbones.
- SANS at Oak Ridge National Laboratory appears to be well used for morphology studies.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.6 for proposed future work.

- All the proposed work is relevant to the overall progress. However, there is no proposal to carry out high temperature, low RH fuel cell testing of developed materials to demonstrate materials property under DOE targets (0.1 S/cm at operating temperatures of ≤120°C). The team should consider testing the materials under DOE conditions to show the success of the project.
- The fundamental question of whether the approach is feasible to enable high temperature operation is not being directly addressed. I would recommend focusing on imidazoles attached to PFSA (work with 3M on this) and measure conductivity vs. RH at various temperatures. Fenton's testing is not recommended. The PIs should test hydrolytic stability in boiling water first. I am unclear on the value of the MD calculations.
- It was good to see that the team will pursue the prescribed conductivity goals and it was good to see that more work will be done on the new electrode structures. I am not convinced that the IL work should be continued.
- Proposed future work should help achieve these modified objectives.
- I am not sure what benefit working to tether imidazoles to available materials polysulfones, polystyrenes, PFSA and perfluorocarboxylic acid will provide when mixtures with untethered imidazoles did not reach the appropriate conductivity. The untethered materials should provide optimum acid-base interactions, while the tethered materials may not.
- The proposed future work strikes a balance between selecting the most promising approaches and advancing to cell and durability testing.

- The future work has improved in the past year. Electrode studies have been given a much lower priority in favor of looking at conductivity / morphology relationships, which is entirely appropriate for a project that has been struggling to find good conductivity with a tethered, IL-derived material.
- While a fundamental approach may be good for understanding conductivity, it may be preferred to simply use the remaining time to address durability with prescribed accelerated stress tests, as opposed to Fenton chemistry studies and model compound studies.

Strengths and weaknesses

Strengths

- The team has researchers with a thorough understanding of the challenges in meeting DOE low RH objectives and who are capable of providing some meaningful insight into the problem. The team has sound coordination between different research groups. The team has access to good analytical and modeling facilities, which is necessary to carry out such a complex research problem.
- If goals are met, it would be extremely valuable for fuel cell development.
- The PIs displayed excellent electrode work.
- The PI's ability to explore limits of various materials to achieve high conductivity was a strength. The research work was thorough and methodical.
- The basic science work done in the project is good and provides good data to the community to help understand proton conduction.
- The PI investigates a wide range of novel polymer approaches.
- The investigation of the proton conductivity mechanisms was a highlight.
- The initial approach of using imidazole-based ionic liquid-derived materials is one of the strengths of this project. The project does not overlap directly with any of the other projects in the portfolio and it occupies research space that is very much of interest.
- The project has shown willingness to address fundamental concerns of how conductivity is influenced by morphology and the repulsion of water.
- The addition of modeling should enable the project to at least report some addition to the community's knowledge of these materials, even if a finished material is not delivered.

Weaknesses

- This project lacks any plan of testing the materials under realistic fuel cell conditions to demonstrate the progress against DOE targets. The proposed plan of testing MEAs at 30°C to determine whether the membrane has practical conductivity of 0.1 S/cm falls severely short of the DOE target of 120°C. The team should test the materials under DOE's proposed conditions to determine their success.
- The fundamental hypothesis of whether the approach is feasible to enable high temperature operation has still not been proven, after 3.5 years. Poor collaborations were used. Electrode studies and
- mechanical/morphological testing have not provided significant guidance to improve materials.
- Ionic liquids were used.
- The chosen platform or technology may not have the highest probability to achieve the ultimate goal of a high temperature PEM that requires no humidification.
- The go/no-go criteria are nebulous, which allows work to continue on materials with poor conductivity. They plan to proceed with MEA and stability testing prior to achieving needed conductivity.
- High membrane conductivities and MEA performance have yet to be demonstrated.
- The overall experimental progress of the project is slow. Despite attempts, the project has not really been able to establish a decent baseline for a proton-conducting, ionic liquid-derived, tethered ionomer.
- Some suggestions that have come to the project from outside sources; e.g., the FreedomCAR (Cooperative Automotive Research) and Fuel Partnership's Fuel Cell Technical Team (FCTT)'s recommendation to try tethering to a PFSA, have only recently been implemented and there is not enough progress to report.
- The slow progress has prevented the project from being able to deliver classes of data for which other projects are fairly judged including data for swelling, durability, gas crossover, etc.

- A solid Go/No-go gate should be used before any MEA development work is done. The membrane needs to meet some hard conductivity target (I suggest they use the interim target the other set of membrane projects needed to pass, 0.1 S/cm or greater at 120°C and 50%RH) before any MEA development work is done with it, and I also would suggest before any oxidative stability testing is done.
- Produce a conductivity curve over the entire RH range this would help in comparing to other materials.
- Focus on imidazoles attached to perfluorosulfonic acid and measure conductivity vs. RH at various temperatures. Eliminate electrode work. Eliminate Fenton's testing and instead test for stability in liquid water. Focus modeling and other characterization work on a systematic series of materials where single variables are controlled independently, so the synthesis guidelines can be developed to improve membrane conductivity.
- Ionic liquids should be included.
- If the project had not added a modeling component to study the limits of conductivity, one would be suggested here. Some focus should remain on how modeling-suggested morphologies could be experimentally realized.
- There should be better rationale provided for why particular backbones or block copolymers are being used. It is too late in the project for approaches that have not been shown to add some morphological / conductivity advantage.
- It would be interesting to see if the water repulsion could be used to a greater advantage. It could possibly lower EW materials.

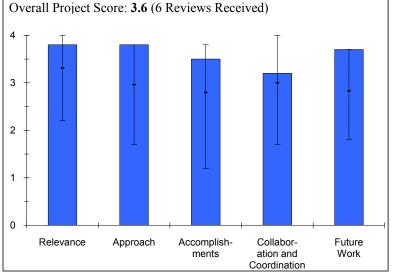
Project # FC-34: Membranes and MEAs for Dry, Hot Operating Conditions

Steven Hamrock; 3M

Brief Summary of Project

The overall project objective is to develop a new proton exchange membrane (PEM) with higher proton conductivity and improved durability under hotter and dryer conditions compared to current membranes. The approach includes 1) testing of new polymers, new membrane additives, proton transport studies, and new membrane fabrication methods, and 2) performance and durability testing.

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>



This project earned a score of **3.8** for its relevance to DOE objectives.

- Development of robust, high temperature, low relative humidity (RH) membranes is vital for the successful commercialization of fuel cells (FCs), especially for vehicle applications as high temperature and low RH allows for a simplification (cheaper) balance of plant (BOP).
- Membrane performance and durability directly affect DOE's objectives and technical targets.
- Polymer membranes with greater operating range, higher performance, improved durability, and lower cost are critical to the FC Program.
- The project is highly relevant to the DOE objectives. The project tasks are a nice mix of theory, polymer synthesis, and membrane characterization work.
- The project is highly relevant to DOE. Development of membranes with high conductivity under hot and dry conditions is directly applicable to DOE membrane targets. Furthermore, through addressing mechanical and chemical stability, the project retains relevance higher than that of several other membrane projects by increasing the probability that new membrane materials developed will be suitable for FC applications.

Question 2: Approach to performing the research and development

This project was rated **3.8** on its approach.

- Slide 13 of Hamrock's presentation clearly shows all of the strategies one could implement in order to improve the mechanical robustness of extremely low equivalent weight (EW), short side chain perfluorosulfonic acid (PFSA) materials.
- The introduction of multi-acidic side chains is a very interesting strategy and the ongoing work on heteropoly acids (HPAs) from Colorado School of Mines (CSM) continues to show promise.
- The approach is systematic, based on sound chemistry and parametric consideration of important variables.
- Appropriate *ex situ* analyses are used.
- Experimental data are used to establish necessary changes to membrane electrode assembly (MEA) materials and structures.
- Focusing on materials that are scalable is a good approach.
- The use of additives is questionable for long term operation.
- The approach covers a wide range of candidate materials, experimental and theoretical studies of the proton transport mechanisms, as well as membrane fabrication and evaluation in conductivity cells and FCs.
- The 3M approach of examining polymer blending, crosslinking, reinforcement, and polymer modifications is comprehensive and, in some respects, innovative. For the most part, the members of the research team have

complementary talents that mesh together nicely. 3M seems to have done a good job in overseeing the various project tasks.

- The competitive kinetics and polymer degradation work do not seem to fit with the more important focus of finding new FC membranes with improved performance at high temperatures and low RH.
- The project is well focused on attacking the key technical barrier of achieving high conductivity under hot and dry conditions. The PI has selected several promising routes toward achieving this goal, and has conducted R&D activities in a well-planned, pragmatic manner. Some of the routes investigated; i.e., inorganic HPAs, seem less promising, and should be lower priority, but given that the project continues to make good progress overall, this is not seen as a significant demerit.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.5** based on accomplishments.

- Synthetic progress on perfluoroimide acid (PFIA) materials is commendable and the results thus far are promising. Based on the EW values of the PFIA materials, conductivity is as expected, but FC polarization curves are lower—MEA optimization is still needed.
- HPA conductivity values are off of the chart, and it will be interesting to see if this technology will translate to a water insoluble, mechanically robust FC MEA.
- 3M has demonstrated a stable MEA for 18,000 hours under voltage and humidity cycling conditions.
- Alternative electrode structures and materials have been successfully matched with new membrane materials.
- It is good to see that the conductivity of the membranes has been conducted using the BekkTech apparatus that allows a standard method and setup to evaluate whether a material has reached DOE goals. The positive results in these tests as a function of EW show the significant improvements made in the new materials compared to Nafion®. (The effect of EW on conductivity though is an obvious result well known in the literature, as is the loss of crystallinity with the lowering of EW.)
- Conductivity exceeding the program target was shown with novel perfluoroimide multi-acid side-chain (MASC) polymers.
- Based on the FC durability test data, combining higher conductivity with high durability appears promising.
- There have been a number of significant accomplishments by the research team. The MASC polymers are impressive, as are the results from the PFIA work. The experimental work (whether it involves polymer synthesis, membrane characterization, etc.) appears to have been carried out carefully with attention to detail.
- The progress of this project continues to be very good. The development of the PFIA polymer is a promising step toward developing a membrane that meets DOE resistance targets. The materials appear to currently meet DOE high temperature, areal resistance targets at the high end of the humidity range (120°C, 80 kPa water), but it doesn't appear to be there yet in terms of meeting the target at the drier end of the humidity range. Given that 3M is still planning to investigate lower EW materials of this family, and given that some approaches; i.e., crosslinking, are still available to allow development of stable materials at even lower EW, 3M appears to be making good progress and has a high likelihood of soon meeting all resistance targets.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.2** for technology transfer and collaboration.

- Collaborations are clearly stated and shown. The program with CSM has been ongoing for quite some time; however, I wonder how integrated the programs of the other collaborators are or if are they more of an afterthought.
- The team includes an automobile original equipment manufacturer (OEM) and several universities.
- Some integration of the collaborators' work is needed to achieve the project goals.
- Some of the collaborative work (the competitive kinetics and polymer degradation work) is not well coordinated with the main thrust of this research project, that being the identification of new membranes that operate at high temperature and low RH.
- Given the large suite of capabilities present at 3M, this project requires a lesser degree of collaboration compared to some of the other membrane projects. Still, it is not clear that 3M is getting much useful help from subcontractors, and I have to wonder if other collaborations could be more fruitful. Collaboration with Case

Western Reserve University (CWRU) and the University of Delaware at Mercy has produced some interesting results, though the degree to which these have guided or influenced polymer development at 3M is unclear. The collaboration with GM is good. The collaboration with CSM is again interesting, but unclear that investigation of this pathway (inorganic HPAs) is necessary or appropriate given how strong progress is toward achievement of targets with the lower risk approaches being pursued at 3M with more conventional organic polymers.

<u>Question 5: Approach to and relevance of proposed future research</u>

This project was rated **3.7** for proposed future work.

- Future work encompasses the further optimization of PFIA and its subsequent incorporation into a MEA for FC testing. At this time, there is not more that can be inputted.
- The project ends in less than a year.
- Continued chemical and mechanical evaluation or characterization and enhancements of MEA materials will continue. More information on how to stabilize low EW materials is needed.
- The downselected membrane(s) will be durability tested under cycling conditions.
- It is not clear from slide 32 what vehicle, cell or stack, will be used in the final testing.
- The results so far are good, but the weaknesses are obvious in that low EW results in betterconductivity but poorer durability. The next steps of "blends, cross-linking, reinforcement, and polymer modifications" are critical and will determine the success of the project.
- A logical plan to continue and complete the tasks is in progress.
- The research team has properly downselected the MASC and PFIA polymers. Future work is correctly focused on both membranes and MEAs.
- The future work presents a reasonable and appropriate pathway to further advancement toward DOE targets and incorporation of novel 3M materials in MEAs.

Strengths and weaknesses

Strengths

- There is very strong project management with the ability of scale up and commercialization. The team has a very strong grasp of polymer synthesis, casting and scale-up. The team is excellent.
- They have a good approach, scalable materials, and good benchmarking tests for conductivity.
- A number of approaches to improving performance and durability have been successfully combined (MASC and additives).
- Building on proven PFSA chemistry has allowed improved conductivity and durability while taking advantage of PFSA's proven track record and manufacturability.
- The team has accomplished impressive research, especially in regards to the MASC and PFIA work.
- A wide range of careful and precise experiments have been carried out.
- The new polymer synthesis work is well thought out.
- 3M has lots of good ideas for attacking membrane resistance targets, using athorough and well-structured approach to investigating the various ideas. The ability of the participants to vigorously attack the conductivity issue while still keeping an eye on mechanical and chemical stability is an important strength.

Weaknesses

- Collaborations with other institutions besides CSM do not seem as strong or vibrant.
- There needs to be some consideration or discussion of potential cost.
- The results so far are good, but the weaknesses are obvious in that low EW results in better conductivity but poorer durability. The next steps of "blends, cross-linking, reinforcement, and polymer modifications" are critical and will determine the success of the project.
- The cost has not yet been discussed.
- It is not clear why the polymer degradation work is required and how the results from this subproject will impact on the MASC and PFIA work. The same can be said for the competitive kinetics work.
- It is not clear why it is necessary to add HPAs to a membrane. The chemical stabilization effect of inorganic oxide/HPA seems to be important. I am not convinced that the additives are needed for enhancing proton

conductivity. With a 50% loss in the additive upon membrane soaking, I am not sure if significant manpower should be devoted to finding a way to improve additive retention.

• Collaborations don't seem to be contributing all that much. Although the number of ideas generated by the project is seen as a plus, narrowing down these ideas and greater degree of downselection of the most promising ones could serve to accelerate progress.

- The project is going as planned with good progress. It is difficult for this reviewer to add to a very well thought out program which is achieving its goals.
- De-emphasize the model compound polymer degradation work and the competitive kinetics work.
- The inorganic HPA work is less significant and less promising, in terms of potential for application in real FC materials, than the MASC/PFIA approaches.

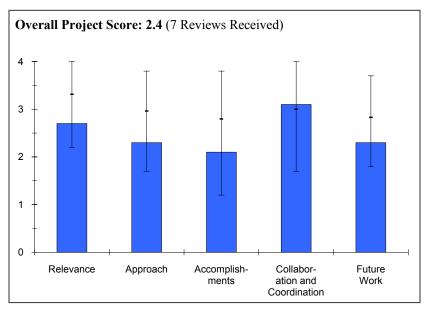
Project # FC-35: Lead Research and Development Activity for DOE's High Temperature, Low Relative Humidity Membrane Program

James Fenton; University of Central Florida

Brief Summary of Project

Project objectives are to: 1) fabricate membrane electrode assemblies (MEAs) from team membranes; 2) test team MEAs for fuel cell performance; 3) standardize methodologies for in-plane and throughplane membrane conductivity measurements; 4) provide High Temperature Membrane Working Group (HTMWG) members with standardized tests and methodologies, and 5) organize HTMWG bi-annual meetings.

<u>**Ouestion 1: Relevance to overall DOE**</u> <u>objectives</u>



This project earned a score of **2.7** for its relevance to DOE objectives.

- The relevance of this project is clear on principle only for a limited number of DOE research, development and demonstration (RD&D) program objectives. It strives to provide a standardized MEA fabrication and testing service for membrane fabricators as part of the HTMWG, with primary output being membrane properties. Currently automotive fuel cell original equipment manufacturers are shying away from needing 120°C stable membranes, so the original tasking of the HTMWG may not be so relevant any longer.
- Coordination of DOE funded membrane development efforts enables common platforms and methods for materials evaluation and comparison. Focusing on MEA development and performance testing does not provide much value.
- This project is a relevant activity to commercialize fuel cell technology for real world applications.
- The project is aligned with DOE goals and targets. Independent testing of high temperature membranes provides needed data to DOE for evaluation of other membrane projects.
- The development of standardized testing methods for conductivity and fabrication and testing of partner MEAs are needed to compare the different membrane technologies.
- This project is of marginal relevance to the Hydrogen Program. Development of membranes capable of operation under hot and dry conditions is highly relevant, but at this stage, further work on membrane development should be the priority. Optimizing MEAs based on recently developed membranes, most of which still require significant additional R&D effort, is of lesser significance at this stage than R&D on the membranes themselves. The fact that the project did not report MEA testing results, but only sent them back to the teams (who may or may not choose to share them), further diminishes the relevance of this project.
- The DOE membrane projects need to measure conductivity, *in situ* performance, and durability of their samples to understand whether or not progress is being made. This project is assigned to do just that. In this sense, this project is perfectly aligned with DOE objectives.
- The relevance of this project is also helped by the need for consistently measured data between projects. Conductivity data have been shown in the past to be sensitive to the exact protocol and equipment being used. This project helps to make measurements common among developers.

Question 2: Approach to performing the research and development

This project was rated 2.3 on its approach.

- The approach may succeed in providing data that could contribute to overcoming a DOE barrier, but there are significant weaknesses that will challenge the concept. The electrode characteristics, which this project is responsible for making, will have some influence on the resulting membrane performance properties. This is difficult to avoid in any approach, unless all the MEA components are optimized for one another. So, it is entirely possible that their specific electrode and method of application may bias one membrane characteristic over another and, therefore, not allow a level playing field when comparing different membrane suppliers. The electrodes have a strong impact on apparent membrane performance and durability, so conclusions from their approach will be limited to their specific catalyst and electrode fabrication methodology.
- There is too much focus on optimizing MEAs for each project. The focus should be on measuring membrane targets as defined in DOE target table: area specific resistance, chemical and mechanical durability, crossover and shorting resistance. The Pt vs Pt/Co electrode study is not relevant for the membrane project and it has been done already. Chemical degradation studies were not done using DOE recommended accelerated stress tests (ASTs)—alternating current voltage, 90°C, and 50% relative humidity (RH). The study of the type and amount of ionomer in electrode is also not critical for the membrane program. The reason for adding carbons in electrodes is unclear.
- The scope of the project appeared unnecessarily broad and risky. Assuming the primary objective is to evaluate various membranes in some standard, common MEA design—the approach should be to adopt a low risk, repeatable path to make MEAs. The winning membranes will require specific optimization before commercialization, which will be done by an MEA manufacturer who will optimize around all operational, durability, and cost constraints anyway. An attempt was made to improve MEA performance using a Pt-Co catalyst. There are known issues with this type of a catalyst, which can ultimately cloud the primary objective of evaluating a new membrane performance in an MEA.
- Independent testing and measurements of conductivity as a function of RH (both in plane and thru plane), and fabrication of MEAs using new membranes, provides needed data to DOE on the status of membrane development work.
- The policy to release data only to membrane providers and not show it at this review makes it difficult to determine how much work they've done. Showing the results without attributing them to anyone could be better.
- The flow chart for MEA testing is reasonable and test results showing optimization for 3M membrane vs. Nafion® suggest they are on the right track. The point they showed for optimization of ionomer loading (400 mA/cm² at 80°C, and 100% RH) is probably not the point of interest for these materials. The membrane projects are designed for high temperatures and low RH; and they should probably compare ionomer preparations at conditions of high temperatures and low RH, and probably high current density if you are going to pick one point but they really should make comparisons at a variety of conditions.
- The MEA testing is to be done at several conditions.
- The program structure is fairly well defined, feasible, and aligned with the technical barriers.
- Some areas, such as examining fluoride emission rate and the effect of Pt-Co vs. Pt, are outside the scope of the project. These efforts are of little value and distract from what should be the immediate focus: developing optimized MEAs based on membranes supplied by the other teams.
- The project has been able to deliver a common source for membrane conductivity results to the different teams that are funded by DOE.
- The project has focused on optimizing ionomer loadings in electrodes, as well other material and processing parameters that would improve MEA performance. However, the objective here is to obtain membrane information, which can be done with or without optimized electrodes. Gas crossover and high frequency resistance (HFR) measurements can provide valuable information, as well as the time or number of cycles until high gas crossover in accelerated stress tests.
- The use of PtCo is not necessary since it introduces additional failure modes. Furthermore, it may help to mask the failure modes that need to be observed from the membranes, as evidenced by the lower fluorine emission rate with PtCo.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.1 based on accomplishments.

- For one year of work, the number of catalyst coated membranes (CCMs) fabricated and tested is quite small at 29, representing about two per month. This is not a high enough testing rate for the number of different membranes to obtain statistically significant results or even begin to explore the number of independent MEA component parameters and combinations that can significantly affect their membrane performance measurements.
- It is not clear from the presentation what progress was made towards any of the DOE membrane property targets.
- After four years of activity, it would be good to see the overall rate of progress of the project towards the DOE objectives and barriers.
- The University of Central Florida (UCF) has made a lot of MEAs, but the materials and preparation methods vary significantly for the various projects. It would be useful to see a comparison of all the HTMWG team's membranes compared in a common MEA design.
- There is still no through-plane resistance data or defined procedures. ASTs for chemical and mechanical durability are not done.
- Almost every aspect of MEA fabrication was explored.
- It is difficult to determine how much work they've done in MEA testing since data for membrane providers isn't shared.
- The work on the specific area resistance measurement protocol doesn't appear to have progressed. It was not made clear why work on the Pt/Co and 3M ionomers will help the performance of the collaborators membranes, some of which contain novel hydrocarbon-based materials.
- It's hard to evaluate the accomplishments of this project when the PI diverts all MEA testing results back to the other teams. Of course, the data should be shared with the teams first, but the point of this project is (or should be) to optimize MEAs and compare them. In addition to the other work presented, the presentation should have a slide showing results from MEA testing with FuelCell Energy materials, a slide with results from Case Western Reserve University (CWRU) materials, etc., and then a table comparing results from each project.
- Results from the comparison of Pt vs. Pt/Co, and comparison of different types of electrodes on commercially available membranes, are not relevant to the barriers at hand, and, for the purpose of this review, are not considered as progress.
- Although results were not provided in the presentation, the presentation indicates that at least some MEA testing using membranes supplied by the teams has been performed, indicating that some progress may have occurred.
- Given the project approach, the project did manage to demonstrate improved iR-free performance with 7 psig backpressures, NR 211, and < 0.5 mg_{Pt}/cm² Pt/C. Although the project should be directed to focus on membrane characteristics, it managed to deliver some results for what it was trying to do.
- The progress on membrane through-plane conductivity was alluded to in some slides, but was not explicitly reported.
- The full cycle of testing has not yet been completed for any given collaborator.
- Attention has been paid to optimizing electrodes to suit different membranes, but apples-to-apples comparisons with common electrodes are valuable as well.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.1** for technology transfer and collaboration.

- The nature of this project relies on collaboration with membrane suppliers and they have a good number. It is not clear to this reviewer, however, why, after four years, they are still in the "exchange e-mail state" with some suppliers, or preliminary discussions of what will be tested.
- UCF is working well with all the membrane developers to evaluate their membranes. UCF is also working with Bekktech and Scribner on conductivity measurements, although no results from Scribenr have been presented.
- There appears to be good collaboration among all partners and stakeholders. For apples-to apples-comparisons, all partners should provide membranes, which then should be made into MEAs using the same electrodes and fabrication processes. This reviewer is not sure why Arizona State University will provide MEA in hardware.
- There is good collaboration with groups providing membranes for testing. There is good collaboration with 3M.

- This is a good group of partners and project collaborators. Degree of coordination with most of the collaborators appears adequate.
- The collaboration is reasonable. Clearly, the main collaboration here is that between the Florida Solar Energy Center (FSEC) and the membrane development teams. The fact that negotiations on testing procedures are still ongoing with some teams suggests that the degree of cooperation could be improved.
- The project relies considerably upon collaborative interactions, but the progress reported reveals that these interactions have been difficult. Nearly all the progress reported this year focuses on electrode optimization carried out at UCF-FSEC, and not on testing of collaborator materials. The slides reveal that collaborators appear unwilling to share samples, or that they wish to change electrode parameters before doing so, which defeats the purpose of obtaining consistent *in situ* results.
- Some collaboration was evidenced with the use of a 3M ionomer for electrode optimization.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.3 for proposed future work.

- The proposed future work appears to be more of the same modus operandi.
- No specific plan was presented of how the results of the testing will feed back to the membrane suppliers to make further improvements, followed by subsequent further characterization to make progress towards overcoming the barriers.
- Progress may also be constrained by the lack of clear guidance on the area specific resistance measurements.
- There was too much focus on MEA optimization. This is a membrane project. Pick a common, stable, MEA platform to use for all membranes and do the testing. Focus on through-plane resistance and chemical and mechanical durability.
- The proposed future work should include endurance testing. Other proposed tasks appear consistent with the goal of the project.
- MEA testing is to be done at several conditions, including high temperature (120°C) and low RH.
- Area-specific resistance (ASR) should be measured for all contributors' membranes.
- The project is supposed to end in ~one year.
- Recommend that the collaboration with 3M on the electrode development and characterization should be deemphasized. Focus should be to develop standardized test methods and to work with the university to test their membranes using standard, rather than state-of-the-art (SOA), materials.
- The proposed future work on membrane characterization and MEA development and testing is good. The discussion of through-plane vs. in-plane characterization is irrelevant. Through-plane is the only thing that matters for fuel cells.
- Continued electrode optimization does not appear to lend itself to the membrane comparisons that this project should deliver.
- Identifying ASR as a "controversy" reveals that the project intends to argue with revised DOE targets. The use of ASR is intended to encapsulate both proton conductivity (a material property) and the ability of the individual projects to fabricate thinner membranes. If either conductivity is low or if the membrane cannot be made thin, then ASR will be high, which is a much more relevant result for research customers than just proton conductivity alone.

Strengths and weaknesses

Strengths

- This has the potential to compare multiple membrane types within one electrode platform and testing protocol. It can provide testing capabilities to some suppliers that may be new to the area without access to their own testing.
- Strong collaboration with most membrane projects.
- Building consensus among all partners.
- Collaborations with membrane providers are strong.
- There is a good group of partners and collaborators on this project.

- A number of exciting materials have been developed by the membrane teams, so this project has some good (albeit, mostly still unoptimized) materials to work with.
- The project benefits from its approach to produce apples-to-apples comparative data for membrane samples.
- The project has a proven history of providing in-plane proton conductivity data.
- Although the need is questionable, the project has proven itself capable of optimizing Pt/C electrodes.

Weaknesses

- It is complicated to work with many different suppliers on a confidential basis.
- The rate of progress appears slow due to the lack of charting progress towards the barrier targets. It would be useful if the various team members who report results in their own projects would allow their reported progress to be compiled to make overall charts of progress towards a specific target.
- Trying to develop and apply their own electrode is limiting their ability to provide a SOA MEA component that is critical to the membrane operation.
- It has lost focus on measuring the membrane properties and comparing to the targets, as defined by DOE. Much of the electrode work has already been done by others.
- They need to keep the project scope simple and focused.
- The goals of the PI mostly do not seem to align with the needs of DOE. The project has been wasting time and resources on studying things like Pt vs. Pt/Co, when it should be focusing on developing electrode structures optimized for each different membrane. Let the catalyst projects look at Pt vs. Pt/Co.
- The project has allowed itself to lose focus on providing membrane characteristics and has detoured into electrode development using Pt alloys, varied ionomer loadings, use of collaborator ionomers, and other activities that take away from learning about membrane performance and durability.
- The project needs to find a way to make delivery of membranes for testing more facile and to make established protocols for MEA assembly and testing more acceptable to collaborators.

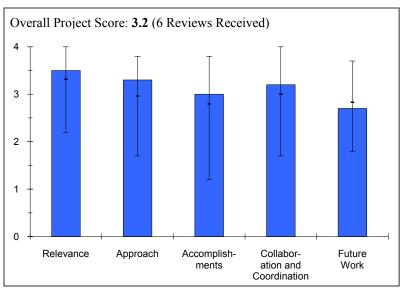
- They should discontinue trying to optimize electrodes on their own. Consider going outside to a wellestablished SOA fuel cell laboratory in industry for their best electrode knowledge, and adopt as much of that art as possible. If this is not possible, then focus more on the *ex situ* membrane fundamental properties that can be measured without complications of how the whole MEA was constructed.
- Stop optimizing MEAs for each program.
- Prioritize through-plane resistance and chemical and mechanical durability testing.
- During the questions and answers part of the presentation, certain technical team members indicated that electrode optimization should be eliminated from the scope of the project, and that the same type of electrode should be used on all membranes. This reviewer strongly disagrees with that recommendation. Membrane-electrode interactions are a critical factor in determining MEA performance, and there is no "one-size-fits-all" ionomer-containing electrode. The only reason this project still exists is so that it can work to optimize performance of novel membranes in fuel cells. Use of the same standard electrode would negate this project's *raison d'être*.
- Studies of different catalyst materials should be deleted from the project scope. While the optimal catalyst may be different for different types of MEAs, this effect is expected to be smaller than the effect of ionomer type, ionomer loading, processing conditions, and fabrication methods. Furthermore, the project should not be investigating optimized electrodes on commercial membranes, except to the very limited extent that these are needed for base lining. Keep the focus tightly on optimizing electrodes for each novel membrane.
- The following should be removed: the use of PtCo and further optimization of electrodes.
- While certain collaborators may have a preferred electrode composition, a common electrode should be used for at least one series of tests. This electrode may be a gas diffusion electrode. Tests with preferred electrode compositions may be done in comparison with an apples-to-apples baseline, if requested.
- *In situ* tests need to be focused on those diagnostics that provide membrane information: HFR, hydrogen crossover, and electrical insulation measurements. These three diagnostics cover the ideal function of the membrane.

Project # FC-36: Dimensionally Stable Membranes

Cortney Mittelsteadt; Giner Electrochemical Systems, LLC

Brief Summary of Project

The ultimate goal of this project is to meet 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 at 80% relative humidity (RH) at room temperature, 3) demonstrate membrane conductivity greater than 0.1 S/cm at 25% RH at 120°C using non-Nafion® materials, 4) demonstrate ability to generate these materials in quantities suitable for automotive stack, 5) build short stack with optimized materials and demonstrate durability, and 6) demonstrate how these



materials can be produced to meet DOE cost targets.

Question 1: Relevance to overall DOE objectives

This project earned a score of 3.5 for its relevance to DOE objectives.

- Development of robust, high temperature, low RH membranes is vital for the successful commercialization of fuel cells (FCs), especially for vehicle applications as high temperature / low RH allows a simplified (cheaper) balance of plant (BOP).
- The project is directly relevant and applicable to DOE's objectives.
- The project is relevant to the objectives of DOE's multi-year R&D plan. Initial activities were very much aligned to DOE's goal. Improvement of low RH membrane conductivity is critical to the success of DOE's hydrogen research initiatives.
- This program is relevant to DOE R&D objectives for a more durable conductive membrane.
- Membrane survivability is key to FC program success.
- The project seeks to find new membranes that operate in a high temperature and low RH FC environment. Such membrane materials are important to the DOE Hydrogen Program.

Question 2: Approach to performing the research and development

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

- The approach of this project is to develop two key aspects of the membrane: 1) extremely low equivalent weight (EW), chemically stabilized PFSA materials, and 2) a mechanically robust reinforcement layer.
- Giner is addressing the issue of low EW (high conductivity), leading to poor mechanical stability by supporting the ionomer in a strong mechanical structure and by modifying the polymer.
- Difficulty in impregnating the structure with ionomer is an issue.
- The porous structure results in a conductivity penalty.
- The approach of making dimensionally stable membranes with two-dimensional laser-drilled hole supports (2DSM) and dimensionally stable membranes with three-dimensional porous supports (3DSM) is good for meeting the program objectives. Determining the conductivity of triflic acid and benzene sulfonic acid (BSA) is a good approach to understand the feasibility of achieving the DOE low RH membrane target using presently available sulfonic acid based ionomeric membranes. The data in slides 9 and 10 show that it's very convincing

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that it's an uphill battle to achieve the DOE target in a practical polymeric membrane, while neat BSA just meets the target. Conversion of BSA in any polymeric form will result in a conductivity penalty.

- The low EW ionomers do meet conductivity and resistance targets. The key to this program is to find a way of making stable membranes. It is not clear if the supports will make these membranes stable enough, but this is the right approach.
- The work keeps cost, performance, and life targets in sight and in balance.
- The PI has correctly noted that FC membranes with next-generation properties (performance) will be composed of low EW polymers, where the membrane polymer is chemically stable and where the membrane is very thin. The PI's approach of embedding an ionomer in a two-dimensional or three-dimensional matrix is not particularly innovative, but it does represent a rational and reasonable approach to making better FC membranes. The key here is to find the right ionomer, i.e., an ionomer with a very high conductivity that is not water soluble, since the inert matrix will effectively dilute the ionomer's ion exchange capacity (IEC) and lower the conductivity.

<u>Ouestion 3: Technical accomplishments and progress toward project and DOE goals</u></u>

This project was rated **3.0** based on accomplishments.

- Technical progress in the two key areas of this project, 1) the ionomer, and 2) the reinforcement layer, is ongoing.
- Unfortunately, Giner's core intellectual property (IP) and unique sales point (USP) is it's 2-DSM material, but they were unable to reduce the cost sufficiently. With that in mind, it still remains unclear what Giner is bringing to this program.
- With the 3-DSM, it is not readily apparent how uniformly the polyelectrolyte is penetrating the reinforcement layer. Furthermore, the inconsistency between the proton conductivity (PC) and polarization (I-V) curves indicate perhaps a thin film of polyelectrolyte on the reinforcement surface, which in turn would affect the proton conductivity results in a positive way (through plane vs. in plane).
- The electrode-membrane interface has been improved, resulting in lower MEA resistivity and higher cell performance.
- Giner has downselected to the 3-DSM approach and stabilized the low EW material.
- Partner Millipore estimates the cost of the porous structure is well within DOE targets.
- The team has made good progress in the development of two- and three-dimensional, stable membranes. The team has used 700 EW membranes and PFSA homo-polymers to develop dimensionally stable membranes. It seems the team's strategy is to lower the EW of the ionomer to achieve low RH and high temperature conductivity. The team should also consider the dissolution behavior of low EW ionomers during humidity cycling. Under fully humidified condition, these low EW ionomers and homo-polymers tend to dissolve and leach out of the reinforcement matrix. So, the team should also include testing of these membranes under automotive humidity cycling conditions. From the data in slides 23 and 24, it seems that the team has hit the conductivity limit that can be achieved by low EW PFSA type material. The team needs to think of an alternative strategy to achieve low RH conductivity, while maintaining the stability of the ionomer under humidity cycling condition. The durability study of 3DSM under electrolyzer conditions would demonstrate the viability of 3DSM membranes for electrolyzer applications.
- Conductivity values are high enough to meet the targets that have been demonstrated. Showing that threedimensional stabilization can allow membranes to meet the durability targets is critical at this point. A method of determining if the 20,000 humidity cycle target can be met is needed.
- Improvements in cycle life were demonstrated. Significant improvements in combined performance, cost, and life are still needed.
- Technical accomplishments this past year have been good. The PI has realized that his two-dimensional matrix was too expensive for commercialization. The move to examine three-dimensional supports was correct. Unfortunately, the conductivity results for the three-dimensional matrix are not as good as those collected with a two-dimensional support (I believe that the two-dimensional support conductivity results may have been skewed by the presence of an ionomer over layer). The project is now moving toward identifying (synthesizing) new high conductivity polymers to impregnate into the three-dimensional support. The problem here is the water solubility of very low EW polymers.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.2** for technology transfer and collaboration.

- The project is very well managed, with clear objectives from the various partners—State University New York Environmental Science Forestry (SUNY-ESF) provides low EW PFSA materials, Florida does PC and FC testing, and General Motors (GM) is the voice of the original equipment manufacturer (OEM).
- However, as previously stated, Giner is no longer using their 2-DSM material; therefore, their reliance on collaborators and suppliers increased significantly. Perhaps Giner should be a little more active with their membrane scale-up and potential catalyst-coated membrane (CCM) fabrication.
- The team includes an automobile OEM, universities, and an industrial manufacturer of porous structures.
- The team consists of good, but very small, academic and industrial collaborations. 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 2DSM and 3DSM membranes.
- It is not clear what GM is doing on this project.
- The project team has been collaborating with OEMs and responding to cost and manufacturability concerns raised in prior years.
- Most of the work was carried out at Giner. The low EW polymer synthesis work was performed at SUNY-ESF. Coordination between the partners seemed to be adequate.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

This project was rated 2.7 for proposed future work.

- Proposed future work covers the key needs of the program; however, it is unclear if enough time or money remains in this project to reach a conclusion. The membrane at this point still requires a lot of optimization, as does the membrane electrode assembly (MEA).
- The project will end within a year.
- Effort on stabilizing the system (with RH cycling) and optimizing the catalyst layer/membrane interface will continue.
- A stack test may not be the best use of remaining scarce resources.
- The proposed plan is sound and fits with the development work. RH cycling and short stack testing is necessary. However, given the difficulty in impregnating the reinforcement matrix with ionomer, a performance reproducibility study using membranes made during different batches should be considered.
- More work should be done on the insoluble 3-DSM. This material looks most promising to meet the targets.
- A better definition of future work plans is needed.
- The success of this project hinges on finding a low EW ionomer to impregnate into the three-dimensional support, where the ionomer is not water soluble and is of sufficiently high conductivity, so that the "effective" conductivity of the composite membrane meets DOE targets. The project PI realizes this point. I am somewhat skeptical that such a polymer will be found (for many years researchers have been searching in vain for such a material).

Strengths and weaknesses

Strengths

- The project management of Giner is a strength.
- The team has access to automobile test protocols and real-world drive protocols at their partner's (GM) test laboratory. The team also has individuals with solid understandings of the field and related challenges in such development work.
- The low EW ionomers do meet conductivity and resistance targets.
- The project builds understanding of low EW material performance and durability, when combined with support structure materials.
- The project PI has a good grasp of the technical challenges that must be overcome next year.
- The PI has performed both physical property characterization experiments and FC tests on his new membrane materials.

Weaknesses

- Unfortunately, Giner's core IP and USP is its 2-DSM material, but they were unable to reduce the cost sufficiently. With that in mind, it remains unclear what Giner is bringing to this program. Perhaps Giner should be a little more active with their membrane scale-up and potential CCM fabrication.
- The team is exploring the avenue of low EW ionomers, which is a common strategy most of the researchers are pursuing. The team should think of some alternative strategy to circumvent the traditional approach to low RH membrane conductivity.
- It is not clear if the supports will make these membranes stable enough.
- A method of determining if the 20,000 humidity cycle target can be met is needed.
- There needs to be a clearer definition of future work.
- The use of a three-dimensional inert polymer matrix with ionomer impregnation is not new and innovative (the PI's composite films are very much like GoreSelect membranes).
- It is doubtful that a water-insoluble ionomer for impregnation will be found with a very low EW. The use of higher EW polymers will result in a composite membrane conductivity that does not meet the DOE target. In moving from a two-dimensional to a three-dimensional matrix, the PI is confronting the same problem that many FC membrane researchers have faced in the past: anything that one does to a charged polymer to prevent water solubility or excessive swelling produces an unwanted drop in proton conductivity.

Specific recommendations and additions or deletions to the work scope

• The project plans cover what is needed for successful completion. However, this reviewer is concerned about the time and money left in this program. Furthermore, the inconsistency between the PC and I-V curves for 3-DSM composite membranes indicate perhaps a thin film of polyelectrolyte on the reinforcement surface, which in turn would affect the proton conductivity results in a positive way (through plane vs. in plane). Further characterization and optimization is needed.

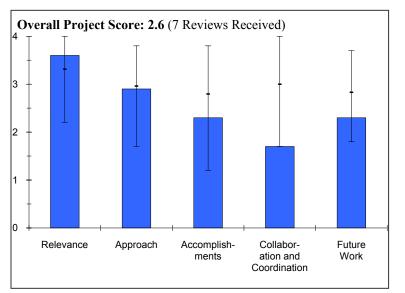
Project # FC-37: Rigid Rod Polyelectrolytes: Effect on Physical Properties Frozen-in Free Volume: High Conductivity at Low RH

Morton Litt; Case Western Reserve University

Brief Summary of Project

Project objectives are to: 1) synthesize polyelectrolytes that reach or exceed DOE low humidity conductivity requirements, 2) use materials and synthetic methods that could lead to cheap proton exchange membranes (PEM), 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.

<u>**Ouestion 1: Relevance to overall DOE**</u> <u>**objectives**</u>



This project earned a score of **3.6** for its relevance to DOE objectives.

- Development of robust, high temperature, low relative humidity (RH) membranes is vital for the successful commercialization of fuel cells (FCs), especially for vehicle applications as high temperature and low RH, allows a simplified (cheaper) balance of plant (BOP).
- Membranes for hot and dry service are relevant to DOE's objectives.
- Membranes with high conductivity at low RH would provide significant performance benefits and enable PEMFC system simplification and cost reduction.
- This program is very relevant to DOE R&D objectives for a more durable conductive membrane.
- This certainly is a relevant project and will help design simpler fuel cell systems for practical purposes. Being able to operate at a higher temperature with no or low humidification, while maintaining high performance (which requires higher membrane conductivity) and efficiency, are key requirements.
- The project is very relevant and shows promise for new materials to meet DOE targets.
- Interesting and relevant to the program. Novel polymer chemistry is being developed to produce materials that are thermally stable and have enhanced conductivity at low RH. It is not clear if this is going to be a cost-effective solution, considering the chemistry and reagents being used.

Question 2: Approach to performing the research and development

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

- While the PI is clearly a world leader in polymer synthesis, this approach clearly lacks a strategy or pathway of getting a high potential or high proton conducting polyelectrolyte into a commercialized mechanical robust membrane. Instead of focusing in on a promising polyelectrolyte and trying to make it more viable for FC applications, the PI moves onto another polymer system.
- The approach of a rigid rod structure "locking in" volume to hold water is valid.
- Apparently, other parts of the system; e.g., mechanical stability, have not been easy to achieve.
- Base polyparaphylene polymers have proven to have very high conductivity. The focus has been to apply a variety of techniques to make mechanically stable membranes via high molecular weight (MW), grafting, and co-polymerization.
- They have taken a very novel approach to high conductivity at low RH.

- This is a very creative, out-of-the-box approach, but it is also a risky one! The PI is approaching this methodically to address issues with mechanical properties and also to address the issue of poor co-polymerization with non-polar co-monomers.
- The approach is good for the preparation of new materials, but it needs work on stabilizing them and making them water insoluble.
- The project approach is generally reasonable. The work to crosslink the polyelectrolyte is understood, but there is a need to do some characterization of membrane properties, *ex situ* polyelectrolyte durability, and membrane electrode assembly (MEA) testing to establish proof of concept that this technology will work in a FC.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.3 based on accomplishments.

- On numerous slides, incredibly high proton conductivity as a function of RH is demonstrated, but no work on stabilizing these materials has taken place. As previously mentioned, instead of focusing in on a promising polyelectrolyte and trying to make it more viable for FC applications, the PI moves onto another polymer system.
- Materials stability is still an issue. By now, cell testing should be underway.
- So far, attempts to make mechanically stable polymers have not worked.
- The conductivity of these materials is very high at low RH percentages and exceeds targets. Demonstrating progress on making membranes with suitable mechanical properties and chemical stability will be important for the final year of the program.
- It is a difficult task and will be a challenging one to find an optimum solution. Good progress was made toward developing water insoluble PEMs with high ion exchange capacity (IEC) and high conductivity by grafting alkyl benzenes on sulfonic acids. Some of them even demonstrated dimensional stability. Issues remain with mechanical properties and other areas.
- The accomplishments are excellent except in the area of working with a collaborator to form stable materials that can be further tested and characterized.
- Some progress has been made toward crosslinking the polyelectrolytes, but more is required.
- No progress has been made on determining the *ex situ* durability of these materials and little progress has been made on membrane properties, outside of solubility and conductivity.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated 1.7 for technology transfer and collaboration.

- There have been no collaborations outside. "Conversations" with outside people is the bare minimum.
- There seems to be active involvement only with other universities.
- The project could benefit from additional polymer synthesis expertise. Approaches from General Motors (GM) were rebuffed because of intellectual property (IP) concerns.
- This program would benefit from greater interaction with a stack or MEA manufacturer for more FC evaluation.
- They appeared to have good collaboration. A slide recognizing various partners is always a good idea!
- Seriously needs some collaborative support and acceleration of developing a stable testable material.
- Other than the Florida Solar Energy Center (FSEC) and Dr. Pintauro's involvement, there is no evidence of solid collaboration with other institutions or industry.

Question 5: Approach to and relevance of proposed future research

This project was rated **2.3** for proposed future work.

- In my opinion, until the PI starts to take proactive steps to stabilize promising polyelectrolytes and works with outside institutions, this project will be nothing more than an academic exercise.
- The project has less than a year to go.
- The path forward is unsure, nebulous, and somewhat Edisonion.

- Working with Pintauro at Vanderbilt to electrospin composite membranes may enable preparation of stable membranes. Otherwise, it looks like more of the same ideas will be tried.
- Developing a water-stable membrane will allow more extensive testing of this important new material. This should be a high priority.
- A significant amount of high risk work is ahead of the PI to successfully complete this project. Completing this task within the remaining budget and on schedule will be challenging.
- Electrospinning and grafting are possible pathways to stabilize the materials and should be pursued with urgency.

Strengths and weaknesses

Strengths

- They have exhibited world-class polymer synthesis.
- Inherently highly conductive materials developed that exceed DOE targets.
- Very novel approach to high conductivity at low RH.
- Out of the box, creative approach!
- Excellent materials development.
- Creative approach and the conductivity results of unoptimized membranes look promising.

Weaknesses

- There is no membrane stabilization in the works. There are no collaborations with other institutions.
- The project seems to be manpower limited.
- They have poor collaborations. Very little progress has been made in the past year.
- This program would benefit from greater interaction with a stack or MEA manufacturer for more FC evaluation.
- Inherently poor mechanical properties of the rigid rod material.
- They lack urgency in preparing a stable, water-insoluble material and collaboration with industry.
- Mechanical properties of the materials may be a problem if there is any degree of swelling of the membrane in the MEA. The approach may not adequately address this and it is not clear that putting it in an inert matrix will solve the problem. Water solubility of the materials also hasn't been fully addressed.

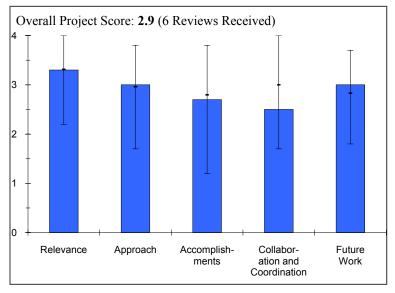
- Pursue membrane stabilization and increase collaborations.
- Professor Litt claimed that copolymerization with non-functionalized monomers and blocking cannot be done, but I would still like to see block polymerization attempts added to the program. Include quantifiable measurements of mechanical properties (tensile testing and swelling).
- This program would benefit from greater interaction with a stack or MEA manufacturer for more FC evaluation.
- Narrow down the scope of the proposed future work.

Project # FC-38: 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 nano capillary network (NCN) proton conducting membranes for hydrogen/air fuel cells (FC) that operate under high temperature, low humidity conditions, with: 1) high proton conductivity, 2) low gas crossover, and 3) good mechanical properties. The 2009-2010 project goal is to prepare NCN proton exchange membranes (PEM) where high ion exchange capacity (IEC) sulfonated polyphenylene (from Morton Litt's group at Case Western Reserve University (CWRU)) replaces the sulfonated polyhedral oligometric silsesquioxane (POSS) and a commercially available uncharged polyphenylsulfone (PPSU) replaces Norland Optical Adhesive (NOA) 63.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.3** for its relevance to DOE objectives.

- Membranes which operate under high temperature and low humidity conditions and maintain high proton conductivity, low gas crossover, and good mechanical properties are important to achieving DOE's FC performance and durability goals. The objective should also contain a membrane cost target that is consistent with DOE's cost targets.
- The project is relevant to the objectives of DOE's multi-year R&D plan. The activities are very much aligned to DOE's goal. Improvement of low relative humidity (RH) membrane conductivity is critical to the success of DOE's hydrogen research initiatives. The NCN method is a unique way to generate nanomorphology mediated block-copolymer morphology.
- The project is aligned with DOE goals and targets. High temperature membranes are an enabling technology that allows system simplification and cost reductions.
- There is good support of DOE program objectives. It would help to outline the benefits of this approach versus using an expanded poly-tetrafluoroethylene (PTFE) scaffold, especially since perfluorosulfonic acid (PFSA) ionomers appear to be the material of choice for the work now.
- The project is relevant to DOE objectives. Development of stable, low resistance membranes is important to the success of the DOE Hydrogen Program, and this project is making a contribution to that effort. The relevance is diminished to some extent by the dependence of the PI on materials supplied by other groups, which are not necessarily being developed with funding from this project. As it stands, the relevance of the project is limited to membrane architectures. The project does not include a strong proton-conductor development component, which is arguably a bigger challenge in developing membranes for hot and dry operation.
- This project represents a concept for creating durable membranes by allocating mechanical stability to a blended component. Because durable membranes are highly relevant to the DOE effort's goals, this project is highly relevant.
- The project has been using both PFSA-based and hydrocarbon ionomers, which imply that the project has not limited itself to proton conduction in one temperature regime, which further extends project relevance.
- Other targets (low gas crossover) are also taken into consideration.

Question 2: Approach to performing the research and development

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

- The research shows promise, but it has not yet focused on a single process methodology, and appears to still be in "scouting" mode. A more in-depth study of the best process should be completed within the next year.
- This is a "out-of-the-box" approach to the solution to low RH membrane conductivity. Changes in the electrospinning material composition and corresponding inert materials is a good approach in exploring the feasibility of obtaining a functional nanocomposite membrane. Mixing inorganic additives to the ionomers to make inorganic additive impregnated ionomeric nanofibers is also a good approach to add different materials to nanofibers. However, the team should carry out studies on the leachability of such additives from ionomeric nanofibers. The approach to make "Hollow Bore" Nafion® nanofibers is an interesting approach.
- This reviewer is not sure about using the materials mentioned.
- This is an interesting and flexible approach that may be applied to a variety of polymer chemistries to make composite materials.
- The approach of the project is to develop membrane architectures that combine a proton-conducting component with an engineering polymer capable of supplying the necessary mechanical properties. This is a sound and reasonable approach. Collaboration with 3M to incorporate low equivalent weight (EW) ionomers is a critical part of this approach. If successful, the approach would succeed in producing membrane materials that meet DOE resistance targets while maintaining good mechanical and chemical stability. The focus, however, seems diffused, with too much emphasis on experimenting with various combinations of proton-conducting and support polymers, and insufficient emphasis on using the knowledge gained to develop high performance membranes.
- Many membranes prepared in the DOE projects are attempting to decouple mechanical stability from the conductive ionomeric phase in order to create greater flexibility in ionomer chemistry, while also providing mechanical stability. This project is consistent with those approaches, while adding the novelty of reversing the usual roles by preparing the ionomer in a "strand-like" phase, while the non-conductive component fills the remaining volume (or is also electrospun).
- The identified approach still leaves some questions unanswered, particularly those related to the cost of scaling up the electrospinning process.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.7 based on accomplishments.

- Significant progress has been made in this project, but the results presented do not demonstrate a significant advantage over current commercially available materials.
- Most of the results presented show that the performance of nanofiber-based membranes depends on the volume fraction of ionomer present in the composite membrane. With Nafion and 3M PFSA, the composite membrane performance depends on the total amount of ionomer present in the composite. Hence, the advantage of having an ionomeric nanofiber matrix has not yet been realized. So the question is whether there is any promise of going for such a route. If the overall performance depends on the total ionomer quantity and the nanofiber entangled matrix doesn't offer any advantage, then pursuing this membrane fabrication route is questionable.. The team should think along this line.
- Replacing NOA with polyphenylsulfone and developing a dual fiber mat and consolidation technology demonstrates the practicality of this approach.
- Swelling data is useful, especially the comparison of Nafion 212 with a 3M825 mat.
- Durability comparison should be done using the same cycles for both Nafion and nanofiber composite.
- No conductivity data at high temperature and low RH for this past year—still need to work toward high temperature, low RH targets with stable materials, or show nanofiber geometry stabilizes materials (CWRU materials or lower equivalent weight PFSAs from 3M).
- A good amount of progress has been made, especially with eliminating the optical adhesive as the matrix.
- Accomplishments this year have been good, but not as good as the previous year. Interesting work has been performed, but it is not clear in all cases that this work represents significant progress toward meeting DOE targets. Replacement of the NOA 63 resin with PPSU is a good step. However, the value of the studies

comparing Nafion fibers in PPSU matrix vs. the inverted PPSU fiber in Nafion matrix is unclear. One would have hoped that the PI would have continued to make progress in decreasing area resistance by optimizing structure, thickness, nature of ionomer, and use of additives, but there is no data or other indication that resistances of current membranes are lower than those from a year ago. Use of a higher EW ionomer like commercial Nafion is acceptable in this project to develop fundamental understanding of the nanoporous network, but it would have been nice to have seen a low resistance membrane incorporating a low EW ionomer to demonstrate that progress has been made over the past year in developing materials that will be useful in FCs. Without such a demonstration, it is not clear that the work done over the past year is actually leading toward the development of viable materials for FCs. The preparation of electrodes based on ionomer nanofibers, in contrast, represents a potentially significant development for FCs. This electrode work should be emphasized going forward.

- Although the concept has been shown to deliver high conductivity results, efforts with more conductive PFSA, and with use of PPSUs for mechanical stability have not yet shown exceptional conductivity. However, what is being compared is essentially Nafion versus less Nafion, so not much can be expected yet.
- Nice progress has been shown for open circuit voltage (OCV) testing with electrospun Nafion (at least 200 hours near 2 mA/cm² hydrogen crossover), and for low in-plane swelling using the 3M ionomer (lower than PFSA baselines).
- Electrospun electrodes are interesting, but have not demonstrated a tangible advantage yet.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **2.5** for technology transfer and collaboration.

- 3M and Nissan are both capable of providing practical industry feedback.
- The team consists of good partners and good collaborative work is expected. So far, no technology transfer scenario is reported by the team. Except obtaining a PFSA ionomer from 3M, not much collaborative work with the partners has been reported.
- Extending collaboration with Nissan would be helpful.
- There is good collaboration with 3M.
- The project aims to develop membrane architectures that are conducive to stable, low resistance operation, but does not address the chemistry of the low EW or otherwise highly conductive ionomers required as the protonconducting component of these architectures. Effective collaboration is required to supply this component. The collaborations with 3M and with CWRU are, therefore, vital to the success of the project. A lack of data with CWRU materials, and a lack of data with more advanced 3M materials, suggests that the collaborations are not yielding as much fruit as one would have hoped.
- As indicated by the low x-y swelling data for the nanofibers generated from 3M 825 EW ionomer, the 3M collaboration is contributing to the progress of this project.
- In the question and answer part of the presentation, it was mentioned that cost analyses are being performed with a company in Tennessee. This collaborator should be identified.
- Some test protocol sharing exists with Nissan, but this does not appear to be an extensive effort.
- This could be an opportunity for greater collaboration as the project attempts to deliver final membrane samples and evaluations.

Question 5: Approach to and relevance of proposed future research

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

- The future work plan is ambitious, considering past progress and the project end date, but the work is relevant and could lead to better membrane and/or electrode manufacturing processes.
- Most of the emphasis of the future work is on further optimization of the nanofiber matrix and nanofiber membrane formation. Very little emphasis is given to MEA fabrication and FC testing. The team should first determine the limit of this technology and see whether the nanofiber route is capable of delivering membranes with low RH conductivity better than corresponding dense membranes made with the same ionomer. Otherwise, the research work will remain far away from DOE's low RH membrane goal. So far, all the data suggests that

the nanofiber membranes perform to the level of available ionomer volume fraction and doesn't show any performance advantage over corresponding dense polymeric membrane.

- Stability of the sulfonated polyphenylenes from CWRU is questionable. A back-up plan is needed for increasing conductivity at high temperatures with low RH.
- Collaborations with others in the high temperature membrane group could be beneficial. The team could look into spinning CSM polyPOMs or using them as additives.
- The future work proposed is logical and is constructed to move the project in the correct direction.
- For the most part, the future work is reasonable, but some parts should be deemphasized. In particular, the work on hollow bore Nafion fibers should not be continued unless the PI has a reasonable path toward producing narrower fibers; from the presentation, I gather that he does not have such a path. However, the PI verbally indicated during the presentation that work on hollow-bore fibers is ending, so for the purpose of scoring the review I will give precedence to this comment and ignore the fact that hollow-bore nanofibers are listed on the future work slide.
- The proposed future work on inert or uncharged polymers has not been sufficiently justified. The benefits of this work are unclear. The inadequacies of polyvinylidene fluoride (PVDF) and polyphenylsulphone (PPSU) were not pointed out, and Vanderbilt should explain if there is reason to believe that other materials will be better.
- The electrode work should be emphasized, as this is a promising application for electrospun nanofibers. The initial work looks very promising and should be pursued aggressively to eliminate potential poisoning components and to maximize performance.
- The proposed new membrane samples are as expected from the context of the presentation. The continued use of 3M PFSAs is the right direction.
- The examination of alternative non-conductive components will likely prove to be useful, although the deficiencies of the incumbent PPSU have not been clearly reported (the x-y swelling appeared to be low).
- The hollow bore materials are worth studying for the possibilities that exist in promoting proton conduction, but the size must be reduced.
- It is not yet quantified what the motivation might be for electrospun electrodes. An oxygen diffusivity measurement might be useful. Some questions may be asked as to whether this electrode structure would tend to be overloaded with ionomer, and, therefore, promote platinum dissolution.

Strengths and weaknesses

Strengths

- The project pursues an alternative route to low gas crossover membrane fabrication. The team is solid, with a good knowledge base of the challenges in this project. The project is exploring an alternate route for making block-copolymer type morphology in membranes. The electrospin process is versatile and can accommodate many different types of ionomers and ionomer mixtures. This gives the team the flexibility in fabricating and evaluating many different types of composites.
- Vanderbilt employed a novel materials engineering approach to improving membranes.
- Vanderbilt used a novel approach toward creating composite membranes, one that is completely different from most of the other approaches out there.
- The electrospinning technique is a good technique to make interesting and potentially useful membrane and electrode structures. The project is not dependent on the development of any one ionomer chemistry, but rather could be applied to just about any ionomer capable of being electrospun. If combined with the best available proton-conducting materials, the project has the potential to produce high performance membranes. Electrodes produced with electrospun fibers would have a promising architecture for good mass transfer, as well as good protonic and ionic conductivity.
- The project manages to present a novel concept that is still consistent with the strategy of many of the other projects.
- The project has shown the capability of creating materials that are highly conductive at 120°C, 50% RH.
- The project has responded well to past feedback and has incorporated PFSAs as candidate ionomers.
- The project has been able to adapt the primary technique of electrospinning quite capably for a variety of materials, and has been able to find ways to work with materials that have been historically tough to work with, through the addition of polyethylene oxide (PEO) or other species.

Weaknesses

- The project should focus more on the feasibility study of nanocapillary methods for obtaining a functional, low RH membrane. So far, none of the performance data suggests that the nanocapillary route to membrane fabrication offers better conductivity than the dense membrane made from a parent ionomer. The team should work more to determine whether the mix of ionomer nanofibers offers any potential of having nano composite membrane with higher low RH conductivity than its parent ionomers.
- The ionomer synthesis is a weak aspect of the project.
- The process is complicated, especially for a dual fiber mat with a polysulfone matrix.
- The lack of a significant ionomer-development component makes the PI dependent on partners, or nonpartnering institutions, for ionomer materials. So far, no results have been presented for the highest performance state-of-the-art or experimental ionomer materials. The project has not demonstrated a high performance membrane in the last year.
- The project still needs to be able to deliver final membrane samples using components that are expected to be durable and highly conductive.
- There are some areas where project decisions should probably be explained more clearly. For example, the reasons for trying different non-conductive materials as mechanical supports were not explained. This reviewer would also like to know if there is any data that supports the use of electrospun electrodes.
- A cost estimate is still needed.
- There is still some opportunity for greater collaboration.
- *In situ* diagnostics for cell tests should be reported; e.g. high frequency resistance (HFR), hydrogen crossover, electrochemical area (ECA), etc.).

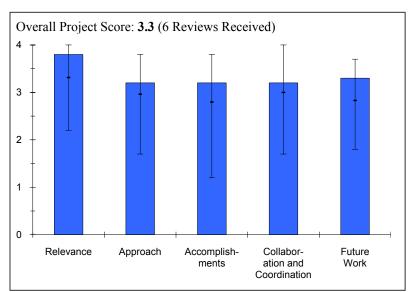
- Include a cost study to prove that an electrospun membrane can be consistent with DOE's cost targets.
- Compare electrospun membrane performance, durability, and cost against current competitive options (W.L. Gore & Associates (Gore), or 3M membranes, not Nafion).
- Add a cost study and a comparison of projected cost for electrospun fibers vs. three-dimensional mats; i.e. the Giner approach.
- Eliminate hollow-bore nanofiber work.
- Without compelling data for the electrospun electrode effort, this part of the project could be truncated.
- A cost estimate should be added and the collaborating party should be listed.
- RH cycling should be performed soon with a 3M-based PFSA for either one of the two methods (ionomer surrounded or uncharged polymer surrounded), just to have some baseline.
- More rationale could be provided for the down selection of uncharged polymers.

Project # FC-39: Novel Approaches to Immobilized Heteropoly Acid (HPA) 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 conductivities (σ) >0.1 S/cm at 120°C and <50% relative humidity (RH). The objective for 2010 is 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 or durability.



<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of **3.8** for its relevance to DOE objectives.

- The development of robust, high temperature, low RH membranes is vital for the successful commercialization of fuel cells (FCs), especially for vehicle applications, as high temperature with low RH allows a simplified (cheaper) balance of plant (BOP).
- The project directly addresses membrane development, one of DOE's key technical issues.
- The development of a membrane that is highly conductive and stable at 120°C would enable significant fuel cell system (FCS) simplification and cost savings.
- The work addresses the very significant challenge of high conductivity, low RH, high temperature membranes.
- This certainly is a very relevant project and will help design simpler FCSs for practical purposes. Being able to operate at a higher temperature with no or low humidification, while maintaining high performance (requires higher membrane conductivity) and efficiency, are key requirements.
- This project is very relevant. The need to provide conductivity in the absence of water is directly addressed. The project has materials that are inherently conductive and hence its full upside potential is very large.

Question 2: Approach to performing the research and development

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

- The concept of immobilizing HPA materials into a polyelectrolyte is a very interesting approach which has fairly high potential.
- PI states that a series of experiments, which were known beforehand not to be viable, were necessary (model compounds which certainly would have dissolved), but this reviewer still does not believe their validity.
- The project has overcome most of the issues expected with HPA-based membranes.
- The Colorado School of Mines (CSM) has employed a unique approach of incorporating "super acids" in a polymer backbone to enable low RH conductivity. Several types of HPA polymer systems have been studied, including those with and without methylene groups. There's been some work on making chemically and mechanically stable films, but more emphasis is needed there.
- The work seeks a novel approach, but with a clear connection to achieving a manufacturable solution.
- The very focused and methodical approach has been yielding promising results.
- The staged approach is good and demonstrates the utility of the HFA additives.

<u>Ouestion 3: Technical accomplishments and progress toward project and DOE goals</u></u>

This project was rated 3.2 based on accomplishments.

- Conductivity as a function of RH is certainly excellent and far exceeds the DOE milestones. Immobilization techniques with 3M appear to be progressing well, and preliminary results look promising, provided the baseline material (pink line on slide 17) does not conduct protons as the PI stated.
- Conductivity has been established and good mechanical properties are claimed, although IP issues prevent full discussion.
- CSM seems to have settled on the HSiW₁₁(vinyl)₂ HPA to develop membrane concepts. I couldn't understand the morphological studies results presented for Poly-POM 85v. There is some encouraging conductivity data on the initial type II copolymers, but these materials are not mechanically stable. The type III copolymers developed with 3M should provide better mechanical stability, but the conductivities are lower than perfluorosulfonic acid (PFSA) ionomers.
- There has been good progress in high temperature conductivity, but there are solubility challenges.
- They were consistently demonstrating high proton conductivity in robust films. Several film chemistries were developed that demonstrate high oxidative stability and promising mechanical properties.
- The progress is good, although one might think 3M could provide more assistance with the polymer synthesis and modification portion of the work.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.2** for technology transfer and collaboration.

- The collaboration with 3M is very strong and clearly is shown throughout the project. In fact, this collaboration is exactly why this program is so strong and should be used as a benchmark for future DOE collaborations. My suggestion is to have the stack developer involved at one point for outside testing and validation.
- The industrial partner provides valuable technical and material support.
- There is a strong collaboration with 3M to develop the copolymers.
- There is a good connection to manufacturing through 3M. There is also a good use of external test house through the Florida Solar Energy Center (FSEC).
- They have exhibited a satisfactory level of collaboration with development partners.
- The addition of General Motors (GM) is good, but they could use more academic collaborations to extend understanding. 3M might not share results.

Question 5: Approach to and relevance of proposed future research

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

- The proposed future work is outstanding and is in line with previous accomplished work. CSM could perhaps add a third party for cell testing and validation.
- The project ends in less than a year.
- The remaining work focuses on morphology, durability, and FC testing.
- Cost needs to be addressed.
- The plan is to develop materials that have both good conductivity and are mechanically robust. It is unclear if any more HPA monomer work is going on or if CSM has settled on the HSiW₁₁(vinyl)₂ monomers for all membrane concepts. Explicit mechanical testing should be included. Some effect should be focused on improving lower temperature conductivity. The project could also benefit from some modeling effort to understand the conduction mechanism in these systems to guide in materials selection.
- These are all reasonable plans. One would like more detail on plans for addressing solubility.
- CSM has well thought out future works. The combined efforts at CSM and 3M should yield an optimum solution.
- It seems like an awful lot to do in the last year. It would be good to break out which organization is doing what. The plans are challenging.

Strengths and weaknesses

Strengths

- The immobilization of HPA into polymer backbone has the potential in achieving key DOE milestones. The collaboration with 3M is excellent.
- They employed a unique approach to meeting low RH conductivity targets. They have a strong collaboration with a polymer and membrane supplier in 3M.
- They used a novel approach that adds to knowledge base.
- Material selection and zooming in with the right platform are both strengths of this project.
- The HPA materials are unique and provide a novel mechanism.

Weaknesses

- They took perhaps too long to get this point. The model compound studies perhaps were not necessary.
- Low temperature conductivity is an issue.
- The materials for type III copolymers were not disclosed, so the value to the FC community is limited. There is a lack of mechanical characterization, understandable swelling data, stability measurements, and modeling effort.
- Solubility needs to be addressed.
- The reviewer didn't get a good feel for projected durability in a real-world environment.
- CSM needs more synthetic help.

- Add a stack developer for outside testing and for validation when a stable membrane is made.
- I think that adding a functional group, such as sulfonic acid, to the copolymers in type III may warrant investigation. Add mechanical/swelling testing and hydrolytic stability testing to scope.
- 3M needs to do more in this project.

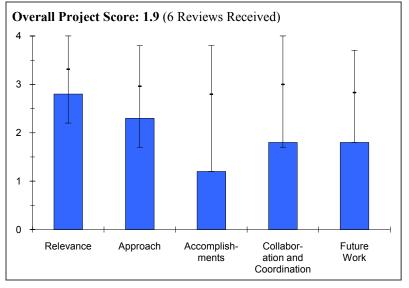
Project # FC-40: High Temperature Membrane with Humidification-Independent Cluster Structure *Ludwig Lipp; FuelCell Energy, Inc.*

Brief Summary of Project

The objectives of this program are to: 1) develop polymer membranes 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 DOE objectives

This project earned a score of **2.8** for its relevance to DOE objectives.



- Polymer membranes with improved conductivity at up to 120°C are relevant to DOE targets, but the objective makes no reference to cost or durability targets.
- The project is relevant to the requirements for high temperature membrane development and meets overall DOE objectives. The goals and objectives of the project are designed satisfactorily. The multi-year plan is in line with DOE research, development & demonstration (RD&D) objectives.
- The project goals are relevant. The presentation spent too much time on this, and not enough time on how they would actually do this.
- The project is aligned with DOE targets and goals. High temperature membranes are an enabling technology that will allow system simplification and cost reductions.
- The development of composite membranes with high conductivity under hot and dry conditions is relevant to the goals and objectives of the Hydrogen Program. The relevance of the project is limited by the refusal of FuelCell Energy, Inc. (FCE) to share information with the public, despite the fact that the work is funded by the public.
- This project is marked "good" for relevance, since it seeks to fabricate high temperature membranes that will meet targets for conductivity, durability, and gas crossover.
- It is well understood within the program that membranes are needed that can perform at temperatures up to 120°C.
- Although there are no compositional details provided as to the materials being used, there is no reason to conclude that this work is not attempting to achieve targets relevant to automotive fuel cell (FC) development.
- Because chemistry is not identified, this project can only be rated "Good" for relevance. For all that the reviewers know, there may be unstable chemistries or high cost processing involved.

Question 2: Approach to performing the research and development

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

- The approach seems well planned. They need to approach references, cost, and durability, which were neglected in the project objective statement.
- The approach of making composite membranes possessing materials with adequate properties is a good idea. However, interaction between these multiple components in the composite under low relative humidity (RH) FC operational condition is the unknown.
- It is very difficult to assess the work. The approach seems okay, but experiencing a lot of different additive or instability issues, as this project is, is asking for trouble.

- It is difficult to judge the approach as the description of the approach is limited. Very little information on the approach was provided.
- The approach of using a low equivalent weight (EW) perfluorosulfonic acid (PFSA), with reinforcement to provide mechanical stability, and additives to provide increased water uptake and increased protonic conductivity, is a good one. The project effectively addresses technical barriers.
- Details regarding the chemistry of the ionomer, reinforcement, and additives have not been provided. Details regarding processing have not been provided. Therefore, without knowledge of the chemistry involved, the approach cannot be judged.
- In the past, distinctions have been made between membrane projects based on approach. Some projects have used ionomers with unstable functional groups, or have used reinforcements that proved to be too costly. This project, however, has attempted to avoid any judgment by not revealing composition or processing. In doing so, it violates the intention of publicly funded research.
- Partners in the project have not been identified, which is completely unprecedented in a DOE effort!

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 1.2 based on accomplishments.

- Qualitative descriptions of challenges and solutions did not provide an adequate explanation of technical accomplishments. More details and data need to be shared in order to prove that the project has made any measurable progress.
- No data have been provided by the team to conduct fair analysis and review of their technical accomplishments. Increasing the synthetic capability of the conductivity enhancer from a one gram (g) batch to a 25 g batch and water retaining additive from two g to eight g is not a huge increase. A 25-g batch size is too small to claim any commercial feasibility of the material. It's understandable that the structure of these additive materials is company confidential; however, the team should at least tell the nature of these materials. From the presentation, it's not known whether these materials are compatible with each other or not. The claims of technical accomplishments in slides 11-17 don't include any data. It's very hard to judge these accomplishments without seeing any data. The cost figures claimed by the team don't have any baseline membrane material against which they have claimed improvements. The team needs to provide more explicit data for the reviewers to understand their progress.
- One really cannot assess this. We are given isolated data points with no graphical presentations, which would give us some ability to assess the quality of the data. The presenter is hiding his data. There has to be more data that can be provided that will not give away intellectual property (IP).
- No data was provided, so it is not possible to determine what progress has been made. More information needs to be provided.
- FCE already had a membrane capable of meeting conductivity targets at last year's AMR. This year, they should have reported MEA testing results. Although indicating that membrane samples have been supplied to the University of Central Florida (UCF) with "promising" initial results, they provided no data and no quantitative information. Shockingly, this is the only 2010 milestone marked as completed. The other three milestones are in progress or are planned. Assessing progress toward milestones is difficult when specific dates are not provided for each milestone, but still, the fact that only one out of four has been achieved by this point in the year (and the assertion that the one that is complete is questionable) is disappointing. FCE does provide various assertions about progress in improving additive retention, lowering additive cost, etc., but evaluation of these assertions is complicated by the fact that FCE has chosen to withhold all data and nearly all technical information from their review presentation.
- Except for the "Project Summary Table" slide, no data were provided.
- The fact that a membrane was fabricated that achieved lower temperature targets was already known from prior years.
- The achievement of a range of 86-148 S/cm at 120°C with 50% RH is not impressive for the following reasons: 1) the target should actually identify 25% RH as the humidity reference in order to avoid water partial pressures over 50 kPa (100 kPa of water is not feasible and pinches oxygen partial pressure), and 2) the composition of the membrane, as well as prospects for expected durability, including consideration of swelling or leaching, is completely unknown. This is unacceptable for a membrane project in this late stage.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **1.8** for technology transfer and collaboration.

- FCE seems to be collaborating with a number of other companies and institutions.
- The partners in this project seem to be nameless. It's hard to assess the capability of the partnering entities without their names. The team needs to provide the names of their partners.
- This is very difficult to assess. It seems they may be keeping some collaborators secret.
- Their collaborators are not identified, and it is unclear how many collaborators there are and what roles they play. More information needs to be provided.
- The fact that FCE is collaborating with a polymer partner and with additive partners, and the fact that changes in the ionomer and in the additive synthesis have been made, suggests that some reasonable level of collaboration is occurring.
- Collaboration can usually be measured based upon the reputation of the collaborators, the work plan to involve them (delineated by tasks), and the results that speak to the collaboration. Here, the collaborators are not named, and their efforts are addressed in abstractions that reference unidentified baselines; e.g., "15x lower material cost" or "6x shorter processing time").
- The collaborators are unknown, and, save for a few hints, their efforts are unknown. There is no way to objectively rate collaboration.

Question 5: Approach to and relevance of proposed future research

This project was rated 1.8 for proposed future work.

- More quantitative data and technical detail is required to evaluate the quality of the proposed future work.
- The future work, as explained by the team, seems to be aligned with their present status of their work. However, due to the lack of data, it's hard to know their present status and correlate it to their future work. The only option that we have, as reviewers, is to believe the claims of the team and reply on their judgment. The team should provide more data to help the reviewers.
- This entire section was too vague.
- The proposed future work is reasonable, although inclusion of more specifics and a clear timeline would have been nice. The existence of alternative development pathways is unclear.
- Any evaluation of future work is meaningless without data pertaining to material composition, swelling, leaching, chemical durability (such as open circuit voltage (OCV) testing), mechanical durability (such as RH cycling), or gas crossover.
- No cost estimates or cost study has been shown for the materials studied here, which means that the worthiness of any milestone associated with selecting a "low cost" membrane design is impossible to judge.
- No data were provided on the 1,000-hour stability test. The test is not identified to show whether it is relevant to targets. DOE-prescribed tests for membranes do not call for a "1,000 hour" metric (OCV test has specified 200 hours or, more recently, 500 hours; the RH cycling test has specified 20,000 cycles).

Strengths and weaknesses

Strengths

- They have a good concept for a multicomponent membrane. Unlike other researchers, who try to develop one material with multiple problem solving features, this team has made a good choice of using different components to address different issues and challenges in the membrane.
- The overall approach is reasonable and FCE seems to be getting good materials from their suppliers. The project has succeeded in producing materials with good conductivity under hot and dry conditions, although most of these accomplishments were realized prior to the current year, and significant progress in the current year has not been conclusively demonstrated.
- In general, the project follows a similar strategy to what would be expected—the development of a high conductivity, water-insoluble ionomer with reinforcement to ensure mechanical durability, and with additives, to enhance proton conduction.
- Conductivity measurements from the project are high relative to other projects.

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Weaknesses

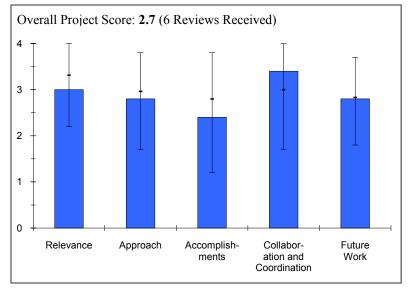
- The qualitative descriptions of challenges and solutions did not provide an adequate explanation of technical accomplishments. More technical details and quantitative data need to be shared in order to prove that the project has made any measurable progress.
- Except for claims of accomplishments, no data has been provided by the team for review. It's absolutely impossible to conduct an outside review without seeing the data. The team needs to disclose more data to help reviewers understand the synergy between these multiple components and to understand the feasibility of maintaining such synergy under low RH FC operational conditions. From the information provided in this presentation, an independent review cannot be done for this project.
- The lack of information on the materials and processes, and the inadequacy of the data, raises large suspicions about the quality of this project.
- The lack of data and any information on the systems being investigated make it impossible to judge the work.
- The value of this project to taxpayers is questionable. FCE has, to date, provided virtually no data and no technical information to the public. Without existence of any public information from this project, should FCE choose not to pursue commercialization of materials developed under this contract, the public will have gained absolutely nothing from the investment of R&D dollars into FCE.
- The complete lack of compositional or processing information leaves the project impossible to judge.
- The cost analysis is missing.
- The lack of compositional information completely removes any possibility for speculation on durability, gas crossover, swelling, and other characteristics that are crucial to understand at this late stage of the project.
- The collaborators have not been identified.
- Durability has not yet been quantified, nor does the future work contain a reasonable plan for doing so.

- More technical details and quantitative data need to be shared in order to prove that the project has made any measurable progress.
- You should be able to share more of your results while maintaining your trade secrets or IP. This project needs to at least reveal some better experimental data to provide some credibility.
- The entire project should be deleted unless FCE shows a willingness to make knowledge gained from this taxpayer-funded project available to the taxpayers.
- The time has come for this project to disclose the chemistry and processing involved with the membrane samples.
- The project needs to show data immediately. Without data, it is impossible to know which efforts need to be pursued more vigorously. Instead, we are forced to take the project at its word when claims are made that molecular weight is being increased to enhance mechanical properties, or that the removal of a hot acid step lowers cost. If only qualitative statements such as these are made, then the statements become meaningless.

Project # FC-41: Novel Approach to Advanced Direct Methanol Fuel Cell Anode Catalysts *Huven 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 DOE's 2010 targets for consumer electronics application. The project goal is to improve the catalytic activity and durability of the platinum ruthenium for the methanol oxidation reaction via optimized catalyst support interactions. A similar approach for oxygen reduction reaction catalysis is advantageous for both DMFC and hydrogen fuel cells (FCs).



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.0** for its relevance to DOE objectives.

- The project demonstrates relevance to the need for moved anode catalysts for DMFC systems. The project objectives are a fit with the need for improved performance and durability supporting the overall goals. While the project is still in the early stages, and it has not moved to more practical catalyst supports, there was no connection drawn to the expected cost of the catalyst compared to the potential benefits over the catalyst's expected life.
- Direct methanol anode catalysis is very important for methanol oxidation. This project perfectly fits the research need. The oriented pyrolytic graphite substrate could increase the catalyst stability under DMFC conditions.
- The project objective is to develop and demonstrate DMFC anode catalyst systems that meet or exceed DOE's 2010 targets for consumer electronics application by improving the catalytic activity and durability of the platinum ruthenium alloy for the methanol oxidation reaction (MOR) via optimized catalyst or support interactions.
- Methanol represents a liquid fuel that represents stored hydrogen. If methanol could be stably oxidized at an anode at high currents, it would be a great advance in FC technology for power sources, and, to date, this feat has not been possible. The failure mechanism of methanol platinum/ruthenium oxidation catalysis is not understood or even known. This project addresses stabilizing the dispersion and chemical composition of the platinum/ruthenium catalyst on a support to attempt to stabilize catalysis of methanol oxidation, and therefore this project is of good merit.
- The assumption is that platinum ruthenium is a suitable and chemically stable catalyst and just needs to be stabilized. This assumption may or may not be true.
- This work should make an effort to analyze stability of the dispersion by microscopy and should analyze catalyst surface composition; e.g., by X-ray photon spectroscopy (XPS) or atomic emissions spectroscopy (AES). In this way the contribution to MOR catalysis of these two effects: 1) dispersion of platinum/ruthenium, and 2) chemical surface composition of platinum/ruthenium catalysts, should be individually trackable. The relevance of this project lies in determining whether catalyst support interactions will improve 1 or 2 or both, and whether MOR activity is stabilized and/or improved.
- The intended relevance is the improvement of durability and the lowering of cost for DMFC for small portable power applications, such as consumer electronics. Development of FCs for consumer electronics is one of the DOE-EERE Fuel Cell Technologies Program (FCT) objectives, as stated in the Multi-Year RD&D plan. However, as described below, it is not clear how the project's approach will address the most prevalent DMFC catalyst degradation mechanism.

• The development of methanol oxidation catalysts with improved activity represents the greatest challenge of DMFC technology. Consequently, this project is relevant to DOE objectives. The advantage of the approach taken is still to be demonstrated, though.

Question 2: Approach to performing the research and development

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

- The initial approach of examining a model catalyst support to examine the benefits of the catalyst fabrication process is good, but the program needs to move to high surface area catalyst supports and membrane electrode assembly (MEA) measurements to find the real benefit, as well as to determine the catalyst performance under real operating conditions. The durability of the catalyst needs to be determined in an MEA to examine if there are any benefits to this approach, particularly with ruthenium dissolution.
- The ion implantation method for the platinum ruthenium catalyst deposition is a good method for fundamental research to investigate the platinum ruthenium catalyst arrangement on the substrate. Nitrogen doped highly ordered pyrolytic graphite (HOPG) shows good performance as a support for platinum ruthenium. I believe the nitrogen replaced the oxygen on the graphite surface.
- The graphite surface contains dangling carbon-oxygen bonds. Nitrogen treatment could be effective even without ion implantation.
- The main problem for graphite support is that the graphite surface is very smooth (thinking as a good lubricate). How the platinum ruthenium catalyst stays on the surface for long term tests is a good question. Graphite surface area is quite low (even if roughing the surface), and the orientation is fixed after the catalyst deposited on electrolyte membrane or gas diffusion layer (GDL). Therefore, in the real FC, the catalyst utilization could be reduced.
- The project is to use ion implantation to stabilize catalyst dispersion (and possibly composition) -- a good first step in a rational attempt to stabilize methanol oxidation catalysis.
- It is a good idea to work on implantation to stabilize a catalyst on a well-defined surface like HOPG, because it will help understand the effects of implantation of the support on catalyst stability. Other low surface area carbon supports should be considered, like glassy carbon and edge plane of HOPG. Possibly molybdenum nitride might be considered, since it spontaneously attracts platinum, so platinum/ruthenium might benefit too. Ultimately translating the low surface area supports to high surface area supports (like nanotubes) is needed for making a practical catalyst, but this translation should be delayed until the effects of implantation on catalyst stability and activity can be understood on a low surface area support.
- The approach to lowering the cost of DMFC anodes is to improve the dispersion of platinum ruthenium on the carbon support and to maintain that dispersion over the life of the device by enhancing the interaction of the catalyst particle with the support through doping of the carbon support with other elements. However, the major degradation mechanism of DMFC anode catalysts is the loss of ruthenium from the alloy. The presentation did not acknowledge this degradation mechanism, nor did the presenter give a clear scientific rationale that doping the support would mitigate this loss mechanism.
- While interesting from the catalyst design point of view (anchoring of catalyst particles to carbon support via nitrogen), the proposed approach does not offer a clear path toward improvement in MOR electrocatalysis.
- To date there has been no evidence of improvement to catalysis of methanol oxidation with this approach. Voltammetric data attest to some possible mass-transport benefits only.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.4 based on accomplishments.

- The team has made good progress with the fundamental catalyst studies and in preparing to make catalysts on high surface area supports for the MEA studies.
- Graphite particles have different shapes and structures. So far, almost a hundred graphite structures have been reported, for example, spherical graphite, planar graphite, horn graphite, worm graphite, etc. Apparently, the research group may not know enough about graphite. Naturally boron graphite and nitrogen graphite are available. I suggest the team consult or work with other researchers to find the best graphite for ion implantation (may not be necessary to use ion implantation).

- The work plan began July 2009 (not much time to report a lot of results), and is well on its way. Preliminary results on HOPG (with a 52x gain in activity) are impressive.
- The material and equipment have been set up to learn if the approach will work. It remains to be seen if they will execute the plan methodically.
- The optimal implantation process has been determined to be a 45-second exposure, and implanted surfaces have been spectroscopically analyzed to show implanted moieties.
- Nitrogen doping was found to be the best doping to date in regards to promoting MOR catalysis.
- Three catalyst deposition methods have been found.
- High surface area catalyst making was shown (but I feel no more work should be done on this until results with low surface area supports find the best treatments, assuming this low surface area work will translate into high surface area catalysts).
- An array cell has been set up.
- Excellent progress toward the stated objectives of the project have been made over the short duration of the project. However, as mentioned in the approach section, it is not clear that the existing scope and approach of the project will overcome the most significant electrocatalytic barriers toward implementation of DMFCs in portable electronics. It is also not clear if the improvements in MOR activity seen with the electrode deposited platinum ruthenium on doped HOPG versus HOPG are due to the effect of the support or to different platinum ruthenium catalyst composition; i.e., X-ray diffraction (XRD) and/or elemental analysis was not presented. The stated purpose of the HOPG studies is for a "better understanding" of the catalyst-support interactions; however, understanding obtained from the HOPG results to date was not clearly stated.
- Nearly halfway through its duration, this project offers little performance data and no proof of the validity of the approach taken. Progress should be accelerated involving, among others, fundamental kinetic analysis under rigorous conditions.
- Data preceding the project inception should not be presented as an accomplishment (Raman spectra in slide 9 were already published in J. Mater. Chem, 2009, 19, 7830, before this project had even begun.).

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.4** for technology transfer and collaboration.

- There are good collaborations on the program for the testing of the catalyst. The program is quite short and the team demonstrates good coordination.
- The partners include Mechanical Technology, Inc. (MTI) and BASF Fuel Cell, which are good for the catalyst scale up.
- The collaborators (outlined below) have great capabilities and that should greatly support the execution and validation and implementation of the program:
 - Develop novel catalyst-doped supports (the National Renewable Energy Laboratory (NREL) and Colorado School of Mines (CSM)).
 - Combinatorial electrode studies (the Jet Propulsion Laboratory (JPL) and NREL).
 - Generate downselected novel catalysts for DMFC MEA (NREL, CSM, and BASF).
 - MEA evaluation (NREL,CSM, and MTI).
- This teaming is impressive.
- The PI has assembled a very good team covering all aspects of the project's approach. The project is new, but has already demonstrated significant collaboration between CSM, JPL, and NREL.
- The team represents a good mix of two national laboratories, a university, and two FC developers.
- The role of MTI is unclear.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.8 for proposed future work.

• The continued combinatorial testing of the catalyst to optimize the performance is a good approach. It is important to move to a high surface area carbon support and to test in an MEA to determine to performance in

an MEA and the durability under practical DMFC conditions, including both on time, start up and shutdown, as well as in the off state.

- As said above, the graphite has too many structures. Without selecting the best graphite, the project may go nowhere.
- There is a plan to extend the work to high surface area supports. Carbon nanotubes seem a logical choice as a well-defined surface for modification by implanting stabilized catalysts for making a high power methanol oxidation anode. The following three tasks seem to be most important:
 - 1) Initiate implantation of other dopants into HOPG (boron, sulfur, iodine).
 - 2) Continue combinatorial electrochemical investigation of various implanted HOPG substrates (type and extent of dopant) (JPL).
 - 3) Investigate different methods to dope carbon (in situ and ex situ) ion implantation, chemical vapor deposition, and pyrolysis.
- I think work on low surface area supports will give the most information at this stage. Alternative carbon supports are important and trying to stabilize platinum/ruthenium on inorganic supports (like molybdenum nitride supports) may be a good idea.
- The immediate proposed future work of characterizing the MOR of platinum ruthenium on doped high surface area carbon is good. The majority of the future work should focus on the high surface area carbon support, rather than HOPG, and on a scalable and cost effective catalyst deposition method.
- Proposed future research lacks a kinetic component that would help validate the approach used.
- By this time, HOPG should have been eliminated and replaced by viable carbons.

Strengths and weaknesses

Strengths

- NREL has a good sound approach to examining the approach for attaching the catalyst and determining the inherent performance of the catalyst. The project has moved quickly toward preparing to make high surface area catalysts, which are important to establishing the real benefits of the catalysis approach in terms of performance, potential costs, and durability.
- Graphite has better corrosion resistance, which is needed to improve catalyst lifetime. However, the researchers need to find a good graphite.
- Team skills and resources are the great strengths.
- NREL assembled a good team with a good approach to anchoring platinum ruthenium on support, providing better dispersions and preventing catalyst particle agglomeration.

Weaknesses

- The project needs to move toward determining the ability to produce high surface area catalysts, to demonstrate the indicated performance benefits that are realized in the MEA at acceptable loadings. The NREL team should determine the durability of the catalyst compared to the commercially available catalyst under the same MEA conditions, including a range of operating conditions including start up, shutdown and off state time (thermal cycling, dry out, etc.).
- Graphite has low surface area and is well orientated, which could reduce the catalyst utilization in a real FC. Also, the binding force between platinum ruthenium with graphite is lower than platinum ruthenium on high surface carbon. Therefore, the platinum ruthenium may not stay on graphite long enough under FC operation.
- Methanol electro-oxidation catalysis failure mechanism is not known, which jeopardizes the project unless the team can clearly learn this mechanism.
- There is a lack of convincing rationale that doping the carbon support will address one of the major degradation mechanisms for platinum ruthenium anode catalysts, which is ruthenium leaching from the alloy.
- The electrochemistry part is weak; and the kinetic analysis is non-existent.
- Electrocatalytic benefits of the approach have not been shown yet.

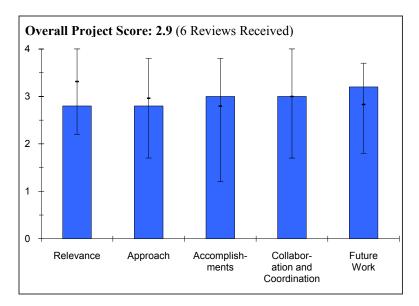
- I recommend increased testing in MEA and if possible testing short stacks to determine the durability under a range of real operating conditions experienced in the predicted devices.
- Select the best graphite to do the later work.

- I recommend work on a catalyst stabilized by ion implantation on low surface area supports, with careful characterization of the catalysis of methanol oxidation using the materials made in this project. The great benefit of this project would be to find the first stable and possibly high activity catalyst for methanol electro-oxidation.
- Add a project task aimed at demonstrating that the alloy composition of platinum ruthenium anchored to doped carbon supports is more stable than the same platinum ruthenium alloy with the same particle size anchored to a support that has not been doped. Down selection for one catalyst deposition process should be made halfway through the project duration, with criteria including scalability and cost of production of the catalyst.
- The validity of the approach has to be demonstrated in an FC as soon as possible using state-of-the-art techniques in MOR electrocatalysis as a reference.
- Catalyst performance should be optimized using practical carbon supports, not HOPG.
- Kinetic analysis of MOR on different catalysts is required. Cyclic voltammetry can be used for general screening of electrocatalytic properties, but steady state techniques should be used for kinetic analysis.

Project # FC-42: Advanced Materials for RSOFC Dual 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) 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-class RSOFC stack demonstration, a) RSOFC dual mode operation of 1,500 hours with more than 10 SOFC/solid oxide electrolyzer cell (SOEC) transitions, b) operating current density of more than 300 mA/cm² in both SOFC and SOEC modes, and c) an overall decay rate of less than 4% per 1,000 hours of operation.



<u>**Ouestion 1: Relevance to overall DOE**</u> <u>objectives</u>

This project earned a score of 2.8 for its relevance to DOE objectives.

- This project exactly fits DOE's objectives for residential hydrogen production and power generation.
- Reversible fuel cells may be part of DOE's new portfolio, but the performance targets proposed for this project are so low that its relevance is questionable.
- This is the most relevant project to DOE as it addresses renewable energy storage with mindfulness of high capital utilization.
- I don't see where this fits under the Multi-Year Program Plan (MYPP) currently on the EERE website (updated April 2009).
- RSOFC can integrate renewable production of electricity and hydrogen when power generation and steam electrolysis are coupled in a system, which can turn intermittent solar and wind energy into "firm power."

Question 2: Approach to performing the research and development

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

- Reversible proton exchange membrane fuel cells (PEMFCs) have been well developed. Reversible SOFC has been demonstrated in single cells.
- This project focuses on performance and lifetime, which are the main hurdles for SOFC commercialization.
- Leveraging Solid State Energy Conversion Alliance (SECA) technology is a good start.
- The technical targets are too low. A reversible SOFC should have a lifetime target of five years minimum and ten years preferred, with an end-of-life performance of, at worst, 80% of the original. Therefore, decay should be approximately 0.5% per 1,000 hours.
- They propose 1,500 hours of operation with ten SOFC/SOEC transitions and to use this in energy storage. For energy storage they should be transitioning between SOFC/SOEC at least every 24 hours.
- They did not look at turndown performance. For integration with renewable sources, turn down will be important.
- Energy storage systems using hydrogen show that one would have an electrolyzer to make hydrogen when electricity is cheap and have a fuel cell (FC) or turbine to make electricity when it's expensive. In this paradigm, an energy storage facility would have to purchase two pieces of capital, and use each of them at a fraction of the time. However, in Versa Power Systems' (VPS's) technology, one would purchase a single piece

of hardware and perform both functions with it, thus utilizing capital to a much larger extent. Capital utilization is one of the most critical aspects to favorable economic performance.

- The project is appropriately focused on degradation mechanisms, with quantitative targets. The experimental approach is sound, building on SOFC-mode progress to date. The team utilizes intra-cell voltage measurements and transmission electron microscopy (TEM) to good effect. They have solid testing protocols.
- Developing high performance and low degradation RSOFC cell and stack technology is critical for the reversible SOFC/SOEC system. This is the focus of the work.
- The approach is excellent and includes building on VPS's strong SOFC cell and stack baseline, leveraging cell and stack advancements from the DOE-SECA-SOFC project, addressing RSOFC degradation mechanisms in SOEC mode with innovative cell and stack repeat unit configurations, conducting parallel materials development activities and integrating them with cell production technology development, and completing RSOFC stack and process designs to address durability, performance, and cost in both SOFC and SOEC operating modes.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.0** based on accomplishments.

- Technical progress as shown is the SOFC test under electrolysis mode. All of those tests showed the degradation, even under steady state conditions. No thermo-cycles were reported. Thermo-cycle is the main challenge for the durability. Without solving the fundamental thermo-cycle durability problem, the scale-up may not be useful.
- Their performance is an improvement over Idaho National Laboratory's (INL's) high temperature electrolysis work.
- The long-term testing showed a degradation rate of 3.8% over 1,000 hours, meeting their milestones, but that degradation rate is still >7 times too high for a commercial system.
- There was no discussion on degradation mechanisms. From the discussion they have some studies in this. It would be nice if they presented the details.
- There was no discussion on materials development.
- The number of cycles is too low.
- They need to report their round-trip efficiency.
- They need to look at turndown operation for both the SOFC and SOEC.
- Outstanding development of better materials to provide higher performance and lower degradation rates.
- The project is solidly ahead of schedule, in part due to the initial lack of SOEC mode data upon which targets and schedule would be established. Nevertheless, combined degradation, SOEC-SOFC transitions, and operating time demonstrate significant progress.
- Two types of RSOFC cells developed have met the electrochemical performance target, and RSOFC; four met both performance and degradation criteria.
- A steady-state single cell test has run in electrolysis for one year with a degradation rate of less than 3% over 1,000 hours.
- A baseline 28-cell stack (kW-class) test has run in electrolysis for over 2,500 hours at 3.8% over a 1,000-hour degradation rate.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.0** for technology transfer and collaboration.

- Their partners include Boeing, SECA, and INL. They did not show what Boeing did or what has been done by SECA.
- They have a large number of "collaborators," but it seems that most of the collaboration is VPS applying technology developed on other projects, such as SECA's and the Defense Advanced Research Projects Agency's (DARPA's) work to this one.
- The company has communicated well with its customer base to drive toward a product set that would land them in a very useful market space.

- This project builds upon established SECA technology and concurrent Boeing RSOFC work. VPS has excellent in-house personnel and equipment for the proposed work.
- Great leveraging: Boeing: collaborated on and funded the initial RSOFC development work through both Boeing and DARPA funded efforts, and anticipated follow-on a DARPA award this calendar year. SECA: as subcontractor to FuelCell Energy in SECA, VPS has advanced SOFC cell and stack technology which has been applied in this program. INL will integrate SOEC technology for hydrogen production with Next Generation High Temperature Nuclear Reactor, and demonstrate suitability of VPS SOEC technology for this application at the kW-class stack level.

<u>Question 5: Approach to and relevance of proposed future research</u>

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

- The proposed work only shows the electrochemical degradation tests without thermo-cycle tests. If the RSOFC failed on several thermo-cycles, the electrochemical degradation tests might not need to occur.
- The future work does address the core issues of degradation.
- Good as proposed—see comments under scope additions below.
- For fiscal year (FY) 2010: This is an excellent plan building on progress and accelerating RSOFC stack development: Complete degradation mechanism study. Conduct single cell tests at various operating conditions (temperature, current, steam utilization). Conduct post-test analysis with detailed microscopic analysis; i.e., TEM, scanning electron microscopy (SEM), and energy dispersive X-ray spectroscopy (EDX). Complete test facility improvements. A potential additional scope could be conducting additional stack testing early in the project.
- For FY 2011: Complete go/no-go decision point test. Complete cell and interconnect materials development. Down-select material systems for RSOFC stack development. Complete the final project metric test with kWclass RSOFC stack.

Strengths and weaknesses

Strengths

- The high-temperature electrolyzer may have better electrochemical efficiency.
- A reversible SOFC for energy storage has the potential for high capital utilization.
- Solid technology that demonstrated progress.

Weaknesses

- Thermo-cycle needs to be addressed.
- The targets are too low and need to be increased as discussed above.
- INL has a great deal of understanding on degradation mechanisms for SOEC. VPS is not using that knowledge.
- Work such as this should always reference a systems study or analysis to show the reason for pursuing the work. Yes, one can certainly envision an energy storage system based upon RSOFC; however, a cost entitlement analysis relative to existing and advanced concepts would establish a basis for the work. Cost is always a consideration.

- Test the thermo-cycle before the electrochemical cycle.
- They need to make the degradation target at most 0.5% per 1,000 hours.
- They need to do real cycling experiments.
- They should define what the roles of the participants are and clearly state what each participant is doing.
- They need to discuss materials development.
- They need to include turndown experiments.

Project # FC-43: Resonance-Stabilized Anion Exchange Polymer Electrolytes

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3

2

Yu Seung Kim: Los Alamos National Laboratory

Brief Summary of Project

The objectives of this project are to: 1) develop anion exchange polymer electrolytes that have high hydroxyl conductivity and stability in high pH conditions, and 2) demonstrate an improved single cell performance of solid-state alkaline fuel cells using the polymer electrolytes and non-precious metal catalyst.

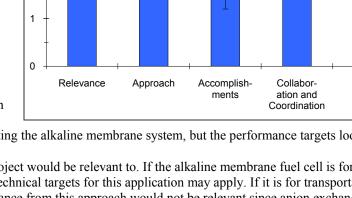
Question 1: Relevance to overall DOE objectives

This project earned a score of 2.9 for its relevance to DOE objectives.

- Work It is not clear how this project fits with Coordination the overall DOE objectives for the Hydrogen Program. The PIs are targeting the alkaline membrane system, but the performance targets look too low.
- It was unclear what application the project would be relevant to. If the alkaline membrane fuel cell is for consumer electronics, then the DOE technical targets for this application may apply. If it is for transportation application, then the fuel cell performance from this approach would not be relevant since anion exchange polymer electrolytes cannot achieve the technical targets. I think that it would be difficult for this approach to achieve even the consumer electronics technical targets.
- The alkaline membrane conductivity, stability, and electrode processing issues are worth studying.
- The conductivity technical target set for the project does not seem significant since it is set at 80°C and the recently reported conductivity values are for 20° and 50°C. It is expected that higher conductivity would be achieved at a higher temperature.
- Alkaline-membrane fuel cells have several advantages, but also several challenges. They could reduce fuel cell • cost and enhance performance.
- Finding electrolytes that allow the use of active inexpensive materials for hydrogen and air fuel cells is ٠ important to the practical use of fuel cells.
- PIs will develop anion exchange polymer electrolytes that have high hydroxyl conductivity and stability in high • pH conditions.
- PIs will demonstrate improved single cell performance of solid-state alkaline fuel cells using polymer electrolytes and non-precious metal catalysts.
- This project is an innovative way to find if stable alkaline membranes can be obtained to achieve these goals, namely, > 50 mS/cm at 80°C, and > 500 h at 80°C in 1 M NaOH solution at $> 200 \text{ mW/cm}^2 \text{ H}_2/\text{air}$ at 80°C.
- I think characterizing membranes in 1 M aqueous KOH is a good first start, but the effects of water activity should be addressed. It may not be important to do address water activity at first, but when a good electrolyte system is found in 1 M aqueous potassium hydroxide, it is relevant to practical systems to scrutinize the effects of decreasing water content and fuel cell-gas humidification.
- This project is for an alkaline fuel cell, which is a peripheral part of the Fuel Cell Technologies Program.
- Alkaline membrane approach may allow for the use of non-precious metal catalysts.
- Supports targets proposed to DOE are: conductivity at > 50 mS/cm at 80°C, stability at >500 hours at 80°C in 1 M NaOH solution, and electrode processing at $> 200 \text{ mW/cm}^2 \text{ H}_2/\text{air}$ at 80°C.

Question 2: Approach to performing the research and development

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



Future

Overall Project Score: 2.6 (6 Reviews Received)

- The in-cell performance targets seem too low. It is good that the team is working on the key ionomer for the electrode to provide sufficient oxygen access and stability.
- The project is interesting and addresses several issues that are relevant for alkaline membrane fuel cells.
- It seems that many new tasks need to be completed within a limited time and with limited funding. The team is trying to develop an anion exchange membrane with higher conductivity and stability, non-precious catalysts for the electrode, optimize the electrode with new catalyst, membrane and ionomer, and fabricate membrane electrode assemblies (MEAs) for fuel cell testing. These are not trivial tasks by themselves. If successful, it could have some impact and have demonstrated some novel concepts. The resources may be spread too thin to achieve significant progress in all areas.
- It is also unclear what percentage of the effort and in what time line will be dedicated to membrane development, non-precious catalyst development, electrode optimization and MEA fabrication. This information would be helpful to determine the degree of difficulty of each task and whether appropriate effort is dedicated to the task.
- The project targets stability, one of the main challenges for the alkaline membrane fuel cell.
- A major degradation route of anion exchange polymer electrolytes is nucleophilic substitution (SN2).
- The project's approach is to make resonance stabilized guanidine functional group in a polymer. This approach should reveal if SN2 is a major degradation route for anion exchange polymer electrolytes and if the resonance structure of guanidine base enhances the cation stability and hydroxyl conductivity or not.
- The approach is very interesting. It is focusing on low cost materials that would enable solid state alkalines. However, most materials in existing alkaline fuel cells are already cheap. The big problem is CO₂.
- Next to reacting with the alkali, alkaline fuel cells historically are lower in power than polymer electrolyte membrane fuel cells (PEMFCs). Their approach may improve the power.
- One of the objectives is to develop a non-precious metal catalyst.
- Approach is very sound and has a contingency plan.
- The two approaches, stability and electrode performance, are correctly identified.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.4 based on accomplishments.

- The project does not appear to be making much progress on obtaining a viable ionomer for the electrode that provides sufficient stability. The performance targets for the stated vehicle application look too low even when taking into account the potential lower catalyst costs that can be realized from the use of non-precious metal catalysts.
- It is a new project, so limited but good progress has been achieved with membrane development.
- Apparently the membrane is not stable even under hot water. Did not see the data from leachants. The proposed degradation mechanism is valid; however, if the electron withdrawing decreases (hydrocarbon guanidine base), it might change the pH of the membrane or reduce the OH⁻ conductivity.
- This is a relatively new project (September 2009 start).
- Amine and guanidine polymer materials have been made.
- Methods set up to track membrane composition and performance to validate if S_N2 is a major degradation route for anion exchange polymer electrolytes and if resonance structure of guanidine base enhances the cation stability and hydroxyl conductivity or not.
- Finding the effects of electron density on the stability is impressive and should be followed up in guiding new materials.
- The improvement in conductivity is a good step.
- The new membranes show promise, but it would be good to see more conductivity and operation data.
- The stability was disappointing, but the PI has ideas on how to improve it.
- One of the objectives is the development of non-precious metal catalysts, but this topic was not discussed in the presentation.
- Significant progress has been made.
- The project started in September 2009. There have been few accomplishments. Conductivity of the perfluorinated guanidine base in boiling water was disappointing.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.4** for technology transfer and collaboration.

- There are limited significant ongoing collaborations. There are a large number of stated occasional collaborations. It was unclear if these were well coordinated to help to address the issues. Increased collaboration is needed to develop the electrodes, improve performance, and ensure that the goals are targeted at the DOE Hydrogen Program objectives for performance and cost.
- The collaboration and partners seem appropriate.
- A very strong team.
- The team is skilled in all aspects of the project and should have all bases covered to do the work.
- There are a lot of partners that are contributing to the work.
- Excellent collaborations.
- Outstanding.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.9 for proposed future work.

- It is important to address the durability and performance of the ionomer if this system is to be practical. More effort may be needed to develop the ionomer and the catalyst to optimize the performance and potentially address the cost and performance objectives for the intended hydrogen / air application.
- It was unclear what non-precious metal catalyst will be studied. What if no good performing non-precious metal is found via high throughput electrochemical screening? Will a Pt catalyst be used instead to achieve the targeted performance?
- The backup plan to use nanostructured thin film (NSTF) materials in electrodes if an appropriate ionomer is not found may or may not be appropriate. The idea is that NSTF materials do not need an ionomer in the electrode. However, the non-precious metal catalysts that will be sputtered onto NSTF materials have to be good ion conductors like Pt.
- There was no time line or decision points specified.
- Before solving the stability issues, MEA fabrication may not be necessary. Also, membrane stability should be tested under hydrogen and oxygen saturated water at elevated temperatures.
- The work is based on prior knowledge of past alkaline membranes and has a hypothesis to improve stability (that is, cation stability based on electron donating spacer), which has long eluded workers trying to make stable anion exchange membranes.
- The future work focuses on the appropriate areas.
- Since so much development needs to occur on increasing the membrane stability, testing in an FC would be premature at this time. But it needs to be done eventually.
- Very good future work plan.
- Focus is polymer chemistry. Suitable plan.

Strengths and weaknesses

Strengths

- Good polymer synthesis expertise. Targets are for a good, potential technology that may address catalyst cost issues.
- The issues being pursued are worth addressing and the general approach is fine.
- The membrane shows CO₂ tolerance with promising cation conductivity. The alkaline fuel cell will drastically reduce the precious metal usage in the fuel cells.
- A great strength is that the team is quite knowledgeable on the subjects of alkaline membranes and technology and methods to characterize materials and material performance.
- Strong team.
- Innovative idea.

- A lot of work can be done on improving alkaline fuel cells. Compared to PEMFC R&D, there is not a lot of work being done in this area.
- Has the potential for alkaline fuel cells that do not use precious metal catalysts.
- Interesting project in polymer chemistry. Excellent technical work just beginning.

Weaknesses

- Projected performance goals at the MEA-level seem too low for the intended vehicle application. Given the lower performance targets, the project does not provide any information about the potential cost improvements that the membrane will provide in terms of lower catalyst costs for the fuel cell system.
- The project tries to tackle too many difficult issues with limited resources to achieve them.
- Relevance is not clear.
- The stability is not very good.
- Alkaline catalysis is well known to give fairly stable high power to fuel cells with a hydrogen anode and an oxygen cathode. However, alkaline fuel cells are not well understood with cathodes running on air with carbon dioxide and other impurities.
- It needs to be appreciated that catalysis work to date is really short term, and electrolyte and catalyst stability in air must be questioned.
- The alkaline fuel cell is CO₂-sensitive. The PIs may develop the best membrane and great non-precious metal catalysts, but unless they address the CO₂ issue, the commercial viability of this work will be limited.
- There is a significant probability that anionic conductivities and stabilities in alkaline solutions of the membranes will not be suitable for applications.
- Should use non-precious metals from the beginning.

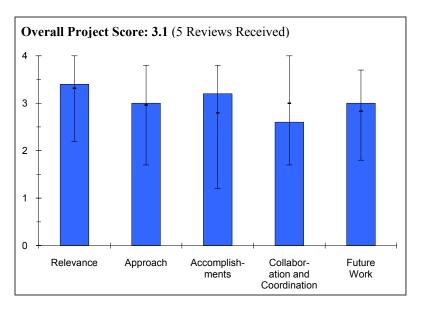
- Increase MEA testing and the involvement of collaborators to ensure the cost and performance targets are aligned with the intended application.
- Focus on 1-2 tasks or issues rather than all of the issues or tasks presented.
- Improve the membrane stability before putting it into a fuel cell.
- The project is just underway. Membranes based on the team's design criteria should be made as soon as possible for testing.
- It may help to try to model studies by making small molecules of a stabilized guanidine and mixing with 1M aqueous KOH base and to characterize performance in a 3-electrode cell as a function of water activity.
- Recommend testing in a fuel cell earlier than what they currently plan. Many materials that look good in characterization tests perform poorly in a fuel cell.
- The PIs need to test with air (including CO₂) in addition to the O₂ testing.

Project # FC-44: Engineered Nano-scale Ceramic Supports for PEM Fuel Cells

Eric L. 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 fuel cell (PEMFC) 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 (MEA) 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) synthesis method/procedure amenable to scale-up, and 7) reasonable synthesis costs.



Question 1: Relevance to overall DOE objectives

This project earned a score of 3.4 for its relevance to DOE objectives.

- This project evaluates alternates to the carbon-supported catalyst types currently in used in PEM electrodes. The evaluations are worthwhile in that corrosion of the cathode supports in a low fuel scenario is a key cell failure mode. A catalyst support that is more corrosion-resistant than the currently used supports, with the same degree of hydrophobicity, should result in a more durable cell.
- This review has adequately covered and summarized the topic.
- Cost and durability are the most critical gaps for fuel cell commercialization, and catalyst durability is one of the most important factors for these critical attributes. Alternative support materials may solve carbon-related materials stability problems.
- Development of high surface area supports alternative to carbon materials is indeed important, as demonstrated clearly by the development of 3M NSTF catalysts. In this project, the PIs have proposed to develop a low cost durable ceramic support that will enhance resistance to corrosion and attenuate Pt agglomeration. Although the development of ceramic supports with such characteristics appear to be feasible, this project needs more convincing data that this type of support will have any benefits to the existing carbon- or 3M-type supports.
- This project is aimed at developing alternative supports for Pt-based catalysts to improve stability of cathode catalysts. While this approach addresses cathode degradation due to support corrosion at high potentials and may also address Pt particle coalescence, it does not address Pt dissolution as was stated in the presentation.
- Supports DOE technical targets.

Question 2: Approach to performing the research and development

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

- The approach appears novel.
- Modeling approach part is not clear. Is this a molecular dynamics to develop a Pourbaix diagram?
- The project is relatively well designed and is based on the required stability and conductivity of supports in PEMFCs. The methods for characterization are also reasonably well chosen, so it appears that it will be possible to establish the important stability and activity properties of such systems. The PIs need to be more specific

about how they, in fact, plan to determine the adsorption sites for Pt particles as well as the methods to follow oxide-induced dissolution of Pt.

- Why do modeling on stability without the presence of catalyst? Catalysts are known to accelerate the processes contributing to support degradation.
- The approach is correct—focus on select support candidate materials using various synthesis methods.

<u>Ouestion 3: Technical accomplishments and progress toward project and DOE goals</u></u>

This project was rated 3.2 based on accomplishments.

- The accomplishments appear to correspond with expectations.
- Current data seem to be promising.
- The project is in its early stages, so it is very difficult to judge real technical accomplishments. Nevertheless, some of the materials synthesized based on Mo-N compounds show promising characteristics, such as exceptional surface area electronic conductivity and stability. In my opinion, this is the better part of the project. However, electrochemical measurements, in particular for the oxygen reduction reaction (ORR), are clearly questionable and need more to be done to make any meaningful conclusions about catalytic activity. Based on polarization curves for the ORR on Pt, it is not clear why the activities are well below the benchmark activities observed for the ORR on Pt catalysts supported on carbon. In addition, for samples prepared at 700°C, a clearly visible peak in the cyclic voltammogram (CV) at ~ 0.4 V may indicate that Mo is still "active," and that this temperature is not the best choice for the sample preparation. The PIs are claiming that they have the evidence for enhanced Pt interaction with Mo₂N support. The 300°C temperature may not be high enough to have any effect on Pt agglomeration; therefore, more careful experiments need to be performed to come to such a conclusion.
- The bulk of data shown was for the Mo₂N system. Good progress has been made in getting this material to a high surface area form. Initial and preliminary data were shown for ORR activities of Pt deposited on high surface area Mo₂N. Though the presentation states that the ORR activity is comparable to commercial (E-Tek) 20 wt.% Pt/C, the raw rotating disk electrode data showed problems with achieving a flat limiting current region over the potential studied. Without a steady-state limiting current, it is impossible to evaluate the ORR kinetic current and thus impossible to make the claim of comparable activity. It is also premature to say that there's evidence for enhanced Pt/support interaction for Mo₂N versus carbon. Also, a better method for determining electronic conductivity of support in powder form should be developed—the two point probe resistance method of the foam is not reliable or quantifiable—and may be skewed by presence of carbon in foam.
- On-schedule to meet all milestones:
 - Synthesis of alternative ceramic supports pushed ahead of schedule.
 - BET analysis of Mo₂N cubic phase indicates exceptional surface area.
 - Pt/Mo₂N CV characterization with 0.5 M solution of H₂SO₄ shows Pt/C-like activity, which is good.
 - Evidence for enhanced platinum interaction with molybdenum nitride support.
- In short, the Mo₂N ceramic support is a strong candidate with high surface area, electronic conductivity, and stability. Evidence for enhanced Pt-support interaction with Pt/Mo₂N, electrochemically active surface area, and demonstrated ORR activities.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **2.6** for technology transfer and collaboration.

- Collaboration appears to be limited to members on the contract. There does not appear to be any commercial involvement to act as a sounding board and provide guidance toward specific applications.
- For the scope of this project, three partner organizations may not be sufficient. The systems chosen to be studied are rather complex and addition of carefully selected theoretical methods may help in the quest to design a new generation of non-carbon supports.
- This project relies mainly on expertise at LANL. The project has a subcontractor with the University of New Mexico, but it was not evident from the presentation that they had done anything yet (problems in establishing subcontract). Once the subcontract is in place, the project would benefit greatly from the XPS characterization capabilities at the University of New Mexico (UNM), which was not included in the description of the tasks.

- Good collaboration.
- Good interaction among UNM, LANL and ORNL.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- The future work appears to be more toward lab techniques for deposition. Is there any thought to scale-up techniques?
- In addition to the current project scope, it would be good to investigate catalyst performance with respect to support materials (interactions) and to understand mechanism of performance enhancement with support materials.
- In general, the proposed future work appears to be reasonable, at least for the synthesize part. I think the project needs much more aggressive electrochemical characterization.
- The proposed future work should include developing a reliable method for quickly screening the electronic conductivities of the newly-developed support materials and for determining their corrosion resistance in the presence of catalyst. The proposed future work should also include an evaluation of the cost effectiveness of the proposed materials and processes for making them. Fast-tracking the go/no-go decision and downselecting classes of materials early in the project is a good idea.
- Future work is logical extension.

Strengths and weaknesses

Strengths

- The strengths are the technical expertise of the researchers.
- The major strength of the project is the unique ability of PIs to synthesize and characterize different ceramic type supports, which might be of potential interest for the Hydrogen Program. The PIs are qualified for this work, but they have to demonstrate that the kind of support they are proposing may eventually compete with the existing carbon supports and especially with 3M NSTF systems.
- The strength of the project is the ability of the LANL team to make high surface area ceramic supports, as one of the major limitations to using the conductive ceramics as supports in catalysis is their low surface area.
- This is important work. Some substrate support development should go in this direction.
- Very promising.

Weaknesses

- The weakness appears to be the lack of commercial guidance on applications.
- Electrochemical characterization is an apparent weak point of this project. The electrochemical results must be more convincing and in line with the state-of-the-art PGM catalysts supported on carbon and/or attached to the 3M supports. Given that this is just a first year of the project, there is a plenty of time to overcome these problems.
- Weaknesses of the project are the lack of experimental evidence behind the claims of improved properties of the ceramic supports versus traditional carbon supports, though it is early in the project. The ultimate proof of the concept is the performance and performance durability in a fuel cell and MEA fabrication and testing is part of the planned near-future activities. However, as LANL knows quite well, it is not straightforward to fabricate high-performing MEAs with a new material set. It would, therefore, speed progress in the downselection of promising materials if quick, reliable, quantifiable, *ex situ* methods were identified for determining important materials properties such as electronic conductivity.
- PI needs to optimize structures at some point.

- In addition to the current project scope, it would be good to investigate catalyst performance with respect to support materials (interactions) and understand mechanism of performance enhancement with support materials.
- Need better method for determining electronic conductivity of support materials. Is carbonaceous material responsible for two-point conductivity observed for nitride support? Add TGA in the presence of catalysts to

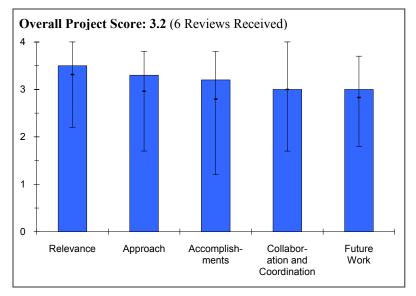
determine mass loss and degradation of catalysts in a humidified air atmosphere. Add an evaluation of cost effectiveness and scalability of materials and materials synthesis techniques. Refine RDE technique / catalyst deposition on disk to obtain data that can be used to evaluate and compare kinetic currents.

Project # FC-45: Effects of Fuel and Air Impurities on PEM Fuel Cell Performance

Fernando Garzon; Los Alamos National Laboratory

Brief Summary of Project

The overall objective of this project is to lower the cost of fuel cell operation by improving performance and increasing lifetime. The objectives are to: 1) understand the effects of fuel cell operation with less than pure fuel and air and simulate "real world" operation, 2) understand how impurities affect DOE fuel cell cost and performance targets, 3) contribute to the scientific understanding of impurity fuel cell component interactions and performance inhibition mechanisms, 4) develop science-based models of impurity effects upon fuel cell performance, 5) perform experimental validation of models, and 6) develop mitigation strategies and methods.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.5** for its relevance to DOE objectives.

- The generation of data on reactive compounds that could be in air is a valid area of research. It is rational to be examining compounds that might be found in the fuel stream and could diffuse across the membrane electrode assembly (MEA) to the cathode.
- The project's objectives of understanding durability and the performance effects of key impurities are well aligned with the DOE Program objectives. The stated objective of forming mitigation strategies for the impurities is also well aligned with improving fuel cell performance, durability and performance.
- Understanding the impact of impurities on the fuel cell operation is critical for the FCT program.
- Understanding impurity effects on the rates of fuel cell reactions is of paramount importance; therefore, this project is (potentially) critical to the Hydrogen Program.
- This project identifies and addresses critical path barriers of cost, durability, and performance.
- In general the group did an outstanding job. They focused on highly relevant materials and investigated useful operating conditions. The project was very informative.

Question 2: Approach to performing the research and development

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

- The approach is valid. For example, according to the EPA (2002), SO_x concentration in a number of U.S. cities is in the range of 10 to 15 ppb. In Europe, this number can be 4 to 5 times greater.
- The approach to understand the effect of the key impurities is both sound and thorough. The testing seems to be well designed and targeted at improved understanding of the behavior of the key impurities.
- The single component impurity testing is well thought out.
- The PIs need to include mixtures of impurities to understand their impact. Perhaps a design of experiment statistical analysis approach would be useful to be able to determine and model the impact of mixtures without having to test every mixture composition.
- The project is relatively well designed and is based on utilization of variety of analytical methods, surface sensitive techniques, and modeling. In my opinion, the addition of a classical rotating disk electrode (RDE) method will be very useful in unraveling the true effects of sulfur poisoning of active catalyst sites supported on

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high surface area carbons. In addition, careful measurements with bulk-like polycrystalline electrodes will be very useful to resolve many puzzling issues associated with Pt-S interactions and poisoning effects on the oxygen reduction reaction (ORR).

- The proposed objective is in line with overall DOE program objectives (NH₃, sulfur compounds), and some impurities studied seem to be of minor relevance (NO_x). More input or guidance from industry may identify impurities of more importance.
- This was a really nice project with an excellent approach.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.2** based on accomplishments.

- The accomplishments are as expected for this point in the investigation.
- The team has made good progress on the understanding of the effects of the impurities individually and in interaction with the fuel cell operating parameters, in particular the effects of ammonia and its interaction with water. There is little discussion or progress against the stated objective of developing mitigation strategies.
- LANL has made good progress in understanding the impact of impurities.
- The results are a good beginning. The team needs to continue with this work and expand it to understand mixtures.
- Since this is the end of a well-funded project, more results were expected. LANL should present a little of the model results and validation.
- The project is in its final stage, so the accomplishments must be rather obvious, which appears they are. The project clearly points to various degrees of contamination of platinum group metal (PGM) catalysts and membranes, which in turn have a strong effect on the polymer electrolyte membrane fuel cell performance. Although it was clearly demonstrated that both SO₂ and NO₂ have a negative effect on the activity, it is not clear how the adsorbed species are affecting the performance. The surface chemistry of NO₂ is rather rich and the nature of N-adsorbed species may depend strongly on the applied potential. Much more fundamental work is needed to resolve these complexities. The same applies for both the effects of NH₄⁺ and other cations present in the membrane. In my opinion, more fundamental work is required to fully understand electrochemistry at three phase interfaces.
- Ammonia and sulfur compound results are important.
- Outstanding technical accomplishments.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.0** for technology transfer and collaboration.

- The noted collaboration looks to be closer to a list of subcontractors or vendors. It is suggested that this data be discussed with John Van Zee of South Carolina and Scott Greenway of Savannah River National Lab. Discussion of this data plus the data they generated as part of the hydrogen purity research might help pull together a clearer picture of NH₃ poisoning.
- The project has good collaborators for the examination of the key impurities and their effects on the performance and durability. The collaborations appear to be well focused on the detailed fundamental understanding of the poisoning effects of the impurities and the interaction with the fuel cell operating parameters.
- They have good collaborations and spread the work around appropriately.
- One would expect more publications from this work from the results and the funding level than what they reported.
- This project is multifaceted, and as such requires a close collaboration between the PIs. Eight partner organizations with highly complementary skills are put together in catalysts development, electrode-structure design, materials characterization, and catalyst fabrication. The degree of collaboration is indeed outstanding and certainly a big part of success of the entire project.
- More collaboration with industry is needed for focused direction.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- The future work appears to be appropriate. Using EPA air quality data may help bound the expected exposures. Similar data from major cities worldwide would be helpful also.
- The future work looks good for addressing further understanding of the effects of the impurities. More focus on mitigation strategies and understanding them would be helpful.
- The future work seems appropriate, but should be expanded to include mixtures of impurities.
- They need to continue model refinement and validation.
- In general, the proposed future work appear to be reasonable, at least for the synthesize part. I think the project needs much more aggressive electrochemical characterization.

Strengths and weaknesses

Strengths

- The strengths of this effort appear to be the researchers' knowledge and techniques.
- Good approach to understanding the fundamental effects of impurities on the performance and durability and their interaction with water in the fuel cell.
- Strong teams. They have the right tools to perform the analysis.
- The major strength of the project is unique ability of PIs to synthesize and characterize different ceramic type supports, which might be of potential interest for the Hydrogen Program. The PIs are qualified for this work, but they have to demonstrate that the kind of support they are proposing may eventually compete with the existing carbon supports and especially with 3M NSTF systems.
- Overall, this is an excellent project and has produced very useful data.

Weaknesses

- An apparent weakness is the lack of the team availing themselves to the synergy of the hydrogen quality researchers.
- No reported work on the development of mitigation strategies for these impurities to improve the durability.
- Electrochemical characterization is an apparent weak point of this project. The electrochemical results must be more convincing and in line with the state-of-the-art PGM catalysts supported on carbon and/or attached to the 3M supports.
- Short list of collaborators.

- The PIs need to look at chlorides.
- More work on understanding mitigation strategies for the impurities to improve the fuel cell durability.
- They need to look at mixtures.
- The researchers should publish more of their work (they have some publications, but with the amount of research performed more publications were expected).
- DOE should consider expanding the impurities to include ones that people do not often think about such as the off gases from fresh blacktop and chlorides.

Project # FC-46: Effects of Impurities on Fuel Cell Performance and Durability

James Goodwin; Clemson University

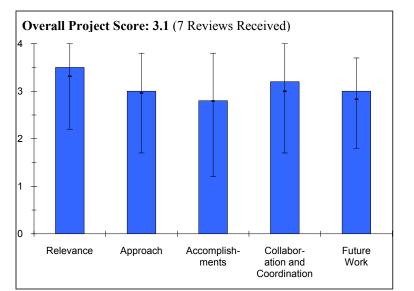
Brief Summary of Project

The overall objectives of this project are to 1) investigate in detail the effects of impurities in the hydrogen fuel and oxygen streams on the operation and durability of fuel cells, 2) determine mechanisms of impurity effects, and 3) suggest ways to overcome impurity effects.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.5** for its relevance to DOE objectives.

• The project addresses two principally different contamination processes that are both key for fuel cell



performance: the impact of contaminants on catalyst performance, and the impact of contaminants on membrane conductivity

- The effect of impurities on membrane performance is of great importance if fuel cell technology is to become practical.
- The study of impurities is an essential topic in moving fuel cell technologies successfully to the marketplace.
- This project to investigate in detail the effects of impurities in the hydrogen fuel and oxygen streams on the operation and durability of fuel cells is relevant no matter what membrane is ultimately used in PEM fuel cells.
- Comprehensive analysis on impurity effectiveness is important for standardization and development of risk mitigation strategy.
- Addresses long term durability due to impurities, both on the air side and fuel borne.
- Topic area is extremely important to fuel cell performance and durability in real-world environments; however, the project could be even more relevant if it were to focus on the study of contaminants not already covered by past and existing projects.
- Understanding the effects of impurities and the development of mitigation strategies are critical to the advancement of fuel cell technology. However, it is unclear that new observations and insights have occurred through this project.

<u>Ouestion 2: Approach to performing the research and development</u>

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

- The PI intends to investigate a large number of contaminants. Each contaminant as well as contaminant mixtures are studied in a very short time frame of 3 months only. These studies are likely to be incomplete and may leave many questions unanswered. Contaminant effects are typically strongly affected by operating conditions and the investigation requires a significant amount of time at realistic contaminant concentrations. Furthermore, no information was given on how the correlation between *ex situ* phenomenological studies and fuel cell studies will be established.
- The introduction of known impurities and careful characterization of their short and long-term effects in fuel cell performance and membrane conductivity is a good approach to learning the effects of impurities.
- Empirical correlation of impurity concentration to performance is important. Formulating mechanisms of impurity effects is important and there have been some trends developed from empirical correlations to develop mechanisms of impurity effects (cations, ammonia and water content on conductivity, surface poisoning to fuel

cell performance, etc.). The project started in 2007, so I would have expected more explanation of mechanisms of impurity effects. Maybe this is forthcoming.

- It is necessary to identify how to distinguish performance degradation by impurity effects from materials degradation by fuel cell operation.
- PI is focused on both near and long term. Correlation experiments between SRNL and Clemson are relevant for comparison of data sets. The approach, as outlined, covers both component level and system level effects and feeds forward to mitigation of detrimental conditions.
- The majority of the contaminants studied in this project have been studied in previous projects. Due to the importance of this topic area to the ultimate success of fuel cells in real-world applications, the effect of understudied contaminants should have been the focus of this project. The effect of Na⁺, Ca²⁺, and Fe³⁺ have already been studied. The effect of CO on MEA performance has been extensively studied in the past. The effect of furan, hydrocarbons, and perchloroethylene are good selections as, to my knowledge, they have not been studied at all or have not been adequately studied in past work.
- The overall project objectives are too broadly stated as "investigate effects of impurities," determine mechanisms, and overcome impurity effects." The objectives for FY 09-10 are more specific, but the choice of some of the study subjects seems repetitive with past work and other ongoing projects. Also, the work seems disjointed compared to the very broadly stated goals.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.8 based on accomplishments.

Ex situ studies on CO only confirmed existing knowledge. The benefit of these studies and the meaning of the received numbers are unclear. Some explanation and/or clarification of whether these numbers are useful for a modeling effort would be helpful.

- Understanding how an increase of the ionic conductivity of the catalyst layer affects the overall resistance of the fuel cell is very important, specifically if it allows one to detect such an effect within the fuel cell. It could also be beneficial to see if *in situ* diagnostics such as electrochemical impedance spectroscopy could detect such an effect.
- Cleaning agents have been identified as potentially important contaminants that may find their way into a fuel cell system. In this study, however, the investigation of cleaning agents seems to be somewhat disconnected from the other studies.
- The presented mitigation studies are not thoroughly discussed. In the case of RH, the PIs create conflict since they suggest a higher RH for one contaminant and a lower RH for another. Obviously you cannot run both these conditions at the same time, although it is likely both contaminants are in the same feed stream.
- For the limited impurities and feed streams studied to date (CO, water, cations, organics like THF, perchloroethylene, ammonia) the team has well characterized impurity effects and shown good understanding of these effects.
- It is good to identify implication of fuel cell operating conditions (humidity).
- Impressive accomplishments in both basic studies of poisoning mechanisms and in the bottom line effect of poisons on measured fuel cell performance and performance recovery. The scope of the number of potential poisons, the importance of this issue, and the limited time to achieve the DOE technical targets indicate that a more prudent use of project resources would be to focus on contaminants that have not been studied and focus on combinations of contaminants, especially airborne contaminants.
- Listed below are comments on each major area of work separately:
 - (1) Effect of Nafion® and %RH on hydrogen activation.
 - It is common knowledge that hydrogen adsorption and oxidation are not severely impacted by these variables.
 - Given that CO has been studied extensively in fuel cells, the impact of water on CO removal in a qualitative sense is known. With only three humidity conditions shown, it is unclear that more is known after these experiments.
 - (2) The esterification approach is a useful tool for assessing cationic contamination that has been developed through this project and validated experimentally.
 - (3) The cationic contamination studies appear repetitive with previous work in terms of choice of cations and specific observations; e.g., the effect of monovalent versus divalent ions is known by now.

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- (4) Some "new" contaminants such as THF and PCE have been screened, but no chemical insight has been gained. We have learned from this work that these species poison the fuel cell, and that some extreme operating conditions can remove them, but not much else.
- (5) Expansion of the contaminant "database": Although such data must be tabulated, ultimately this information should be synthesized into useful models or groupings that allow for prediction, which has not occurred.
- (6) Mitigation strategies: two of the suggested strategies, increasing RH to increase conductivity during NH3 poisoning or using more Pt to prevent CO poisoning (the latter is impractical), are already known. As for the third strategy of lowering humidity for CO removal, one would expect that the effect of humidity on CO poisoning has also been determined based on the extremely long history of the study of this molecule in fuel cells. No strategy was experimentally validated to be pragmatic.
- Overall, the work did not generate many new, predictive insights or any validated mitigation strategies.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.2** for technology transfer and collaboration.

- Collaboration between university, National Laboratory, and industry partners provides a very good mixture.
- The team is a great strength of the project.
- The team is well situated to have one segment do the impurity effect experiments with good control in the laboratory and good prospects for having another segment of the team validating these results in practical environments.
- How are these results are connected to the codes and standards team efforts?
- Good triad of collaborators: laboratories, academia, and industry.
- Collaboration and coordination of the project team was evident with defined roles for each partner in the team.
- Although university, national laboratories, and industry are represented here, there seems to be duplication of work performed in other impurities projects and, importantly, when work on well-studied topics has been performed (such as CO, Nafion®, cationic contamination, etc.), the degree of understanding has not been significantly advanced over previous reports.
- The exact roles of John Deere and Argonne National Laboratory remain somewhat unclear.

Question 5: Approach to and relevance of proposed future research

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

- Logical continuation of the project is suggested for most tasks. However, the future work should address the gaps in the research to make this project more valuable to the fuel cell community. This includes developing and confirming applicable mitigation strategies, and correlating the phenomenological studies with fuel cell experiments in a useful applicable way.
- The plan is being well implemented to learn the effects of impurities on fuel cell performance and stability.
- At this stage, I expect the team should emphasize the proposed future work to "correlate phenomenological results with actual fuel cell runs."
- Builds nicely on past accomplishments.
- The proposed future work on PCE, CO₂, ethane, ethylene, and acetaldehyde is well planned and very relevant. The proposed future work on ammonia and CO has minimal value in providing new information on these contaminants and in addressing fuel cell performance barriers.
- The project is nearing completion, and, although studies on "poisoning mechanisms and rates" have been promised, this type of work has not been well represented in the project to date. Degradation rates on a variety of contaminants will be obtained to "provide Argonne National Laboratory Modeling Group with ... data on NH₃ poisoning of Nafion® and CO and PCE." Such information is needed but does not constitute a comprehensive, insightful contribution to the subject of impurity studies.

Strengths and weaknesses

Strengths

- Sound approach using some novel ideas.
- The methodology, resources, and team skills are the greatest strength of the project.
- Comprehensive set of characterization techniques with complementary fuel cell and components studies. Study of hydrocarbon contaminants.
- Communication with DOE seems to be strong.
- The analytical test developed to assess conductivity in electrodes may prove useful to the community.

Weaknesses

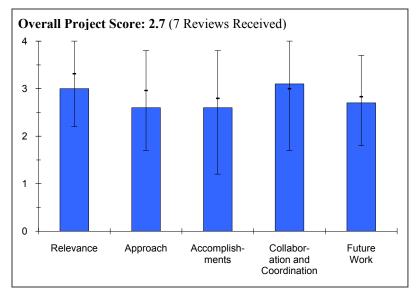
- Correlation of *ex situ* results to *in situ* fuel cell results is largely missing. The benefit of the *ex situ* experiments is unclear.
- Mitigation strategy section is non-existent and needs more attention.
- The modeling component of the project is very limited and for a lot of the data, it is unclear how the results will be used and if they will be used at all.
- Approach is not followed through far enough to exploit the full potential of beneficial information that the ideas in this project had to offer.
- A limited set of impurities has been selected. The overall effect of studied impurity on fuel cell response is being well characterized and this is useful information. However, how the mechanisms of the effects of the impurities (in particular cooperative interactions of mixtures of impurities) are being determined was not clear to me. As a result, the team is clearly finding how a selected impurity affects fuel cell performance, but may be getting limited use out of its work. The effects of a particular impurity may be discovered, but this knowledge may not be extendable to other similar materials unless the mechanism is determined (e.g., is an activated hydrocarbon like PCE an organic or chloride impurity?). This approach may change as mechanistic trends emerge. The team should try to learn trends.
- Duplication of past contaminant work, dwelling on ammonia and carbon monoxide impurities.
- Many older topics are addressed such as effect of Nafion® and water on CO without any notable advance in understanding or mitigation.
- When working on heavily examined topics, the burden is on the researcher to demonstrate clearly what the advances in knowledge are.

- It would be wise to try to isolate the moiety that is causing trouble in a complex impurity. For example, in the case of perchloroethylene, it would be wise to see if an equimolar amount of chloride gives the same impurity effect.
- Delete further work on ammonia, carbon monoxide, and other contaminants already studied in past projects. Add airborne impurities to project scope.
- Perhaps any work using CO should discontinue. It is unclear what is new here.
- New chemical insight should be generated. Which contaminants act as simply site blockers? Which can be removed by potential pulses? Which have irreversible effects? Are there any non-additive effects when more than one contaminant is present? How can one deal with cation contamination once it is present? Such questions that generate new information should guide the project.

Project # FC-47: The Effects of Impurities on Fuel Cell Performance and Durability *Trent Molter: University of Connecticut*

Brief Summary of Project

The overall objective of this project is to develop an understanding of the effects of various H₂ impurities on fuel cell performance and durability-critically important for automotive fuel quality. Specific task objectives are to: 1) identify contaminants in both fuel and oxidant streams; 2) develop analytical methods to study contaminants; 3) develop contaminant analytical models that explain contaminant effects; 4) construct that quantify that material degradation; 5) validate contaminant models through single cell experimentation; 6) develop and validate novel technologies for mitigating the effects of contamination; and 7) conduct outreach activities.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.0** for its relevance to DOE objectives.

- The study of impurities is an essential topic in moving fuel cell technologies successfully to the marketplace.
- Impurities are specifically important for the performance and durability of fuel cell vehicles.
- This activity is highly relevant for a hydrogen economy. The testing is in support of a hydrogen fuel standard, the lynch pin for a hydrogen infrastructure. This research validates JARI data while continuing the screening of related compounds of interest to industry.
- The investigation of the effects of the key impurities on the fuel cell performance and durability is well aligned with the DOE objectives. The proposed objectives of developing mitigation strategies and providing the information to the industry through outreach are well aligned with the DOE objectives.
- Although understanding impurity effects on the rates of fuel cell reactions is of paramount importance in the quest for full utilization of PEMFC systems, in my opinion, this work is not critical to the Hydrogen Program.
- Good focus on the automotive component of fuel quality.
- Though contamination is a very relevant area of study, and the authors had input from the fuel quality team for potential impurities to study, the conditions were not very relevant. Nearly all experiments were performed at 100% RH, which would likely flush the contaminant species from the system, and 800 mA/cm² is too high a current density for the contaminants being studied. It is not surprising that the authors observed little effect from many of their experiments, the impurities not flushed off were likely oxidized at high potentials. It is recommended that the authors probe different operating conditions.
- The stated work scope of the project is certainly relevant to Hydrogen Program goals. However, it is not clear based on this presentation that the objectives related to model development, mechanistic understanding, and development of impurity mitigation strategies have been achieved. Perhaps some of these significant remaining tasks can be addressed in the final half year if external data sets are also used.

Question 2: Approach to performing the research and development

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

• The overall project approach is sound.

The project focuses on the effect of contaminants in the anode feed stream at contaminant concentrations identified by DOE, industry, and working groups. This is a good approach for creating valuable data of interest to the community.

- The project's intent is to use the data for model development. Modeling is one essential part of increasing understanding of the effect of contaminants.
- The approach is tried and true and is compatible with the research done by the other labs involved in this type of testing and funded by DOE, NEDO, and the European Union.
- The use of industry input and standardized testing provides a good approach to the understanding of the effects of the impurities. The initial examination of high levels of impurities to screen for significant effects is good.
- The project is not well designed and needs major improvement to be able to add substantially to this DOE Hydrogen Program. Although testing the impacts of different hydrocarbons and halogenated compounds in systems of importance for fuel cell developers is important, the true effects of these impurities should have been tested in a simple electrochemical cell. With this approach, the data collection (and understanding) would be much more improved. For example, this methodology would allow utilization of infrared spectroscopy, which is indispensable for such systems. I do not think that NMR is as useful as IR. To use solely cyclic voltammetry for analyzing the coverage and the nature of contaminants is insufficient, as seen with the great uncertainty about CO adsorption even for as simple system as formic acid oxidation. Recovery, i.e., oxidation of CO produced in a course of HCOOH oxidation, is well understood and the PIs should consult the existing literature about this "problem." The same applies for NH₃ contamination. Simple laboratory testing is required to unravel the effect of NH₃ on modifying catalytic properties.
- The project builds on and leverages past work and it sticks to standard protocols where possible. I recommend focus on specific impurities identified by DOE, industry, and working groups. The project plan and participant roles are well defined.
- It does not make sense for the authors are running for 100 hours to establish a baseline and then starting to introduce contaminant species. It seems like a waste of test stand time. Perhaps they should develop a better break-in protocol and then run a baseline for only a couple hours.
- At the end of the third year of a project, it is fair to look at what has been accomplished rather than what is promised to evaluate the approach. If one looks at what has been finished, the core of the approach seems to have been to run 100-h experiments on a variety of contaminants. Without any supporting experiments, little insight can be gained from this type of work.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.6 based on accomplishments.

- This project addresses a significant amount of necessary labor and time-intensive footwork for understanding hydrogen fuel contaminant effects. The results are important, but scientifically not very interesting.
- Measurements of a variety of hydrocarbons and halogenates have been performed. While most impurities that were tested have little to no effect, a more detailed study was conducted on formic acid which showed an effect at high but not at low concentrations. Such studies would further increase the value of the project when combined with predictive models.
- The study of contaminant mixtures was missing.
- The amount of directly applicable data generated over the duration of this project is impressive.
- The project has made good progress in understanding the effects of the impurities. The PI did not report any significant progress against the goal of developing migration strategies for the key impurities.
- I am not convinced that accomplishments are significant. Most of the data presented so far are unconvincing and even without this project, the fuel cell community has already been aware of contamination problems induced by CO adsorption, ammonia decomposition, and negative cation effects on the membrane properties.
- Critical issues defined beyond standard program rhetoric. Need of additional coordination between labs is critical to eliminate duplication of effort. Standardization of protocols and hardware is critical for comparison of results.
- Impurity investigation is methodical and well documented.
- The rate of progress seems really slow. The experiments are very long and testing conditions do not seem that well thought out, especially for a three-year-old project.

- Most of the experiments presented were 100-h fuel cell tests at constant current density followed by introduction of a contaminant and continuation of the test for another 100 h. While such experiments contribute the building of a database of contaminant behavior under consistent conditions, which is one type of valuable information, they are not a sufficient basis for model development and or mitigation strategy development. For example, perturbations such as humidity changes and competition between contaminants would aid in generalizing the results. Comparisons between contaminants should also be made in the context of some governing model that leads to predictions that can be validated.
- It is unclear what new information has been gained from the formic acid study, especially the cyclic voltammetry section. If the loss in ECSA had been correlated to the voltage loss in some way, then that would have added more value.
- Aside from the above details, the stated goals of "develop(ing) contaminant analytical models, construct(ing) material state of change models, and develop(ing) and validat(ing) novel technologies for mitigating the effect[s] of contamination" have not been addressed to any meaningful degree by the end of year three of a four-year project.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.1** for technology transfer and collaboration.

- Collaboration between two university institutes and two industry partners provides a good mixture. It remains a little bit unclear how much the individual parties contributed to the project.
- I recommend collaborating with / participating in a work group addressing hydrogen fuel contaminants and
- with a national laboratory.
- The collaboration with JARI, CEA, HNEI, LANL, the University of South Carolina, Clemson/SRNL, NREL and ANL is refreshing. The outreach to industry on the selection and prioritization of impurities was a pleasant development. The effort by UConn is the gold standard on collaboration.
- The PI has built a good team of participants for the project and appears to be working on outreach and industry interaction to ensure understanding of the key impurities.
- Collaboration between partner organizations is good.
- The collaborative group is far and away the most diverse of the fuel impurity investigations.
- Since the use of partner data for the creation of models and mitigation strategies is implicit, and these goals have not been achieved, one should assume that the collaboration level is also insufficient.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.7 for proposed future work.

- The base idea that future work should concentrate on completing the contaminant database, model development, and the development of mitigation strategies was presented.
- Detailed plans were not presented and left open questions such as which contaminants will be focused on, which will be used for modeling, for which will mitigation strategies be developed?
- Mixture effects were not included.
- The long term effects of contaminants are also important although they may be out of the scope of this work.
- Real-world operating scenarios during exposure to contaminant mixtures would be another important future work item.
- The future work is appropriate to the stage of the project. More data on halogenated compounds and diols (heat transfer fluids) would be useful.
- The proposed approach of moving towards testing mitigation strategies is an important objective in meeting the stated project objectives in the next year.
- In my opinion, the proposed future work may lead to improvements, but needs much better focus on overcoming barriers.
- Plans are methodical, well documented and not the typical scatter of proposed future work.
- I would recommend the group discuss testing conditions and proposed mitigation experiments with the Tech Team to get input on the experiments. I would also recommend asking the Tech Team to suggest a better break-in protocol for future experiments.

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• Since most of the major goals (including modeling and mitigation) have not been addressed, the project is in a state of catching up. It is not very likely that three years of work can be finished in only one.

Strengths and weaknesses

Strengths

- Not much scientific new ground is explored. Mostly labor intensive though important footwork is performed.
- This project focuses on filling the gaps that were present to create a comprehensive overview of the hydrogen fuel contaminant impacts.
- The strengths of effort is the collaboration with other labs, leveraging of industry expertise, and the researchers' techniques.
- Good fundamental understanding of the effects of the impurities and their effects on the performance. Good industry interaction on understanding the important impurities.
- The strength of the project is systematic collection of data related to poisoning effects of hydrocarbons and halogenated compounds. Also, a positive side of the project is the potential technology transfer of data and a strong collaboration with industrial partners.
- Well thought out. Well organized. Well documented.

Weaknesses

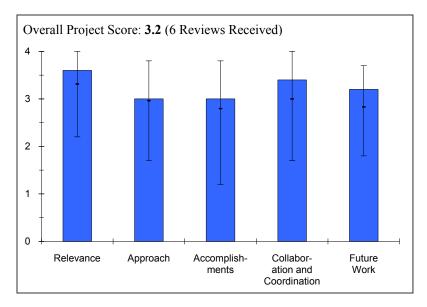
- Although this project is focused on testing impurity effects in real fuel cell systems, much more fundamental work is needed to be able to understand the formation of spectator species and how to remove them to recover activity and stability of fuel cell interfaces.
- There is no higher level organization of the data sets into predictive models. If this is to be a contaminant screening exercise, then that should be clearly stated from the beginning.

- Need to move forward on the development of mitigation strategies for the impurities examined.
- Increase the scientific output of this project by concentrating on the mechanism of contaminants that showed a performance effect, modeling of these mechanisms with the goal to provide a predictive model, and identifying reactions that may occur in the cell even for contaminants that do not show any performance impact. This last activity may particularly be important for understanding contaminant mixtures.
- Continuation of activities in this area would be money well spent.

Project # FC-48: Effect of System and Air Contaminants on PEMFC Performance and Durability *Huyen Dinh; National Renewable Energy Laboratory*

Brief Summary of Project

The overall objective of this project is to decrease the cost associated with system components without compromising function, fuel cell (FC) performance, or durability. Objectives are to: 1) identify and quantify system derived contaminants, 2) develop ex situ and in situ test methods to study system components, 3) identify severity of system contaminants and their impact on operating conditions, 4) identify poisoning mechanisms and investigate mitigation strategies, 5) develop models/predictive capability, 6) develop material/component catalogues based on system contaminant potential to guide system developers on future material selection, and 7) disseminate knowledge gained to the community.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.6** for its relevance to DOE objectives.

- The project is very relevant to this program. The off gassing of many compounds used in the manufacturing of a vehicle has the potential to become an airborne contaminant that would adversely affect the performance of the FC.
- To identify contaminants which have an effect on FC performance and durability is important. It is necessary to obtain general knowledge for possible contaminants rather than specific materials which they are using in the current system. This should not be an engineering project; it should be a research project to obtain fundamental knowledge about possible contaminants and their mechanisms.
- FC system contaminants can have a significant effect on FC durability, but there has been little quantitative work performed to date. This project fills that gap.
- Relative to other DOE-funded impurity projects, which are often overlapping and redundant, focusing on system impurities is new and relevant.

Question 2: Approach to performing the research and development

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

- The approach is valid. It is suggested that NREL go through the same round robin time testing that the Japan Automobile Research Institute (JARI), the Hawaii Natural Energy Institute (HNEI), the Los Alamos National Laboratory (LANL), and the Universities of South Carolina (USC), Clemson, and Connecticut did. This testing would verify repeatability and reproducibility, as was done on the fuel quality effort.
- The project seems to focus on materials that are used in the current system and to see whether or not they would be contaminants. It is necessary to dig up one more detail to obtain fundamental knowledge about possible contaminants and their mechanisms.
- The addressing of system derived contamination has been overlooked in the past. As commercialization proceeds, this possible avenue of contamination must be investigated more closely with the direct input of industry collaborators.
- The focus has been prioritized by employing in-house knowledge and experience from General Motors (GM).

- The approach is sound and well-considered.
- Additional details of the testing protocols used for different system contaminants would help to even better evaluate the approach taken.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.0 based on accomplishments.

- The accomplishments are appropriate for this point in the program.
- The project has just begun, but significant progress is already being made.
- This project has shown good progress at the development of test methods.
- The lack of any electrochemistry and FC data is a drawback.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.4** for technology transfer and collaboration.

- The list of collaborators is impressive.
- They have taken an excellent team approach.
- The team is strong with well-defined roles of participants. The level of involvement to date of several partnerorganizations in the project; e.g., that of LANL, USC, and the University of Hawaii (UH), has not been clearly highlighted in the PI's presentation.

Question 5: Approach to and relevance of proposed future research

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

- The proposed work flow is rational.
- The proposed future work plan appears well-focused.
- The proposed future work plan does not seem to have been very well thought out. Most materials should have been selected and acquired by now. The same applies to the review of prior work.

Strengths and weaknesses

Strengths

- The strengths of this project are the collaborations.
- NREL assembled an excellent team with excellent capabilities and a well-focused effort.
- The focus on system materials and good teamwork are definite strengths of this project.

Weaknesses

- Weaknesses do not appear to be evident.
- Impact of contaminants on FC performance has not started yet.

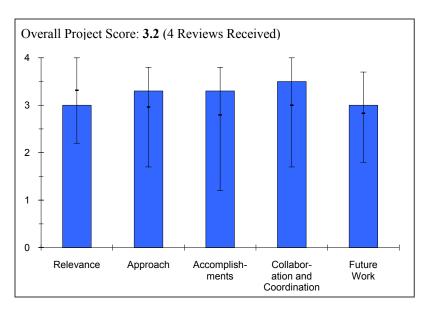
- The project seems to be focusing on materials which are used in the current system and to see whether or not these materials could be contaminants. It is necessary to dig up one more detail to obtain fundamental knowledge about possible contaminants and their mechanisms.
- Electrochemical ("half-cell") and FC testing should start as soon as possible.
- The development of a preliminary model for impurities with different impacts on the key FC components (catalyst, membrane) should be initiated and subjected to experimental verification, in parallel to the stability testing of system materials.

Project # FC-50: Economic Analysis of Stationary PEM Fuel Cell Systems

Kathya Mahadevan; Battelle

Brief Summary of Project

The overall project objective is to assist DOE in developing fuel cell (FC) systems by analyzing the technical, economic, and market drivers of polymer electrolyte membrane fuel cell (PEMFC) adoption for stationary applications. Support in 2009 included two tasks: 1) developing technical targets for a 5-kW direct hydrogen PEMFC for backup power by developing a manufacturing cost analysis at varying levels of production, and 2) developing an economic and market opportunity analysis for micro-combined heat and power (CHP) PEMFCs to identify key target markets and value proposition for PEMFCs.



<u>Ouestion 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of 3.0 for its relevance to DOE objectives.

- This project is an ongoing study of the cost of stationary FC systems targeted at the 5-kW output power level. It is relevant to the new responsibility of the Fuel Cell Technologies Program to focus on near-term, early market FC opportunities.
- This project addresses all barriers in the multi-year plan through economic analysis (task 9) of small stationary PEMFCs. Both manufacturing cost and micro-CHP analysis are considered.
- This project is relevant to the DOE program objectives, particularly since DOE has increased itsfocus on stationary and early-market applications.
- The project addresses the DOE goals for boron and fluorine.

Question 2: Approach to performing the research and development

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

- The approach of developing a "generic" baseline system design that is not dependent on a specific FC supplier design is good. Likewise, doing costing for 2,000, 10,000, and 100,000 units per year is important and provides costing information with demand flexibility and economy of scale effects to be analyzed.
- Input was received from the extensive stakeholder and user community. Battelle used a very logical approach, starting from system design to base case analysis (2,000 units, 5 kW), with future case cost and analysis refinement. Some of this was done based on their own experience, which could have some error bars associated with it, although that is mostly from external input. Battelle used well-accepted Boothroyd-Dewhurst estimating software for calculating the manufacturing cost.
- Previous studies by Battelle under this contract identifiedpotential markets and opportunities for FCs in nonautomotive applications. In addition to the market analysis, this year's effort includes a cost analysis for micro-CHP systems. The Battelle approach is similar to the approach taken by Directed Technologies, Inc. (DTI) for estimating the manufacturing cost for automotive FC systems—the design for manufacturing and assembly (DFMA®) analysis. Their assumptions regarding system configuration and manufacturing techniques have been vetted with FC developers.

- The approach is solid. Using leading cost estimation software makes sense. The weakness lies in the current limitation to one design. Incipient sensitivity analyses were presented. This should be extended in the future work.
- All data delivered are concrete data without consideration of the statistical variations. The Monte Carlo simulation allows handling of statistically distributed cost data easily. It might be considered for implementation, since some cost contributions are well known and others are highly speculative. This method allows for a better feeling for the variation of the cost prediction.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.3** based on accomplishments.

- This has been a long-term project (~6 years) and has shown steady progress toward meeting its goals and objectives.
- There is good technical progress, but it is a very long project (started 2003). Battelle presented believable numbers for the base case scenario. The scrap or reject rate assumptions (up to 30% for some parts) seem quite high. They should go down when going from 2,000 to 100,000 units. The balance of plant (BOP) breakdown cost summary was quite good. The stack component manufacturing cost breakdown (including tooling, processing, set-up, and raw materials) was revealing. There was a good capital cost analysis for a manufacturing plant, which was useful for Columbus, Ohio, but may not be useful in high cost areas.
- As of the date of slide submission to the AMR for fiscal year (FY) 2010, funding had not been received. Accomplishments include completion of the manufacturing cost analysis for 2000 5-kW systems, and the definition of market requirements for these systems.
- The achievement is very good. The analyses dig deep into the manufacturing process and consider the setup of a manufacturing plant.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.5** for technology transfer and collaboration.

- There are extensive collaborations securing input from FC users—more than 400. Many publications and reports are making the findings of this study available to the general FC community.
- They conducted extensive analysis using input from more than 60 FC industry stakeholders and more than 400 current or candidate users.
- Collaboration primarily consisted of vetting the project assumptions with numerous component and system developers in this market space.
- Sixty partners is the utmost number for a project to handle.

Question 5: Approach to and relevance of proposed future research

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

- It was not clear how long this study would continue, but the proposed focus on early adopters and market opportunities seems appropriate.
- The project is wrapping up. PI and coworkers are in the final stages of future case cost analysis modeling and refining technical targets.
- The project is ending in October 2010. The most important deliverable to be completed is a cost-benefit analysis of FC systems vs. the competing alternatives.
- Since the cost calculations are based on one selected case that can be referred to as a "base case", it would make sense for the future work or perhaps for a future project to add variations to this base case since cost is very sensitive to the systems design and not only to the stack.

Strengths and weaknesses

Strengths

- Battelle has extensive experience base in stationary system design and costing.
- Battelle exhibits good use of DFMA tool in a ground up analysis to evaluate manufacturing cost. There is good effort to obtain input from a variety of external sources.
- Battelle's market analyses have correctly identified market pull opportunities for back-up power and material handling vehicles.
- Battelle employed industry standard methods in developing a manufacturing cost estimate of micro-CHP systems.
- There is only a generic estimation of the cost, including production cost. Many industrial partners are involved.

Weaknesses

- None apparent.
- The sensitivity analysis could have large error factors.
- Micro-CHP systems, for the most part, will operate on natural gas or other common fuels, not bottled hydrogen, as depicted in the system schematic. A reformer should be included in the system cost.
- The estimation was based on one base case and no statistical data was incorporated.

Specific recommendations and additions or deletions to the work scope

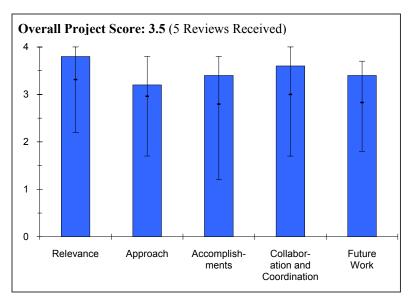
• Complete the project this year.

Project # FC-51: Fuel Cell Testing at the Argonne Fuel Cell Test Facility

Ira Bloom; Argonne National Laboratory

Brief Summary of Project

The overall objectives of this project are to 1) provide DOE with an independent assessment of state-of-the-art fuel cell technology; and 2) benchmark commercial fuel cell technology developments. ANL will: 1) develop standardized test procedures for the evaluation of 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.



<u>Ouestion 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of 3.8 for its relevance to DOE objectives.

- This project is very useful to evaluate in a reliable and reproducible way the performance and lifetime of the different stacks and systems developed in the different projects. With respect to results obtained in round-robin testing, standardized fuel cell testing procedures are really needed.
- It is very important to have standardized test procedures to get through the marketing hype so that informed decisions can be made based on validated data. This project supports this objective.
- This is a very relevant project, to verify manufacturer performance and R&D improvements.
- The project objectives are to assess independently a wide power range of fuel cell deliverables and benchmark commercial fuel cell technology development. These project objectives address performance and durability of the DOE Hydrogen Program.

Question 2: Approach to performing the research and development

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

- The proposed approach to test, on the one hand, different stacks and systems using existing U.S. protocols, and, on the other hand, to compare those protocols with international protocols is adequate.
- The test procedures refer to transportation applications. But, since DOE objectives have evolved, in particular for fuel cell applications, protocols for stationary applications should be added.
- The use of an old stack is questionable. By definition, it is old technology. We should really be using the best available technology. An older stack that has been sitting on the shelf for years could have unknown shelf life issues that could give imprecise data or lead to incorrect conclusions. Recommend that the Program provide enough funding to the project to allow it to purchase current technology and not be forced to use cast offs.
- The project is able to provide very detailed stack performance characteristics. With the current resources, it is adequate to show performance, although it is not able to do it in a statistically significant manner (no repeat measurement for multiple stacks). Given that each stack has a large number of cells, it is still adequate for the task.
- This project develops standardized test protocols for evaluation of different state-of-the-art fuel cell technologies with the ultimate goal to address fuel cell commercialization barriers such as durability and performance.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.4 based on accomplishments.

- The presented results are good. However, more details on the test conditions (pressure, temperature, RH, etc. and for current-voltage (i-V) curves (dwell time, sampling time, etc.)) would provide all the needed parameters to evaluate the data. References to the protocols used could also be given.
- For system testing, the results showed that the tested systems are working well. A long-term graph would have been appreciated. As previously mentioned, the graphs are lacking information to be well understood.
- For protocol comparisons, the comparison between DOE and the Fuel Cell Testing, Safety and Quality Assurance (FCTES^{QA}) i-V protocols is very interesting. It shows that, as for single cells, if the testing conditions are stable, the values remain comparable even for different ways of recording.
- Now that it has been demonstrated that the two tests are comparable, the partners should work to develop a common standard.
- The team demonstrated an extensive evaluation of two 5-kW stacks as planned. The results were used to verify the standardized tests and the comparison of two protocols have been initiated. To date, the project has generated performance reference points for dynamic cycling tests based on two test protocols as originally planned.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.6** for technology transfer and collaboration.

- Collaboration with other institutions seems correct. Participation in the European project FCTES^{QA} and in TC105 WG11 is appreciated and should continue in the future.
- All the major players are involved.
- The PI has been able to set up relationships with key industry manufacturers.
- There is a good selection of partners (domestic and international) to fulfill the project objectives.

Question 5: Approach to and relevance of proposed future research

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

- No specific slide on the future work is presented.
- Testing FCTES^{QA} protocol A of the dynamic load cycling should lead to interesting results for stacks.
- Good plans competently executed.
- The future research objective is clear. The team will complete accelerated durability tests under the FCTES^{QA} dynamic cycling profile. This test will finalize the benchmarking of commercial stacks and at the end, the report will be delivered to DOE.

Strengths and weaknesses

Strengths

- The strengths of the project are being able to test full systems and to compare existing testing procedures from U.S. and international as European stack testing procedures. Implication in international standardization, in particular TC105 WG11, is also appreciated.
- Achieved the objectives on schedule. Well planned and executed.
- Comparison of international fuel cell testing procedures.
- The experimental procedures are well defined, executed, and the results clearly presented.

Weaknesses

- The main weaknesses of this project are the lack of different stacks as an old 2002 stack has been used and the lack of details of the testing conditions which leads to a more difficult evaluation of the quality of the work.
- Use of an old stack is false economy.

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- Old stack hardware was used for testing.
- Standardized test protocols should be application-specific rather than generic.

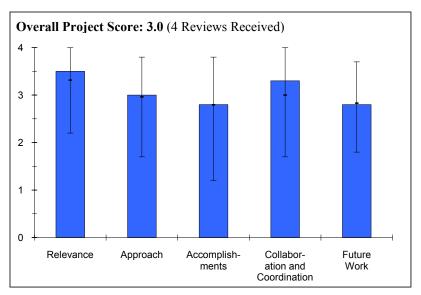
- Regarding the DOE objective evolution, in particular for the fuel cell applications, protocols for stationary applications should be integrated. Procedures already exist (IEC62282) and a new version is under discussion for small polymer electrolyte fuel cell systems (<10 kW, TC105 WG4). Applying it may be useful for experimental validation
- For protocol comparisons, the comparison between DOE and the European FCTES^{QA} protocols should go on as further testing procedures should be released in the following months. Moreover, a new work item proposal has been proposed to TC105 to establish i-V curve procedure as an IS. Experimental comparisons will be very helpful to establish valuable international procedures.
- Please provide an aggregated public presentation of the results. I realize the results are considered proprietary. However, an aggregated industry result could be presented in lines with the feedback provided by NREL's vehicle Tech Val program. This feedback would be of value for manufacturers as well as the research base to see, for example, the distribution of polarization curves or operating parameters in industry.
- Protocol verification tests are recommended for dynamic cycling tests. It is suggested to reverse the order of FCTES^{QA} and dynamic stress tests to exclude the effects of the experimental order.

Project # FC-52: Technical Assistance to Developers

Tommy Rockward; Los Alamos National Laboratory

Brief Summary of Project

This project supports Los Alamos National Laboratory (LANL) technical assistance to fuel cell component and system developers as directed by DOE. LANL is expected to include testing of materials and participate in the further development and validation of a single cell test protocols with the U.S. Fuel Cell Council. This task also covers technical assistance to Working Group 12, USCAR, and the Fuel Cell Technical Team (FCTT). This assistance includes making technical experts available to the FCTT as questions arise, focused single cell testing to support the development of targets and test protocols, and regular participation in working groups and review meetings.



Question 1: Relevance to overall DOE objectives

This project earned a score of 3.5 for its relevance to DOE objectives.

- This project provides the expertise and equipment of LANL to companies and agencies. This is a smart way to leverage the DOE investment and to provide to organizations answers to technical questions. If executed well, it has the potential to make very valuable contributions to the Hydrogen Program.
- A solid resource for all fuel cell developers.
- A good idea to provide these services, some of which are sophisticated and may not be available to new and resource-limited developers.

Question 2: Approach to performing the research and development

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

- It was not clear how clients are solicited, but those that seek support appear to be well supported. It would be useful to have feedback reports from each client—did they receive value for the effort?
- Delivers services to a wide range of customers. Offers both consulting support and on-site training.
- The array of services is broad. Should consider incorporating similar services for other types of fuel cells beyond PEM.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.8 based on accomplishments.

- Need feedback from clients to make a determination of value and progress, otherwise it is just a collection of facts with no appreciation or interpretation of results.
- Good variety of services delivered in past year.
- Most capabilities appear at a mature level. Provided assistance to a wide array of developers and researchers.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.3** for technology transfer and collaboration.

- This project is all about collaboration. Several clients were supported. Could they have done more? How do they reach out to potential clients—do they advertise the service? How are priorities established.
- Wide variety of customers served.
- Appears good collaboration among developers and service provider.

<u>Question 5: Approach to and relevance of proposed future research</u>

This project was rated 2.8 for proposed future work.

- No plan was identified. Need to coordinate with overall strategic objectives of the Hydrogen Program to ensure the efforts are "strategic."
- Continues current work. Would like to see plans for expanding services or reaching or communicating capabilities more customers, to increase number of customers served; e.g., what is the continuous improvement plan.
- Planned classes on hands-on training for various techniques.

Strengths and weaknesses

Strengths

- LANL has excellent people and equipment resources. This project has the potential to leverage them in a strategic way.
- A resource that is used by many fuel cell entities.
- Support from LANL.

Weaknesses

- Not clear if the effort is "strategic." Needs to work with DOE program to ensure "clients" have relevant and important issues to solve that are important to the Hydrogen Program. This project should not be a "make work" project.
- The quality of the review presentation was very disappointing. Hard to follow. Did not follow the format. A very half-hearted effort. LANL can do much better than this.
- Possibly the need for more communication of capabilities and increased utilization.
- Currently appears to be limited just to PEM.

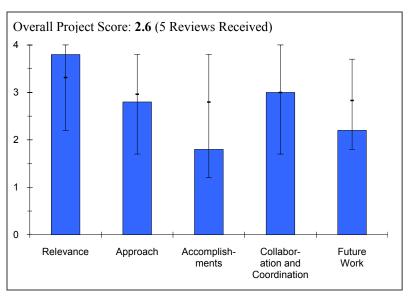
- Develop a strategic test plan in coordination with DOE that targets clients rather than a first come, first served approach.
- Add continuous improvement plan—how will you reach more people each year or what new services will be provided in future year. Also, what is your feedback mechanism (beyond AMR) to determine if the services you are providing are what is, or will, be needed by fuel cell developers?
- Include other fuel cell technologies. Help developers study decay mechanisms, impact of impurity and variability of operating conditions.

Project # FC-59: Improved, Low Cost, Durable Fuel Cell Membranes

James Goldbach; Arkema

Brief Summary of Project

The objectives of this project are to: 1) develop a membrane capable of operating at 80°C at low relative humidity (RH) (25%-50%), 2) develop a membrane capable of operating at temperatures up to 120°C and ultra-low RH of inlet gases at <1.5 kPa, 3) elucidate ionomer and membrane failure and degradation mechanisms via *ex situ* and *in situ* accelerated testing and develop mitigation strategies for any identified degradation mechanism, and 4) use commercially-available matrix materials as a low cost approach.



<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of **3.8** for its relevance to DOE objectives.

- Membrane development is in line with DOE objectives.
- Low cost, durable fuel cell (FC) membranes are necessary to meet DOE cost and durability targets.
- The project is in line with DOE goals and targets. The project targets use low cost, high temperature, and low RH membranes, which would enable system simplification and cost reduction.
- Developing a polymer with good mechanical properties and with high conductivity under hot and dry conditions is relevant to DOE objectives.
- Development of membranes that can operate over a wide temperature range, including 120°C and at low RH is crucial for being able to simplify the balance-of-plant (BOP) and lower the cost and size of the thermal subsystem. This project seeks to address these concerns.
- The project also maintains relevance by attempting a low cost approach and by seeking to understand failure modes associated with chemical and mechanical durability.

Question 2: Approach to performing the research and development

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

- The approach of blending a structural polymer such as polyvinylidene fluoride (PVDF) with a conductive ionomer is valid. However, the loss of conductivity resulting from the low conductivity of the structural polymer must be recovered by the properties of the ionomer. Much of the project has involved an unfruitful search for an ionomer to meet or exceed Nafion®.
- Blending a highly conductive, low cost ionomer with a stable non-conductive polymer is a solid approach.
- The approach to make blends with PVDF may have cost benefits. The approach to make blends with Virginia Tech block copolymers that do not meet the conductivity targets themselves is questionable—blending with PVDF will reduce the conductivity further. Biphenylsulfone: hydrogen form (BPSH) 60 does not meet conductivity targets, so blending with that does not make sense. Crosslinking highly sulfonated (BPSH 100) type materials to improve stability appears to be a better approach.
- The project is well designed and well organized, with a clear focus on simultaneously improving conductivity and mechanical properties. The blending approach is a reasonable approach to take in addressing these issues.
- The approach begins with the attempt to combine a low cost material with mechanical strength Kynar with a conductive polyelectrolyte. In this sense, the project approach is adequate relative to the other membrane projects in the program.

- The approach falls down when the following is considered: why would the polyelectrolytes used be likely to be more conductive than Nafion? No morphological or conductivity data are given to justify why conductivity should be higher, either before or after mixing polyelectrolyte with Kynar.
- Given the low performance shown by BPSH in prior projects, it is unclear how this route would assist with high temperature conductivity.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 1.8 based on accomplishments.

- Very little progress is evident since last year. The performance and conductivity of the Kynar/M70 blend are still significantly below that of Nafion 211.
- The initial scale up of the fabrication process was successful.
- No durability data were presented for M70.
- Unfortunately, the polyelectrolytes are not conductive enough, and the membranes still cannot compete with state-of-art perfluorosulfonic acid (PFSA) membranes at dry conditions. Also, no analysis was given to prove the low cost aspect.
- Arkema has improved performance at low RH, but performance is still below that of Nafion 212. Arkema did not show high temperature (120°C) performance. The project showed some improvement in durability in open circuit voltage (OCV) hold tests and should demonstrate this improvement in RH cycling and membrane electrode assembly (MEA) durability tests as well.
- Although the approach is interesting, the results speak for themselves. In two and a half years of work, the project has yet to generate a material with conductivity as high as the baseline Nafion. Progress has been made in improving the conductivity, but the project is now reaching a dead end. Increasing the acidity of the proton-donating groups is apparently the only route forward for this material, but the chances of success of this route are nil.
- Although progress has been made relative to the past year's results, the fact remains that the improved materials still perform worse than NR211 at all RH at 80°C.
- The most positive aspect of the project is low gas crossover for M70 and M43.
- BPSH-based membranes showed even lower conductivity than the M-series samples.
- Microscopy data were obtained, but it is not clear how these data will lead the way toward a morphology-based conductivity improvement.
- The OCV and RH cycling testing on M43 was completed last year; no new durability testing on M70.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.0** for technology transfer and collaboration.

- Collaborations include a catalyst developer or manufacturer as well as experts in cell testing and microscopic characterization.
- Ionomers from the Virginia Polytechnic Institute and State University (Virginia Tech) were evaluated. Johnson Matthey Fuel Cells, Inc. (JMFC) made the MEAs. ORNL provided transmission electron microscopes (TEMs), but I don't see how that added value to the project.
- The collaborations between partners are good.
- The McGrath group is an excellent choice for collaboration.
- Regardless of results, collaboration clearly exists and is evident throughout the project. No contributors appear to be listed gratuitously.
- The Oak Ridge National Laboratory (ORNL) contributed to understanding morphology, which is a topic that should be well explored in order to understand whether theoretical limitations of conductivity are close to being reached.
- The Virginia Tech collaboration is evident in the provision of alternative ionomers, as well as in nuclear magnetic resonance (NMR) studies.
- The collaboration with JMFC is less obvious. It appears this likely has to do with MEA preparation.
- M70 x-y swelling data appear to be missing.

<u>Question 5: Approach to and relevance of proposed future research</u>

This project was rated **2.2** for proposed future work.

- The project is virtually finished. Testing and characterization will continue.
- There's not much time left. Test M70 for durability.
- The project is set to end in September. The proposed work should bring it to a logical conclusion.
- The future work plans may be reasonable for improving the properties of the existing materials, but the chances of meeting DOE targets using these materials, or any derivatives of the materials that could conceivably be developed before the end of the project, are poor. The materials may be of interest in other applications, but from the standpoint of making progress toward DOE targets for hot and dry operation, the project is at a dead end.
- The future work section mentions some worthwhile efforts such as continued OCV and RH cycling. However, the project has existed for some time, and it would be interesting to know if the project is bumping up against theoretical limitations. Rather than just saying that membrane formulations will be "optimized," it would be refreshing to hear about specific prospects for improving one of these four aspects: 1) morphology to align conductive groups better, 2) acidity of conductive groups, 3) possibility of improving water retention without increasing swelling, and 4) all of the above in the context of minimizing PVDF.

Strengths and weaknesses

Strengths

- The membrane mechanical and chemical durability target achieved with M43 (last year's data). That they even did the tests should be to their credit.
- The potential cost benefit to PFSA is a strength.
- The project combined the resources of some knowledgeable polymer experts to investigate high conductivity without sacrificing mechanical properties.
- Use of a low cost component (Kynar) for mechanical strength is a strength.
- Arkema's willingness to adapt alternative ionomers within the concept of mixing with Kynar is a strength.
- Arkema's prior year use of durability protocols (OCV and RH cycling) for evaluating membranes was impressive.
- The collaborations are used well within the proposed work plan.
- The materials show low gas crossover.
- Arkema's reporting of in situ diagnostics (high frequency resistance (HFR) and hydrogen crossover) for in situ tests is a strength.

Weaknesses

- The ionomer chemistry was not disclosed, so the FC community cannot benefit from the findings.
- Blends have had poor conductivity.
- Arkema failed to incorporate alternative development pathways. The primary strategy for achieving high conductivity under hot and dry conditions has failed. A path forward has not been demonstrated.
- There has been continued optimization without really attacking the issues regarding the boundaries of proton conductivity.
- Arkema is still not obtaining the desired conductivity measurements.
- There are likely better alternative ionomers that could be used than the BPSH series. The concept may work better with other polyelectrolytes.

- It is too late to change the scope. Provide some cost estimate to justify the basis for the low cost claim.
- With the limited time available, all durability and FC testing work should be curtailed, and resources should be oriented toward increasing conductivity.

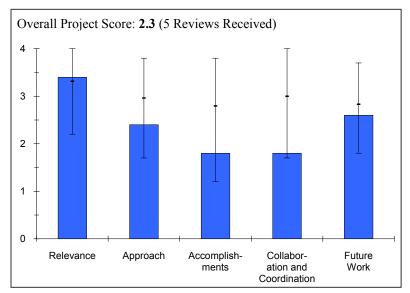
- The project could benefit through interaction with a theoretical group that could predict conductivity as a function of morphology, acidity, water uptake, etc. This would help to avoid studies with ionomers that will not perform well at low RH.
- Efforts involving FC polarization tests or production scale up could be minimized in favor of providing a full understanding of what material properties and components can be used to increase conductivity without sacrificing crossover or durability.

Project # FC-60: Protic Salt Polymer Membranes: High Temperature Water-Free Proton-Conducting Membranes

Dominic Gervasio; Arizona State University

Brief Summary of Project

The objective of this program is to make a solid water-insoluble anhydrous protonconducting electrolyte membrane that has: 1) good membrane performance, including a) proton conductivity that surpasses the 2009 target of > 0.1 S/cm at 120°C and 50% relative humidity (RH), b) effectively having no co-transport of molecular species with a proton, and c) good mechanical strength and chemical stability; and 2) good electrode performance, with the assisting catalyst to promote reduced activation overvoltage.



<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of 3.4 for its relevance to DOE objectives.

- Membranes with high conductivity at high temperature and low RH are relevant to DOE objectives.
- The development of an anhydrous proton conductor could enable significant system simplification and cost reduction of polymer electrolyte membrane fuel cell (PEMFC) systems.
- The project is mostly aligned with DOE targets and goals. High temperature membranes are an enabling technology that will allow system simplification and cost reductions.
- The project mostly aligns with DOE objectives. The ionic liquid studies are not directly relevant to the program, but to the extent that these studies could lead to immobilized ionic conductors, they are still relevant. The relevance is limited by the poor progress to date—a project that doesn't meet objectives or advance understanding cannot be said to be relevant.
- The DOE Hydrogen Program is still in search of membranes that can operate at reduced humidity and wider temperature range, including temperatures as high as 120°C. The intention of this project is to fabricate membranes capable of both.
- There are no aspects of the project that deviate from the intentions of the DOE program. Both low water and no water proton conduction are relevant to the intentions of simplifying the balance-of-plant (BOP) and reducing the cost and size of the thermal loop.

Question 2: Approach to performing the research and development

This project was rated **2.4** on its approach.

- The approach has changed dramatically since the beginning of the project from protic ionic liquid (PIL) to solid conductors based on indium tin phosphate (ITP). The ITP has excellent conductivity, but is brittle and somewhat porous. These issues are being addressed by adding a polymer to seal the pores and provide flexibility.
- The doped ITP should be the sole focus for the rest of the program.
- PILs present a unique approach to developing anhydrous proton conductors. Although the focus seemed to switch to ceramic proton conductors, such as ITP blended with organic polymers. More focus on actually making membranes is required.

- The idea of immobilizing ionic liquids for proton conduction at low RH is good. Work on non-platinum catalysts and FCs with PILs is outside the scope of high temperature low RH membranes. The work lacked focus on developing membranes and barriers to developing membranes that conduct at low RH.
- The project has lacked a clear and consistent focus. The lack of a well-organized approach has decreased the prospects that relevant and useful materials or discoveries will be produced.
- The approach seeks to generate proton-conducting membranes that require little or no water by deriving materials from functional groups found in PILs. Since PILs are known to have high proton conductivity, and since it may be possible to make membranes that are water insoluble, the approach is well conceived.
- Although it is reasonable for the approach to screen samples with thermal gravimetric analysis (TGA), there is no mention of durability experiments given a successfully conductive membrane.
- There is also no mention of preliminary ionic liquid conductivity that would be prerequisite to fabricating a membrane derived from the given ionic liquid.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 1.8 based on accomplishments.

- The original protic liquid approach was not successful in meeting project and DOE technical targets.
- Progress demonstrating the doped ITP has been slow.
- Liquid PILs do not meet DOE performance targets. One would not expect an improvement with ILs tethered to solid polymer backbones.
- Ceramic (ITP) shows good conductivity but cannot be made as an impermeable membrane. ITP polymer blends have lower conductivity. ITP has not been proven to be stable in an FC environment or even liquid water. No conductivity vs. RH data or mechanical properties was shown.
- Immobilized ionic liquids had poor conductivity. ITP showed some promise and had conductivity that met intermediate targets, but it does not have good mechanical properties. No improvement was made within the past year on improving mechanics or conductivity.
- No independent testing or verification of ITP conductivity was presented. There was no discussion or mention of durability of this material under FC conditions (RH and potential cycling).
- It is not clear that any of the materials developed and investigated will prove applicable to FCs. The project has yet to demonstrate a membrane capable of meeting the 0.02 Ω-cm² at 120°C at 40-80 kPa water target, discounting the "pure" ITP material, which does not constitute a membrane. Having failed thus far to demonstrate achievement of the earlier 0.1 S/cm at 120°C and 50% RH in a membrane (again discounting non-membrane ITP material), the project is more than one year behind schedule, and odds of achieving even this interim target before the end of the project are low.
- The 2-fluoropyridinium triflate (2-FPTf) was touted as being a stable ionic liquid, but conductivity is too low for applications that must operate at times around 40°C (~ 6x10⁻³ S/cm). Polarization curves bear this out and also show mixed potential possibilities on catalysts at lower temperatures.
- Conductivity of ITP at low temperature is also too low (0.01-0.03 S/cm). ITP is to be blended with polyphosphazenes, but polyphosphazene conductivity does not appear to be reported.
- An attempt was made to report higher oxygen reduction activity in ionic liquid presence. However, the experiment was not noted to be a rotating disk electrode (RDE) (no revolutions per minute (RPM) value shown) and no limiting current reference could be used to understand the four electrons versus two electrons reduction. Mixed potentials were evident on polarization curves, which could imply that reduction currents could be associated with desorption of ionic liquid species from platinum, not actual oxygen reduction.
- In general, the project should be demonstrating more progress at this stage with respect to membrane conductivity, and all the properties expected with a membrane sample (swelling, gas crossover, durability, etc.).

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated 1.8 for technology transfer and collaboration.

- Collaborations are with strong companies, but their public history in PEMFCs for automobiles is small.
- A membrane electrode assembly (MEA) or stack developer might prove beneficial.
- It's not clear at all what role the subcontractors play.

- It is not clear what Boeing's role was. Collaboration between other partners was good.
- The role of collaborators in the project is unclear. A "collaborations" slide was included, but based on the content of the rest of the slides, it seems that this is basically a solo effort.
- Technical roles for the University of Akron and Boeing are not entirely clear from the presentation materials, except for the fact that they are mentioned as collaborators.
- Interactions with the project have not evidenced a collaborative spirit. Instead, suggestions have been cast aside to find ILs or ceramics that demonstrate high conductivity throughout the entire temperature range of interest (including temperatures as low as ambient).
- Beyond Akron or Boeing, there is little evidence of collaboration beyond submissions to Bekktech and University of Central Florida (UCF).

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- Doped ITP will be further studied and tested.
- Making and testing membranes of ITP blended with organic polymers such as polyphosphazene should be interesting. Future focus should be on conductivity, hydrolytic, and mechanical stability of these materials.
- Plans to look at blends or composites of ITP and polymers or stabilized ionic liquids may help overcome mechanical issues.
- The project is unlikely to meet the original goals, but under the circumstances, if work is continued on the project, then the proposed work is reasonable. An emphasis should be placed on validating ITP results, given the revelation that free phosphates played a role in earlier, apparently erroneous, reports of high conductivity in neat ITP.
- The future work points to ITP polymer blends as a possible material with which to deliver membrane results. Unfortunately, the conductivity of ITP itself is low at lower temperatures. The prospects of increasing the conductivity with use of a polyphosphazene are questionable.
- The evaluation of platinum surface area in the context of ITP suggests that somehow ITP could be expected to increase surface area. The intention of whether the ITP is to be used as an ionomer or not is unclear. Otherwise, this activity does not seem reasonable, and it does not speak to the specific activity of the catalyst or Tafel phenomena. Again, it was not made clear in the results that the reduction current observed in the presence of ionic liquids were not related to desorption processes or 2-electron reductions.

Strengths and weaknesses

Strengths

- The unique approach to making anhydrous proton conductors separates this project from others in the high temperature membrane working group (HTMWG) program.
- The PI has some interesting and unconventional ideas. It is nice to see outside-the-box thinking.
- The initial approach of attempting to create membranes from immobilized ionic liquids was well founded.
- Prior to fabricating membranes, the project attempted to screen conductivity of some of the conductive species.
- The project appears to have a certain degree of curiosity about interactions of the conductive species with other parts of the MEA.

Weaknesses

- There were poor collaborations. The project could benefit from more expertise on polymer chemistry and membrane processing. There were no responses to reviewers' comments from previous years.
- The project lacked focus. Immobilized ionic liquids had low conductivity.
- The approach was scattered and disorganized, and the experimental work seems sloppy (free phosphate in ITP that in earlier reviews was asserted to be free of such residual species).
- The project has not focused on either delivering membranes or delivering a verdict against the use of proposed materials. Instead, it attempts to make the best of what it has, despite the fact that further development may lead to a dead end.

- Screening measurements—such as proton conductivity—on conductive species have not been interpreted as motivation to look at more conductive materials. Instead, membrane fabrication and FC testing are carried forward for materials that are not worthwhile.
- Deeper interpretation of kinetic data is needed to make the case that ionic liquids assist with oxygen reduction.

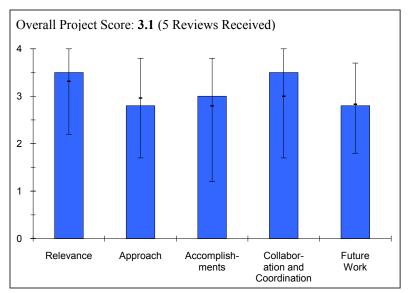
- Try blending ITP with Nafion[®]. That may at least help with low temperature conductivity. Conduct more stability testing and test the mechanical properties of membranes. Hold off on FC testing until conductivity and stability are proven. Test conductivity vs. RH.
- The value of this project to the hydrogen program is questionable. None of the approaches being pursued are likely to contribute significantly to the objectives of the program, so everything here looks like a good candidate for deletion, starting with the non-platinum catalyst component.
- If there were enough time remaining, some degree of proton conductivity modeling (as function of temperature) might be helpful toward directing the project to more promising species.
- Collaboration with electrocatalyst experts, such as Argonne National Laboratory personnel, would be helpful toward understanding whether the reverse sweep currents from voltammetry in oxygen are meaningful.
- In general, continued surveying of conductivity throughout a wide temperature range is useful. In the meantime, FC testing could be eliminated until worthwhile membranes are fabricated.

Project # FC-76: Biomass Fuel Cell Systems

Neal Sullivan; Colorado School of Mines

Brief Summary of Project

Overall objective for this project is to improve durability and performance of solid oxide fuel cell (SOFC) systems while lowering costs. Task 1 is to develop SOFC materials for robust operation on biofuels, including sulfur- and redox-tolerant materials to broaden SOFC operating windows and nickel-free, perovskite-based anodes with novel cell architectures. Task 2 is fuel processing of bio-derived fuels, including methane from anaerobic digesters of waste-water treatment plants, fuelprocessing for biomass-derived liquid fuels (butanol), and decreasing the cost of fuelprocessing balance-of-plant (BOP) hardware. Task 3 includes modeling and simulation to: 1) develop chemically



reacting flow models of fuel-processing hardware, 2) use model-predictive controls to integrate system hardware, 3) conduct thermal modeling of hot-zone system components, and 4) employ system modeling to explore the benefits of balance of planar-component integration.

Question 1: Relevance to overall DOE objectives

This project earned a score of 3.5 for its relevance to DOE objectives.

- This is a really excellent project. It supports biomass to hydrogen, which is a very important hydrogen pathway.
- Colorado School of Mines (CSM) is looking at a 1-kW system using renewable hydrogen sources.
- CSM stated the application was for military 1- kW battery charger and/or generation set. DOE should not be funding work for the military. I doubt this is their only application. CSM needs to be clearer about their applications. In addition, this size range is on the border line for use of non-jet propulsion type 8 (JP8) fuels. If CSM scales it up (which they should), the Department of Defense will want to use a real logistics fuel in their proposed application.
- This technology is in line with DOE program objectives, as it directly supports SOFC development, while also providing expertise and training at university settings.
- The FC section of the Multi-Year Project Plan (MYPP) on the Fuel Cell Technologies Program (FCT) website indicates the program goal is to "develop and demonstrate FC power system technologies for transportation, stationary and portable power applications." This project is supportive of that goal; however, the MYPP objectives supporting that goal do not relate well to this project and appear quite dated. The technical approach section in the MYPP indicates that SOFC work is pursued under the Solid State Energy Conversion Alliance (SECA).

Question 2: Approach to performing the research and development

This project was rated 2.8 on its approach.

- The project has developed novel ceramic heat exchangers. There is extensive use of modeling, both as part of the design effort and then for conducting validation based on experimental results. Excellent collaboration with W.L. Gore & Associates (Gore) on ceramics really leverages both the monetary investment by DOE, but, more importantly, brings the expertise of Gore to bear on solving the problem.
- CSM has a solid team.

- CSM was unaware of others who had done work using ceramic microchannel reactors previously.
- CSM does not discuss the sealing issue, which will be critical for this project, especially since their target applications will be thermal cycling.
- Anaerobic biogas has a lot of impurities. CSM needs to discuss how they will handle the impurities, especially sulfur.
- Partial oxidation reactors have in the past produced small amounts of ammonia. CSM needs to analyze their reformate for this.
- CSM is not using water in their reformer. If the reformate is run to the reactor dry, it will coke up eventually.
- CSM is using a tubular SOFC configuration. CSM will have power density problems, which is especially important for transportable power generation. The low power density will be exasperated by their catalyst selection, which has never demonstrated high power in the literature.
- This approach is quite interesting. However, the mass and energy balance of the system was not shown. It was unclear why partial oxidation was used and what product market the device is pursuing.
- There has been considerable strontium titanate-based SOFC anode work done elsewhere, including SECA at the Pacific Northwest National Laboratory (PNNL). There does not appear to have been a thorough assessment prior to defining this work. Furthermore, R&D should focus on the technical or cost barriers to commercial viability, such as the cost or performance advantages of the work which, if successful, should be quantified to include specific targets; e.g., desired hydrogen sulfide tolerance, etc.).Similarly, the anaerobic digester gas (ADG) reforming work is not novel and does not advance the state-of-the-art, as the technology is readily available and in use (e.g., FuelCell Energy's (FCE's) molten carbonate fuel cell (MCFC) installations). The butanol work is more interesting; however, here again there is a solid base of existing R&D (injector design, catalyst development, etc.) looking at reforming pump diesel and JP8 (far more difficult fuels). It would be logical to evaluate butanol reforming using state-of-the-art technology prior to developing a R&D plan.
- High temperature heat exchangers are a high cost element of the SOFC system. BOP super alloy-based heat exchangers (HEX) are very expensive on a \$/kW basis, with questionable life. Ceramic HEX have been pursued by others; e.g., Blasch and Acumentrics in their hybrid design, among others. A cost target (\$/kW net system) should be established, and the actual projected unit cost used as one metric to track technical progress. The work to date has shown nominal manufacturability and technical viability. The attempts to integrate HEX/reformer dilute the effort and are overly aggressive—proving that the suitability of HEX function alone would be an important success.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.0 based on accomplishments.

- CSM has achieved excellent results in a short time frame. Some problems have been identified, but solutions have also been developed.
- The modeling work is completed.
- They have some initial data on their reformer.
- This is a newer project, so only a limited amount has been done.
- I was surprised that they did not know about other ceramic reactors and heat exchangers that have been built. They need to do a better literature review so they can learn from what others have done.
- The FC testing has all been short term. It will be interesting when they begin some long-term testing.
- Transportable generators need high power density, which was not demonstrated.
- As stated above, the effort does support the DOE/EERE goal as delineated in the latest MYPP. However, the technical and cost targets and justification for the individual elements of this project are ill-defined. There is not a clear relationship between commercialization barriers and the "problems" being addressed within this project.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.5** for technology transfer and collaboration.

- There is close collaboration with Gore. A great relationship that has been competently leveraged.
- They have good collaboration with Coorstek.

- They need to contact Ceramatec (part of Coorstek) to learn about the tape casting and reactor design experience that Ceramatec has.
- The university was working closely with Coortek.
- CoorsTek is a well-regarded ceramics company and is an excellent partner/supplier for the ceramic HEX components. I understand that many of the details regarding the manufacturing and cost are held proprietary, lessening the value of the overall effort.
- Other organizations; e.g., PCI, could be approached with respect to diesel reforming and its applicability to butanol.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.8 for proposed future work.

- The approach is very focused and aggressive.
- They need to develop a critical path to be able to achieve their goals. SOFC with perovskite electrodes have been demonstrated.
- They do not mention sealing. This will be more difficult than they think, especially if they are going to thermally cycle the heat exchangers and SOFC, which their stated applications demand.
- They need to address the contaminant issues. There is nothing in their future work dealing with this.
- Design and construction of a 1 kW SOFC stack will be difficult. This project is over ambitious for the funding amount.
- The ceramic HEX work addresses a substantial need with respect to low cost SOFC systems. CSM can address both manufactured cost and robustness or reliability. See previous comments with respect to the lack of clear quantified technical objectives or targets.

Strengths and weaknesses

Strengths

- Great leverage of partner resources. CSM has a focused plan that is well executed. CSM is hitting their targets and have good leadership.
- Colorado School of Mines has some very good modelers with experience modeling microchannel reactors and heat exchangers.
- The team is strong.
- Coorstek has great ceramic capabilities.
- The project team appears to have excellent facilities and equipment, as well as capable personnel.
- In particular, the ceramic HEX work with CoorsTek's participation addresses a critical need for high temperature FC systems.

Weaknesses

- Their system efficiency would be higher if they used steam reforming.
- They need to develop a critical path.
- The sealing of ceramics is very difficult. They need to start work on this immediately. If they have the technology for sealing, they should show data on thermally cycling it.
- Tubular SOFC have low power density, as does the catalyst they have chosen.
- The effort appears diluted given the funding—anode development, biofuel reforming (ADG and butanol), ceramic HEX, and modeling. Additional focus on specific objectives, with respect to the ceramic HEX for example, could yield more definitive results.
- The project elements on anode development and fuel processing are not positioned to advance the state-of-theart.
- The project lacks definitive, quantifiable technical and cost targets that tie to program-level metrics; e.g., \$/kW, desired fuel and associated sulfur tolerance, etc.

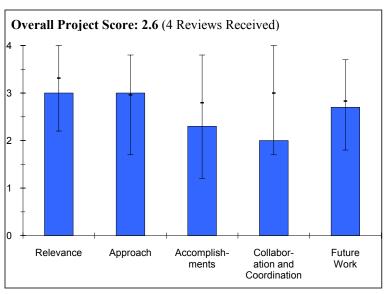
- Provide more funding and turn them loose.
- CSM needs to include impurities in their biogas reforming or explain how they will clean the gas.
- They need to work on the sealing of ceramics. CSM needs to demonstrate the ability of their ceramic reactors, heat exchangers, and stacks to thermally cycle.
- CSM is going to coke their reactor unless they add some water to the system either by pumping it in or doing anode gas recycle, but CSM is not planning on either. This issueneeds to be addressed.
- It was unclear what market segment this device is pursuing. The presentation was for a nearly complete system. It would be beneficial to focus the development with some market study and understanding of product specifications being pursued.
- If this is a biomass FC system, it would be needed to show why partial oxidation was selected. We did not see the performance of the partial oxidation portion of the process. Process flow diagram and mass and energy balance would be very useful to have. Additional consideration of BoP is necessary to provide guidance on product pressure drop and efficiency optimization.

Project # FC-77: Fuel Cell Coolant Optimization and Scale-up

Satish Mohapatra; Dynalene

Brief Summary of Project

The overall objective of this project is to optimize and then scale-up the process to make Dynalene fuel cell (FC) coolant with a great deal of reproducibility. One key ingredient of the coolant (a nanoparticle) will be produced in 100 L batches in a pilotscale operation and the effect of various process parameters on the size and charge density of the particles will be determined. Another technical objective of this project is to optimize the filtration process for the nanoparticles to minimize the cleaning time for different-scale operations.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.0** for its relevance to DOE objectives.

- This project directed toward the development of a long-term, low electrical conductivity coolant for FC operation is relevant to improving FC performance and durability.
- This project addresses FC system thermal and water management issues, which is a barrier listed in the Multi-Year Program Plan (MYPP).
- The coolant is applicable for future elevated temperature polymer electrolyte membrane (PEM) operation up to 122°C.
- The project addresses the scale up of an already developed coolant. Hence, it has high industrial relevance.

Question 2: Approach to performing the research and development

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

- This project builds on Small Business Innovation Research (SBIR) Phase I and II projects, and the current approach is directed toward scaling up the production of the developed coolant. It is a very focused approach to prove the availability of this product on a timely schedule for expanded application by FC suppliers.
- The project uses a low-conductivity fluid consisting of glycol, water, corrosion inhibitors, and nanoparticles to prevent the buildup of ions in coolant fluid. This allows the system to run without a deionizing filter, decreasing complexity and cost. The nanoparticles were not identified, so the effect of these materials on cost cannot be evaluated. Also, once the nanoparticles become saturated, the ionic concentration in the fluid will rise. This could be a major flaw. The emphasis in this project is on smaller FCs, 1-100 kW.
- The PI was not able to come and it was not possible to get all technical details answered by the substitute.
- A technical approach for the nano-particle production is taken. Such work is needed and often underestimated in order to attain stable production processes.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.3 based on accomplishments.

• The progress to date seems to be adequate, although past progress was supported largely by SBIR funding.

- Started out as a Phase I and II SBIR project. The current goal is to scale up from 0.5 L to a 10-100 L pilot plant. Reviewer would have expected the project to be at this point after the Phase II SBIR. It appears to have optimized the coolant fluid composition, although this was not presented.
- The progress seems to be good since a pilot plant is under development. Little to no technical information about the progress is provided. For that reason, the progress cannot be evaluated and was rated fair.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **2.0** for technology transfer and collaboration.

- Collaboration with a university partner (subcontractor) was noted, but the role of the partner or nature of the collaboration was not apparent.
- No collaborations were identified.
- At this stage, the project does not need extensive collaboration. The existing collaboration with Lehigh University seems to be sufficient.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.7 for proposed future work.

- The future work plans focus on demonstrating the scale-up of the coolant production and are consistent with the project goals and objectives.
- The goal is to scale up to a 10-100 L plant by the end of August and resume research at these larger scales.
- Too little information is provided in order to evaluate the future work.

Strengths and weaknesses

Strengths

- They have experience based on previous SBIR support.
- This is a potentially valuable coolant that will be more compatible with smaller FCs and can simplify the system.
- They are very advanced in their status and are close to production.

Weaknesses

- None apparent.
- The saturation of nanoparticles is potentially a limiting factor. No cost comparison with other coolants was provided. One would need to see a cost comparison with standard coolants (using a deionizing filter) and see long-term test data to fully evaluate coolant effectiveness.
- No original equipment manufacturer (OEM) or system integrator data about the acceptance of the coolant was provided. No cost information was provided. No benchmark to existing cooling oils was provided.

- Independent validation and performance evaluation in an operating FC system by a National laboratory would help to establish the claimed benefits of this coolant.
- If the project is to continue, it should get a partnership with an FC manufacturer.
- No information about potential applications was provided. No tests by system integrators or OEMs were quoted. It should be made clear that the fluid really suits coolant requirements, a fact that is stated by the developer, but not proven by any facts in the poster.