2015 — Fuel Cells
Summary of Annual Merit Review of the Fuel Cells Sub-Program

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

Reviewers felt that there was a good balance between near-, mid-, and long-term research and development (R&D) in the Fuel Cells sub-program. Reviewers agreed that cost and durability are the major technical challenges and praised the sub-program's approach to identifying and addressing these issues. Reviewers also agreed that current year successes and overall sub-program progress were clearly described. However, some reviewers commented that this year's successes were not explicitly benchmarked against the prior year to demonstrate progress in specific technologies. A strength of the sub-program, as noted by most reviewers, is its well-structured, focused, and well-managed projects. In particular, reviewers commented that the catalyst cost reduction projects were exceptionally strong this year. Reviewers praised the sub-program's robust project teams and identified the sub-program's collaboration with academia, industry, the national laboratories, and the community—for example, via the U.S. Council for Automotive Research—as crucial in focusing R&D on the most relevant challenges. Some reviewers expressed a desire to see more validation in single-cell or short-stack experiments in addition to the excellent materials advances achieved so far.

Fuel Cells Funding:

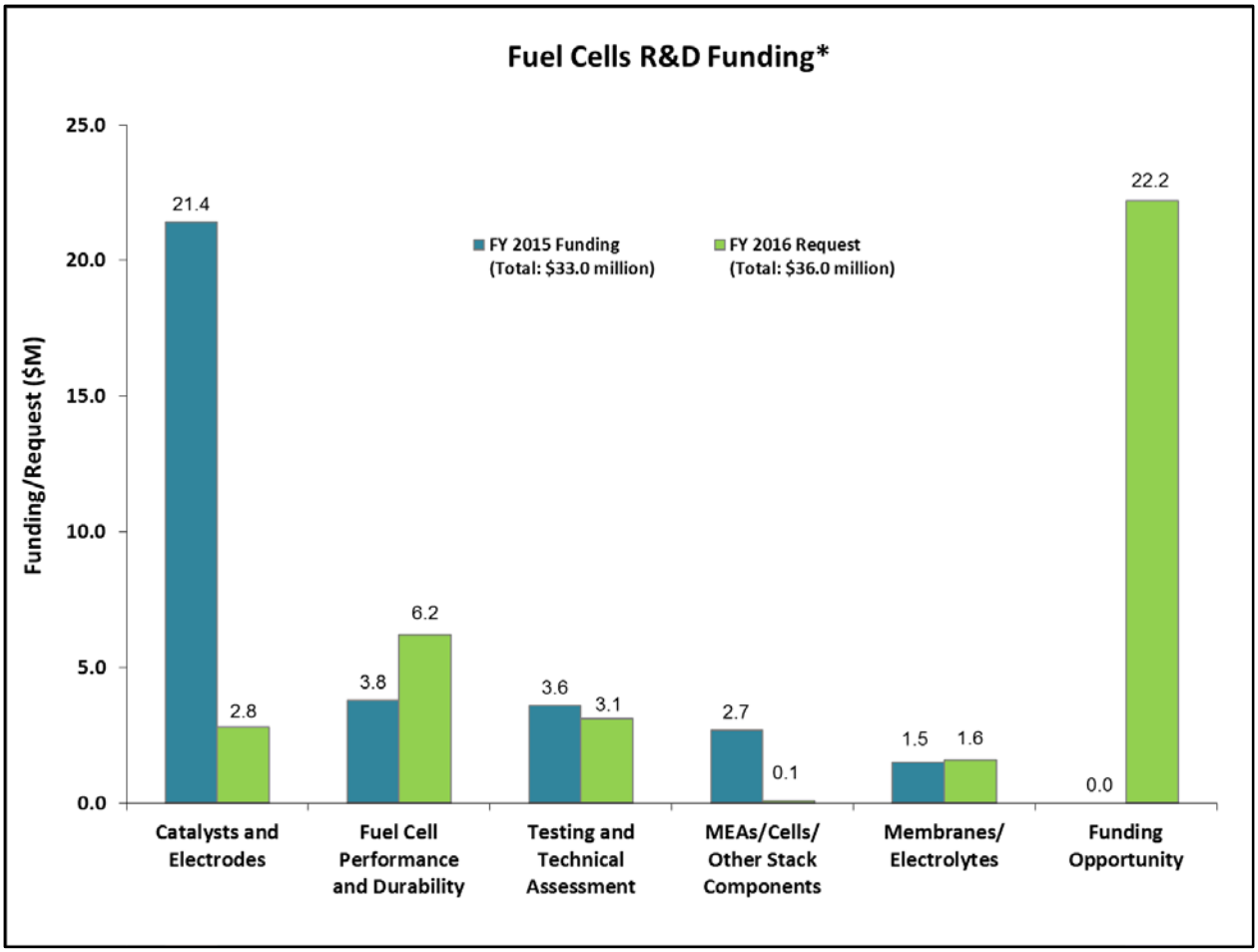
The sub-program received $33 million in fiscal year (FY) 2015, as shown in the chart on the following page. The request for FY 2016 is $36 million. The sub-program focuses on reducing fuel cell costs and improving durability. Efforts include approaches that will achieve increased activity and utilization of platinum-group-metal (PGM) and PGM-alloy catalysts, non-PGM catalysts for long-term applications, ion exchange membranes with enhanced performance and stability at reduced cost, improved integration of catalysts and membranes into membrane electrode assemblies (MEAs), and a better understanding of degradation mechanisms and mass transport. The FY 2015 funding opportunity announcement will result in $12 million in funding for new catalysts/electrodes projects. In addition, $1.8 million in FY 2015 funding will result in five new projects under the incubator program. There is no funding in FY 2015 or planned funding in FY 2016 for balance-of-plant (BOP) components projects.

Majority of Reviewer Comments and Recommendations:

At this year's review, 48 projects funded by the Fuel Cells sub-program were presented, and 34 were reviewed. Projects were reviewed by between three and eight reviewers, with a median of seven experts reviewing each project. Reviewer scores for these projects ranged from 2.7 to 3.6, with an average score of 3.1. This year's highest score of 3.6 and average score of 3.1 were similar to last year's highest and average scores of 3.6 and 3.2, respectively. The lowest score of 2.7 for all projects reviewed in 2015 was a modest improvement over 2014's low score of 2.6.

Catalysts and Electrodes: The scores for the 10 catalyst projects ranged from 2.7 to 3.4, with an average of 3.0. Reviewers praised the highest-rated project for its approach to modeling and characterization of a novel engineered ionomer topology within the catalyst layer, as well as the progress made with the model. In addition, reviewers recognized the importance of reducing fuel cell cost by reducing platinum (Pt) loading in this project. For the lowest-scoring project, which tied for the lowest score in the sub-program, reviewers felt that the results for the selected non-PGM catalysts do not yet meet the performance requirements for use in fuel cells, and that the project should use characterization techniques beyond rotating disk electrode measurements.

Fuel Cell Performance and Durability: Four projects were reviewed, with three projects receiving a score of 3.3 and the remaining project receiving a score of 3.0, for an average score of 3.2. Reviewers praised the highest-rated projects for the strength and design of their approaches, their relevance to and alignment with sub-program goals, and their strong teams and collaboration. In addition, reviewers commented that these projects provide results that industry could use immediately and lauded the use of both in situ and ex situ experimental techniques. However, reviewers recommended that the projects use more state-of-the-art materials and that the researchers review their use of serpentine flow fields, where appropriate. Reviewers felt that the lower-scoring project's approach is



∗ Subject to appropriations, project go/no-go decisions, and competitive selections. Exact amounts will be determined based on research and development progress in each area and the relative merit and applicability of projects competitively selected through planned funding opportunity announcements.

reasonable, addresses known durability issues in polymer electrolyte membrane fuel cells, and relies on effective collaboration; however, they expressed concern that the project does not demonstrate or elucidate an adequate understanding of the mechanisms behind the contaminants, and that it does not use MEAs with state-of-the-art Pt loadings.

Testing and Technical Assessment: Eight projects were reviewed and received scores between 3.0 and 3.6, with an average score of 3.4. The highest-rated project in this topic area was the highest-rated project in the sub-program. Reviewers stated that the project is well integrated into the sub-program, providing a critical capability that consistently delivers excellent progress from a highly collaborative team. While the reviewers generally thought the lowest-rated project demonstrates excellent analysis and collaboration, they identified a number of aspects that are inconsistent with real-world applications, and some questioned the project's contribution to cost reduction and customer acceptance.

MEAs, Cells, and Other Stack Components: Four projects were reviewed in this area, with three projects receiving a score of 3.1 and the remaining project receiving a score of 2.7, for an average score of 2.9. Reviewers felt the highest-rated projects are well aligned with the sub-program goals and have strong team members who collaborate well. Reviewers noted that these projects either demonstrate good progress toward the sub-program goals or contribute important findings that are relevant to the fuel cell community as a whole. For the lowest-scoring project, which tied for the lowest score in the sub-program, reviewers were impressed with the project team's ability to quickly engineer, develop, and deploy a semi-automated system for MEA fabrication, but they expressed serious

reservations regarding the project's performance on hydrogen/air and recommended identifying the right industry collaborators to resolve these performance issues before proceeding.

Membranes/Electrolytes: The seven membrane projects reviewed received scores between 2.7 and 3.5, with an average score of 3.1. Reviewers found the highest-rated project to be an effective collaboration among very strong team members that uses a systematic approach to address a challenge relevant to the cost, durability, and performance challenges faced by the sub-program. They noted that the project has met most of its milestones, including a key achievement of 0.1 S/cm at 50% relative humidity and 90°C. Reviewers expressed concern that insufficient details were revealed about the lowest-rated project's approach, prohibiting reviewers from making an accurate assessment of the project's approach or results. They were impressed with the conductivity but concerned that it came from thick membranes for which the project team had done limited or no mechanical properties or stability testing.

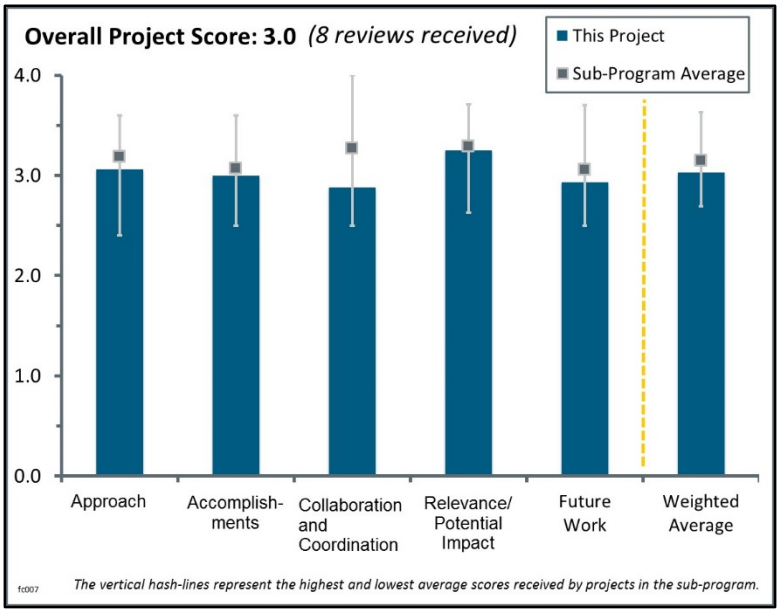
BOP Components: One project, development of a compressor/expander unit, was reviewed in this topic area. It received a score of 2.8. Reviewers regarded the focus of this project as very important for successfully launching fuel cell electric vehicles. While reviewers lauded the team's collaboration and expressed interest in validation with a full system, they expressed a greater concern that the project has not met the targets, that there did not appear to be a plan to meet the targets, and that the validation planned did not include the expander.

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

Bryan Pivovar; National Renewable Energy Laboratory

Brief Summary of Project:

National Renewable Energy Laboratory (NREL) has pursued synthesis of novel extended thin-film electrocatalyst structures (ETFECSs) for improved cost, performance, and durability. A focus is on increasing Pt mass activity and increasing durability via post treatment processing. NREL will then incorporate these novel cathode catalyst materials and structures into membrane electrode assemblies (MEAs) to meet U.S. Department of Energy (DOE) technical targets for fuel cell cost, performance, and durability.



Question 1: Approach to performing the work

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

- The approach this year, focusing on the use of several post-treatments and their combinations and interactions, was well performed and understood. There was enough discussion on why the treatments were chosen, and the method and new ideas during the process were excellent. The only part for which there was not enough justification was the reason for choosing Pt/Ni instead of other catalysts. It is clear that the principal investigator (PI) had to make a choice, as it is impossible to test all the catalysts, but some other catalysts with similar mass activities could be evaluated too.
- With the project almost completed, the team appears to have made significant progress toward making electrocatalysts with potential to meet DOE's goals. The annealing approaches appear to have been successful in addressing durability issues. The only concern is that the team pursued a non-scalable galvanic displacement process to demonstrate feasibility and now has only a few months remaining in the project to demonstrate targeted performance and durability with electrocatalysts made with the more scalable atomic layer deposition (ALD) process. Hindsight is 20/20, but it may have been more appropriate to have initiated work with the ALD process much earlier.
- The current approach is very good overall. The nanowire electrocatalyst approach has led to very high activity and durability, key fuel cell commercialization barriers. One issue with the approach is that most activity and durability characterization appears to be occurring in rotating disk electrodes (RDEs) and not in membrane electrode assembly (MEA) electrodes.
- The concept of nanowire as catalyst/support is sound since it promotes charge/mass transport and, in principle, improves the platinum utilization. Using a transition metal-based core is expected to produce a more stable catalyst in terms of oxidation corrosion resistance compared to carbon. However, the potential for membrane performance degradation by leached transition metal ions, such as Ni^{+2}, should have been a major consideration. The PI should have elaborated on the design principle and scientific hypothesis, particularly the concept and steps taken to address concerns raised during last year's DOE Hydrogen and Fuel Cells Program Annual Merit Review (AMR).
- Using inexpensive commercial Ni noodles decorated with Pt was originally an interesting and worthwhile idea. The majority of the work over the years appears to have focused on RDE testing. Despite good RDE results, transitioning to MEA testing has been slow and late, given the time frame of the project. The fiscal year (FY) 2014 MEA results appear a bit haphazard, and since it appears that the FY 2015 results require

further catalyst treatments to provide suitable results, it is clear that the catalysts should have seen MEAs earlier and more forcefully in the effort.

- It is an effective approach to increase the electrochemically available surface area (ECA) in the electrochemical solution and achieve good durability. However, the project needs to prove the reproducibility to overcome transition metal ion contamination of the MEA and increase the ionomer/catalyst interface or effective ECA in an MEA in actual fuel cell operation between anode and cathode.
- The approach of making extended, continuous Pt nanostructures has been shown effective in 3M's nanostructured-thin-film-type catalysts in typical 25- and 50-cm^2 single cells. However, it is not clear how the team will solve the issue of obtaining a pinhole-free extended Pt surface having high mass activity and stability under accelerated stress test conditions. It is unclear from the approach how the 2020 DOE target for platinum group metal (PGM) total content can be achieved with the 0.2 mg Pt/cm^2 currently used in 5- and 25-cm^2 cells shown in slides 16 and 17.
- The project is structured around three sequenced stages to (1) improve catalyst mass activity, (2) demonstrate fuel cell performance with the new catalyst, and (3) optimize the fuel cell fabrication techniques.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The high mass activity materials were developed by galvanic displacement in 2014 but showed poor durability. Therefore the work of the third and fourth quarters (Q3–Q4) of 2014 and Q1 2015 focused on addressing durability concerns of transition metal ion contamination by acid leach followed by O_2 anneal of the platinum nanowire (PtNW) catalysts. ALD processing for Pt alloy NW synthesis was investigated and resulted in faster and easier synthesis over galvanic displacement. Yet the specific activity of Pt alloy NW is not as high as with galvanic displacement. The resulting MEA exhibited higher specific activity than, and similar performance to, reference Pt/C, and fuel cells with PtNiNW showed greater stability over reference Pt/C.
- Significant progress has been made in evaluating the mass and specific activities of ETFECS catalysts both in rotating ring-disk electrodes and MEAs. It would be interesting to know the H_2/air fuel cell performance of MEAs subjected to 0.6–1.0 V and 1.0–1.5 V cycling protocols. However, considering the current project end date of September 2015, the team may not have sufficient time to engineer MEAs for H_2/air fuel cell operation unless the project is extended.
- There has been excellent progress toward generation of high mass activity electrocatalysts that are also durable against potential cycling. Progress toward high-performance hydrogen/air electrodes seems to be substantially lacking; the key focus for future work needs to be electrode integration. Use of RDEs for durability is less aggressive than in MEAs, at least in part because of the substantially reduced cell temperature (room temperature rather than 80°C, as specified in the DOE protocols). Stability against the DOE support cycle should also be assessed and reported.
- In electrochemical solution, the increased ECA leads to high mass activity. The leached catalyst shows good durability. Progress has been made for catalyst durability in the MEA electrode in H_2/N_2. The actual fuel cell operation data will be required to demonstrate the catalyst's performance not only for activation polarization but also for the ohmic and mass transfer polarization so that the contamination issue can be fully understood.
- The team has established a new class of electro-catalysts with potential for meeting DOE's performance and durability targets. However, the performance and durability needs to be replicated with materials made by scalable processes; otherwise, the project will have no lasting impact.
- Judging simply from the results achieved and presented, the project has achieved the DOE goals of mass activity and durability.
- The team has demonstrated an excellent stability of pre-leached PtNiNW during a 6000 cycle (1–1.5 V) cycling test against the benchmark Pt/C catalyst. High mass activity was also achieved at RDE level. Although such mass activity is encouraging, it is essential that it has to be achieved at MEA level.

Unfortunately, this is not the case here. Considering that DOE has provided nearly $10 million in funding over the lifespan of this project, one could not argue that the return on investment is good in this project.
- The primary emphasis of this last year of the project should have been to obtain promising fuel cell performance with these types of catalysts. As shown on slide 15 for previous MEA results, rather poor performance is obtained with low loadings and/or without carbon added. New for FY 2015 is an acid-leached PtNi NW MEA with improved mass and specific activities in oxygen (slide 16), but the MEA performance, as shown by a simple polarization curve on air, is not provided, so it is not clear that any actual fuel cell performance increase was obtained by the leaching. It appears that results for only a single additional MEA are shown for FY 2015 beyond those performed in FY 2014, and even then it is not clear that any MEA improvement was obtained.

Question 3: Collaboration and coordination with other institutions

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

- The team has established strong collaboration with universities and industry. The objective of including the task "electrospinning of polymer nanowires for electrode incorporation" by one of the subcontractors toward the end of the project is unclear.
- There is good collaboration, but the team may need to closely work with the characterization partners to collect more information about the catalyst, electrode, and membrane.
- The team is excellent, with complementary skills.
- The partnership has significantly evolved along the project duration from 11 partners in 2013, to 5 in 2014, to 3 remaining partners in 2015. While the role of the leaving partners may have been limited to being subcontractors, it is unclear why they detached from the collaboration up to the final stages. In 2014–2015, the interactions and scientific dissemination of the project were ensured by six publications (done or submitted), five presentations in the United States, and one presentation in China.
- The collaborations with university partners seem adequate for this type of project. There was no indication in the presentation materials or in the presentation itself that General Motors (GM) has been an active participant in this project.
- While collaborators are listed, their contributions are not very much in evidence from the presentation slides. Either they are not contributing much, or the contributions are not being attributed.
- It is unclear what the partners are contributing to the project.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.3** for its relevance/potential impact.

- The project is critical to the Hydrogen and Fuel Cells Program, and it fully supports DOE research, development, and demonstration objectives. In particular it looks at PGM content of 0.125 g/kW, mass activity of 0.44 A/mg, and mass activity loss <10%. The project aims at developing high performing, durable, and low-cost MEA fuel cell technology competitive with battery and internal combustion engine technologies.
- This approach has the potential to be highly relevant to DOE cost and durability goals. Demonstration of high-activity MEA electrodes with even a substantial fraction of what is reported in RDEs would significantly advance the state of the art.
- The project does provide an alternative route to obtain catalyst alloys that achieve higher mass activity and durability—especially the way that it is achieved, with the removal of carbon as the support. This is key to solving some issues, such as start-up/shut-down corrosion. The potential to achieve the research and development targets is high, provided that the catalyst can be synthesized in larger volumes, which is an issue that the project has been trying to address since its inception.
- The project has a clear objective of developing highly active and stable oxygen reduction reaction (ORR) electrocatalysts in accordance with the DOE goals for automotive catalysts.

- The project directly addresses high mass activity and good durability for a low-cost fuel cell catalyst, which aligns with DOE goals.
- This project has a very high potential for impact, given the demonstrated performance and durability levels. However, the key word here is "potential" since the potential will be realized only if the performance and durability are demonstrated on electrocatalysts made using a scalable process.
- Although the knowledge generated through this project is helpful, it is unlikely the catalyst or concept can be implemented in the actual fuel cell system in comparison with some other Pt alloy catalysts under development.
- Based on the underlying issues with Ni contamination and the relatively poor and sparse fuel cell results (and perhaps the need to still require carbon in the catalyst layer), the catalyst does not appear to offer meaningful competition to current technology or other catalyst systems in the pipeline. It was an interesting and worthy approach to investigate, but it appears to have run its course.

Question 5: Proposed future work

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

- The proposed future work of "focus on reproducibility/increased batch size" is a key area on which to focus energies, now that it has been shown that the catalyst meets the minimum requirements at a laboratory scale; therefore, this is nicely proposed. What is not clear in this proposed work is how and with whom this will be done. To scale a new approach like this, an industrial catalyst partner is highly desired, but none appears in the list of collaborations. Of course the project is at its end, but if there is a need for further funding, integrating a catalyst manufacturer is recommended. On the topic of "electrode structure/fuel cell studies," this is a large body of work. Even though this is an area that needs to be addressed, it was unclear why there was a decision already on optimization of catalyst ink and composition. The right process is first to characterize the new catalyst in other typical automotive operating conditions, such as hot operation, cold operation, pressures, stoichs, concentrations, and humidity; and from there, to derive what the issues are with the catalyst and propose the necessary electrode optimization work.
- The project has identified the remaining barriers: improved synthesis of catalysts at larger scale and investigation of fuel cell behavior to meet the performance targets. The cost assessment is not sufficiently considered, although costs are mentioned as a barrier by the project; hence the financial benefits should be measured at some point.
- There is very little time left on this project, and the team's stated plans for focusing the remaining effort on demonstrating fuel cell performance on electrocatalysts made by the ALD process is appropriate. The project should abandon any work on scaling up the galvanic displacement process unless the team can identify a viable scale-up approach.
- The project is in its last year, so future work covers only the remainder of FY 2015. Work needs to be done on the catalyst layers, as the polarization curves on slide 15 indicate that there are curious effects going on as the loading is increased. Apparently, substantial transport improvement is obtained when high surface area carbon (HSC) is added to the electrode, which is unfortunately counter to the objective of removing carbon materials from the active area (indeed, slide 29 seems to indicate that the carbon durability is as much of a problem as it is with Pt/C catalysts).
- ALD coating may not be an effective way because the catalyst nanomaterial cannot be coated with a thick ALD layer; otherwise, the catalyst's mass activity will be reduced. If the ALD layer is thin enough, it cannot maintain its coverage on the catalyst's surface, especially when the catalyst is leached or annealed or utilized in the fuel cells. The team should carefully think about it. The team has considered the scale-up and reproducibility issues, as well as the MEA fabrication issue.
- The proposed future work is reasonable and sound. However, such tasks should have already been completed within the allocated budget and effort.
- The team had proposed to "isolate the overpotential losses in MEAs made with ETFECS materials" as the future work in the 2014 AMR meeting, with no significant progress made thereafter. Considering the time left in the project, the team may not have sufficient time to complete all of the proposed future work.
- MEA electrode optimization should be the primary focus.

Project strengths:

- The excellent team made significant strides this year. Very good activity was demonstrated at the RDE level. Very good durability improvement over Pt/C has been shown.
- The project has a very good approach, and the team has made necessary go/no-go decisions to identify a catalyst system with high mass activity and stability. Since the beginning of the project, the team has made significant collaborations with academic institutions, national laboratories, a catalyst developer, and original equipment manufacturers.
- The project team has identified an extraordinarily promising route to high-activity, durable (cyclic) alloy electrocatalysts using pragmatic approaches.
- The project involves demonstration of a new class of electrocatalyst materials with potential for meeting DOE performance and durability targets, as well as identification of annealing approaches to optimize performance and durability.
- Lack of carbon supports does eliminate one of the biggest failure modes. A new rational approach for depositing the catalyst does provide a new area of optimization in electrode structure.
- The team has long-term perspective and a commitment to converge toward DOE targets.
- The team has good resources.

Project weaknesses:

- (1) There is no catalyst industry partner to scale up the processes proposed (as they are not typically used by catalyst suppliers). (2) Even though GM is cited as an automotive partner for development, it is not clear whether the catalyst is being tested—or testing is planned—under other typical automotive conditions such as hot operation or cold operation, conditions under which this type of catalyst structure would frequently fall short of requirements.
- Mass activity is still lower than DOE targets at the MEA level. The transition metal leaching issue was not addressed.
- There has been insufficient consideration of cost assessment for the catalyst fabrication techniques.
- (1) The contribution of each collaborator is not clearly mentioned in the presentation slides. (2) Since the 2014 AMR meeting, the team in fact had ample time for electrode optimization and testing the durability of a promising candidate material under H_2/air fuel cell operating conditions.
- Correlation of the characterization and performance needs to be further analyzed.
- The team is not focusing on the key issue of MEA electrodes.
- Too much work has been performed using a non-scalable synthesis process.
- After this project's many years, ETFECS materials are not much closer to becoming practical fuel cell catalysts.

Recommendations for additions/deletions to project scope:

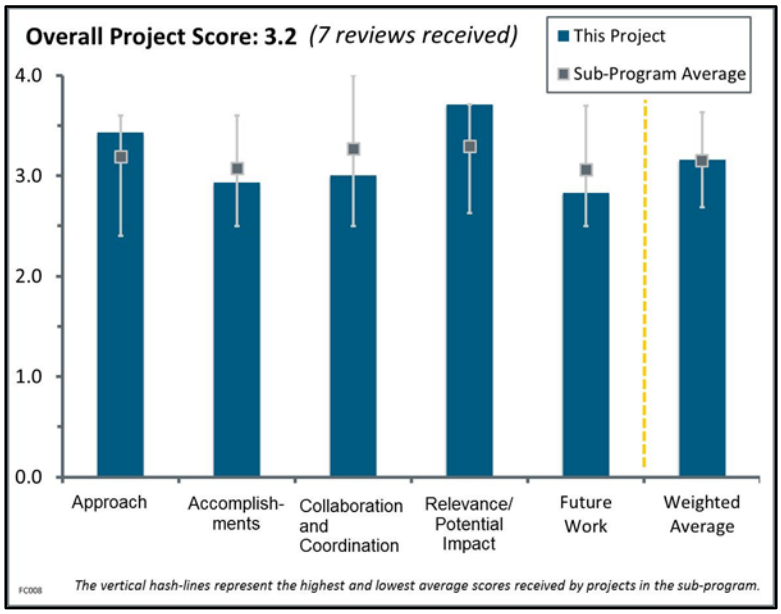
- The project should (1) add a catalyst industry partner now that it seems that the catalyst is ready to be scaled up, (2) perform more characterization work under other automotive requirements, and (3) investigate the effect of the post-treatment work proven successful with PtNi on other alloys with similar mass activity (such as PtCo).
- The project should be extended one year and funded sufficiently to allow a significant focus on electrode optimization only. Collaboration with others that have significant electrode design expertise (either in DOE or in industry) should be stressed.
- The project should abandon work on galvanic displacement and focus all remaining resources on getting the fuel cell test results that provide "proof" that the new electrocatalyst materials are worth pursuing so that the technology is advanced by a fuel cell MEA manufacturer.
- The project should include a cost–benefit study of the manufacturing at the fuel cell level (Pt reduction and fabrication costs).
- The project ends in September 2015. Hence, no additions/deletions to project scope are suggested.
- The project should be wrapped up. No further development in this direction is recommended.
- The effort should probably end, along with the FY 2015 funding.

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

Vojislav Stamenkovic; Argonne National Laboratory

Brief Summary of Project:

The overall objective of this project is to develop highly efficient and durable multi-metallic PtMN nanosegregated catalysts for the oxygen reduction reaction with ultra-low platinum content. Argonne National Laboratory (ANL) will conduct a materials-by-design approach to design, characterize, understand, synthesize/fabricate, test and develop advanced nanosegregated multi-metallic nanoparticles and nanostructured thin metal films.



Question 1: Approach to performing the work

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

- The team went from analyzing the catalytic phenomena during the oxygen reduction reaction (ORR) that damage Pt/C catalysts to the approach of mixed metals. The team systematically investigated the effects when adding other metals, supported that by modeling, and synthesized new catalysts. In a first stage, the focus was on non-Pt metal additions, then nanostructuring came in until the Ni-framed Pt catalysts were achieved. This very scientific approach is a good example of how successful use-inspired basic research can be if it is clearly targeted and a strong team of academia and industry is formed. Hence, this project is considered outstanding.
- The project approach overall has been excellent, with significant achievements made in new catalyst designs, characterization, and understanding of activity and stability impacts. The project team has achieved what appears to be a scalable catalyst design with ability to control surface structure and morphology. The current work on understanding fundamentals required for the next step in membrane electrode assembly (MEA) design is good, but while still at the rotating disk electrode (RDE) level, e.g., ionomer adsorption. At the MEA level, more work will be required on the voltage loss breakdown and modeling of the results in order to achieve understanding of catalyst layer optimization opportunities.
- Enhancement of ORR catalyst activity is imperative for the automotive fuel cell. This project is looking at the atomic-level structure of the ORR catalyst to enhance the performance. The novelty of this approach is outstanding.
- The approach to the work in past years has been consistently novel and outstanding—catalyst species have ranged from non-perylene modifications of 3M nanostructured thin films to the nanoframe catalyst that has been the subject of recent work. The remaining approach to the project is to scale up synthesis and perform fuel cell measurements. In the course of doing this, the investigators understood that re-confirmation of the nanoframe structure would be needed after fabricating the MEA, and they also understood that ionomer might have an effect on activity, so they redid RDE experiments with ionomer. This year's approach was compromised by lack of an industrial collaborator, which may have assisted with earlier delivery of results, as well as with analysis of voltage losses in the cell. Furthermore, the scale-up of catalyst fabrication was only up to 60 milligrams, which could have been improved with industrial collaboration.
- The research team members are world leaders in the nanoengineering of Pt particles. The approach is very interesting from a science perspective. From an industrial processing viewpoint, it is significantly more challenging than conventional synthesis. There is little doubt that the approach improves the electrocatalyst performance under RDE conditions.

- No presentation materials on this project were available on PeerNet, nor was the presentation included on the presentation flash drive handed out at the U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program Annual Merit Review. This makes proper review more difficult. Sufficient respect should be shown for everyone in the review process to get presentations in on time. There were no dramatic new results (since the submission deadline) in the live presentation that might have partially justified the lack of a pre-submitted presentation document. The presentation included a comment that the project had been forced to change focus, moving from fundamental to more applied work. Argonne National Laboratory (ANL) is one of the few places in the world equipped to do really fundamental work on electrocatalysts, including work on single crystals. That fundamental work has had a profound impact on the field of electrocatalysis as a whole. A major mission of the national laboratories is to do technology-supporting experiments that can be done nowhere else. Even national laboratory research funded by the DOE Office of Energy Efficiency and Renewable Energy (EERE), which overall has more of an applied cast than DOE Basic Energy Sciences (BES) work, should support this mission of the laboratories. The very long delays in getting the catalyst scale-up laboratory running at ANL demonstrate the inefficiencies inherent in diverting national laboratory resources to what is perceived as more applied work.
- The general approach is to discover new catalyst configurations and structures that promise high activity. Superior MEA performance and durability should have been the major objective of this project in order to demonstrate the value of these highly touted catalysts (some of which were developed prior to this particular project) to the fuel cell DOE program and community. Considering the status of the technologies, both at the beginning of this five-year project and now in its last year, the approach has fallen short in carrying the catalysts to a level of development that is useful for fuel cell manufacturers.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The project technical targets are three times the DOE targets for mass and specific activity and for lower total platinum group metal content. Much of the presentation was focused on prior years' accomplishments. In the 2014 reporting year, a huge step was achieved due to the discovery of the highly active nanoframe structure. In the current reporting year, the work appeared to focus on the catalyst scale-up and MEA testing. The results in these areas show a work in progress. The MEA performance with the nanoframes achieved a mass activity of ~3x that of Pt, which exceeds the DOE target, but the MEA performance falls short of targets. However, this was at a very low Pt loading. There is still a fundamental gap in achieving high current density results, which is disappointing, but maybe not surprising. The scale-up has been challenging; the catalyst produced is still highly active, but approximately half of the activity was lost. In an MEA, the nanoframes do not appear to be any better than the nanostructured thin film, but they might provide additional opportunities for catalyst layer design, and there is certainly considerable room for an additional upside. It is difficult to assess whether the targets will be achievable in the project timeframe.
- The 2015 accomplishments include limited scale-up of the nanoframes and successful determination of MEA performance. The MEA results were on small 5 cm^2 cells, most likely because of the difficulty in scaling up the process. The translation of high-performance RDE results into high-performance MEAs has been very challenging for all of the new catalyst systems. More work will be required to determine the suitability of the approach in fuel cell stacks.
- The project focused extremely well on the barriers and DOE targets. Notably, some of the targets were exceeded, and one was even exceeded by more than an order of magnitude.
- Accomplishments in the 2014 presentation, particularly the nanoframe concept, were outstanding. Compared with the 2014 presentation, it is hard to see significant progress in the 2015 presentation. There are several technical accomplishments throughout this project from 2009 to 2014: surface segregation of Pt alloy, ternary alloy, mesostructured thin film, and nanoframes. For the rest of the project term, it is suggested that the project investigate to show a ranking of these concepts for the project target, e.g., which concept is the most promising for the automotive fuel cell. Although the nanoframe catalyst shows a high performance at the low current density, it does not show a high performance at the higher current densities. Performance at the high current density is also important for increasing area-specific power density to

reduce the fuel cell cost. This could be out of the original scope of the project (maybe this could be another project). It is suggested that the project investigate the cause for the rest of this project term.

- Progress measured over the entire time of this project has been excellent, but it is unclear how much has been accomplished in the past year. The presentation said that this year's talk would concentrate on nanowires, but relatively little was said on this point other than removing the final anneal to prevent Au from coming to the surface and disrupting ORR, plus recent addition of a new component which could not be disclosed and for which no data were shown. It is good that Au's negative effects on ORR in the near-surface region are now being acknowledged by the project, not just the claimed improvements in durability. Nanoframes have now been tested in 5 cm^2 MEAs, but the discussion of this provided little beyond the fact that the basic nanoframe structure was maintained but that further loss of Ni was observed. The scale-up milestone was described as 80% done, but there were few clear data on scaled-up catalysts in the talk. It is not clear that a 5 cm^2 MEA is considered a scale-up. The mentioned, but not shown, very recent data showing a 30% improvement in MEA performance upon addition of an ionic liquid to an MEA indicate a start on determining whether ionic liquids could provide practical benefits in MEAs.
- Scale-up mass is still low. It was good to see the reporting of how much lower the catalyst activity goes with scale-up. The project should focus on why scale-up decreases activity. In situ mass activity measurements show that mass activity is still three times higher than the baseline. Fuel cell tests show that performance for the nanoframe catalysts at high current density is low. Despite the low Pt loading (0.035 mg/cm^2), the inflection of the polarization curve shows that mass transport losses begin to decrease performance around 0.4 A/cm^2. It may be interesting to see a polarization curve at higher Pt loading. Breakdown of performance losses is needed. The project also needs to share parameters related to the rest of the MEA (membrane thickness, anode catalyst loading, etc.). Although Ni leaching was detected, there should have been greater delivery with respect to addressing this, especially with another project at ANL addressing Ni leaching from dealloyed PtNi nanoparticles.
- The primary fiscal year (FY) 2015 accomplishments were on MEA testing with the nanoframes, in which considerable improvements in the kinetic region were observed over Pt/C. Higher currents resulted in poor performance. It was mentioned that transport limitations need to be overcome, but a curious inflection is obtained, indicating that other issues may be involved. The scale-up results with nanoframes are not particularly encouraging. This is a long way from commercial relevance.

Question 3: Collaboration and coordination with other institutions

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

- The collaboration between the catalyst synthesis and the MEA fabrication and fuel cell testing was excellent. This approach needs to be duplicated for all promising catalysts first investigated by RDE measurements.
- A very strong team of the leading groups in academia (of ANL and Oak Ridge National Laboratory [ORNL]) and one of the leading companies (3M) was formed. The structure brought in the necessary proficiency without bringing in too many partners.
- Collaboration between the different divisions of ANL on this project now seems to be very good. There was not much discussion on outside collaborations within the presentation.
- With the exception of characterization (which is excellent), it appears that collaboration is mostly within ANL. A group with more MEA design experience could be beneficial at this stage in the catalyst development.
- There are only three partners, and they are all national laboratories. In the last year, a supplier or a developer would have added valuable collaboration, but this did not occur. ORNL has provided services with respect to imaging the catalysts. Los Alamos National Laboratory (LANL) provided fuel cell testing of the catalysts.
- No highly effective collaboration can be seen in this project expect atomic-scale microscopic analysis with ORNL. More effective collaboration is expected for durability (LANL) and cell fabrication and testing (ANL). In the cell fabrication area, collaboration with an industry partner is suggested.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.7** for its relevance/potential impact.

- The relevance of this project is very high. A highly active catalyst could have the ability to considerably change the affordability of commercial fuel cell vehicles by decreasing the size of the stack and decreasing the overall amount of precious metals used in the stack. Surface-segregated PtNi with a Pt skin surface has been known for many years to be a highly active surface for oxygen reduction. This project seeks not only to make a nanoparticle version of this but also to do it with a novel morphology that can accommodate further design features in the catalyst layer. The project is committed to understanding whether activity shown ex situ can be confirmed in an operating fuel cell.
- Enhancement of ORR catalyst activity is imperative for the automotive fuel cell. This project is looking at atomic-level structure of the ORR catalyst to enhance the performance. The novelty of this approach is outstanding.
- The work to translate a significant portion of the thin film Pt3Ni(111) specific activity into a manufacturable and stable nanoparticulate catalyst is highly relevant to achieving the ultimate Pt loading targets for high-volume commercial fuel cells and meeting program targets.
- The relevance for automotive fuel cells is very high. The main barriers were successfully targeted. The results are getting even more relevant as the first two Asian companies launched the market introduction of fuel cell vehicles in 2014 and early 2015.
- This project historically has generated new paths towards higher activity and improved durability, which has influenced catalyst development throughout the industry. The fundamental work within this project has improved understanding of what factors are important to determining the activity and durability of a catalyst. It is not clear whether the new work this past year will be as influential as the earlier work, but the new efforts to control the negative effects of Au while maintaining Au's apparent durability benefits could prove fruitful.
- The relevance of the project is good. Much has been learned about how the microstructure and composition of Pt catalysts influence electrocatalyst activity. As for potential impact, that remains to be seen. After the disappointing performance witnessed by nanostructured thin film catalysts under real-world fuel cell conditions, judgments need to be withheld until extensive MEA optimization and fuel cell performance and durability testing are performed.
- The substantial number of presentations and high-visibility publications will have an effect on literature and citations. Less certain is that the efforts will have an impact on industry and commercial fuel cells.

Question 5: Proposed future work

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

- For the final year of the project, "alternative approaches to highly active and stable catalysts" would not likely be a task. The project already has much to settle with nanoframe performance in a fuel cell. Scale-up of the catalyst is needed, but it would be helpful to have a supplier involved. Perhaps this could be addressed in a later funding period. Catalyst stability does need to be addressed, but during both fabrication and operation. Further studies need to be done to understand the sources of voltage loss. Testing at higher Pt loading would also be helpful. It is good to see that the mesostructured thin films may be carried forward in a future funding period.
- It is absolutely imperative that the catalyst synthesis be scaled up to real-world quantities of materials (grams at least) and fuel cell MEA optimization be performed. Extensive testing of the materials in larger cells under DOE protocols is required.
- The future work is appropriate, but it is not clear whether the plans for MEA development are sufficient to fully realize the potential of these outstanding catalysts. Additional focus is also required to achieve scale-up of these catalysts.
- The plans to finish up the project, while not emphasized in the talk, seem appropriate.

- MEA development and testing need more emphasis.
- This project is ending in September 2015, and the remaining time is not enough to do something new.

Project strengths:

- The project has had a remarkable degree of success in past years, showing activity for PtNi nanoframes that is 35 times baseline catalysts and 15 times the DOE target. Investigators have extensive backgrounds and were involved in the original discovery of the PtNi(111) activity with a Pt skin surface. ANL and the other laboratories involved have considerable access to analytical capability.
- The outstanding capabilities of the project team have led to world-leading advancements in fuel cell catalyst design ability for performance and stability. The overall approach of structured, step-by-step improvement in understanding, as well as some serendipity in discoveries, has been highly effective.
- The fundamental studies of the factors leading to high ORR activity and durability under this project have been critical to developments throughout the field. ANL has one of the few laboratories in the world equipped to study single-crystal surfaces of alloy catalysts and the experience to interpret the results in a way that can provide unambiguous insights to the fundamentals of activity and durability.
- This is a world-class team in electrocatalyst synthesis in partnership with world-class MEA fabrication and fuel cell testing. The very novel synthesis approaches are used with excellent diagnostic and theoretical analysis. There is strong leveraging of BES and EERE capabilities.
- The strength of the project is based on its very basic scientific approach with a clear devotion to solving the issues of the ORR in view of cost reduction and lifetime enhancement. This also shows that longer-termed projects such as this one (over a five-year term) provide an opportunity for researchers to solve intricate problems in multistep approaches.
- The project has been very creative in developing new configurations and types of catalysts.
- The novelty of the atomic-level catalyst structure is a project strength.

Project weaknesses:

- The project is very strong, and no notable weaknesses can be identified. Minor issues should not be criticized just for the sake of completeness.
- There is very little MEA-level assessment and no optimization yet. Although the assessment at the MEA level needs to be done, care should be taken not to draw conclusions too quickly on the MEA performance. It is likely that much work will be required to realize the full potential of these catalysts at the MEA level.
- Lack of scale-up of materials synthesis thus far is a weakness. There is a need for extensive MEA performance optimization.
- Scale-up of the high-surface-area catalysts developed under this project has been very slow and raises questions of whether national laboratories should attempt such scale-up on their own rather than seeking experienced outside collaborators. Redirection of the project from its initial fundamental focus to greater attention to high-surface-area “practical” catalysts has diluted its impact on broader electrocatalyst development.
- The project lacks industrial collaboration. In the past year, the project was slow to produce fuel cell testing results. Results that have been reported have shown low performance without also exploring wide ranges of Pt loading or operating conditions. The synthesis scale reaches only milligram-size batches. The project still needs to explore why activity decreases with increases in fabrication scale.
- The project lacks proper collaboration, particularly in the fabrication of the cell area (transforming catalyst concept to real cell).
- The project has not been effectively focused on developing a product or technology that is useful for commercial fuel cells. It has been more of a BES-type of discovery project.

Recommendations for additions/deletions to project scope:

- Continued funding is needed to determine the viability of the novel electrocatalysts. The future research plan is well conceived and needs more years of EERE support.
- The project should restore the greater emphasis on fundamental work, including that on single crystals. Scale-up would likely be more efficient if undertaken with experienced outside suppliers rather than by

attempting it within ANL. National laboratory time is expensive and should be directed toward the unique capabilities of the laboratories rather than toward immediate commercialization.

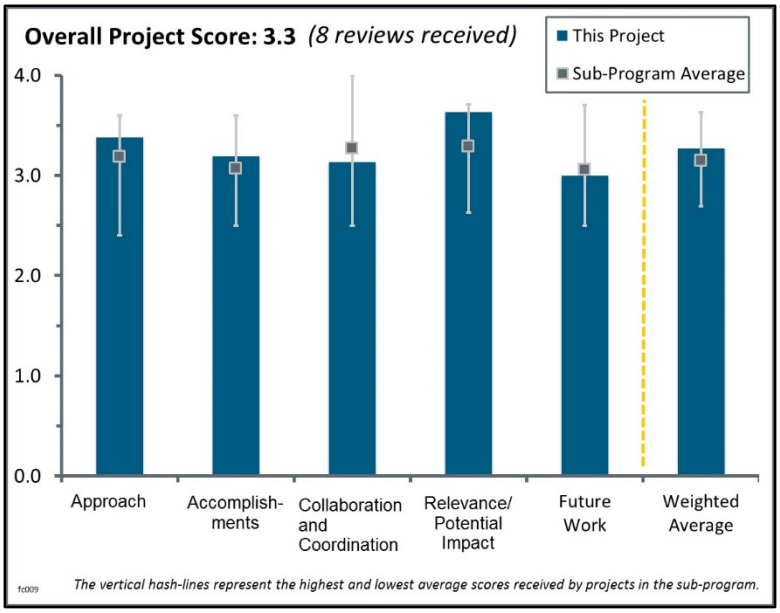
- Additions should include industrial partners (supplier and developer), an emphasis on gram-scale production, fuel cell testing that includes a range of Pt loading and operating conditions, and extensive in situ diagnostics to understand the sources of voltage loss. The remainder of the project should focus on fuel cell testing of PtNi nanoframes. At least for this project, there should be no further work on new catalysts.
- For the cell fabrication area, collaboration with an industry partner is suggested.

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

Radoslav Adzic; Brookhaven National Laboratory

Brief Summary of Project:

The overall objective of this project is to synthesize a high-performance platinum monolayer (ML) on stable, inexpensive metal or alloy nanostructure electrocatalysts for the oxygen reduction reaction (ORR). For fiscal year (FY) 2015, Brookhaven National Laboratory (BNL) conducted electrodeposition of inexpensive refractory metal alloy nanoparticles on gas diffusion layers to fabricate electrodes of 5, 25, and 50 cm^2 and carried out membrane electrode assembly tests at BNL and General Motors (GM). BNL also further developed the nitriding method to stabilize cores and the stabilization method involving the addition of Au to cores. Finally, BNL sought to demonstrate the suitability of graphene as a support for Pt ML catalysts.



Question 1: Approach to performing the work

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

- Enhancing the ORR activity for the automotive fuel cell is imperative. The novelty of the approach (synthesizing a Pt ML) sets a trend for the ORR catalyst research, such as surface segregation and de-alloying.
- The approach for this project has been well thought out since the beginning: synthesizing a Pt ML on inexpensive support metals.
- This team continues its creative and productive development of new cathode catalysts. The primary challenge in the past has been lack of membrane electrode assembly (MEA) integration. The team has made much progress in this area, although the excellent half-cell performance of the materials does not translate all that well in MEAs. The team will need to address these sources of loss in MEAs.
- The approach is strongly focused on meeting U.S. Department of Energy (DOE) targets on Pt loading, activity, and durability
- The project pursues numerous synthesis approaches for developing improved catalysts. Each individual segment progresses at its own pace and will ultimately be tested in MEAs.
- The approach to catalyst development is good. However, it is not clear how the IrRuOx fits into the range of materials.
- In general, BNL's Pt ML approach provides an excellent building block to achieve maximum Pt utilization and, hence, shows the best opportunity for achieving both high oxygen reduction activity and high power performance with ultralow platinum group metal (PGM) electrodes. The project revisits on stabilization of the core with Au and nitride are encouraging. The focus on obtaining MEA data on new catalysts is welcome, y. Yet MEA fabrication appeared to be a weakness. Despite early promising MEA results on electrodeposited Pt/Pd/WNi, its MEA performance is still depressingly poor. Other MEA data with dispersed catalyst also look poor. It is important to determine whether these materials have an intrinsic problem or not. It is not clear if BNL has a plan and resources to investigate and improve upon these issues. The project also includes many other studies, some of which are questionable and perhaps even serve as

distractions. For example, the electrodeposited dendritic Pd is a huge waste of PGM and will create an electrode structure that floods readily and has poor O_2 transport. The PtPd dendritic catalyst is also susceptible to sintering and dissolution. Fe porphyrin/graphene is not particularly new and will cause severe membrane degradation. The study on graphene might be interesting, but as with any new support, oxygen transport in this layer can be an issue and requires proper MEA testing and analyses.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

This project was rated **3.2** for its accomplishments and progress.

- The team has made very good progress toward the design of highly active and durable catalysts in electrochemical cells. Several novel and promising highly active catalysts (dendritic PtPd, PtML on nitride-stabilized cores, and Fe porhyrine supported on graphene) have been developed. Performance in the air still needs to be optimized.
- The project made excellent progress this year, with lots of MEA testing showing very promising results for PGM loading as low as 0.07 mg_{PGM}/cm^2. Although results indicate that air performance needs to be improved, ORR activity in MEA is close to the 2020 target.
- Overall, good progress has been made. Several candidates appear to yield mass activity approaching or exceeding DOE targets, and there appears to be a larger focus on MEA testing for evaluation of the electrocatalysts' activity and durability. Relevant MEA H_2/air performance is still substantially lower than state-of-the-art electrodes and needs increased focus.
- Catalyst stability using nitriding strategies is impressive, and the fact that the team has been moving much further toward MEA integration is good. Some of the other areas seem a bit tangential. It was not clear why the team moved to graphene-based supports, and their results in this area are not that impressive.
- The new syntheses (Pt ML on Pd/WNi/ gas diffusion layer) shows progress for durability. It is suggested that BNL show an effect of Pd or underpotential deposition Cu displacement. It seems to be a Pd effect. It keeps total PGM (including Pd) low. The PdNi core shows a possibility to reduce Pd content (lower cost). It is suggested to show the merit of graphene as a support (performance and durability), and determine which is more promising; FeP on graphene or reduced graphene oxide.
- The project accomplished high mass activity and good stability in the rotating disk electrode (RDE).
- MEA testing, which was lacking in this project, was accomplished with several catalysts. There is work ahead to achieve high performance catalyst layers. Regarding the MEA testing, results are reported in several instances for pressures, e.g., 150 kPa. Usually, kilopascal values are understood to be absolute pressures (e.g., 150 kPa is ~22 psia or ~7.4 psig). However, slide 8 reports results at 0 kPa, which obviously means that the kilopascal values are on top of atmospheric pressure. These pressures are significantly higher than typically inferred for data at 150 kPa.
- It was not clear if the project made any progress against DOE goals this year. Note that the project did not adhere closely with the DOE-recommended testing methods. Tricks such as pure O_2 and high pressure were used to show better-looking results.

Question 3: Collaboration and coordination with other institutions

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

- There is very strong collaboration among BNL, industry (GM and IRD), and other institutes. GM has demonstrated excellent work by testing these catalysts in MEAs. The H_2/O_2 and H_2/air tests in 50 cm^2 MEA are impressive. This is a significant leap from the last couple of years and is excellent progress compared to other projects on low-PGM catalysts. Toyota's active involvement is unclear. It looks like most of the MEA tests are done at BNL and GM.
- There is excellent coordination with industrial partners.
- Including industry partners adds a variety of technical approaches to fabricate the catalyst structure.
- This is a very well-coordinated effort between BNL and industry. Collaboration with theorists are indicated on slide 21, but none of the theoretical results are presented.
- Collaborations appear to be numerous and reasonably productive.

- The listed collaborators are excellent, but it is unclear what the work distribution is.
- It was not clear if there is any collaboration.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.6** for its relevance/potential impact.

- The catalysts developed by the team demonstrate very high activity and durability at very low Pt loading, which aligns very well with Hydrogen and Fuel Cells Program objectives.
- Objectives of this project are very relevant to the Fuel Cell Technology Office and DOE research, development, and deployment objectives.
- It is imperative to enhance the ORR activity for the automotive fuel cell. Novelty of the approach (synthesizing Pt ML) sets a trend for the ORR catalyst research, such as surface segregation and de-alloying.
- If the Pt ML catalyst could be made stably, it could significantly reduce the stack cost and reliance on PGM, predominantly thanks to its high Pt utilization.
- The project is highly aligned with the key barriers of cost and durability. Relevance will be improved when high performance MEA electrodes can be demonstrated.
- Successfully implementing catalysts into low-loading, high-performance and durability MEAs would have a significant effect on the fuel cell industry.
- It was not clear if the complex chemistries used here will ever translate to industrial-scale implementation. The team has made a lot of progress toward MEA integration and larger amounts of material are being synthesized. There is much to be learned of fundamental importance here.
- The project matches the DOE objective of low PGM and high stability.

Question 5: Proposed future work

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

- Future work is focused on the improvement of catalyst stability and durability in the fuel cell environment, which is critical for this sub-program. Novel strategies are proposed that could further improve catalyst durability. The number of novel systems needs to be reduced in order to have an opportunity for the complete optimization of at least for one system.
- Proposed future work on MEAs and stack tests is very good, but shedding light on what causes drops in performance in MEAs would be great. Also, avoiding adding Au for stabilizing the core would help to reduce the PGM cost.
- Synthesis, MEAs, and stack tests are reasonable, and moving toward incorporation of refractory elements is an area of interest for many. However, the effects of nitriding at high pressure and/or reactive spray deposition were unexplained.
- The stack tests with GM would be an important demonstration of the future viability of these catalysts.
- The focus on durability of core and support materials and performance enhancement is understandable.
- The project continues to focus on stabilization of core materials using multiple approaches. MEA development appears to be relying on a non-funded partner (i.e., GM). New catalyst developments also appear to require increasing amounts of MEA knowledge and resources going forward. It is not very clear if relying only on in-kind contributors will be an adequate approach.
- It would be good to see the evaluation of catalyst stability in full-size MEAs. Optimization of air performance is recommended as is developing an approach to material scale-up.
- It is recommended that for FY 2015 the project focus on electrode development to allow demonstration of the project electocatalysts' performance in relevant formats. Working with other groups with electrode expertise is warranted.

Project strengths:

- The team is very creative in formulating novel approaches for catalyst synthesis. The team standards are so high that extremely high activities obtained in electrochemical cells do not surprise anybody anymore.
- There is very strong collaboration among BNL, academia, and industry. Objectives and approaches are very well thought out. There is lots of MEA testing, and nearing stack testing indicates closeness to commercialization
- The knowledgeable team, state-of-the-art nanoparticle characterization capability, practicality, and product-oriented culture are all project strengths.
- A project strength is the high degree of sophistication in synthesis.
- The team appears to be making progress on many fronts.
- The project has demonstrated electrocatalysts with high activity and durability in RDE.
- The novelty of the Pt ML concept and the inclusion of industry collaborators are project strengths.

Project weaknesses:

- Industry needs to be more involved in order to help the team with optimization of catalyst layers. The project covers too many novel systems instead of focusing on the full optimization of just a few of them.
- The team does not have a complete understanding of degradation in MEA and its countermeasures.
- It is questionable whether the strategies here are viable in mass production.
- MEA knowledge and resources are lacking. It is recommended that the project align to DOE-suggested methods.
- Perhaps the project is too disjointed. It is not clear what the value is of yet another catalyst that performs well in KOH or of Ru/Ir oxygen evolution catalysts or of graphene supports.
- The performance in MEA electrodes is poor.

Recommendations for additions/deletions to project scope:

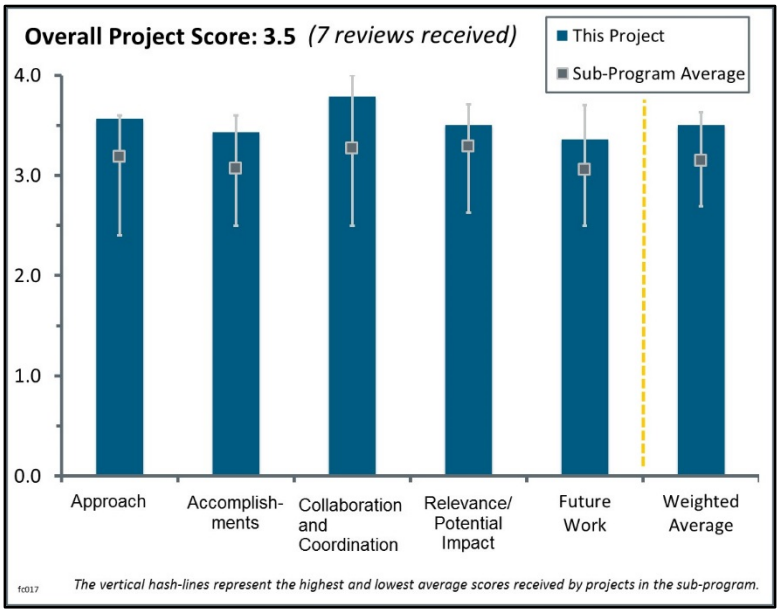
- The team should focus on the two systems that look the most promising and perform their complete analysis and optimization. One of the systems that looks the most promising is the nitride-stabilized cores.
- It is suggested that BNL show the merit of graphene as a support (performance and durability), and determine which is more promising; FeP on graphene or reduced graphene oxide.
- Prioritizing the approaches is recommended.

Project # FC-017: Fuel Cells Systems Analysis

Rajesh Ahluwalia; Argonne National Laboratory

Brief Summary of Project:

The overall objective of this project is to develop a validated system model and use it to assess design-point, part-load, and dynamic performance of automotive and stationary fuel cell systems. Argonne National Laboratory (ANL) will support the U.S. Department of Energy (DOE) in (1) setting technical targets and directing component development, (2) establishing metrics for gauging progress of research and development (R&D) projects, and (3) providing data and specifications to DOE projects on high-volume manufacturing cost estimation.



Question 1: Approach to performing the work

This project was rated **3.6** for its approach.

- This project stands alone as the most comprehensive fuel cell system model in the public domain. The investigators are actively refining their work using input from key stakeholders. The model represents the current state of the art. In terms of approach, it does at least seem as if it would be difficult for someone outside the project to exercise the model. It is not clear whether the model will be disseminated as a user tool.
- The project addresses system-level operation with a projection for cost. This is a very useful methodology for cascading developments in components and stack configurations to the commercialization targets for fuel cell systems.
- Overall, the project appears aligned with the industry direction and is capturing the developments of new materials (nanostructured thin film [NSTF]) that make this very relevant as a tool for industry. Working closely with industry to try to achieve adoption of the model within automotive fuel cell development community is encouraged.
- The linkage of GCTool and Automomie models is an effective approach to analyze performance of the fuel cell system. The model has a dynamic performance capability. Technical assumptions rely on discussion with the U.S. DRIVE Partnership Technical Teams to leverage up-to-date industry perspectives. The project leverages Strategic Analysis, Inc.'s (SA's) cost analysis for optimization of the system architecture.
- The approach is appropriate for the objectives outlined. The teaming is appropriate, as are the objectives. The specific high-level technical tasks fit the objectives, leading to solid contributions.
- Some modification may be appropriate for the actual technical evaluations.
- The project has a very logical breakdown of efforts covering stack, air, water, fuel, and thermal subsystems.
- The approach and goals are well organized and carefully adapted to the needs of fuel cell researchers and developers. The model and approach, based on publicly available materials and components that are considered to be state-of-the-art, are becoming more realistic and in line with industry expectations every year. The use of the outcomes for cost analysis (FC-018) is therefore valuable. However, ANL should analyze the sensibility of fuel cell system architecture itself. Original equipment manufacturer (OEM) systems are not unique.
- The underlying task of this project is to create a computer-based simulation (a "model") of a hypothetical polymer electrolyte membrane (PEM) fuel cell system, given detailed and authoritative data on the behavior of its many components. This is a fine approach for gaining insight into the emergent behavior of the system as a whole and for the stated purposes of the project, which include gaining insight into

attainable technical targets and how the overall performance and cost of the system is affected by its individual parts—i.e., a sensitivity analysis. This can guide DOE toward focusing efforts on improving certain components in ways that could have the largest impact. Overall, the approach of the project, which assimilates large amounts of component-level data in an organized, thoughtful, and meaningful way, is well grounded.

- However, at various points, the presentation refers to the resultant model as "validated." The author may believe the model is "validated" because of the rigor in which the component-level data is vetted. Nonetheless, there is little or no evidence that the system model itself is "validated", i.e., that predictions of the model are tested against real-world data that are separate and independent of the data from which the model is built. While it is always difficult to know how accurate model predictions might be, the lack of "validation" in this sense means that it is even more difficult to judge how much weight (trust) one might ascribe to the many detailed estimates and predictions that are the outputs of the model here.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

This project was rated **3.4** for its accomplishments and progress.

- The project has made excellent progress on investigations related to the NSTF catalyst, and the focus on cell-to-cell performance variability is excellent. Comparisons between PtNi and Pt provide valuable insights to the industry and the principal investigator (PI) is encouraged to begin making a broader assessment of additional catalyst materials, including variation of the supports where data is available.
- Very good progress has been made. ANL has demonstrated a significant amount of work looking at electrode variabilities, NSTF degradation, and Roots air supply system. Representative and best-in-class results have been introduced this year. NSTF degradation testing has been linked to fluoride emission rate (FER). Deeper understanding of mechanisms may be needed, and, in particular, the impact of the temperature should be studied.
- As presented, the results and progress are consistent with expectations for the project funding and overall magnitude of the project scope. The number of components, subsystems, and their interactions is overwhelming. The team did a good job prioritizing the evaluations.
- The project added an alternate catalyst (de-alloyed PtNi dispersed on carbon support) to the NSTF catalyst membrane electrode assembly [MEA]).
- The correlation between performance degradation and fluoride release rate (membrane degradation) was identified.
- The PI reported accomplishments over the past year, including analyses of stack performances with different catalyst systems, air management (compressors and expanders), water flow, and waste heat management. These all seem to be of genuine interest to collaborators and important to the Fuel Cells R&D sub-program overall. In each case, the project stands out as being "data driven," with many quantitative results (e.g., graphs). Nonetheless, these results may be most useful as qualitative comparisons among alternatives, rather than rigorous predictions of quantitative outcomes. \Tthe lack of explicit validation and a general lack of uncertainty analysis—i.e., propagating uncertainties from the underlying data through to the predictions of the model—is noted.
- There is good progress toward the end-of-life (EOL) analysis. This should continue to be the main focus; there is still a lot of work to be done. The investigators must not only consider EOL performance data from idealized experiments but also consider issues associated with system integration. Perhaps data from the National Renewable Energy Laboratory (NREL) contamination studies could be considered.
- In reference to slide 7, work on cell degradation and cell-to-cell performance variability could benefit from system-level experiments that use actual stacks to measure cell flow variations with manifold water accumulation. In reference to slide 17, it is not clear how much effort should be applied to studying NSTF when it appears this electrode approach is still far from meeting DOE targets. In reference to slides 18 and 19, it is not clear how much effort should be applied to studying the Roots compressor/expander because it appears this air management approach is still far from meeting DOE targets.

Question 3: Collaboration and coordination with other institutions

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

- The collaboration among ANL, SA, component suppliers, OEMs, and laboratories is of high quality and enables ANL to build the model and provide key feedback to the partners. The coordination of the project also appears to be very good.
- The level of collaboration is good and well thought out with respect to the collaborators expertise. The feedback from the U.S. DRIVE Technical Teams and the partnerships with MEA suppliers is critical to the success of generating valuable data. The appropriate teams have been engaged to generate relevant data.
- This project is highly collaborative, both in the sourcing of component-level data for the model and in the demands for system-level analyses of varying alternatives. Slide 21 is representative.
- There is clearly a high degree of collaboration with other groups based on the data being fed into the project from the collaborators. However, the PI is encouraged to ensure that the process for the model is not to be a "simulation house" and to actively work on achieving adoption of the simulation software within the collaborator institutions. The collaborations include a cross-section of fuel cell stack level developers and fuel cell material/component developers, which provided an excellent backdrop for the applicability of the work in the project.
- This project leverages various collaborations and is supported by many in-kind works. Effective collaboration has been seen with the U.S. DRIVE Technical Teams.
- There is very strong collaboration with OEM partners. It is not clear if there are opportunities for incorporating the down-the-channel stack model being developed by Lawrence Berkeley National Laboratory.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.5** for its relevance/potential impact.

- The impact of system modeling is potentially huge, and modeling has the potential to significantly advance progress toward DOE research, development, and demonstration goals and objectives. The impact will be even greater when the model includes different components and different architectures. The dissemination of the modeling tools may also increase the impact, as it would allow developers to evaluate their new materials, components, or system architectures at an early stage. Validation of the model on a real system (e.g., using data from TV-001) would definitively enhance its impact.
- The model/simulation package will provide a very useful tool in evaluating a projected system cost for new materials and operational modes. This is a very important part of reaching the DOE targets from the perspective of down-selecting or targeting the efforts of other R&D projects. The use of this tool on material development projects as a final stage assessment of impact toward overall cost/performance is encouraged.
- This project is important to identify a technology direction and technology selection for the goal of automotive fuel cell systems.
- This project will be very impactful because of the team's close collaboration with industrial partners.
- The impact of this project will be improved by describing the EOL performance of a fully integrated system.
- The PI's tests and ultimate data collection and analyses are absolutely relevant. In particular, the degradation mechanisms and their impact on system performance and lifetime are highly relevant. However, it is not clear if there is a comprehensive understanding of the makeup of the MEA so that clear performance differences can be ascertained during performance testing and if using the test conditions with MEAs with different ionomer loadings or the like would potentially impact the "sweet spot" of performance for that given MEA. It is unclear if the gas diffusion layer (GDL) is the same in all tests or if there is a microporous layer. It is also unclear if the catalyst layer membrane interface is the same. Although these are details, they can impact performance and failure mode behavior. The percentage of polytetrafluoroethylene (PTFE) in the GDL is another issue. The humidity tests would be impacted by the

GDL and the microporous layer's PTFE content, as would the plate flow-field used. The FER tests are important and insightful. From prior published results, is it clear that all perfluorosulfonic acid membranes evolve F^- ions over the first 100 hours or so. It was not clear that any pre-preparatory or pre-operation period took place before measuring the F^- ion release rates. J. McElroy presented work in this area. Furthermore, if an ionomer is used in the electrode layer, it is not clear if F^- ions are released from this component. A long-hold at a specific V is not a degradation mechanism; it facilitates a degradation mechanism. The chemical or electrochemical degradation mechanism would be helpful to understand as a function of V. There appeared to be no data relative to stationary applications and/or operating conditions. They are different. Still, the tests on the various new catalyst systems are good and helpful to evaluate many aspects of their viability.

- Even with all the uncertainties, the system cost variability data are insightful.
- The relevance of this project arises from its ability to demonstrate a system-level impact of component-level changes. While this is a very useful, and indeed necessary capability to have within the DOE Hydrogen & Fuel Cell Program (the Program), it needs be understood that the model only provides estimates, not actual real-world results. Models are useful in that they allow alternatives to be explored without the time and expense of building physical systems, but ultimately physical systems need to be constructed to bear out the predictions of the model, and to refine it. Also, while analytical models may illuminate areas where technology innovations would be most needed or are the most successful, in and of themselves they do not create new innovations.

Question 5: Proposed future work

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

- The proposed future work (slide 22) seems appropriate and well balanced, although the reviewer offers a caution. The future work includes analyses of both system performance and manufacturing costs. This project has, as its strength, the ability to model system performance. Other projects (e.g., FC-018) are perhaps better optimized for modelling manufacturing costs. Therefore, the future work should be more heavily weighted toward performance analyses.
- The proposed work is in line with the objectives of the project. It is important not to focus on a given MEA and instead introduce alternate MEAs with advanced alloy catalysts. Introduction of bipolar plates and flow fields for low-pressure drops, uniform air/fuel distribution, and low cell-to-stack performance differentials are appreciated. Predictive degradation models might be introduced to reduce experimental testing.
- The future plans seem reasonable, but it is not clear how much of this project's activities should be coupled to NSTF electrode development and Eaton's Roots compressor/expander.
- The future work is appropriate as long as there are inputs from the partners. Further refinement of the tests without changing the relevance or direction will provide valuable data overall. The tests on the catalysts and related performance should remain a high priority.
- The proposed work addressed a wide cross-section of important areas. However, it may be too aggressive depending on the resources in the project.

Project strengths:

- The model appears quite complete and takes into account many mechanisms involved in the operation of a fuel cell system. The parallel cost estimation with SA is appreciated. Another strength is the high level of collaboration with other institutions and various suppliers, which facilitate the study of each subsystem/material performance and cost.
- The validated fuel cell system model and collaboration of Autonomie, cost model, and U.S. DRIVE Technical Teams are all project strengths. The availability of data from the DOE project and industry partners (in-kind) bases is another strength.
- Comprehensive tools and analyses on relevant components and test hardware are in place and being utilized. The team's qualifications are a project strength. It sounds like there is a good level of communication with the partners.

- The project has excellent collaboration activities and a highly relevant system-level analysis from an industry perspective. The focus on trade-offs and new material ensures the project is aligned with the current research direction and system-level configuration needs.
- The strength of this project arises from its ability to demonstrate a system-level impact of component-level changes. This helps enable the Program to keep an eye on the big picture, rather than just a lot of technical minutia.
- The project has great collaboration and a relevant model.
- The modeling represents the latest developments in application-scale technology.

Project weaknesses:

- This is a project with a broad scope and with many variables to take into consideration. This is not a weakness, but it adds to the difficulty of testing the appropriate components, conditions, etc. to yield valuable data. The team needs to stay focused and closely aligned with its partners to keep such an effort from turning into a "weakness" by testing scenarios unrelated to the primary task.
- The team may be devoting too much effort to understanding the performance of components that are not fully vetted at the subsystem level.
- The model relies on performance and durability validation data that lack consensus. It is recommended that these validation datasets be cross-checked with literature and other OEMs. In some cases, the electrodes being used are not optimized. Additionally, system-level mitigation strategies should be incorporated in the durability experiments. In many cases, the patent literature is a good source for determining these methods.
- The team relies too much on the GCTool fuel cell system model. The recent fuel cell stack and system shows higher performance by improving stack architecture, including flow field configurations. It is beyond a non-dimensional model.
- The main weakness of this project is the lack of experimental validation on a real stack or system. As mentioned during the presentation, the investigated system does not exist today, even if it were to be assembled. The data on performance and durability rely on 50 cm^2 single cell testing.
- It is hard to know the actual predictive power of the model (the GCTool), i.e., the uncertainties of its predictions. It is recommended that the team not refer to the model as "validated" unless systematic validation efforts are added to the project.
- The project is considering the durability considerations in system analysis, and it is not clear what mechanisms of durability are included in the model. More effort should be made to provide a high-level overview of the included physics (transport and kinetic) and the required data inputs for the model to allow a more detailed assessment of what is "under the hood."
- The statistical variation of the component properties is also an area of discussion that was not addressed in the cell-to-cell performance variability. Both operational and material sensitivities are important from a cell-to-cell variability perspective, and it would be ideal to understand the impact of both, especially given the fact that the project scope includes future work on assessing implications of high-volume manufacturing.

Recommendations for additions/deletions to project scope:

- Continue to focus on EOL and system-level tradeoffs.
- Consider scaling back NSTF and Eaton collaborations until these components are closer to meeting their individual targets.
- The model should be validated against stack data and system data. As it may be difficult due to intellectual property issues, one option might be to use a system design from the first car generation in TV-001 and to compare the results from the model and the "on the road" driving data assessed by NREL. Regarding stacks, some representative automotive stacks are "available" (see FC-021) for characterization.
- It is suggested that the team input the benchmark information of the latest high-performance automotive fuel cell system, such as Toyota's Mirai fuel cell electric vehicle.
- It could be a time to reconsider the use of GCTool for fuel cell system model. It is basically a non-dimensional model and does not simulate performance and operating conditions with the fuel cell stack architecture (flow field design, etc.). Toyota's latest fuel cell system in the vehicle showed significantly higher performance at the high current density region that leads to a cost reduction. Also, it has no

humidifier. Consider the alternate fuel cell model, e.g., the stack model to cover the implication of stack architecture.

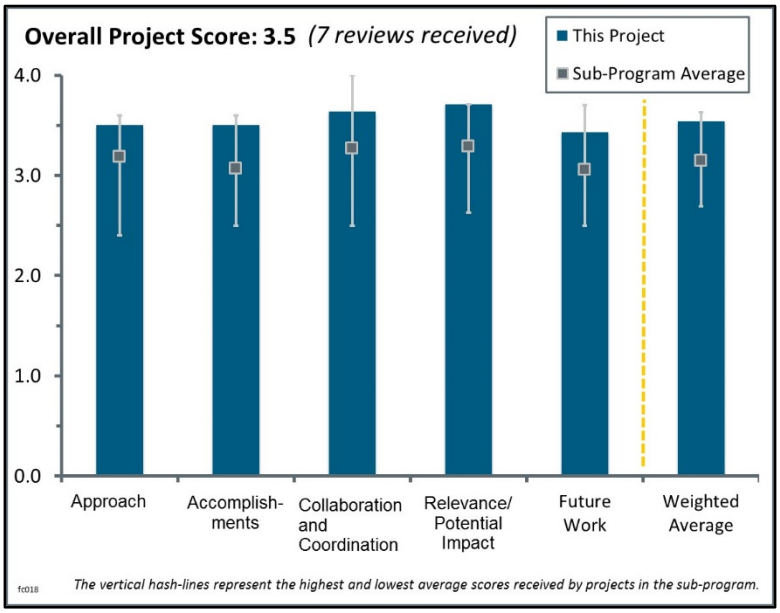
- Use of de-alloyed PtNi needs a system mitigation strategy in the fuel cell system, because the instability of catalyst material (support) at the high half-cell potential.

Project # FC-018: Fuel Cell Vehicle and Bus Cost Analysis

Brian James; Strategic Analysis, Inc.

Brief Summary of Project:

The objectives of this project are to (1) project a future cost of automotive and bus fuel cell systems at high manufacturing rates, (2) identify low-cost pathways to achieve the U.S. Department of Energy (DOE) 2020 goal of $40/kW$_{net}$ (automotive) at 500,000 systems per year, (3) focus on low-volume production (1,000–5,000 systems/year) and near-term applications, and (4) identify fuel cell system cost drivers to facilitate Fuel Cell Technologies Office programmatic decisions.



Question 1: Approach to performing the work

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

- The overall purpose of this project is to develop the knowledge base from which to derive rigorous and detailed cost estimates for the production (manufacturing) of fuel cell systems. This bridges the gap from the understanding of what is technically possible to the understanding of what is economically feasible—that is, what has a realistic chance of being adopted in the marketplace. This approach is vital for maintaining the relevance of the fuel cell research program. More specific to the project itself, its approach is to review technological advances and to map their production steps into a Design for Manufacturing and Assembly (DFMA) framework. This is a fine approach to achieve the goals.
- This is one of the most focused projects in the portfolio. The cost analysis is based on current cell and stack data and system analyses. Very frequent input from stakeholders keeps the approach fresh and timely.
- This approach has been vetted over a number of years and is as good as it has been in the past.
- The analysis methods used are good, and the principal investigator (PI) is responsive to good suggestions.
- The design of the study is very methodical and is valuable for being such a detailed study. However, it would help if there were some guidance or interpretation on taking the findings and applying them. In particular, publications from the project seem to analyze a specific system design. Guidelines for determining costs for other, perhaps similar, systems would be helpful to the community. For example, for the design of an 80 kW system, there should be a discussion of the applicable vehicle platforms and how the costs (at the vehicle level in particular) may vary for vehicle classes that are larger or smaller. The system-level discussion is also particularly useful to elucidate the linearity or nonlinearity of variation in and between the components.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The project's technical accomplishments this year were focused on new analyses of alternative catalyst systems, air handling systems (compressors and/or expanders), and various manufacturing approaches for bipolar plates.

- This cost analysis is based on recent data and analysis and guides DOE efforts to reduce cost. The cost analysis is frequently updated to incorporate the latest findings. The 2015 analysis covered lower production volumes.
- The project has completed the cost estimate for the current year of technology status.
- The accomplishments for this year focused heavily on crossover points at which one manufacturing technology becomes favorable over another. This was helpful to demonstrating the limitations of various options, but it also left open significant questions about which options may be more likely, especially when disparities were large at low volumes but not as large at high volumes. Perhaps further industry collaboration, particularly receiving feedback related to these messages, will provide value to the project and help solidify the total system cost projections.
- Additional areas for potentially lowering manufacturing costs have been identified. Feedback from the 2014 review was incorporated to include lower-volume projections. Some potential cost improvement is shown, but progress toward DOE goals is limited.
- Good progress has been made this past year, but even more could have been done. Also, it is not clear why most, if not all, of the future work shown on slide 21 could not have already been done, as the project has plenty of funding relative to the schedule (i.e., there is a question about why the pace of the work is not faster).

Question 3: Collaboration and coordination with other institutions

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

- There is significant collaboration across DOE, the national laboratories, and industry. The project is also seeking to include Toyota. This work also links to other DOE work, including Los Alamos National Laboratory's polyaniline research.
- The list of collaborators is long and broad. In addition to ongoing baseline system cost analysis, Strategic Analysis, Inc. (SA) provides cost analysis for PIs working on specific components such as compressors or bipolar plates.
- This project is highly collaborative, particularly in the development of detailed manufacturing data.
- This project makes good use of relationships and information provided by industry suppliers.
- SA does seek out industry input when it takes on modeling a new process, which helps make the results reasonably realistic.
- There seems to be heavy industry collaboration to verify the assumed costs, processes, and so forth, but the collaboration did not seem to receive as much attention as expected. Also, the list of collaborators seemed to be limited to only those directly involved in this year's accomplishments. While there is logic in that, it would have been helpful also to provide a more comprehensive list to discuss the collaborations for all system components. Additionally, it would be useful for the study to reach out to policymakers to help determine additional useful outputs that this study could provide. It is clearly a wealth of information, but it does not seem yet to be presented or disseminated in ways that will directly address challenges faced by policymakers.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.7** for its relevance/potential impact.

- The Fuel Cells sub-program needs to set cost targets for stack systems that are competitive with internal combustion engines. This project provides an analytical framework to determine where and how cost reductions might be achieved and to demonstrate program progress with respect to the cost targets. This project provides ongoing insight into where high costs are, helping DOE apply resources logically and intelligently.
- This project is key to helping the DOE sub-program prioritize what should be done to further reduce costs. It is also the type of project that can be done only with DOE funding (i.e., industry is unlikely to share cost modeling results publicly).

- Results show slow advancement to lower automotive production cost. The analysis of cost breakout at differing volumes is useful to both DOE and industry.
- The volume of production appears to be higher than would be realistic in any relevant time frame. Manufacturing volumes of up to 500,000 units per year per original equipment manufacturer (OEM) is unreasonably high. More focus should be made in regard to the reasonable expected volume of production per OEM, for example, by looking at the market share of each major OEM and project volumes of sales as a fraction of common product lines. Perhaps volume data could be extracted from the Polk database*)*. This effort of analysis is done to help guide research and development targets and development. In communicating with OEMs, it is important to ask directly what areas of technology need improvement in the public domain.
- The point made during the presentation that this work helps elucidate the areas in which research may be needed to meet DOE cost targets was particularly effective. It would be helpful for the research to identify or at least suggest options for getting the current-status $43/kW down to the DOE $40/kW goal. This could have been a valuable output that was not presented.

Question 5: Proposed future work

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

- The future work seems well aligned with Hydrogen and Fuel Cells Program needs, including further analysis of low-volume production costs, further analysis of emerging component technologies, and further analysis of a bus fuel cell system.
- All of the work listed on slide 21 is good.
- The project will perform a cost comparison of catalyst preparation methods. Several cell, stack, and/or system components will be costed.
- Buses remain a large potential future market, and the limited modeling systems may inhibit the value of this work to the market.
- It is particularly curious that hydrogen storage is left out of the project, unless it was implied in the general balance of plant (although it did not appear in the sub-bullets). Having an entirely complete analysis of the entire fuel cell electric vehicle's (FCEV's) system and components will be much more valuable than an analysis that covers 95% of the vehicle's systems. Also, a look into how these costs might change based on vehicle platform (which may be implied by the baseline sensitivity analysis) would be extremely valuable.

Project strengths:

- The detailed and methodical analysis utilized at all levels of the analysis is exemplary. It clearly requires varied expertise and significant coordination with manufacturers and other researchers to pull this information together in a way that it can all be informative within a single framework. This is a major accomplishment and possibly the project's greatest strength. Additionally, the project does seem to be appropriately responsive to new developments, looking to provide answers for projected costs of some technologies that are much newer and may be more difficult to analyze.
- The project provides a systematic and effective framework for providing cost projections and for analysis of how costs can be reduced.
- This project incorporates learning and feedback into future work and is very focused on finding the lowest-cost components and assessing more state-of-the-art options. Incorporating the impact of manufacturing volumes is valuable.
- In addition to cost analysis, SA evaluates manufacturing approaches for applicability to fuel cell components.
- Including a more detailed analysis of low-volume cost projections is an excellent addition.

Project weaknesses:

- As with all modeling projects, there is always a risk that the model will diverge from reality in ways that are not readily apparent. Therefore, validation against known systems should be an important part of the

project. The PI indicated intent to perform a comparison with the ENE FARM Panasonic stationary fuel cell systems and to do some benchmarking against the Toyota Mirai. This is to be encouraged.

- It will be difficult to accomplish the program objective with feedback and input from only one automotive OEM.
- The weakness of the project really lies in its transferability. It is not immediately clear how others outside DOE could best take advantage of the results of this study. The value for the DOE is clear, in determining progress to targets and potential paths. However, the wealth of information developed by this project likely has much broader application if it is appropriately brought to bear on other questions, such as the likely costs of fleets of varied vehicles (state or national), costs of implementation of policies and regulations, and so forth. This does seem a little outside the project's stated scope, but it seems close enough and a great opportunity to make the best use of the work.
- The biggest weakness of this project is that it does not have any benchmarking with current prices of actual fuel cells. This study seems to be operating in a parallel universe, and although it is rigorously grounded in its calculations, there has never been any kind of a reality check. Such a benchmarking would greatly enhance the believability of the project's results.
- The model does not seem to account for improvements in performance and potential reduction and/or increase in weight. If a component is taken out of the system, weight would be reduced, and the car would need less power, less hydrogen storage, and so on. Compounding effects like that can have major impacts.
- The project does not address how early-market PEM fuel cell products (e.g., forklift trucks and telecommunications power) may affect costs for road vehicles in low volumes (i.e., initial introduction of FCEVs).

Recommendations for additions/deletions to project scope:

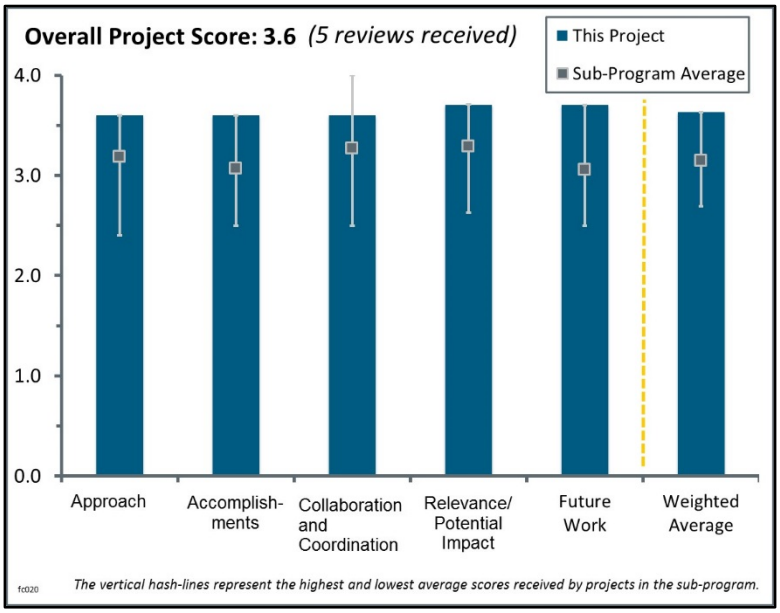
- Non-platinum-group-metal cost modeling should include the impact of the total kilowatts required for a vehicle that must utilize two stacks instead of just one. In other words, the system will need >80 kW to have performance equivalent to an 80 kW system that is significantly smaller and lighter. A cost analysis should be conducted using an estimation of what the early FCEVs are actually doing, as was shown on slides 16 and 17. This exercise could be an excellent verification of SA's low-volume cost projections. It would also be interesting to evaluate how the cost of these early FCEVs could be affected by various changes in key parameters (e.g., what the impact is of limiting peak temperature to 80°C). In addition, it may motivate changes in the current model constraints (e.g., it is not clear why the heat gain per temperature change ($Q/\Delta T$) is being held at 1.45 if a value of 2 is preferable for cost).
- The project should consider analyzing the known production supply chain. For example, if there are one or two suppliers of gas diffusion layers (GDLs), GDL cost could be estimated based off the total volume of that production. On the other hand, stack plates may be produced in one OEM per production line, yielding lower volume or production for that part of the fuel cell system.
- Now that Plug Power is manufacturing thousands of units, it would be nice to have some comparison of what this project's model suggests (hundreds of dollars per kilowatt) and what real fuel cell systems are selling for (thousands of dollars per kilowatt), with some explanation of the differences. It is difficult to understand why this has not already been done.
- Generalization of the results would be a good addition to the project scope. Instead of stating the analyzed base system and its cost, the project should, additionally, discuss what kind of vehicle this system likely represents, how the system and costs are likely to change for other vehicle classes and/or platforms (even if just in a representative sense for the class), or perhaps how the component and system costs vary with major design decisions (such as power output or degree of hybridization).
- It would be nice to be able to run costs as a function of platinum cost, understanding that platinum cost is variable.
- Toyota and/or other automotive OEMs should be added to make sure this work is aligned with industry, not behind it.

Project # FC-020: Characterization of Fuel Cell Materials

Karren More; Oak Ridge National Laboratory

Brief Summary of Project:

The objectives of this project are to (1) identify, develop, and optimize novel high-resolution imaging and compositional/chemical analysis techniques and unique specimen preparation methodologies for the micro- to Angstrom-scale characterization of materials composing fuel cell membrane electrode assemblies (MEAs); (2) understand fundamental relationships between MEA material constituents and correlate these data with stability and performance as per guidance/input from the broad fuel cell community; and (3) integrate microstructural characterization within other U.S. Department of Energy (DOE) projects.



Question 1: Approach to performing the work

This project was rated **3.6** for its approach.

- The principal investigator's project and approach is to develop and apply state-of-the-art electron microscopy techniques in the service of the Hydrogen and Fuel Cells Program (the Program). In other words, the approach of this project is to establish unique expertise in using electron microscopy to measure and characterize what is happening in fuel cell components at the atomic level. The work is outstanding and well integrated into the Program as demonstrated, in part, by the fact that both the Program Director and the Team Leader for the Fuel Cell Technologies Office's (FCTO's) Fuel Cells sub-program drew from the accomplishments of this project in their plenary talks.
- This is a critical capability for the development of fuel cells because the microstructural characterization of catalysts layers and critical interfaces allow a true scientific understanding of the real problems that still need to be solved in these devices. Because of this, it enables the fuel cell community to develop more durable and more highly performing devices. The project is very well integrated into the portfolio of DOE projects.
- The project implementation and development of microscopy is excellent.
- The proposed approach is appropriate because it is collaborative with numerous partners and aims to benefit the entire fuel cell community by providing a better understanding of materials evolution in the MEA components. This ongoing characterization project without milestones is also appropriate—as long as the work is following the community's needs—because it supports other DOE projects and provides the fuel cell community with access to state-of-the-art microscopic capabilities.
- The project's characterization really helps the fuel cell community in general. It is encouraged to work with more partners to explore more innovative ideas outside national laboratories.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- This was a very productive year, which focused on catalysts, catalyst supports, and ionomer thin films. For example, the project demonstrated that electron microscopy can map ionomer distribution by using the fluorine on the ionomer to provide contrast, and thus clearly diagnose the dispersion of ionomer in relationship to the microscopic particles of catalyst.
- The Oak Ridge National Laboratory microscopy team continues to develop critical morphological information for all partners. As usual, a huge amount of data has been collected and disseminated to partners using state-of-the-art transmission electron microscopy (TEM) techniques.
- The project remains as productive as in the previous years. The team has made excellent progress with characterizing the ionomer layer while overcoming some practical limitations related to sample preparation and characterization of operating conditions. This progress has been used to investigate deeper into the ionomer distribution and how its degradation depends on the catalyst layers. Still, some uncertainties arise:
    - It is not clear whether there is any explanation for these agglomerate formations depending on given conditions (e.g., carbon support nature). The importance of the ink preparation process in avoiding it is also unclear.
    - It is not clear whether these observations were enough to build a bridge between the ionomer distribution and the associated performance or the active area.
    - It is not clear whether the unchanged ionomer distribution on Pt/low surface area carbon catalyst layer is reproducible or whether it is linked to the given materials and the investigated testing conditions.
    - In the case in which there is strong carbon corrosion, it is not clear whether the catalyst layer becomes compact and stops working. The added value of looking at the ionomer is not clear.
    - Regarding slide 16, it is not clear whether the protocol of ink preparation with graphene is well mastered or whether it needs to be improved.
- The microscopic characterization indeed helps to understand the catalyst, support, and ionomer degradation. The team may need to further correlate other surface properties to its microscopic morphology characterization.
- While this project has gone a long way to answer many questions, it has done so using a very specialized set of tools. There should be more tie-in to more sample averaged techniques, such as x-ray diffraction (XRD) for particle sizes, porosities for pore sizes, nuclear magnetic resonance (NMR) or others for Nafion particle sizes, etc.

Question 3: Collaboration and coordination with other institutions

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

- This project is highly collaborative, with service to the fuel cell community being one of its missions. The collaboration enables it to more quickly meet the project targets. International collaboration is very positive. As an example, the work with the French Alternative Energies and Atomic Energy Commission on energy-dispersive X-ray spectroscopy map microscopic analysis on ionomer dispersion has been widely used this year.
- The project has an excellent set of collaborators, including universities, national laboratories, industrial laboratories, component manufacturers, and original equipment manufacturers. The facility is open to additional collaborations, so this area of the project also continues to improve.
- This project is inherently collaborative in that its purpose is to develop analytic capability while relying on other researchers to provide material samples for testing and study. The many partners and collaborators with this project are summarized on slide 2 and slide 5.
- It has collaborated with many partners. It should work with them deeply.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.7** for its relevance/potential impact.

- Understanding fundamental relationships between microstructure over the range scales and performance is of the highest importance to making any progress in the commercialization of fuel cells.
- This is an extremely relevant project and is critical to advancing technology toward the goals of the FCTO Multi-Year Research, Demonstration, and Development Plan.
- In order to improve fuel cell durability, it is necessary to first understand the behavior and durability of the components at a microscopic (indeed, atomic) level. This project brings such diagnostics into the Program.
- If the DOE goals are dissemination of expertise, there should be some mechanism to transfer the techniques to the community at large (e.g., protocols of samples preparation).
- The project has good relevance but just needs to have more impact.

Question 5: Proposed future work

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

- The future work is right on the money. The fuel cell community absolutely needs more information about where the ionomer is in the catalyst layer, and more work on non-platinum-group-metal (PGM) catalysts and hetero-atom substituted carbons that could help the community understand how these emerging materials will work is always going to be important.
- The proposed future work, as outlined on slide 21, is an appropriate blend of follow-up to previous work (e.g., continuing development of tomographic techniques) and new research with new collaborations (e.g., improving the understanding of non-PGM catalysts).
- The proposed future work is appropriate. Studies on ionomer distribution and degradation should continue with an emphasis on understanding the correlation between ionomer and MEA performances. Close collaboration with FC-127 (Borup) is encouraged. These observations should be performed on different materials (e.g., carbon support, catalyst supplier, and ink preparation protocol) in order to differentiate what is general from what is specific to a given electrode.
- Electron tomography development and application are a good way to reveal catalysts, ionomers, and their interfaces.

Project strengths:

- The project has good national and international collaborations and provides a valuable and continuous service to the fuel cell community. The project offers excellent characterization capabilities and expertise in the fuel cell materials.
- This project is a stable source of technical excellence and helps to buttress many aspects of the Program.
- Cutting-edge development and implementation of electron microscopy to MEAs are project strengths.
- Projects strengths include the state-of-the-art microscopy applied to real problems in fuel cell science and engineering.
- The team has very strong characterization skills.

Project weaknesses:

- This project has no particular weaknesses.
- No weaknesses were specifically identified.
- There should be more tie-in to more sample-averaged techniques, such as XRD for particle sizes, porosimetry for pore sizes, and NMR or others for Nafion particle sizes. Also, a user-accessible database for distributing the data would be helpful.
- The team does not have enough understanding of the impacts of the surface property changes on performance.

Recommendations for additions/deletions to project scope:

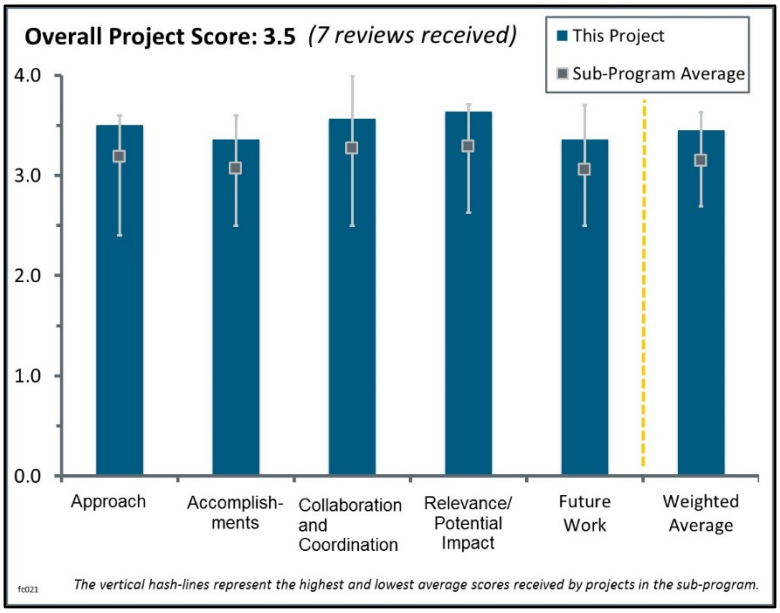
- There should be more tie-in to more sample-averaged techniques, such as XRD for particle sizes, porosimetry for pore sizes, and NMR or others for Nafion particle sizes. Perhaps a set of “representative” MEA samples can be analyzed to address MEA manufacturer comments on the structure of the MEA used in Nafion analysis.
- More in situ characterization of catalysts is recommended.

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

Muhammad Arif; National Institute of Standards and Technology

Brief Summary of Project:

The objectives of this project are to (1) study water transport in single cells and stacks, (2) enable the fuel cell community to study water transport phenomena, (3) tailor neutron imaging to the needs of the fuel cell community, and (4) improve the spatial resolution to provide more detail of the water content in commercial membrane electrode assemblies (MEAs).



Question 1: Approach to performing the work

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

- The National Institute of Standards and Technology (NIST) project for neutron imaging studies of water transport addresses a key performance issue for fuel cells—water management. Water management also has a large impact on cost and durability. The approach (i.e., to maintain a national user facility, develop and maintain fuel cell testing infrastructure, and pursue improvements in resolution) is effective, and the project is sharply focused on addressing fuel cell issues. The project is cost effective for the U.S. Department of Energy (DOE), with a large portion of funding coming from NIST and industry; in addition, the project is leveraging work at the National Aeronautics and Space Administration (NASA) on Wolter optics. The new mail-in service should prove useful for groups that do not have the time or resources to send someone to NIST to run experiments.
- For low-temperature polymer electrolyte membrane fuel cells (PEMFCs), the management of liquid water remains one of the primary areas of concern on the cell and stack levels from the perspectives of both performance and durability. This project is sufficiently focused on providing the tools necessary for industry and academia to understand the distribution and amount of liquid phase present in an operating fuel cell. The approach to continually improve the spatial resolution is directly in line with the needs of the research community, given that an understanding of the water content in the membranes, catalyst layers, and microporous layers is critical for future progress.
- The NIST team has had, throughout the duration of the project, an excellent approach to providing and improving upon a valuable tool for the fuel cell community. As is expected for a characterization project, it is well integrated with the material and cell development and characterization efforts within the Fuel Cell R&D sub-program. The team's approach of continually attempting to improve the resolution of the technique is excellent. In addition, their approach of making this valuable tool more accessible to the community by adding a sample mail-in service demonstrates their commitment to having an impact on the sub-program.
- Neutron imaging of fuel cells and components is a promising approach to image water in fuel cells in situ. This project is taking an excellent approach in developing fuel cells and understanding water transportation in single cells and stacks.
- The mail-in service will certainly save a lot of money and time for users. Continuing improvement in spatial resolution is great, too. The approach taken to try to address the current challenges in fuel cells, such as water and thermal management, is very effective.
- The approach is good and one of the primary pathways through which water in PEMFCs can be identified.
- The overall approach is good because neutron imaging is a key technique for fuel cell water visualization. The use of multiple techniques is a good direction, but it is not clear how much information will be gained

by the combined X-ray studies for cell studies; perhaps information will be gained for component ones, but that does not seem to be the focus.
- The need for better resolution is critical. The mail-in service is a nice idea, although not enough details are given, and it is not clear how much beamtime is dedicated to this service, who will run the actual experiments, and what type of real-time feedback is available.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

This project was rated **3.4** for its accomplishments and progress.

- With the improvement in spatial resolution, actual stack size cells can be accommodated for imaging. The study of the thickness effect of non-platinum group metal (PGM) catalysts to understand er management is very helpful and insightful. The stack imaging experiment with Commissariat à l'énergie atomique was interesting.
- The mail-in service is a very good approach for users because it minimizes the time they spend at NIST. The work related to the non-PGM cathode thickness effects is very good and relevant. Also, the work related to water imaging in a stack is relevant and important. The work related to the neutron lens based on Wolter optics features a very good approach.
- NIST has added a sample mail-in service this year, which is a great benefit to the community and also reduces the cost to the Hydrogen and Fuel Cells Program of this capability, making an impact on fuel cell system development. NIST also added electrochemical impedance spectroscopy (EIS) and an environmental chamber this year, which are excellent added capabilities. Efforts this year in technique development focused on achieving less than 1 µ resolution, and excellent progress has been made. NIST is also adding capability of X-ray imaging.
- The new accomplishments, such as evaluating a full-size cell, are a step forward in approaching real systems.
- The addition of a large-scale test stand should prove beneficial. The first open user experiments to image a stack have been performed. Progress on the new high-resolution cold neutron imaging instrument, which would provide the ability to differentiate between water and ice, is good, and the facility is expected to be operational in July. Resolution improvements to 10 micron are expected by the end of 2017 and to 1 micron by the end of 2018. Acquisition time has been improved using high-resolution image intensifiers. Initial resolution was less than expected, but the vendor is working to correct the issue. The progress on increasing resolution using slit imaging is good, with improved gratings being prepared.
- Interesting work is presented, looking at the differences in water content using the NPGM cathode and the thickness effect. It would have been interesting to see these data with a baseline PGM cathode of similar thickness as a side-by-side comparison. The extraction of the thermal conductivity should be aligned with more detailed and fundamental approaches to ensure the methodology gives consistent results. It would be ideal to report on the progress of the path toward improved spatial resolution with a Gantt chart or a visual of the time frame expected for completion.
- It is tough to determine what is new in a lot of the presented work. Although this presentation is more for the facilities and technique, it would be good to see some aggregated data from the studies or findings that were enabled by the data.

Question 3: Collaboration and coordination with other institutions

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

- The collaborations are very good. Collaborations with NASA on the Wolter optics and Pusan National University in South Korea on preparing gratings have been essential for improving the resolution. Collaborations with General Motors, Los Alamos National Laboratory, and other users are apparent.
- The project features outstanding collaboration with NASA and the Massachusetts Institute of Technology to utilize their lens capabilities for the neutron microscope optics. Also, collaboration with colleagues in South Korea for improved gratings is noted. NIST continued its collaborative efforts with users in the fuel cell community.

- The project seems well coordinated and has excellent cooperation with relevant industrial partners, universities, and national laboratories.
- The project features excellent collaboration with academia, laboratories, and industry. The new mail-in service can be valuable.
- The project features excellent collaborations. NIST is a user facility. It would be good to see how collaborations impacted the findings.
- It would be good to include information regarding the proposal process for access to the facility and the tests that are planned or are in queue. Overall, the project has an extensive list of collaborators, and the focus is primarily on a "service-based" user facility model.
- An unusual number of partners were listed in the presentation. It is hard to gauge what each partner contributed.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.6** for its relevance/potential impact.

- This project is critical to the Hydrogen and Fuel Cells Program (the Program) and has the potential to significantly advance progress toward DOE research, development, and demonstration goals and objectives. Water management issues are key, and this facility is essential for providing in situ measurements to determine where the water is during fuel cell operation.
- Using neutron imaging to understand the water management and localized thermal conductivity can be impactful in designing or optimizing catalyst layers. Identifying the effect of channel geometry on thermal transport from cathode to anode is very insightful.
- The project is targeting the main challenges in fuel cell development: improving performance and durability and identifying water transport in single cells and stacks. Simultaneous in situ neutron/ X-ray analysis is unique -20 keV–90 keV is also a good area for water imaging.
- Water management is highly relevant, especially with the new issues experienced with high-activity, low-loaded catalysts that suffer from additional losses at high current densities.
- The relevance and impact of this work are excellent, based on the impact this characterization capability has on the Program and the fuel cell community.
- Neutron imaging is a critical technique for use in the fuel cell community. The impact of the actual studies is a bit hard to judge.

Question 5: Proposed future work

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

- The proposed future work to increase resolution is appropriate and will help to differentiate boundaries between the gas diffusion layer, catalyst layer, and membrane. Improved resolution will also allow greater resolution in time and provide more accurate determination of how changes in operating conditions affect water distribution. The proposed addition of simultaneous X-ray imaging should prove beneficial in correlating local aging and degradation with water content.
- The addition of EIS will be pretty helpful, and the proposed combined X-ray and neutron imaging will be very interesting because both the porous catalyst layers and water management can be studied simultaneously.
- Improving the resolution is an excellent idea and will lead to greater understanding of water management and identification of local water transport issues.
- Regarding the new high-resolution cold neutron imaging instrument: the goal to achieve a resolution of 1 micron by the end of 2018—but with extremely long exposure times (20 min for 1 micron resolution)—is only suitable for stationary phenomena, but it would still be unique.
- The impact of neutron imaging techniques on the Program is dependent on the resolution of the technique. NIST continues to work toward improving the resolution, and this is an important goal for all future work in this project.

- There is a need for higher spatial and temporal resolution. It seems that this is progressing, but the slides are quite similar to what has been presented before, and it is tough to see if the progress is on track. There are a lot of ways to improve the techniques.
- Information on the status of work completed related the new detectors should be included.

Project strengths:

- The ability to resolve ice and water will present significant value to automotive research and development activities. NIST facilities represent a significant focal point for diagnostics related to *in situ* water content analysis, and the use of the facilities by a broad cross-section of groups ensures a high impact of the work on the detectors and technique capability.
- Neutron imaging is a key technique for the community. Being able to measure water is important in understanding operating fuel cells..
- The project's strengths include its approach, NIST's expertise, its collaborations, and that 50 cm^2—stack-size cells can be accommodated.
- The project features a unique technique for identifying water in PEMFCs. It can be applied to full-scale platforms. Cells can now be mailed in.
- The project's strengths include the excellent research and good cooperation with industrial and institutional partners overseen by the project team.
- Strengths include the unique capabilities of NIST's neutron source and the leveraging of funds from NIST and work at NASA.

Project weaknesses:

- In actual fuel cell operation, water management issues are specific to a given flow field, which is typically proprietary. Although general water management can be understood, specifics for a real stack and a real cell cannot be extrapolated based on these data in subscale cells.
- The project should include a strategic plan on what the use of a higher resolution detector will allow from a fuel cell design activity and what type of processes could be quantified with the higher resolution capability.
- A target of 10 s exposure time with 10 microns resolution (to be achieved by 2017) is not short enough for really dynamic processes.
- The impact to the field is dependent on getting collaborations and key samples, as well as the results of the identified experiments.

Recommendations for additions/deletions to project scope:

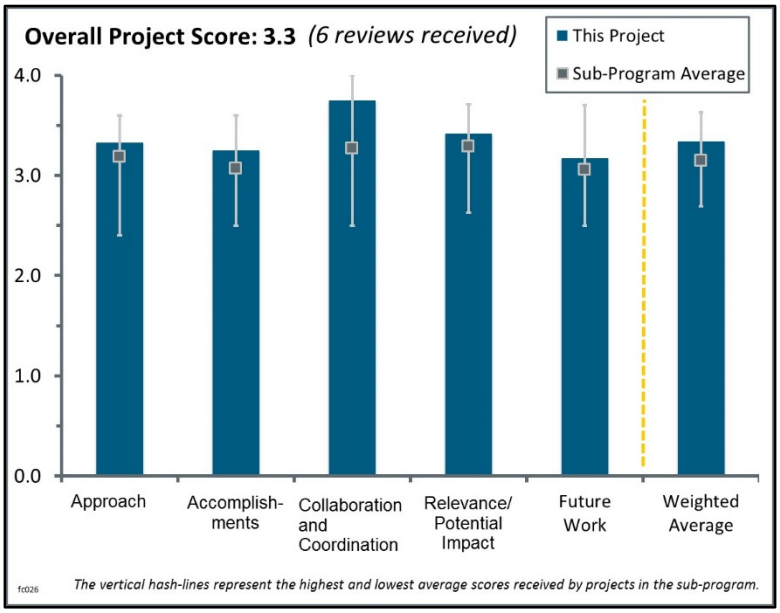
- The project should include a translation from water thickness into a value of local saturation within the MEA; this would make the data more translatable for use in analyses and provide better correlation to performance.
- It would be interesting to see the effect of MEA durability cycling on water management. Also, it would be interesting to compare neutron imaging to X-ray tomography of MEAs.
- At the given resolution, the need to image ice versus water is not required. It would be good to see more progress in size and time resolution.

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

Adam Weber; Lawrence Berkeley National Laboratory

Brief Summary of Project:

The overall objectives of this project are to understand transport phenomena and water and thermal management at low and subzero temperatures using state-of-the-art materials and to elucidate the associated degradation mechanisms due to cold and cool operation. Lawrence Berkeley National Laboratory (LBNL) will (1) examine water management with thin-film catalyst layers, (2) examine water management and key phenomena in the various fuel cell components, and (3) enable optimization strategies to be developed to overcome observed bottlenecks.



Question 1: Approach to performing the work

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

- The approach is sharply focused on resolving durability and performance issues in polymer electrolyte membrane fuel cells employing very thin-film catalyst layers. The approach is well designed because it combines modeling tools and experimental validation in order to understand and mitigate degradation mechanisms caused by cold start and water management at low temperatures.
- The approach is good, and the work plan is thoughtfully distributed among the partners. State-of-the-art materials such as catalysts, ionomers, and gas diffusion layers (GDLs) are used for the study.
- The approach is well structured and includes analytical and modeling techniques that are well developed by this project and previous researchers to develop a thorough understanding of the phenomena. The work plan clearly identifies responsibilities of the organizations but does not identify the task that pulls all of the information together to explain the results of the experimental and modeling efforts. The deliverable should be identified.
- Transport-related barriers are clearly being addressed in a manner that is organized by a state-of-the-art model. However, durability is listed, and it was not apparent how this is being addressed or integrated in the model.
- The general approach of applying ex situ and in situ diagnostics to develop inputs for a cell model that is then applied to explain trends observed in operando is sound. However, there is risk in using conventional serpentine flow fields to develop fundamental data for modeling (e.g., slide 14). At high inlet relative humidity the channel U-bends will likely accumulate water, resulting in “short-circuiting” the flow through the GDL. A pure counter-flow flow field, such as that used by General Motors (GM) and Los Alamos National Laboratory (LANL), would be more appropriate for these experiments and more relevant to transport processes in automotive fuel cell hardware.
- Using adiabatic cell model for freeze start-up is not a practical way to model the cell in a stack. While the middle cell may be approximated as adiabatic when there is no coolant flow, the cells at the end are not adiabatic and are highly relevant for freeze starts. The approach should have contrasted the two extremes for a freeze start.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- Very good progress has been made toward understanding low-temperature performance of nanostructured thin film (NSTF), including catalyst layer resistances and ionomer mass-transport resistances at low loadings.
- The team has made solid progress on understanding the impact of various GDL/membrane properties on the performance of NSTF electrodes.
- The integration of GDL properties into the model and detailed evaluation of NSTF is very good.
- Great progress on several important issues was reported. However, the project has not yet revealed insights regarding NSTF electrodes during cold start. A fundamental description of limitations will enable developers to evaluate the feasibility of applied solutions. The team should continue to focus on interfaces.
- The accomplishments identify the methodologies to be used, and these methodologies have sufficient track records to lead to a successful project. The early modeling results suggest improvements in characterizing observed behaviors. There was no indication that wetting properties or contact angles of water droplets with GDL were considered. If it is assumed that the GDLs are completely wettable, this may not be a valid assumption at the anode. Measurements of catalyst uptake in thick catalyst layer by neutron imaging may not adequately describe the thin catalyst layer water uptake because of differences in porosity and pore size distribution. It is not clear how this is explained. Local catalyst layer resistance increases with lower Pt loading, which could be due to changes in the wettability of the catalyst layer. Pt is more wettable than carbon. If carbon does not have an oxide surface, it is not clear how wettable carbon is. There was no discussion from this perspective; the approach emphasizes electrochemical engineering but does not discuss chemical properties of the materials, unless it was somehow hidden in the analysis. For NSTF catalyst layers in which no carbon is present, it is not clear whether increased resistance would be due to limited "connected" paths in the catalyst layer. It is not clear whether this has this been considered. For a project that started in 2009, a more detailed analysis is expected.
- In regard to slide 14, there are concerns with using serpentine flow field segmented cell data for an along-the-channel model, especially for conditions with significant liquid water. Also, it is not clear whether any effort has been made to compare this model to that developed by the GM-led project that ended in 2014. The GM-led project also developed a publicly available database that could provide additional validation data. In regard to slide 16, it should be emphasized that the presentation of the data points is not indicative of the neutron imaging system's effective spatial resolution but is based on the physical pixel size in the imaging hardware.

Question 3: Collaboration and coordination with other institutions

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

- There is strong collaboration. This project also organizes the transport working group, where a diverse group of stakeholders share their perspectives. This project has been very quick to integrate research needs identified by the working group.
- There is a very well-coordinated effort among LBNL, LANL, 3M, and the United Technologies Research Center.
- The project has a strong and diverse group, which now includes more involvement from automotive original equipment manufacturers to enhance the applicability of the research outcomes.
- An extremely strong team has been organized with expertise in most, if not all, of the critical areas that would lead to an understanding of water distribution in the cell.
- Collaboration among the team members is excellent and evident from the results.
- There is a very broad spectrum of collaborators. It is unclear what the major contribution of each team is.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.4** for its relevance/potential impact.

- This work is excellent and pushing the envelope in understanding fuel cell transport processes. Also, it is very pleasing to see that a technology transfer activity has been included.
- The careful analysis of the contribution of different components to water management issues can help researchers develop mitigation strategies and avoid failures during NSTF operation at low temperatures.
- Understanding the water transport phenomena and water and thermal management at low and subzero temperatures is of prime importance. Different GDLs and ionomers are being used, but the NSTF catalyst system is being studied the most.
- If successful, the project should explain properties of the NSTF catalyst, but it is unclear whether this catalyst is characteristic of real-world catalysts.
- This project seems to be taking on all transport-related issues in fuel cell research. If the investigators continue to integrate current material advancements and fundamental understanding in their model, it will remain highly relevant. However, as the scope continues to increase, there is concern that too much is being attempted with limited resources. The quality of validation will define the lasting impact of this project, and good validation experiments are very tedious. It seems that most of the validation is represented by only a single experimental data set, and this is not sufficient.

Question 5: Proposed future work

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

- Continued NSTF work is important, but shifting even more of the resources toward low-Pt electrodes of conventional design is encouraged.
- Investigation of traditional catalyst layer resistances at low catalyst loadings needs to be complemented by investigation of NSTF resistances.
- The proposed future work addresses the addition of traditional catalysts. Use of the model to determine critical parameters to guide the material development make more sense.
- The “Path Forward” slide identifies problems for NSTF compared to Pt/C systems. It is not clear what the mean pore diameter of the NSTF catalyst layers is.
- Understanding the cold and freeze start performance is critical to enabling the NSTF electrode. However, it is unclear from the proposed work whether the performance gap between conventional and NSTF electrodes can be closed. The team should at least exercise the model to define the parameter space wherein the low-temperature (<20°C) performance of NSTF electrodes are comparable to the traditional carbon-supported electrodes.
- Although it is difficult in a transport/modeling project, the investigators should attempt to define more specific metrics. Much of the future work is simply carried over from last year.

Project strengths:

- This project has a strong team that seeks input from the key stakeholders to remain relevant. The team is continuing to develop collaborations and sharing knowledge in the public domain.
- Project strengths include the very efficient combination of modeling tools, advanced materials diagnostics, and consultations with industry.
- This world-class team is clearly pushing the envelope on the fundamental understanding of fuel cell transport processes.
- Project strengths include the strong team and well-thought-out work plan. The project started focusing on the low-platinum-group-metal catalyst layer.
- Project strengths include the solid modeling and experimental team.
- The project has a strong team and is well funded.

Project weaknesses:

- The project should move away from conventional serpentine flow fields for experiments intended to resolve spatially varying phenomena.
- The project is focused mostly on NSTF. Other catalyst systems were not studied. The catalyst layer resistance is shown to increase with loading, which is expected, and for thinner catalyst layers, resistance increases a lot. However, nothing was mentioned regarding different supports/carbons in a catalyst layer with similar thicknesses. In addition, most of the state-of-the-art low-loading membrane electrode assemblies (MEAs) are shooting for 0.1 mg/cm^2 loading. It would be nice if the team could add this, too.
- The dependence on NSTF is a weakness. While the title says "Fuel Cell Fundamentals at Low and Subzero Temperatures," the work is focused on NSTF.
- The modeling lacks predictive capability.
- It seems that transport in the catalyst layer is the Achilles' heel for a robust model. Lack of validation methods may limit progress. Additionally, this project needs more focus on cell-level validation through parametric studies.
- After several years, there is no indication that the NSTF flooding issues are understood and can be resolved.

Recommendations for additions/deletions to project scope:

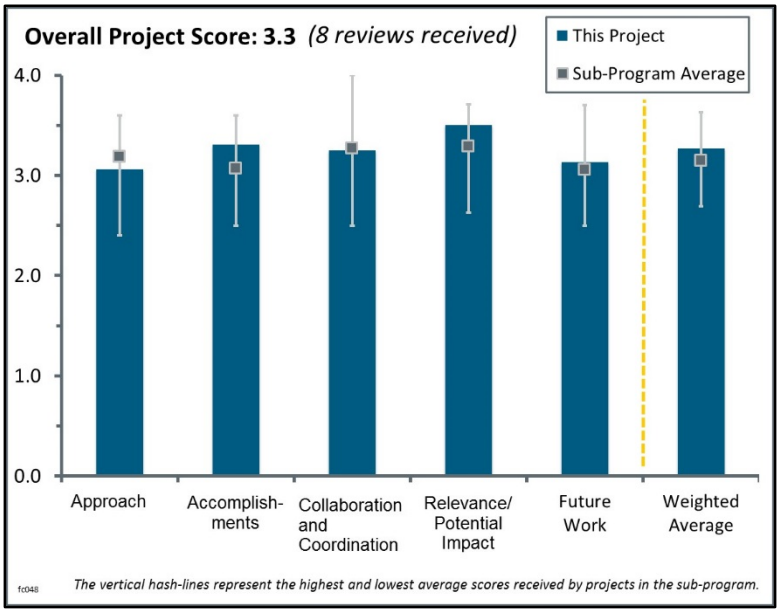
- The effect of loading on the catalyst layer is studied, but most of the state-of-the art low-loading MEAs are shooting for 0.1 mg/cm^2 loading. It would be nice if the team could add this, too. Also, it would be great if the team can perform similar studies for catalysts with different Pt weight percentages but similar Pt loading in MEA in mg/cm^2. Hence, Pt loading in MEA would be similar, but catalyst layer thickness would be different.
- The team should avoid work with thick catalyst layers; the results bring more questions than answers.
- The project should scale back more on the NSTF-focused work.

Project # FC-048: Effect of System Contaminants on Polymer Electrolyte Membrane Fuel Cell Performance and Durability

Huyen Dinh; National Renewable Energy Laboratory

Brief Summary of Project:

The objectives of this project are to (1) understand the extent to which fuel cell performance and durability are impacted by materials used in system balance of plant (BOP) components, (2) identify and quantify contaminants derived from BOP materials, (3) develop an understanding of fundamental degradation mechanisms and performance recoverability resulting from the presence of these system contaminants, and (4) be a resource to the fuel cell community by disseminating findings.



Question 1: Approach to performing the work

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

- The project followed through on the approach outlined in the previous year's future work to quantify leachates, study the effect of leaching parameters, perform mechanistic studies using organic and anionic compounds, and study the effect of both isolated and mixed contaminants. The approach establishes patterns and practices for future contaminant studies.
- One of the objectives of the work is to provide guidance to system developers for BOP material selection. This is solid and relevant work—the current Pt loadings in advanced fuel cell stacks have decreased significantly, so the impact of system contaminants needs to be studied. The selection of caprolactam and sulfate as two model compounds derived from structural plastics is a good approach because the insights obtained from fundamental studies involving these model compounds may be generalized to other compounds with similar functional groups.
- The proposed approach follows a well-thought-out, logical progression from developing identification/quantification procedures for organic contaminants to identifying the degradation products of common fuel cell system components and finally the effect of those degradation components on fuel cell performance and its ability to recover. The establishment and curation of a fuel cell contaminant database at the National Renewable Energy Laboratory (NREL) is an extremely useful tool for the fuel cell community at large. The types of materials and contaminants studied were down-selected with the help of General Motors (GM); however, it would be very useful to look at a broader range of plastics and materials, as well as at greases or sealing compounds commonly found in piping and fabrication, to identify other organic/inorganic materials that could have an impact. This project is slightly different than others that are directly aiming for specific U.S. Department of Energy (DOE) performance targets. For this project, the barriers listed are durability and cost, and technically these are not directly addressed. The results presented, however, will aid future informed/intelligent design of fuel cell components, leading to improved stack durability and lower cost.
- The strengths of the approach include the following: (1) the use of actual cell testing where possible; (2) the identification of model compounds that can represent numerous possible structural materials, assembly aids, or lubricants; (3) the decision to disseminate data on a website that is easy for development teams to use; (4) the consistent reporting of data with standard metrics (delta V1 and delta V2) that are comparable; (5) the use of low relative humidity (RH) operating conditions so that contaminant species are not easily washed away, creating "false positives"; and (6) the standard screening for which contaminants cause

recoverable losses and which cause non-recoverable losses. The use of leachant solutions to determine the appropriate concentration of model contaminants was appropriate. The use of an integral cell (based on stoich) allowed the researchers to encapsulate contamination at both high and low reactant concentrations at inlet and outlet of the cell, respectively, and it also allowed for the detection of mass transport losses. It may be interesting to take 1–2 contaminants and determine whether different cell temperatures would have a profound effect, or whether different gas concentration (hydrogen or oxygen) would also have an effect. The reported data for this year appear to be on 0.4 mgPt/cm^2 cathode catalyst layer loadings. Lower loadings would be preferred (lower loadings were used in past years, but not this past year). While 0.1 mgPt/cm^2 may be aggressive (close to target), a loading on the order of 0.2–0.3 mgPt/cm^2 would be preferred as a baseline versus 0.4 mgPt/cm^2. Pt loading needs to appear in the fuel cell performance plots shown on the NREL website.

- The approach is a survey of the effect on fuel cell performance of leachants from practical structural materials. The project features a direct study of fuel cell performance as well as other characterizations, such as voltammetry, rotating disk electrode (RDE) polarization curves, and quartz crystal microbalance and chromatographic techniques, to develop an understanding of both time dependence (exposure time and reversibility when exposure is removed in time) and the mechanism of leachant effects. Such an understanding is needed, mainly under conditions of normal use for fuel cells, but also during storage (at least 150°C as for batteries). The team may want to think more about extreme conditions to try to learn the effects of heat (e.g., on leaching rate and decomposition products from otherwise benign materials) and environmental conditions such as automotive exhaust, salt spray, etc. Developing a website on contaminants is a good idea.
- The approach is a good start at identifying contaminants that could impact performance. The assumption that standard original equipment manufacturer (OEM) plastics can be used with fuel cell power plants should be reevaluated. Furthermore, if a leachate is identified and shown to impact the performance, it is unclear why there is a need to study the degradation mechanism. The team should spend the time finding more suitable materials. The Japanese combined heat and power units have significant lifetime results. It would be good to know what materials they use. The value of the approach is good. Identifying potential chemical species in any of the subsystems is valuable. Setting up a fast screening approach would be invaluable. The team should not hold the current density at 0.2 A/cm^2—that is too low. The team should use 0.4 A/cm^2 or higher.
- The approach to identify leachate from materials used in fuel cell systems, and then dose them to an operating fuel cell, is good and showed a clear response from the fuel cell. However, it is a very slow and labor-intensive approach, and thus only two materials were studied. In reality, the number of materials that need to be examined is large and impractical to analyze using this approach. Perhaps a more rapid screening approach could be envisioned.
- The approach in pursuing the influence of contaminants on fuel cell performance is appropriate and is capable of addressing the main goals in this project. The choice of analytical techniques is adequate and is properly utilized throughout the project.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- One output from this project is the user-friendly and interactive website that contains a database of the contaminants studied by the team. The website provides a material screening tool that is a very good resource for system developers. The gas chromatography mass spectrometry (GCMS) method that the team developed to quantify the concentration of contaminants in the leachates allowed the team to obtain a range of concentrations that would serve as more realistic doses for future infusion experiments. More realistic doses of contaminants will provide more relevant information to system developers. The team has identified the major species in the two structural plastics—polyamide (PA) and polyphthalamide (PPA)—that have commercial relevance to automotive applications. This is good progress because the researchers have found that the less expensive material (i.e., PA) leaches out more contaminants. Without this kind of knowledge, system developers would almost certainly use the cheaper plastics in order to lower costs.

- This project is very well focused and executed to overcome major obstacles in fuel cell performance coming from the impurities that could be found in real systems. The principal investigator has demonstrated significant progress in identifying the contaminants that could be present and has made impressive effort in quantifying the correlation between impurities and performance. This project has a very important role in addressing the DOE mission, and findings from this report are a valuable asset for the stakeholders from industry and research institutions.
- To this point, this project has successfully accomplished many of its objectives, including development of an identification/quantification procedure for system component degradation products and their impact on fuel cell performance. Half-cell was used in combination with electrochemical quartz crystal microbalance (EQCM) to identify the specific adsorption of degradation components of plastics and ionomers. No specific interaction of the sulfonate component of perfluorosulfonic acid (PFSA) ionomer and membranes was found on Pt; this result is in contrast with much of the literature, and an explanation for this should be presented. Accomplishments to date are well in line with project goals.
- The results were clear—the causes and effects were directly noted in a meaningful way; for example, the effect of impurities on the main fuel cell reactions, hydrogen oxidation and oxygen reduction, was measured as a function of impurity concentration and exposure time, both in a fuel cell and using ex situ techniques such as RDE.
- The project did a very good job of distinguishing whether contamination was associated with high-frequency resistance increases, which is intended to be indicative of membrane contamination. For PPA, PA, and caprolactam, there did not appear to be a loss of more than 6 mV (or less) at 0.2 A/cm^2. The project was responsive to prior feedback that low contamination at low cell RH should be studied. This should have prevented washout of contaminants. The lack of voltage loss with exposure of 10 mM H_2SO_4 prompted considerable discussion. The project team used methods that relied on a liquid carrier (infusion or EQCM), and some suggestions were made that more harmful contaminants would not be water soluble. That said, the project team did attempt to observe contamination from longer chain molecules that contain sulfonic acid groups. EQCM showed little effect by a chain with a sulfonic acid group alone, but it showed an additive effect of the sulfonic acid group when combined with a carboxylic acid group in the same molecule.
- The project team identified major organics in leachates and developed GCMS methods to quantify concentrations, demonstrating that solid phase micro-extraction can be used to observe trace species. There was speculation that caprolactam was contaminating the ionomer; it would have been nice to see more concrete evidence via model x-ray photoelectron spectroscopy (XPS) studies or similar methods. The team showed interactions between individual leachates, with the surprise finding that the presence of sulfate actually reduces the deleterious effects of caprolactam. It was not entirely clear why membrane degradation compounds were being studied—the project seemed focused on contaminants originating from the BOP.
- The overall progress and accomplishments are good, especially the ability to identify the leachates. The ability to see the impact on an electrode is good. Identifying chemical species is challenging. Knowing the percentage of leachates not identified but present would also be of value. Contributing to the NREL website is very positive. It is not clear whether PFSA is the membrane or the ionomer, or both. The impact of fluoride ion or fluorine-containing polymer fragments on electrode performance is also unclear. Few (side chain) sulfonic acid degradation issues were seen.
- Only two materials were studied from two suppliers. The same materials from other suppliers could have different impacts on the fuel cell system.

Question 3: Collaboration and coordination with other institutions

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

- The project is one of the better projects in the DOE portfolio for reflecting a spirit of collaboration. First of all, the project team collaborated with an automotive OEM (GM) in order to obtain an understanding of which materials are relevant, and, therefore, which contaminants are relevant. Second, the project has been highly responsive to reviewer feedback, particularly feedback to study sulfates and sulfonic acid groups and to use low RH conditions. The collaboration with the Colorado School of Mines (CSM) was seen from the EQCM data. These data showed the effect of membrane degradation products that were difficult to analyze under fuel cell conditions. This collaboration assisted with understanding why sulfuric acid did not have an

effect on cell performance, but other membrane degradation products would, especially some containing sulfonic acid groups. 3M provided membrane degradation products for the CSM collaboration. Some added background would be useful to help reviewers understand whether these degradation products would be the ones expected under GM drive cycle conditions.

- The involvement of GM ensures that the contaminants being investigated are relevant to automotive fuel cells. GM's involvement also makes the contaminant concentrations used in the infusion experiments relevant and reliable.
- Collaboration with partners, including GM and 3M, has been integral to the successful progression of the project to this point.
- Collaborations in this project are well executed and coordinated between GM, CSM, and NREL.
- The collaboration and coordination is consistent with the magnitude of the effort. Teaming with GM is a positive.
- The team leverages academia, such as Ryan Richards of CSM; materials suppliers, such as 3M, for samples; and end users, such as GM, for advice. There was more information about these collaborations in the archived slides than in the oral presentation, but it is clear there are adequate inputs about contaminants from many points of view.
- There is good engagement with current partners, including industrial partners at 3M and GM. The project would benefit from increased interaction with other industrial partners and OEMs.
- The presenters only assumed the parts per million (ppm) level of contamination introduced into the fuel cell. In the spirit of collaboration with GM, the authors should analyze the inlet of a real fuel cell system using the suspect materials and see what the ppm level of contamination is in a real fuel cell system.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.5** for its relevance/potential impact.

- This project is very important because the performance gains obtained in other DOE-funded projects, such as low Pt loading and advanced Pt alloy catalysts, may be negated if system contaminants degrade fuel cell performance and durability significantly. With the findings from this project, performance degradation due to contaminants may be avoided.
- The team is focused on meeting the needs of the Hydrogen and Fuel Cells Program (the Program) and will advance progress by avoiding materials that can lead to poor performance of fuel cells. It will also avoid diversions in making progress on performance development. "Real" performance can be masked, since real-world fuel cell performance is dominated by the effects of contaminants leaching either from structural materials or from the environment.
- Attempts to achieve system-level specific power targets prompt the investigation of lightweight materials that may not be compatible with the fuel cell stack. The same is true with respect to meeting system cost targets. Results of the predecessor project (funded from 2009 to 2013) have yielded a website that has become useful to developers and has helped to reduce the time needed to research answers to material selection questions. While contamination-related lifetime limitations are rarely addressed publicly, because of competitive concerns (unlike matters such as carbon corrosion, Pt dissolution, etc.), the practical reality is that these limitations can be encountered within a vehicle program, and having a general resource available to address contamination can be helpful in ways that DOE may never consider.
- This work is extremely relevant. It is important in clearly understanding long life objectives, cost, performance, etc.
- The goals and results of this work align well with the objectives of DOE and the Program. With the majority of research and development focusing on membrane electrode assembly (MEA) and stack development, little effort is focused on addressing the impact and interaction with the components that make up the balance of systems. Identification of the detrimental effects of some of the compounds that make up those components is of critical importance for addressing the longevity of the polymer electrolyte membrane (PEM) fuel cell. What is missing is a more comprehensive survey of potential balance of systems components and development of strategies to either remove or mitigate their impact on fuel cell performance.

- This project already made progress toward the DOE goals by listing accomplishments that provide important guidelines for developers in addressing the DOE technical targets.
- Over the course of the project, a variety of contaminants have been studied and information has been disseminated via the website. This year, advanced methodologies were introduced for identifying and quantifying organic and anionic contaminants that could be utilized by the broader community. Ongoing use and development of the website should be tracked and reported in future reviews.
- The relationship to actual operating fuel cell systems is not clear. As shown on slide 8, it is not clear that exposing 1 ml of deionized (DI) water to 1.5 cm^2 of sample material for 1,000 hours at 90°C has a reliably known relationship to the dosing level of contaminant used during fuel cell tests. For example, in a real system using the studied material, in 1,000 hours, it could be that 10,000 ml of DI water passed the 1.5 cm^2 surface of the sample material, not 1 ml. Therefore, the contamination level in the actual fuel cell inlet could be 10,000 times less than the presenter used in the dosing level for the fuel cell tests. The authors should measure the real ppm level of air inlet contamination in an operating fuel cell system when using the evaluated materials; one suspects it could be much, much lower than the authors used in the dosing experiments. It is not clear how long this level of contamination exists in a real fuel cell, and whether it is confined to the first 100 hours of life. The relationship of where the material is used in the system and the resulting level of contamination is missing. Instead of reporting the cell voltage response to contaminants, the authors should present power output reduction at a fixed cell voltage.

Question 5: Proposed future work

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

- The proposed future work is well aligned with the progress made in this fiscal year and will continue to make progress toward DOE goals.
- Understanding the underlying mechanisms of how the contaminants impact fuel cell performance, particularly possible contaminant-ionomer interactions, is of high importance.
- Lower Pt loading is mentioned in the future work, which is needed. The direction with advanced catalysts is consistent with developments in the rest of the DOE portfolio. To be further consistent with the portfolio, it may be helpful to include 3M polyfluoroimid acid (PFIA) fragments, as well as contaminant effects on PFIA. The directions to look at the effects on ionomer in catalyst layers, and to look at volatiles that evolve from structural materials, are good. Because species such as PtNi likely affect catalyst layer ionomer, it would be wise not to neglect this effect.. Volatiles may come from materials other than structural materials as well. More emphasis on operating conditions may be useful, particularly with respect to temperature and reactant gas concentrations. More studies varying RH are also recommended. High current density and mass transport losses due to contamination appear to have been deemphasized in this year's presentation. Some attention should still be paid to contaminants that might limit performance by changing the surface energy of porous media in the cell.
- Performing mechanistic studies on mixtures of model compounds is a good extension of the work done this fiscal year. In the actual fuel cell system, the contaminants will be mixtures of different species, so findings and insights from these studies will be more relevant to OEMs and system developers. Studying the effect of contaminants on low Pt loading electrodes and advanced alloy catalysts is also a natural extension of the work because these catalysts may be more susceptible to organic contaminants. Also, because it is known that OEMs are actively working on low Pt loadings for their next-generation fuel cell stacks (~0.1 to 0.2 mg/cm^2 Pt), these types of studies would provide timely and relevant information.
- The proposed future work appears sufficient and addresses the interaction of different compounds as well as their effect on loading-optimized MEAs and catalyst layer ionomer. Expanded survey of materials and potential leachates, as well as the impact of water quality, would be helpful. It would also be useful to provide some insight into the development of contaminant mitigation and recovery strategies.
- Overall the future plans are good, but before worrying mixtures, the team may want to consider the effects of extreme conditions to try to learn the effects of heat (on leaching rate and decomposition products from otherwise benign materials) and environmental conditions such as automotive exhaust, salt spray, etc.
- The proposed work is good. The team should consider focusing more on the identification process and the impact on performance than the mechanistic studies.

- A more direct relationship between materials used and their impact in a real fuel cell system should be included. For example, even for the materials studied in this project, it is not clear whether these materials could be used in the fuel cell system at different locations where the impact could be much less or none.

Project strengths:

- The project is responsive to reviewer feedback—it addressed low RH conditions and the possible role of sulfonic acid group or sulfate poisoning, and it used diagnostic techniques such as electrochemical impedance spectroscopy to distinguish ohmic losses from kinetic losses. The project uses consistent metrics (delta V1 and delta V2) to derive consistent comparisons between contaminants for both the time period during which contamination occurs and during recovery. The project has disseminated data in a fashion that is immediately useful to developers. The website can be accessed quickly and is user friendly.
- The dissemination of research findings via NREL's updated website is excellent. This will help system developers in designing BOP components and avoiding fuel cell performance degradation. Insights obtained from the team's fundamental studies may be generalized to other plastics/components with similar functional groups.
- The project successfully developed highly sensitive and quantitative analytical tools for the identification of potential contaminants as well as their direct impact on PEM fuel cell performance. Another project strength is the development and curation of the contaminants database, which is a very useful resource for the fuel cell community at large.
- Project strengths include the well-balanced approach, well-chosen and efficient team work, and systematic evaluation of contaminants.
- The project has done well in developing methods and tools for studying and quantifying BOP contaminants that can be utilized in future studies.
- The project features good teaming and a good methodology for studying the effects of contaminants, and it is systematic.
- The project features a good team, and there is excellent analytical equipment available.
- The project features good analytical tools and analyses. The team is good, including the partners. The team has good electrochemical knowledge. Focusing on the "model" compounds is a good start from a process perspective. It may become a weakness, however, if a broader approach is not taken. Leachates should be identified and tested, but not over-studied.

Project weaknesses:

- The baseline cathode catalyst layer loading of 0.4 mg Pt/cm^2 is still too high. Although the project attempts to use ex situ means with model compounds to address contaminants that may not be water soluble, there is still an open question about what water-insoluble contaminants might be able to do to a fuel cell if there was some way that they would enter the cell. Assignment of contaminant effects on cyclic voltammetry features are often attributed to the contaminant, but there may be some possibility that the contaminant could itself degrade or cause other materials to degrade, which would then lead to the features noted. The project could use greater emphasis on high current phenomena.
- Not a lot of consideration is given to environmental effects such as heat, ambient environment, etc. interacting with the system. For example, heating during storage of a fuel cell can decompose materials, accelerating leaching and even decomposing materials to make contaminants; some environmental materials, such as salt, can ion exchange with membranes, causing a drop in conductivity and poisoning catalysts.
- The project has a limited scope that is focused on only two potential contamination compounds. It is unclear why sulfonate appears not to adsorb. Previous half-cell studies have shown very clear evidence for the adsorption of sulfonate from Nafion onto Pt.
- The project lacks specific mitigation strategies for dealing with contaminants. The focus has become too narrow, with only two contaminants used in this study.
- This could be a huge project with significant value. The team needs to focus on the right materials or the scope may become a weakness.
- The relationship to a real fuel cell system is missing,
- The list of contaminants could be expanded.

Recommendations for additions/deletions to project scope:

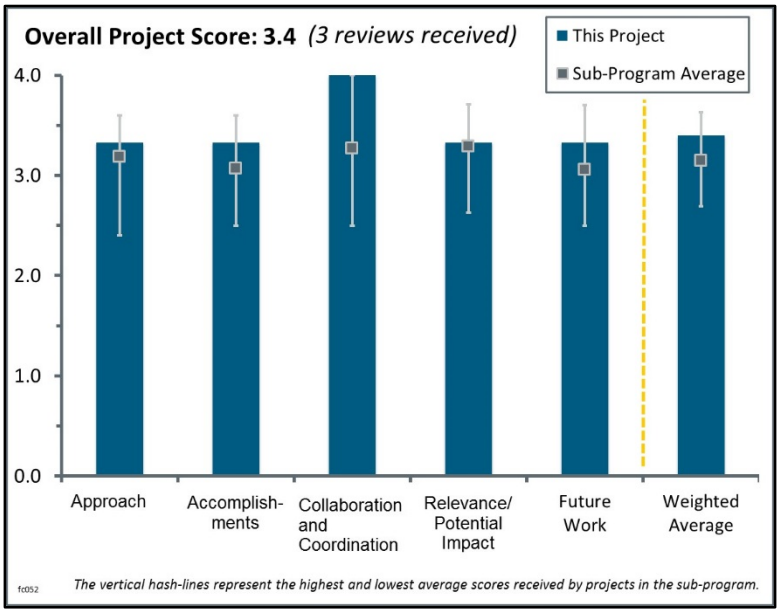
- The team should (1) increase its emphasis on high current phenomena; (2) use limiting current techniques to understand mass transport losses imposed by a contaminant; (3) study contamination effects on advanced catalysts; (4) study contamination effects derived from the breakdown of PFIA membranes or other advanced membranes from the portfolio; (5) study volatile or water-insoluble contaminants; and (6) study all contaminants at greater range of operating conditions, including temperature, reactant gas concentrations, and RH. The team should remove the 0.4 $mgPt/cm^2$ cathode catalyst layer loading and replace it with a lower baseline loading.
- Limiting current measurements in single cells should also be performed in order to determine the effect, if any, of the contaminants on the pressure-independent gas transport resistance, as reported in recent GM, Toyota, and Nissan papers on oxygen gas transport resistance in the cathode. The team should also investigate and report the effect of contaminants on the proton resistance in the cathode in the near future.
- The project team should mainly consider the interaction of the environment—such as heat, salt, etc.—on decomposing materials and leachants from these materials to make poisons that affect fuel cell performance. The team should look at the effect of free sulfate on the anode ($H_2 + H_2SO_4$ yields elemental sulfur, a poison to Pt catalysts).
- The researchers should determine the effect of different sources and qualities of water. They should expand analysis to include a larger library of materials and compounds. They should also develop contamination mitigation strategies and/or methodologies for faster and more complete recovery.
- Considering the project's relevance and systematic approach in addressing the barriers, it would be very useful to include chloride anions to the list of contaminants.
- Adding input from more fuel cell developers to the project would increase the range, depth, and overall impact of the work on the fuel cell community.
- The team should identify, analysis, test, develop a database for leachants, and then move on. The team should not over-study the "whys" in this specific project.
- The project's weaknesses could be mitigated by closer interaction with GM.

Project # FC-052: Technical Assistance to Developers

Tommy Rockward; Los Alamos National Laboratory

Brief Summary of Project:

Los Alamos National Laboratory (LANL) will test catalyst materials and participate in the further development and validation of single-cell design and test protocols. LANL will also provide technical assistance to working groups, the U.S. Council for Automotive Research (USCAR), and the USCAR/U.S. DRIVE Partnership Fuel Cell Technical Team.



Question 1: Approach to performing the work

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

- This work is an excellent initiative by the U.S. Department of Energy (DOE) and LANL that provides a mechanism where developers (e.g., start-up companies and academic laboratories) can take advantage of LANL's technical expertise and fuel cell research equipment/capabilities.
- The mechanism for applying for consideration is straightforward and involves submitting a formal request to DOE and LANL.
- A mechanism is in place to protect the confidential information/intellectual property of the organization seeking technical assistance.
- It is apparent that LANL used sound technical approaches in the tasks it executed in support of fuel cell developers.
- The issue of "Approach" is not relevant here in some respects because LANL essentially responds to the Fuel Cell Technologies Office's requests. However, LANL's approaches to each of the tasks assigned do seem appropriate.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- LANL was able to assist several developers with various studies. For example, institutions such as Pajarito Powder (a start-up company), Indiana University – Purdue University Indianapolis (IUPUI) (an academic institution), and Argonne National Laboratory (ANL) (a premier national laboratory working on advanced fuel cell catalysts) have sought assistance with membrane electrode assembly (MEA) testing of their materials. The project has also helped an original equipment manufacturer (Ford) in its efforts to sputter-deposit Pt onto powder substrates (using LANL's vacuum deposition system).
- Particularly noteworthy is the MEA testing of ANL's advanced PtNi nanoframe catalysts in this project, which shows the impact and usefulness of this technical assistance project. LANL has expertise in the fabrication and testing of MEAs, while ANL is known for its fundamental science and rotating disk electrode (RDE) testing expertise. Through this program, MEA data were generated using ANL's state-of-the-art nanoframe catalyst, an important step in validating the potential of these high-activity, novel catalysts.

- The team appears to have accomplished an impressive amount of work for the budget allocated. (Note: In collaborations with non-U.S. entities, it should be clearly stated that funding is also provided from these sources.)
- This is an extremely difficult project to assess on the basis of progress toward overall DOE goals. DOE made considerable investments at LANL over the past several years, and LANL has world-class facilities and capabilities. Using LANL in this technical support capacity is a great way of extracting further value from that investment. It is especially gratifying to see small businesses taking advantage of LANL's capabilities in this project because it would otherwise be very difficult for them to do so.

Question 3: Collaboration and coordination with other institutions

This project was rated **4.0** for its collaboration and coordination.

- This project necessitates close collaboration and coordination with the institutions that require technical assistance, and from the quality and variety of results presented, it looks like there is good coordination between LANL and the organizations involved.
- This project, by definition, is collaborative in nature.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.3** for its relevance/potential impact.

- This project may provide a way for organizations who are synthesizing high-activity, novel catalyst nanoparticles (e.g., shape-controlled octahedral alloy catalysts) to obtain reliable MEA performance data and not just rely on RDE testing. This project may also help entities who are actively working on new catalyst layer design/architecture using LANL's x-ray tomography capabilities.
- LANL's availability to assist, as needed, can have significant impact on certain projects.
- The potential impact is determined more by the impact of the projects that the program supports, rather than the program's specific tasks. Assuming that LANL is supporting high-impact projects and is performing project-critical tasks, then the potential impact is high.

Question 5: Proposed future work

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

- LANL's plans for currently defined efforts are sound.
- The future work is mostly extensions of the current collaborations. It would be good to see a report on the number of organizations that have formally requested assistance and the number of requests that were approved.

Project strengths:

- This work is an excellent initiative by DOE and LANL in providing a mechanism where developers (e.g., start-up companies and academic laboratories) can take advantage of LANL's technical expertise and fuel cell research equipment/capabilities.
- This project leverages the world-class capabilities and facilities that have been established over several years of DOE investments.
- This project allows others to utilize and benefit significantly from LANL's valuable capabilities.

Project weaknesses:

- *[No reviewers provided feedback.]*

Recommendations for additions/deletions to project scope:

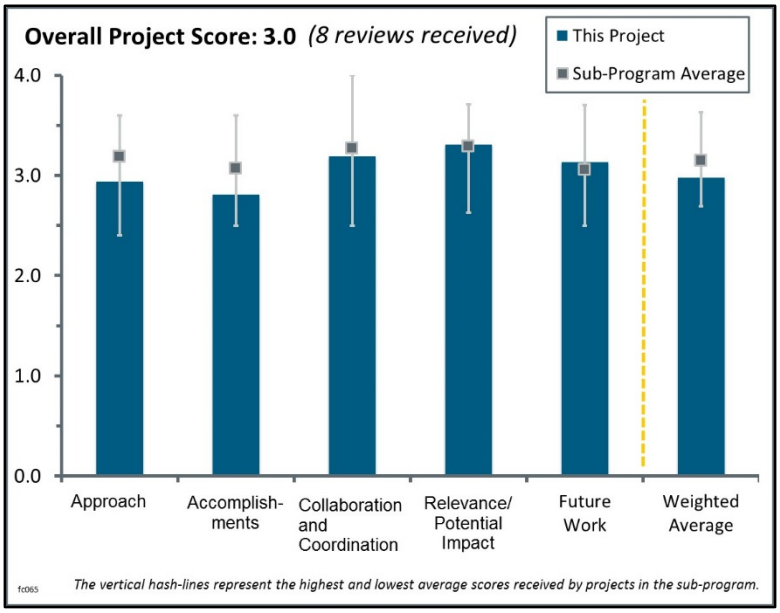
- There are no recommendations from the technical side. From the managerial side, it might be a good idea to have the companies that receive LANL technical support complete a survey that assesses the quality of work performed by LANL and the value of this work to the recipient companies. Committing to this survey could be considered a prerequisite to obtaining the support. DOE (and LANL) should be tasked with working out the details of the survey (e.g., whether the survey should be designed and executed by DOE or LANL).
- The team should add Brookhaven National Laboratory's (BNL's) project (FC-126) to the list—to better understand why the air performance of its MEAs is poor.

Project # FC-065: The Effect of Airborne Contaminants on Fuel Cell Performance and Durability

Jean St-Pierre; Hawaii Natural Energy Institute

Brief Summary of Project:

The overall objectives of this project are (1) to identify and mitigate the airborne contaminants adversely impacting fuel cell system performance and durability and (2) to identify materials, design, operation, and maintenance recommendations to maximize performance. The FY15 project objective is to demonstrate mitigation for species that do not lead to a performance recovery by ceasing contaminant exposure.



Question 1: Approach to performing the work

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

- The approach is well laid out; a Gantt chart describes the approach. The sequence of determining/characterizing the effect of various contaminants on the fuel cell performance, the long-term effects of the contaminant, and the effect of recovery methods (acidic or cleaning solution) is a reasonable approach to evaluate the poisoning effect. Furthermore, the coupling of contaminants (organic and cation) is very important because some effects may be magnifying while others may be dampening, and this will be key to understanding the real-world application scenarios.
- The approach, which uses a selection of contaminants based on probability of being in the air and the effect on membrane electrode assembly (MEA), is correct, particularly as it helps avoid overlap with air contaminants that have already been studied. The table indicating the rationale of the investigated species is appreciated. The systematic approach to identify and characterize potential airborne contaminants is correct. It will lead to an improved understanding of the types of poisoning and potential recovery methods. However, air contaminant mixtures should also have been investigated before the end of the project, as it is well known that not all effects are additive. Investigating the contamination effect on low Pt loading is appreciated. This effort shall guide not only MEA or system developers but also the development of advanced air filters.
- The approach in this project is well balanced with the milestones and effectively addresses fuel cell technical barriers and limitations that might originate from airborne contaminants.
- The approach is generally effective; the principal investigator (PI) is looking at the right class of materials. Nonetheless, it would be beneficial to see greater emphasis on the following:
    - Lower catalyst loadings relevant to current state-of-the-art and future directions (~0.1 mg/cm^2)
    - More focus on real-world contaminant levels (it is not clear if the mechanisms and recovery procedures are the same.)
    - Greater examination of contamination while under voltage cycling that is reflective of real-world operation.
- As a whole, the project has had an effective approach. However, the steps used to poison the MEA with Ca^{+2} are not comparable to how the MEA would be poisoned in real-world applications (especially utilizing a surfactant to help the Ca^{+2} ions penetrate the MEA and saturate the catalyst layer). This could have a large impact on where the Ca^{+2} ends up in the MEA and therefore on how the Ca^{+2} impacts performance. A method of exposure of the MEA to the impurity that is more similar to what would be expected in real-

world applications should be employed. Catalyst loadings have, for the most part, been high. More low-loading studies would be beneficial.

- The project focuses on studying air contaminants for fuel cells. This is clearly an area of merit but not one that has been critically limiting the technology to date, and therefore it is of limited impact. Additionally, the choice of eight specific contaminants has not been strongly justified, and in many cases, the proposed contaminants seem highly unlikely to be major concerns for fuel cell systems. The team also investigated recovery techniques that required difficult implementation (e.g., the addition of isopropanol) to be valuable in deployed systems. It is good to see a focus on low Pt loading.
- The approach used by the PI and his team is rather standard, following U.S. Department of Energy (DOE) and Fuel Cell Technical Team recommendations. Cells are subjected to individual contaminants, performance loss is determined, contaminant is removed, and finally the performance recovery and its degree are studied. While interesting from the point of view of determining performance loss under various conditions and proposing methods of (partial) performance recovery, the project falls short of providing understanding of the mechanism of processes behind the observed phenomena. That includes providing a convincing, science-based rationale for some of the recovery techniques used, e.g., the role of iso-propanol in the performance recovery after Ca^{+2} contamination.
- The work lacks depth and the deeper understanding of the mechanisms that the team is trying address. This may be due to the team trying to do too much or due to the team lacking the ability to draw deeper conclusions from its tests. The results shown do not indicate that a proper design of experiments was crafted or executed. Test parameters—such as acid concentration, time of exposure, and delivery point of exposure—had little to no variation, and no justification as to why the test parameters were chosen was given. In addition, little to no post mortem analysis was conducted to determine why the chosen techniques did not fully recover performance and what the mechanism behind the remaining unrecoverable losses was.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The accomplishments made in this project are invaluable for further advancement of fuel cell technology. The project is very well executed with carefully chosen systems that complement previous projects related to contaminants. This funding cycle was particularly focused on the effect of contamination by Ca cations, and the problem was addressed by systematic evaluation of MEA performance with the addition of Ca, which was followed by acid wash and MEA performance recovery. In addition, long-term evaluation tests were performed in the presence of acetonitrile. The PI provided performance decay rates and contaminant tolerance limits that can serve as guidelines for fuel cell developers.
- This study has generally been effective in determining the effects of contaminants on fuel cell performance. The work is focused on two critical issues: durability and the ability to withstand environmental contaminants the fuel cells would come into contact with in the real world. Ionomer content could have an impact on how Ca^{+2} is distributed and its effect on performance. The effect of ionomer content in the catalyst layer on Ca^{+2} poisoning was not investigated. Mitigation strategies running under conditions where more water was being formed (rather than the relatively low current density operation point) would be of interest to see if Ca^{+2} is flushed out with operation. The PI looked at long-term tests with acetonitrile and showed decreased fluoride emission with acetonitrile present. Tests under voltage cycling conditions are of interest to compare with constant current measurements.
- There seemed to be less progress this year as compared to the previous; in part, this was due to recognized issues with dosing aqueous cation contaminants to the electrode/membrane. The project team did address some of previous year's reviewers' concerns (e.g., the 0.1 mg/cm^2 catalyst), but it still should get more emphasis.
- Some lead-in discussion of the cell hardware used in the project would be useful in understanding the relative levels of cell behavior. Based on the data and mechanism for Ca^{+2} poisoning, it would be ideal to present the transient data as the primary analysis rather than a "steady-state" curve, because it is not clear based on the data presented that the system has reached a "steady" behavior with the poisoning behavior. Essentially, it is unclear what the lower plateau level that the poisoned performance will reach is. The work

on acetonitrile is particularly interesting, and more work on the presence of additional Fenton's Reagent in the membrane would be relevant in the comment on radical scavenging.

- The team has performed well this year with its focus on the gas diffusion layer (GDL) effect on the Ca^{+2} effect and has made several guidelines and discoveries that will benefit the fuel cell community as a whole. However, the PI should have explained why there was a focus on Ca cation and if this species could be seen as a reference for cation contamination similar to how CO is seen to have a "Canary effect" on H_2 fuel quality. Regarding the GDL effect on Ca^{+2}, it is not clear if it is only a GDL effect or if the microporous layer is also involved. Perhaps the effect would be the same with catalyst-coated backing MEAs. Acidic flushing has some positive effect. Nevertheless, it might be difficult to implement it in a real system. Long-term contamination testing on a low Pt loading MEA is interesting if the investigated MEA is representative. The used MEA shows a huge degradation rate in reference conditions and, therefore, creates doubt regarding the reliability of the acetonitrile impact. The team should contact MEA producers providing state-of-the-art low Pt loading MEAs.
- The number of contaminants studied in the past year has been quite modest. Most of the attention has been focused on Ca^{+2} as an important contaminant of the ionomer. The role of acetonitrile in ionomer degradation has been the other focus area, as fewer data have been reported. The mitigation strategies used after the cation contamination have led to recovery of only partial performance.
- Efforts in past year were highly focused on Ca^{+2} and acetonitrile. Ca^{+2} as a divalent ion that could show up as a road deicer has some value, but specific tests did not give meaningful new insight into ionic contamination. Earlier work by Greszler et al., Kienitz et al., Mikkola et al., and/or Weber et al. (including models) exhibited much clearer fundamental understanding of ionic contamination and more detailed experimental studies delineating loss mechanisms and recovery. Acetonitrile has very limited interest in fuel cell systems as a contaminant because no data have been presented that suggests it is likely to be a specific exposure concern to fuel cell systems.
- Comments from last year's reviewers have not been adequately addressed. The mechanisms are not fully understood. A quick check on fuelcelletc.com shows that MEAs with Pt loadings down to 0.03 mg/cm^2 are readily available for purchase. Only one approach for recovery of foreign cations was tried (adding sulfuric acid to the inlet stream). Although this showed some success at recovering fuel cell performance, this test is likely not a feasible option in functioning fuel cell electric vehicles (FCEVs). No other alternatives were suggested or attempted.

Question 3: Collaboration and coordination with other institutions

This project was rated **3.2** for its collaboration and coordination.

- Collaboration with project partners and with University of Connecticut (UConn) has been good. The Hawaii Natural Energy Institute has done a good job of collaborating with the fuel cell community as a whole to determine which contaminants to investigate.
- This project involves several collaborators outside the team of four organizations that are under the DOE contract. The "other collaborators" have well defined roles that benefit the research in the project and enhance its relevance to original equipment manufacturers (OEMs) and the fuel cell community in general.
- Collaborations are well coordinated and executed among the participants in this project.
- There are many organizations involved in this project, but the collaboration seems very good. Partners are well coordinated with the leading PI. In addition to OEMs, air filter manufacturers should be included in the project team.
- Collaboration with other groups is significant, and it is recommended that the team consider a few directed case studies based on upstream analysis and testing for a few selected collaborators and their field-based systems.
- The funded subcontractors are limited, with only UConn having a clearly defined role in the project as presented. The industrial consultants' roles were not clearly defined. The team has been in the area of contaminant testing for some time and has external connections highlighted on the collaborators slide that do provide added value.
- It seems that most of the work described in this presentation was conducted by UConn, which is the team responsible for foreign cation research. There needs to be a better description of what parts of the team did what work over the last year.

- The team perhaps still needs more input from OEMs for recommendations on specific contaminants (and concentration levels) to focus efforts on. It is suggested that the team canvass the OEMs, if it has not already done so. The team may also need more input from fuel and fueling station suppliers.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.3** for its relevance/potential impact.

- DOE technical targets would be effectively achieved by the findings and recommendations from this this project. Airborne contaminants are recognized as one of the major obstacles in fuel cell technology, considering the constant exposure of cathodes to the air. This project does not only quantify the effect of contaminants but also offers plausible solutions to overcome the suppressing effects on fuel cell performance.
- This project is in line with Fuel Cell Technologies Office (FCTO) goals as described in the FCTO Multi-Year Research, Demonstration, and Development Plan. The effect of airborne contaminants on fuel cell systems could potentially be a huge issue with the rollout of a fuel cell fleet with different contaminant effects depending on the location. It is at least as critical as fuel quality. The role of impurities in electrocatalysis is important and will be more important because the catalyst loading has to decrease to meet cost targets.
- The goal of understanding contamination effects on fuel cell performance and developing mitigation and recovery strategies is very important to the success of commercial FCEVs. The most important part of this project has been identifying which contaminants cause recoverable versus unrecoverable performance loss.
- Implementation of fuel cell power plants (automotive or stationary) will be subject to the local jurisdiction's air quality, and the effect of the air quality will be a critical piece of the final market viability for the fuel cell system.
- The project is relevant and addresses DOE Hydrogen and Fuel Cells Program needs. Impurities studied are relevant and have been suggested by fuel cell industry representatives.
- Airborne contaminants are of significant relevance to enabling low-loaded, durable cathode catalysts and membranes. Understanding mechanisms, acceptable contamination levels (for filter requirements/design), mechanisms, and potential mitigation strategies is vital for competitive fuel cell commercialization.
- This research is relevant to DOE goals because it addresses one of the main challenges facing polymer electrolyte fuel cells for automotive applications. The impact has been moderate to date; proposed mitigation techniques seem to be either impractical (e.g., ex situ and take a very long time), partially effective, or both.
- Air contaminants (particularly those investigated in this project) have not been demonstrated as limiting elements to fuel cell deployment through cost, performance, or durability. The experimental results add incrementally to the knowledge base of the community in the area of contaminants. The project is rightfully focusing on low loadings capable of meeting DOE's future targets but has not demonstrated any specific progress toward meeting any specific target.

Question 5: Proposed future work

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

- The project is approaching the end date. The proposed future work is appropriate to complete the project.
- Future work would continue to effectively address performance limitations that originate from airborne contaminants.
- The proposal to demonstrate successful mitigation of the impact of important airborne contaminants is a reasonable goal. The team has identified some experiments it is likely to try, but it does not have any backup plans if those approaches do not work. The team also needs a better and more thorough design of experiments to know what techniques will be most useful and that the techniques have been optimized to some degree.

- The amount and scope of proposed future work is adequate given the time left until the project's completion in December 2015.
- The mitigation strategies to be investigated have to be realistic regarding an embedded fuel cell system. Determining the impact of contaminant mixtures on low Pt loading cathode sounds good if the investigated mixtures are representative of severe environmental conditions. A rationale for that should be presented.
- The PI generally lays out the proper future work (lower platinum group metal loadings, contaminant concentrations, and mechanisms).
- The proposed future work is limited, as would be expected for a project nearing completion. The focus on the four "most important" air contaminants seems reasonable, but it is not clear what these are or the specific criteria used to determine them. It appears these four will be a subset of the eight presented, and these eight are not of the highest interest of different contaminants for consideration, because contamination concerns are a function of both the impact of a contaminant and its likelihood (and type of) exposure to the fuel cell. This project is reaching a logical endpoint in its study.
- A significant focus of the path forward should be mitigation strategies on the system or the MEA level. This work should be done directly in collaboration with an automotive application developer and a catalyst/MEA material developer. This focus will take the large amount of effort and data generated into this project and ensure that it culminates into a strategy to enhance the wider adoption of fuel cells in various applications.

Project strengths:

- The main strength is the approach to testing fuel cell performance with selected contaminants in a way that is complementary with already studied species. Other strengths include the following:
    - The obtained database of contaminants
    - The identification of electrochemical and chemical reaction pathways
    - The short-term publication of the results for the community
- Project strengths include the team's strong background in contaminants, history of performing related work, connections within the contaminants community, and the ability to bring in the U.S. Department of Defense to this specific area, as many of its concerns would be greater than DOE's concerns.
- Project strengths include good collaborations and a good effort to get industry input regarding which contaminants should be investigated.
- Project strengths include a well-balanced approach, a well-chosen and efficient team, and the systematic evaluation of contaminants.
- A large amount of data seems to have been collected. Tests provide a preliminary indication of which contaminants cause recoverable versus unrecoverable performance loss.
- The project is well laid out and systematic. The data collection covers a broad range of contaminants that are directly relevant to potential application locations. The data should be put out in a contamination guide or review paper as a go-to resource.
- The PI has a strong background in the subject. Ballard can supply the OEM perspective.

Project weaknesses:

- The team should work to extract kinetics and mechanisms so that the data are translatable into either system or MEA/unit cell level models. This would allow for projections, impacts, and trade-off analyses to be completed and would enable a better understanding of the cost of recovery procedures.
- The team needs access to consistent, reliable low-loaded state-of-the-art catalysts. The team needs greater input from the other OEMs regarding which contaminants are of greatest priority.
- A more thorough focus on the correlation between the oxygen reduction reaction (ORR) kinetics and contaminants is needed.
- Contaminants contained in this study have relatively low interest/relevance for primary fuel cell target applications. Fundamental understanding and insight gained from the work to date have not been as strong or complete as was desired/expected.
- Insufficient attention to the mechanism of performance loss and performance recovery is the main weakness of this project. That applies in particular to the impact of certain mitigation strategies, such as the

impact of acetonitrile, a known strong poison of Pt active sites through chemisorption, on the electrode kinetics.

- The project still needs to complete testing at low Pt loading on the cathode and, therefore, to link with other DOE projects. An investigation of the impact of contaminant mixtures is lacking, but will be integrated into the ongoing work. The mitigation strategies do not appear very realistic for implementation into a real system. First mitigation strategies may be validated in short stacks. There is no link with air filter manufacturers to relax the MEA constraints and to quantify the cast balance between robust MEAs and advanced air filters.
- Project weaknesses include a lack of deeper understanding, a lack of statistical analysis of results, and a lack of thoroughness and creativity in experimental techniques.

Recommendations for additions/deletions to project scope:

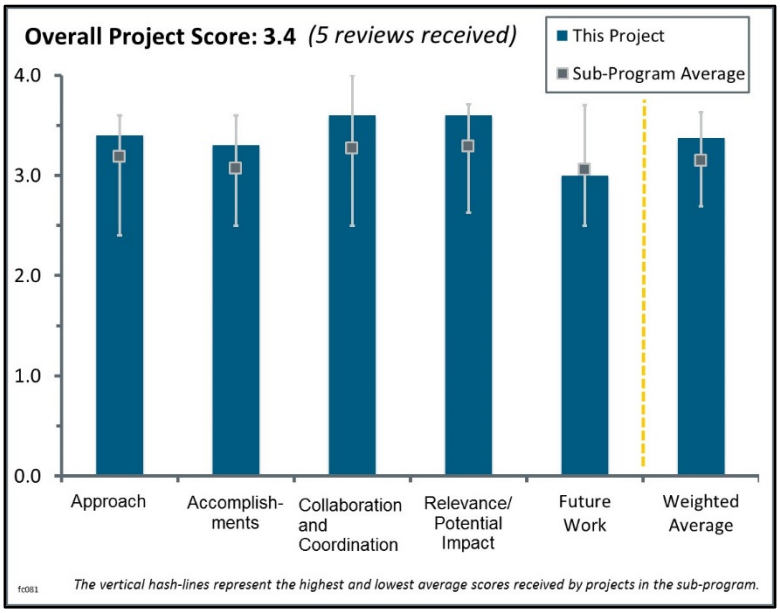
- The project is scheduled to end at the end of the calendar year; it is a logical place to end this specific effort.
- The PI could expand their evaluation of the impact that contaminants have on the ORR kinetics as well as the mechanism.
- It would be interesting to investigate if or how air contamination affects anode performance and durability. If it does, it is not clear what impact it might have on the fuel quality specification. Discussions between these two research groups on possible cross effects of air/fuel contaminants are encouraged.
- The team should add collaborations to system modeling design groups and unit cell/MEA level modeling groups. Additional focus on mechanisms and the extraction of exchange, reaction rates (kinetics), etc. would make the data and work more translatable for further work and use by other groups.
- More fundamental insight into the contamination mechanisms and, especially, into the recovery mechanism is strongly recommended. For example, there seems to be no understanding as to why acid treatment fails to result in full performance recovery after Ca^{+2} contamination. The catalyst performance loss in the presence of acetonitrile, which is entirely neglected in research to date, should be determined and weighed against the gains brought about by the reduction in the rate of polymer degradation.
- There should be less focus on cation work, which has been adequately covered in the existing literature.
- Studies into mitigation strategies need to be more thorough. Pre-test and post-mortem analysis of likely mechanisms of performance loss needs to be carried out. This should include measuring cell characteristics (such as exchange current density, Pt utilization, high frequency resistance, hydrogen crossover, and limiting current) before testing, after contamination, and after the recovery effort. In addition, these tests should be run at multiple fuel cell operating conditions because some fuel cells may operate differently under hot or cold and dry or wet conditions.

Project # FC-081: Fuel Cell Technology Status: Degradation

Jennifer Kurtz; National Renewable Energy Laboratory

Brief Summary of Project:

The fiscal year 2015 objectives of this project are to (1) receive and analyze new laboratory fuel cell durability data and (2) update and publish the durability results. The National Renewable Energy Laboratory will provide an independent assessment and status of state of the art in fuel cell durability for different applications by uniformly applying analysis methods to developers' voluntarily supplied data from laboratory testing.



Question 1: Approach to performing the work

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

- This project makes use of excellent statistical analysis to report what would otherwise be a scattered number of data reported by original equipment manufacturers (OEMs) in other non-technical forums. The project has found creative ways to deal with typical problems on how to report degradation, which is not an easy task, given the variety of definitions and design parameters currently present in the industry.
- The voluntary component is hampering the thoroughness of the analysis, though this is no fault of the principal investigators. It does, however, influence the impact of this project within the grand scheme of things. Good correlation between lab tests (time to 10% loss) and real-life durability lifetime is lacking. Long-term trend analysis may be able to project durability improvements to fuel cell systems over the years, perhaps even predicting near-future performance, which would be of benefit for investors in the fuel cell industry (including the U.S. Department of Energy [DOE]). On an individual level, the partners who voluntarily supply their data are rewarded with a good analysis of where they fit in the current competitive field, thereby stimulating industry participation.
- Though the generation of results has been dependent on voluntary OEM participation, there have been sufficient data generated for analysis. The only means of improving the approach appears to be if vehicle or test rig data were available for the DOE reference stack/system.
- This analysis is not so useful unless the performance and durability statistics are shown with technical information such as fuel cell system design/materials/system architectures. Getting meaningful technical information from fuel cell developers on a voluntary basis seems to be a limitation.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- In general, the project does not solve any particular material or design problem, but it does provide guidance as to whether the industry is on track and how far the industry is from the overall targets. This is key to DOE's understanding of where to direct money for specific projects that address degradation problems.
- The project supports progress toward project and general DOE goals—it speaks to status vs. targets and to general technology progression over the past few years.
- With the resources available, the project is effective.

- No significant updates have been seen since the last review (2013).

Question 3: Collaboration and coordination with other institutions

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

- There is very good collaboration with other institutions, as is evident in the data that are available only from external sources. The project is entirely dependent on external collaborations and data from other institutions.
- Asking OEMs for this type of data is no easy task, and the team was successful in convincing them to provide enough information to have a representative body of data.
- Within the limitation of access to technical information, this project seems to have good relationships with the fuel cell system developers providing information.
- The voluntary participation model may hamper the thoroughness of the evaluation and may even skew the results slightly if either the over- or under-performing systems are omitted.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.6** for its relevance/potential impact.

- Long-term trend analysis may be able to project durability improvements to fuel cell systems over the years, perhaps even predicting near-future performance, which would be of benefit for investors in the fuel cell industry (including DOE). On an individual level, the (industrial?) partners who voluntarily supply their data are rewarded with a good analysis of where they fit in the current competitive field, thereby stimulating industry participation.
- Technical benchmarking is very important for commercialization of automotive fuel cells.
- Status against DOE targets is tracked with in-service data. Project assumptions for high volume are unrealistic based on the current state of the market and market growth.

Question 5: Proposed future work

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

- Electrolyzer data will be very useful now that the focus in the fuel cell field has shifted to include the supply of hydrogen.
- Regarding the proposed future work listed by the principal investigator, it is not clear that the plan to “alternate between durability and cost/price” is necessary unless (1) the work load to get both types of data is too much for the current funding or (2) there is not enough information. Yearly updates are interesting in both areas. In addition, publishing a report on the cost analysis method is recommended.
- As the project relies on external data, future work is dependent on further developing collaborations, which may limit the scope of future work.

Project strengths:

- There is a neutral approach to collecting and publishing data that would otherwise be confidential and not accessible. The project also has a great statistical methodology.
- Valuable data analysis gives investors (industry partners and DOE) a good picture of the status in fuel cell durability. It seems this would be very helpful to the partners in evaluating their own positions within this field.
- The project provides real-world data, collecting information about the progress of the technology and providing overall progress guidance for DOE projects.
- The project analyzes the state of the fuel cell industry against the DOE targets for fuel cell technology from data provided by external collaborations.

- The relationship with fuel cell developers to receive technical information is good.

Project weaknesses:

- The data are grouped by application but not by technology. Information is not provided about the progress of each technology to meet the application needs. This is very important for DOE and commercial clients in technology selection.
- There is limited ability to receive technically meaningful information for performance and durability analysis. Any analysis of this kind without technical information about design/materials/system architecture is not useful.
- The fact that the data sharing is voluntary, combined with the fear of misplacement of confidential data, may lead to omission of valuable segments of the industry, which in turn may lead to inaccurate trends.
- Providing the data is voluntary, which limits the quantification of the results.

Recommendations for additions/deletions to project scope:

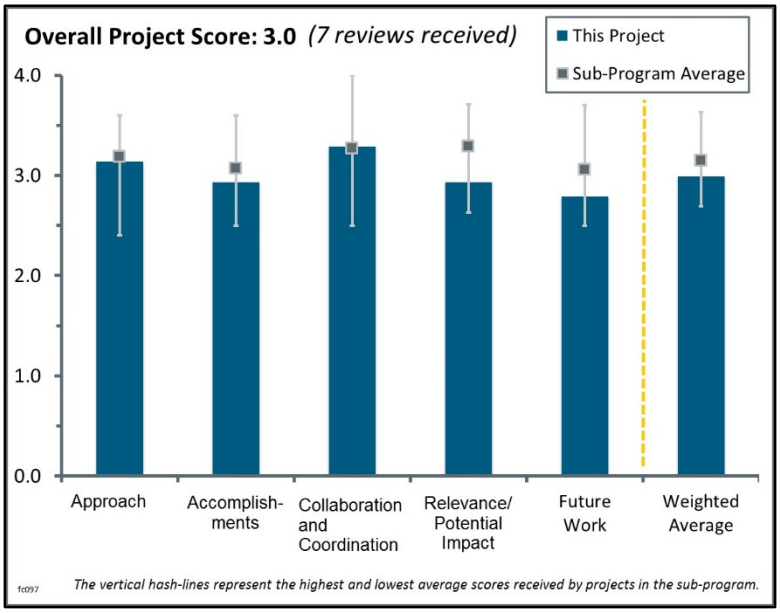
- The need for benchmarking is high. However, any analysis of performance and durability without technical information about design/materials/system architecture is not useful. Data acquisition on a voluntary basis shows critical limitations. It is suggested that this project change to a benchmarking project. Recently, two fuel cell vehicles were released to the market, and another fuel cell vehicle is supposed to be released shortly. Actual benchmarking on these vehicles would be able to show performance and durability with detailed technical information. Collaboration with U.S. DRIVE Partnership Technical Team is expected.
- Incentives (maybe even financial incentives) must be created so that OEMs will publish more data.
- Adding electrolyzers and reformate fuel cells to the analysis would be helpful.
- The project should break down the data into technologies for each application.

Project # FC-097: Stationary and Emerging Market Fuel Cell System Cost Analysis—Primary Power and Combined Heat and Power Applications

Vincent Contini; Battelle

Brief Summary of Project:

The overall objective of this project is to assist the U.S. Department of Energy (DOE) in developing fuel cell systems for stationary and emerging markets by developing independent model and cost estimates. The project goals are to (1) identify major contributors to fuel cell system cost; (2) quantify potential cost reduction based on technological improvements; (3) identify major contributors to fuel cell system manufacturing cost; (4) identify areas for manufacturing research and development (R&D) to improve quality and/or throughput; and (5) provide a basis for consideration of transition from other industries.



Question 1: Approach to performing the work

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

- The researchers did a good job of addressing the makeup of many of the costs and attempted to project into the future based on the design for manufacture and assembly (DFMA) models. It is difficult to assess the overall accuracy of the DFMA model for out-year projections, and it would have been useful to see a discussion on how these were determined. Perhaps there is a comparable industry that has validated the cost reductions of volume manufacturing. It would have been good to see the cost of the catalyst backed out of the overall polymer electrolyte membrane (PEM) membrane electrode assembly (MEA) cost and an estimate of the value of platinum recovery added as a credit to see its impact on MEA cost. It is believed that more than 95% of platinum can be recovered. On a different topic, an essential element of customer acceptance is the cost after sales support and how quickly a unit can be repaired. An estimate is needed for mean time between failures and mean time to repair, as well as what the repair mode is (e.g., repair by replacement). Also needed is an estimate of how much the manufacturer cost for field tech support will be. This can be a huge cost. For example, a field call to commission an electrolyzer can cost more than $20,000. The project needs to include the hourly rate, cost of air travel, and per diem. It adds up fast. The key to overcoming this is to have a robust product that has been extensively tested before it is shipped. The project needs to allocate a significant warranty budget, especially for products that are early in their product cycles, as well as a budget for spare parts. The trend today, it seems, is to lay the cost of spare parts on the customer rather than the vendor. Manufacturers do not want a large spare parts inventory on their shelves. The penalty for the customer is long lead times for spare parts—the wait can be up to six weeks—which means the customer's system is out of action.
- Project barriers are well identified; project goals based off those barriers are feasible and use information and data from other participants.
- The conservative approach to production volumes is more realistic. The analysis includes operating costs in addition to manufacturing costs, and both grid-connected and grid-independent operation. It is not clear why the researchers designed their own system rather than operating and measuring the performance of existing commercial systems of various technologies and capacities.
- The project has a thorough and systematic organization of the manufacturing cost analysis from early evaluation of markets to development of system design, cost modeling, and analysis. A limitation could be

system design because Battelle has limited experience in the design of fuel cell systems and is dependent on original equipment manufacturers.

- The following is a list of items that stand out:
    - Batteries are used only in cases of black-start capability and off-grid operation. Those are rather exotic applications, especially for urban markets. The majority of urban markets are rather unlikely to have such systems and would not justify the expense.
    - The preferential oxidation (PROX) blower is unnecessary. Slipstream from the cathode can be taken to supply a PROX catalyst.
    - Combined heat and power (CHP) efficiency for low-temperature (LT)-PEM is unrealistic for U.S. market hydronic systems. U.S. systems operate with high-temperature loops, which do not lend themselves to high recovery of low-quality heat.
    - CHP heat utilization looks optimistic. Considering seasonal requirements as well as daily fluctuations in demand, numbers can be as low as ~12% of actual utilization. It is often economically unjustifiable to install heat recovery for LT-PEM or solid oxide electrolyzer cell systems. Hence, Bloom Energy does not even make an attempt to recover waste heat, even though the company sells systems outside California as well.
    - 0.4 A/cm^2 for PEM is very low, even for stationary systems. For longevity, such systems are operated at higher current densities to allow lower voltage of operation and reduce carbon corrosion.
    - Both PEM systems and solid oxide fuel cell (SOFC) systems are highly sensitive to complete shutdowns for both thermal and atmospheric cycling. A shutdown reformer unit "inhales" exhaust oxygen as gasses in it cool down and contract. In this process, the atmosphere changes from a reducing environment to an oxidizing environment, causing a myriad of material issues. Thermal cycling itself also causes differential thermal expansion of oxidation layers versus metal layers and thus flaking of metal components and catalysts. Catalysts themselves also go through an oxidation/reduction cycle during each shutdown, which is really not desirable.
    - It was unclear whether the project used any thermodynamic process modeling. If not, there is plenty to tap into, and these models are absolutely necessary for component sizing. It is also very important to model interaction with hydronic systems and estimate the realistic heat recovery from different systems. If the project has not done this, it should not be done at this point. Doing it right is a very involved process, and the information can be seen from previous studies. The information should, however, be examined to determine realistic heat recoveries. The project should not just "take someone's word for it."
- The approach includes market assessment, system design, cost modeling, and sensitivity and life cycle cost analysis. Incumbent and potential processes are analyzed for applicability.
- The project aligns with DOE's objectives for current cost status, per the project's assumptions.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- Project accomplishments so far have been informative and provided useful data. It would be good to include additional LT-PEM stack developer parameters. The progress to date and the process developed is sound, but loading other parameters for various technologies could provide an additional layer of detail that many would find useful.
- Various PEM and SOFC systems and production levels have been analyzed. Dominant cost items have been identified, and an attractive value proposition has been established under certain (utility rate) conditions.
- The team members have made good progress on what they have selected to evaluate, but they should consider other areas to ensure they are looking at a complete picture of what it takes to make a product viable. They have a major hole in the customer acceptance objective. Customers will not accept products that are not robust and that are difficult and expensive to repair. While the team has focused on a technical systems design to identify major technical components, they should consider conducting a total product

systems requirements analysis that looks at all aspects of what it takes to launch a robust product into the market.

- Overall efficiency of 80% for LT-PEM seems excessive. The low-quality heat from the PEM system makes thermal recovery limited. Even as a preheater for hot water, this would require a close proximity to the water heater. It is unclear whether Battelle did the analysis for heat transfer or whether it accepted inputs from LT-PEM producers. The use of silicone seals is questionable for most PEM systems. It is unclear how dependent the PEM life cycle cost analysis is on the CHP or what the life cycle cost would be for the PEM system if CHP were eliminated. It is also unclear why annual operations and maintenance costs are the same for PEM and SOFC systems. Because there are fewer parts in the SOFC, there should be fewer maintenance requirements.
- Analysis results show that SOFC CHP is substantially cheaper to manufacture than PEM CHP—much of industry has already arrived at this conclusion. Reviewer comments from 2014 were appropriately incorporated. Analyzing large-scale PEM may not drive toward meeting DOE efficiency and performance targets.
- It is unclear how this analysis is leading toward cost reduction and customer acceptance. The results are valuable, but the connection was not made toward addressing those barriers.

Question 3: Collaboration and coordination with other institutions

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

- Using real data to conduct analyses shows that there is excellent collaboration. The opportunity to run the analysis multiple times with various parameters adjusted could be very useful.
- The project shows strong collaboration across research and industry. The inclusion of fuel cell system manufacturers with more efficient technology may have changed the direction of this program.
- The team includes fuel cell technology (cells and balance of plant [BOP]) developers, manufacturers, and integrators, as well as systems analysis organizations.
- Battelle has established high-quality collaborators.
- The collaborations are representative of the systems analyzed.
- The project has some excellent collaboration partners but can get more out of them. Panasonic would provide excellent input on what it takes to launch a product into the marketplace, which would help to address the customer acceptance issues. Ballard should also be able to provide input from the point of view of supporting stacks and BOP. Johnson-Matthey Fuel Cells Inc. can provide input on platinum recovery economics.
- The collaboration companies list could be improved. People in the United States are not making small CHP systems, but there are plenty of “consulting” folks who were in the trenches during the early 2000s building such systems. They should be consulted for lessons learned. Until a couple of years ago, for example, Plug Power was making LT-PEM CHP and high-temperature (HT) PEM CHP systems. Plug Power is still around and has in-house staff who should be asked to review and provide input on such models.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **2.9** for its relevance/potential impact.

- The results of the analysis provide valuable input to DOE funding decisions (focus and level) based on Technology Readiness Levels and low-volume cost estimates.
- The project is doing great at identifying cost drivers and break-even times for “average” system architectures.
- The project identifies where cost issues exist and can be used for decisions on funding support.
- It would be good to see a greater effort on specific areas upon which the DOE Fuel Cell Technologies Office can focus its future R&D. To say that electronics and power conversion dominate system cost is too general. The team should investigate which components, specifically, need to be addressed. After spending three years examining these systems, the project should have a good feel for what needs to be fixed. For

example, there is broad consensus that inverters and converters need to be much more robust and cheaper. The project should engage with US Hybrid, a collaborator and an expert in this field, for input. Panasonic and Ballard can be much more specific as to where the effort should be applied.
- The results of this project show a substantial cost differential for SOFC and HT PEM and have the potential to inform DOE about direction of future stationary CHP Fuel Cell Technologies Office (FCTO) funding. Under the current DOE structure, without a focus on stationary systems, and with SOFC in the Office of Fossil Energy, it is unclear where this information will be used.
- The results are valuable for status reporting. How the analysis contributes to cost reduction and customer acceptance is not clear.
- This is an excellent effort and excellent work, but it has been done many times. The benefit is unclear. While Japan shows progress in CHP/residential systems, that work is largely backed up with policy/ subsidies.

Question 5: Proposed future work

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

- The future work seems aligned with the project cycles so far. It would be good to see how it fits with the legacy budget period work and other similar DOE-funded projects.
- While there is benefit in all of the cases being run through the process, running specific cases with real performance data would be more useful.
- The proposed future work slide was too high-level and not specific enough. Some more thought could have been put into future work instead of “more of the same but bigger.” The future work plan needs to be developed to address issues and recommendations identified by reviewers, in addition to evaluating the larger systems.
- A 2014 reviewer comment objected to HT-PEM, and Battelle agreed. HT-PEM “did not make the cut for this year’s CHP and Primary Power analysis.” However, HT-PEM seems to show up on the proposed future work slide, so the status is unclear.
- The project has identified a pathway to complete the cost analysis for several applications. The project should explain why there has not been a greater acceptance of fuel cell systems, given the two-year payback identified.
- The plan for large-scale primary power PEM evaluation seems disconnected from the most efficient and cost-effective stationary systems (e.g., SOFC, molten-carbonate fuel cell, and phosphoric acid fuel cell). This begs the question of whether this evaluation may be taking place simply because Ballard is a collaborator, rather than because the technology with the greatest potential to meet DOE targets was selected.

Project strengths:

- The following are project strengths: good technical systems analysis, good identification of current cost elements, good use of modeling programs, and good industry participation at the technical level.
- The process and details put into the analysis are comprehensive. The amount of data and the cost breakdowns are also useful.
- The project has a good team and a good approach to the cost analysis.
- This project shows the benefits of SOFCs and demonstrates the value of this technology to the FCTO.
- The details in the results are a strength.

Project weaknesses:

- The project does not adequately identify specific areas for future R&D investment, nor does it address real-world customer acceptance issues and associated costs, including after-sales product support.
- It is not clear how other industry organizations will use the information resulting from this project because they are already manufacturing at low volumes and have hundreds/thousands of systems in the field.
- Feeding more relevant information into the process could provide more useful data.

- The project does not clearly show how the analysis results are addressing the barriers.
- System design and the use of low-quality heat water for CHP should be revisited.

Recommendations for additions/deletions to project scope:

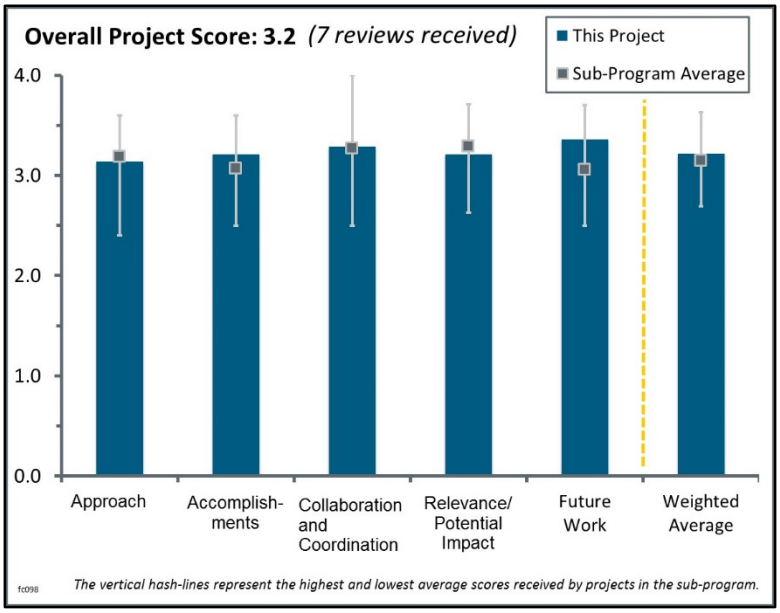
- It would be good to see this work tied into the National Renewable Energy Laboratory's (NREL's) Energy Systems Integration Facility—systems were evaluated in the field, but there may be synergy with the NREL center. Measurement of existing systems and reporting on their success could be more useful to drive deployment than modeling systems (similar to the data collection program).
- Perhaps using the operational data received from the fast food restaurant with real fuel cell performance data could show how technical advances accelerate the return on investment.
- The project should develop a total system requirements model that includes after-sales product support.
- Takeaway messages on the slides would be helpful, as well as what impact this analysis has had.
- The project should revisit low-quality heat for PEM CHP.

Project # FC-098: A Total Cost of Ownership Model for Design and Manufacturing Optimization of Fuel Cells in Stationary and Emerging Market Applications

Max Wei; Lawrence Berkeley National Laboratory

Brief Summary of Project:

The overall objective of this project is to develop a total cost of ownership (TCO) modeling tool for design and manufacturing of fuel cells in stationary and materials handling systems in emerging markets. Lawrence Berkeley National Laboratory (LBNL) will expand the modeling framework to include life cycle analysis and possible ancillary financial benefits, including carbon credits, health/environmental externalities, end-of-life recycling, and reduced costs for building operation.



Question 1: Approach to performing the work

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

- The work is good and does a reasonable job of walking along the footprints of previous analysis methodology.
    - It is not clear whether there is thermal modeling behind the figure on slide 15. It seems that there is not; in addition, there has not been research on published piping and instrumentation diagrams.
    - It is not clear why there is not a fuel blower—in residential systems, the fuel supply is only ~4–30 inches of water pressure.
    - It is not clear how the anode exhaust is entering the fuel pre-treatment. The pressure in the pre-treatment unit would be higher than the pressure of the anode exhaust because of the dynamic pressure drop of the process. A three-way valve is not the right unit operation there.
    - Burner exhaust would have lower mass flow than fresh air (because oxygen is removed in the cathode). In the "react. air heat exchanger," lower temperature differential on the cold stream than on the hot stream is to be expected, yet the cold stream changes by 550 K while the hot stream changes by 440 K. There is a bit of heat capacity differential due to the temperature of the stream, but it looks wrong.
    - In greenhouse gas (GHG) emissions off-set, the project team should consider not grid-mix but removing the generation equipment with most "pressure" against it—coal. If grid equipment is being displaced based on GHG reduction pressure, combined cycle plants would not be shut down—at least, not at first.
- The work is focused on the cost barriers and benefits of fuel cell combined heat and power (CHP) technology. This type of study can be very helpful in guiding research to address the main cost drivers and identify the appropriate markets and locations so initial introductions of the technology are successful and show the technology's benefits. The addition of life cycle impact for environmental and health externalities to the cost model is important because these benefits are often ignored. It is not clear how locations for the TCO model were chosen. The cities mentioned are all large cities and appear to fall mostly in the two extremes for expected heating requirements. Some indication of how the locations are selected would be beneficial.
- The "Total Cost of Ownership Model" as presented on slide 6 is a good overall approach and is supported in more detail on slides 7, 8, and 9. However, there should be equal detail for the "Other Costs" elements. These could be presented in a separate slide or added to slide 9. As it stands now, the "Other Costs"

element appears to have been forgotten or not attributed the priority it deserves. Customer acceptance is critical to a successful product cycle, and expensive maintenance and after-sales support is not going to foster that acceptance.

- The analysis outputs are directly supporting the U.S. Department of Energy's (DOE's) goal for understanding fuel cell costs for targeted and impactful direction on lower-cost research.
- It is not clear how the focus on 10 kW and 50 kW was established. It would have been useful to start with a market analysis to understand what size systems have the greatest addressable market. The expanded life cycle assessment is valuable and commercially relevant, although there are existing benchmarks and standards that could have been used, such as those from the California Air Resources Board, Itron reports, etc. The TCO values electricity but not natural gas, so there is divergence between a predicted market with high electricity costs and the actual market evolving, in which there are high electricity costs and low natural gas costs.
- LBNL costs should be compared to Battelle and Strategic Analysis, Inc. (SA) estimates as they become available. It would be good to include environmental costs, but that seldom plays into a buy decision except in very localized areas. Therefore, it would be useful to isolate the realized (out-of-pocket) cost from the total cost including environmental externalities.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

This project was rated **3.2** for its accomplishments and progress.

- This is a good analysis that is on track to meeting project goals. The work confirms other cost evaluations that have found that the balance of plant (BOP) is key to cost reduction, and power electronics are a major component of BOP.
- The solid oxide fuel cell (SOFC) CHP direct cost model was completed, along with the TCO model for the high-temperature (HT) polymer electrolyte membrane (PEM) CHP system. The addition of regions in the TCO model for the HT-PEM CHP case is valuable.
- This project demonstrates that the SOFC system is close to meeting DOE goals. The inclusion of an incentive review can help DOE look at target costs that are reasonable for the markets.
- The results for TCO will be highly dependent on the system performance. The principal investigator (PI) has validated fuel cell performance against current systems. The PI's comparison of real learning curves from the Japanese micro-CHP fuel cell experience to the model cost reductions with increasing scales is commendable. Some sensitivity analysis to performance parameters may be beneficial (similar to the SA sensitivity study on power density). The PI's assertion that Chicago and Minneapolis are "regions with higher-carbon-intensity electricity" was surprising. Illinois (including the Chicago region) has a substantial amount of nuclear energy production and, according to the U.S. Energy Information Administration, was in the top third for lowest CO_2 intensity of energy supply (kilograms of energy-related CO_2/million Btu). Illinois and Minnesota were both below the national average for CO_2 intensity of their energy supplies. (In 2011, the average for the United States was 55.3 kg/MBtu; Illinois was 50.9 kg/MBtu and Minnesota was 52.9 kg/MBtu. Illinois was only slightly above California, at 49.5 kg CO_2/MBtu.) Kentucky, West Virginia, Wyoming, Indiana, and Utah were the highest (81, 76, 75, 72, and 72 kg CO_2/MBtu, respectively). The data can be found at www.eia.gov/environment/emissions/state/analysis/. The PI should check and provide a reference for his CO_2 intensity and emissions data. Case studies from a high-CO_2-intensity region (e.g., Kentucky, West Virginia, Wyoming, Indiana, or Utah), a state with "average" CO_2 intensity, and a state with low CO_2 intensity would be beneficial (versus two case studies from regions with similar CO_2 intensity). If Chicago and Minneapolis are below average in terms of CO_2 intensity (as the 2000–2011 data show), this means fuel cell CHP will be beneficial for much more of the nation than if they are high-CO_2-intensity regions. The conclusion that fuel cell CHP is most favorable in regions with higher carbon intensity electricity seems counterintuitive. It seems that the cost of electricity, or more accurately, spark spread, would be more important than carbon intensity in determining whether fuel cell CHP is favorable, and areas of high-carbon intensity (with cheap local coal) would be less favorable. Carbon intensity appears, financially, to have a minor impact because savings from GHG credits appear to be much lower than electricity costs on a dollar/kilowatt-hour basis.

- Most of the work to date seems to have been focused on the "Manufacturing Cost Model." This has been detailed and thorough. The "Lifecycle Cost Model" to date seems to have been focused on fuel and electricity costs. What is missing is "Other Costs," such as product maintenance and warranty costs. It appears the project plans to address these elements in its last year. This seems a bit late because the project was started in 2011, and it would be useful to get some advance insight into what these costs might be. For example, on slide 12, it is interesting to note that the reformer and compressor/blower's projected lives are only 10 years. This means there must be a replacement. The project should determine how big an effort is required to change out these components and how much this adds to the life cycle cost. Perhaps this indicates the opportunity to conduct cost trade-offs for designing these components to last for the system life of the fuel cell stacks. The components have the potential to pose a big "hassle factor" and cost for the customer.

Question 3: Collaboration and coordination with other institutions

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

- Additional partners have been added, creating a strong and diverse set of partners.
- There is a broad range of collaborators, including industry representatives.
- The collaboration with SA is good. Feedback from industry for the SOFC (from VersaPower and SOFC Power) and PEM fuel cell models is taking place (Ballard, Altergy, and Panasonic were mentioned last year). Some interactions with the technology validation projects at the National Renewable Energy Laboratory and Pacific Northwest National Laboratory may be beneficial for providing additional input/validation for fuel cell performance assumptions. Some basic assumptions, such as Pt price, should be coordinated between all the cost modeling projects.
- The project has some excellent collaboration partners but can get more out of them. To address the customer acceptance issues that have been raised, the Japanese fuel cell micro-CHP and Italian programs could provide excellent input on what it takes to launch a product into the marketplace. To address some of the BOP issues, the project should engage US Hybrid.
- Additional partners who are working on related areas could be included and parameters shared. Much of this analysis has already been completed by stationary fuel cell companies and third parties. The California Public Utilities Commission recently released an Itron report on the 2013 impact of the Self-Generation Incentive Program. It measures environmental impacts and GHG reductions of fuel cell systems, as well as system performance and capacity factor. These criteria and calculations could be benchmarked for this analysis. Most of the new partners and collaborators are academic, not from industry.
- There should be more collaboration with companies that actually build systems in-house.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.2** for its relevance/potential impact.

- The project is relevant and aligns with DOE Fuel Cell Technologies Office goals and objectives for micro-CHP and medium-scale CHP fuel cell systems. The project helps determine where the costs are in current systems and where the opportunities for cost reductions are, as well as helps identify how these systems would compete in the market and where deployments may make economic sense now. These studies help initial technology validation and market transformation work target the appropriate applications and locations.
- The project advances the DOE goals by valuing the environmental, societal, and financial benefits of fuel cells. The connection to policy is important—it can inform policy initiatives and the direction of DOE support.
- This project confirms findings of other cost-estimating groups and adds the impact of environmental factors. However, environmental factors are not typically part of the buy decision, so a TCO without those factors needs to be included as part of the results. Environmental costs are also controversial, so a bit more attention to that aspect would be appreciated.

- These are values that are used for the status and breakdown of costs, with the specified assumptions on volume.
- Greater effort is recommended on identifying very specific areas upon which the Fuel Cell Technologies Office can focus its future research and development (R&D). On slide 6, a key output is the identification of specific areas in which R&D investment needs to be applied, for example, system components such as compressors and reformers that match the system life of the fuel cells. On slide 26, "Answers to 2014 Reviewer Comments" under BOP opportunities, the project needs to be much more specific, and it is worth a slide on its own to identify the specifics. The project should consult with US Hybrid on this aspect and get its input.
- This exercise can be done by building more on old evaluations and examining technology changes instead of fully reworking the analysis. Efforts may be better spent in tackling technological problems encountered by past "waves" of fuel cell companies. It is well understood already what the roadblocks have been and what performance targets need to be to achieve market penetration.

Question 5: Proposed future work

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

- The planned scenario/sensitivity analysis including energy prices and incentives will be very useful. Future work should include a plan for dissemination of results.
- The proposed future work is appropriate. Investigating systems including absorption chillers is a good idea because much of the United States has low space-heating demands and high cooling demands. Case studies of key cost reduction opportunities in BOP are appropriate because BOP costs are high.
- The future work is consistent with the project cycles and continuation.
- The project has a good plan but needs to enhance the end-user product acceptance barriers. If the issues and recommendations raised in the previous evaluation areas are accepted as having merit, then the future work plan needs to be developed to address these issues.
- The project should leverage existing models, such as the FC Power model for TCO/life cycle engineering modeling. Much work went into getting such models through peer review with DOE, and they should be used for consistency and efficiency of analysis.

Project strengths:

- Project strengths are the manufacturing cost model, CHP system designs and functional specifications, and the broad group of collaborators and reviewers.
- It is important to review the TCO of fuel cell systems to account for their additional environmental and societal benefits.
- The inclusion of environmental and health savings is a strength and has not been done in most other similar efforts.
- The project involves a thorough cost analysis with significant relevance to deployment of CHP systems.

Project weaknesses:

- The project should be cautious about trying to do too much on design for manufacture and assembly (DFMA). Others (e.g., Battelle and SA) are doing that in significant detail. Therefore, LBNL can back off of that aspect by taking advantage of others' work. That will allow LBNL to focus more on its specific strengths: life cycle cost modeling and valuing environmental externalities. Additional discussion of how the environmental externalities are valued would be helpful. This is a controversial area, and some discussion of the "error bars" associated with how environmental costs are counted would be useful. There was some mention of different values for environmental costs but very little backup or explanation—this could be stronger.
- Recognition of the need to address "Other Costs" needs to be translated into specific action; more emphasis is needed on identifying specific areas for additional R&D. Expertise on what it takes to launch and sustain a product into the marketplace needs to be included.

- Rather than following a no-go decision and a redirection of funds, the project continues to analyze HT-PEM because the work was already planned.

Recommendations for additions/deletions to project scope:

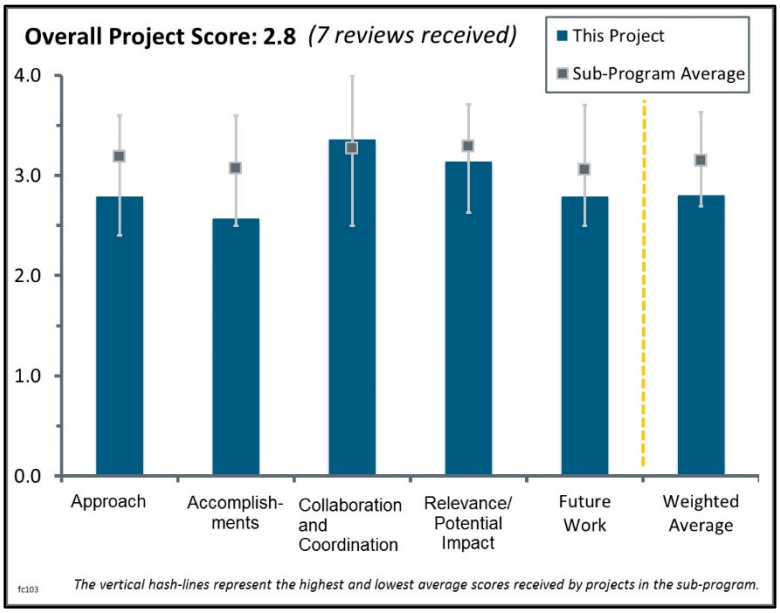
- Overall, this is well-executed and very interesting work.
- It would be good to see the material condensed with takeaways on the slides for simpler interpretation. Every detail of the work is not needed to demonstrate the work and progress. It is not clear how this information can be used for targeted R&D or submitted to fuel cell manufacturers for product improvements and cost reduction.
- The project should address the areas raised in the evaluation, including business issues. US Hybrid should be engaged to assist on BOP issues.
- Because the project is nearing completion, it would be difficult to make significant changes at this point.

Project # FC-103: Roots Air Management System with Integrated Expander

Dale Stretch; Eaton Corporation

Brief Summary of Project:

The primary objective of this project is to demonstrate key improvements to compressor/expander efficiency, including (1) compressor/expander efficiency at 25% flow of >65/70% by 2017, (2) combined motor/controller efficiency at 100% flow of >90% by 2017, and (3) compressor/expander input power at 100% flow of <8/14 kW by 2017. Secondary objectives include conducting a cost reduction analysis and developing fully tested and validated air management system hardware capable of meeting 2017 project targets.



Question 1: Approach to performing the work

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

- The approach is good. It will leverage the broad efficiency map of Eaton's Twin Vortices Series (TVS™) compressor to improve the overall drive-cycle fuel economy and integrate the expander, compressor, and motor to reduce system cost and increase system efficiency (this is a new approach, similar to a traditional turbocharger).
- Eaton has leveraged its automotive supplier production expertise in this project—this is a project strength. While the team members have cited most of the key issues, it is unclear whether they have addressed—or plan to address—them all, including potential fuel cell contamination and the impact of high relative humidity (RH) and liquid water on expander durability.
- The approach is appropriate for the project objective and provides a clear path to solve a real-world problem. More information on the testing for validation would be helpful.
- The approach follows a reasonable product development cycle: modeling/computational fluid dynamics (CFD) to subsystem development/validation to full 80 kW module validation. Although progress has clearly been made, the compressor/expander efficiencies are far from the targets, as is the cost.
- The project employs straightforward design analysis and engineering. This should have revealed the unlikely single-shaft design. The project seems to focus on beating the incumbent centrifugal compressor instead of meeting U.S. Department of Energy (DOE) targets.
- The approach of using a plastic expander has not been successful. It is not clear which barriers can be overcome by continuing to focus on this approach. The project approach does not include the motor/controller, which has the largest potential impact on cost reduction of the air handling system.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The project has made significant progress toward completing its deliverables and milestones.
- The project team moved the bar forward on a number of the metrics compared to the 2011 "baseline" (e.g., compressor-only power, cost, and idle to mid-range power performance). However, the project also fell behind on a few elements, including noise and full power compressor plus expander power, which has an

impact on fuel cell system cost to recover the deficit. The ability to achieve higher compression ratios, especially with the expander, has not yet been demonstrated. System durability, including exposure of the expander to liquid water, and the potential impact of contaminants has also not been demonstrated.
- There has been steady improvement in mid-range power consumption for the compressor/expander, but there is a performance deficit at full power. All performance metrics for the compressor/expander appear to be far from the 2017 targets.
- Several meaningful improvements over incumbent technology were realized, but not all DOE targets were met. The Eaton cost at 500,000 units is higher than that of the incumbent technology (slide 14).
- While moderate progress has been made over the past year, there are still huge gaps to be addressed to meet the critical cost and efficiency targets.
- Only 5 of the 21 targets have been met since 2011.

Question 3: Collaboration and coordination with other institutions

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

- External partnerships with Ballard and Strategic Analysis, Inc. (SA) are useful and will generate meaningful data and relevant cost information, respectively. Other partnerships appear to be providing relevant simulation data that have been incorporated into project results and progress toward achieving targets.
- Collaboration on this project is excellent and provides an appropriate mix of academia, national laboratories, and industry.
- The collaborations with Argonne National Laboratory and SA on system and cost modeling have been productive. Ballard will be doing some validation testing, but it is unclear whether these tests will include the expander. (The slides stating that Ballard testing will include the expander contradict answers during the session discussion.)
- The following summarizes the collaborations:
    - Ballard is an industry subcontractor within the DOE Hydrogen and Fuel Cells Program (the Program). Ballard's responsibilities for this project are to (1) provide fuel cell original equipment manufacturer (OEM) input into the design and specifications of the air management system, and (2) integrate, test, and validate the Eaton compressor/expander with a 75 kW Ballard HD6 stack.
    - Kettering University is a university subcontractor within the Program. Kettering's responsibilities are to provide critical analytical support, including expander CFD analysis, critical speed analysis of compressor/expander design, and critical speed analysis iterations of Eaton's compressor only.
    - Electricore, Inc. is an industry subcontractor within the Program. Electricore is providing administrative project management.
    - Argonne National Laboratory (ANL) is a federal laboratory subcontractor outside the Program. ANL is responsible for providing critical simulation and modeling support of the fuel cell system to assist in optimizing the roots air system with the Ballard HD6 module.
    - SA is an industry subcontractor outside the Program. The company is developing fuel cell system cost, utilizing the manufacturing cost of a roots-based air management system.
- A fuel cell integrator was properly incorporated at all stages, from initial component specifications to the final test. Use of CFD was demonstrated in the early stages of compressor design. From the performance, it appears the project supplier is more than competent, but this cannot be fully evaluated without knowing the source or the design's potential for large-scale manufacturability.
- Collaborators are working on cost analysis, CFD analysis, system analysis, and system testing.
- The partnerships appear to be strong, but the project should include a motor/controller partner because these components seem to be the primary barriers to meeting the cost target.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.1** for its relevance/potential impact.

- The project addresses a clear need related to fuel cell systems and, if successful, can improve the overall balance of plant (BOP). The BOP contributes to a significant portion of the overall system cost and represents a significant problem in system reliability.
- A fully tested and validated (Technology Readiness Level [TRL] 6) air management system hardware capable of meeting 2017 project targets by project conclusion supports Program goals.
- Development of low-cost air handling equipment is one of the biggest enablers of meeting automotive fuel cell cost targets.
- Compressor cost is a key contributor to overall fuel cell system cost and thus is of great relevance to the Program's goals.
- More focus is needed on the motor/controller to achieve the efficiency and cost targets; it is stated that these components account for 71% of cost. Although the work on low-cost plastic components is important, last year's presentation stated that the total cost impact of plastic versus aluminum is small. Therefore, it is not clear that the seemingly significant effort applied to developing plastic rotors is justified.
- BOP component development is not one of DOE's top priorities.

Question 5: Proposed future work

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

- The proposed future work is good, but better-defined validation testing could make this excellent. More definition on addressing the noise of the current system would also be useful.
- Performance and validation testing with Ballard is critical to determining both the technology's viability and whether DOE should invest further to get efficiency and cost closer to fuel cell system targets.
- Integration and testing with fuel cell systems is the key next step, as proposed. However, it appeared from the question-and-answer session that the expander was not incorporated in the unit to be tested with the Ballard fuel cell system. If so, this is not acceptable. The complete compressor/expander needs to be tested in-system. (The presenter's answer may have been misinterpreted.) The following were both mentioned in the Technology Advancement slide but were not listed as future work:
    - Should demonstrate 3.0 PR compressor.
    - Should demonstrate an expander (PR ~2.3–2.6) consistent with the above 3.0 PR compressor and material compatible with its environment (deionized water). It is not clear whether a plastic part can do this.
- The future work should include an emphasis on noise reduction, which should align well with focusing on improving efficiency. Future test validation at Ballard is planned for an 80 kW system, which is good. Unfortunately, the unit being tested at Ballard apparently does not include the expander, so it is unclear what the validation plan is for the full "roots air management system with integrated expander."
- Performance and durability testing on Ballard stacks is planned.
- The critical speed may be too low with overhung expander rotors. Integration with polymer electrolyte membrane fuel cells may not go as well as hoped. Direct coupling of the expander to the compressor may result in fuel cell stack pressure and flow rate control issues. The system might not operate at optimal conditions.
- The project will complete testing to objectives; however, it does not appear that project targets will be met.

Project strengths:

- This is the only DOE project on air handling equipment. There are strong collaborations with modeling and cost projection teams. There is a plan for validation at TRL 7, so the unit should be readily commercialized if it meets requirements.
- Eaton's considerable experience with roots machines and large-scale manufacture is a plus, as is the company's integration with a fuel cell system manufacturer.
- The strengths of the project include the project team and the relevance of the problem being attacked.
- The project is conducting full-scale validation with a strong OEM partner.
- Broad relevant collaborations are a project strength.

Project weaknesses:

- The chosen (roots) technology will inherently be noisier and more bulky than a dynamic (centrifugal) air machine. Issues with potential bearing contamination also remain.
- The project is still far from performance metrics, with no clear path for closing the gaps. The significant cost of the motor/controller (beyond the scope of this project) is a serious concern.
- There is no clear path to the project meeting cost and durability targets.
- There does not appear to be a path for significant reductions in motor and controller costs.
- Eaton may never completely meet the targets.
- Better definition of the testing would help in understanding the true impact.

Recommendations for additions/deletions to project scope:

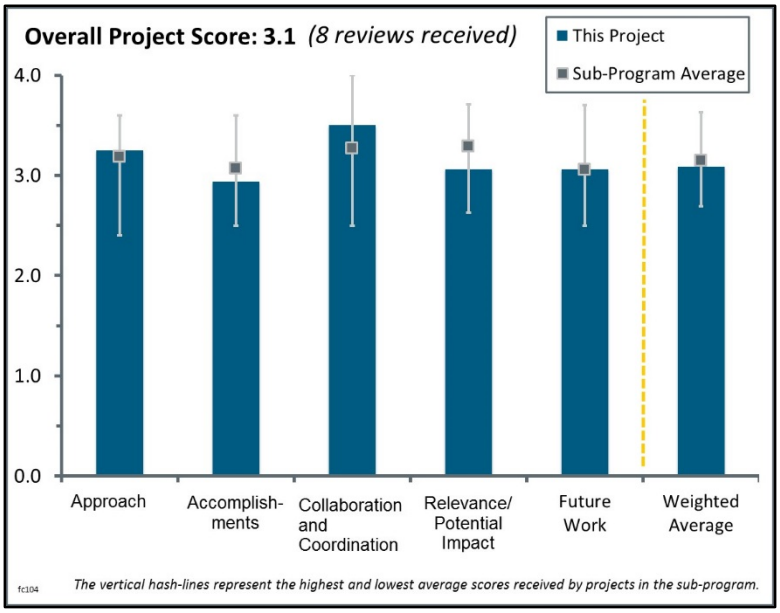
- The focus in the remaining six months should be on:
    - Integrated testing with the Ballard fuel cell system (including the expander).
    - Demonstrating expander compatibility with deionized liquid water.
    - Demonstrating the bearing technology's compatibility with fuel cell systems (analyzing for contaminants and perhaps consulting with the National Renewable Energy Laboratory and Hawaii Natural Energy Institute contaminant programs).
    - Demonstrating the viability of a low-cost plastic expander at PR >2–2.5.
    - Demonstrating a compressor capable of PR~3.
- A focus on cost reduction should be included, as should system testing on the full compressor-expander module, including the expander.
- The project should minimize effort on plastic component fabrication and focus more on closing performance gaps.

Project # FC-104: High-Performance, Durable, Low-Cost Membrane Electrode Assemblies for Transportation Applications

Andrew Steinbach; 3M

Brief Summary of Project:

The overall objective of this project is to develop a durable, low-cost, robust, high-performance membrane electrode assembly (MEA) for transportation applications that is able to meet or exceed U.S. Department of Energy 2020 MEA targets. The objectives for fiscal year (FY) 2015 are to (1) improve MEA robustness for cold start-up and local transient via materials optimization, characterization, and modeling; (2) evaluate candidate MEA and component durability to identify gaps; (3) improve activity, durability, and rated power capability of Pt_3Ni_7/nanostructured thin-film (NSTF) cathodes via post-process optimization and characterization; and (4) integrate MEAs with high activity, rated power, and durability with reduced cost.



Question 1: Approach to performing the work

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

- The project structure is robust. The initial work focuses on developing components (e.g., catalysts, gas diffusion layers [GDLs], and membranes) until they reach durability, performance, and cost thresholds. The components are then assembled into MEAs of various configurations. Best-of-class MEAs are then selected for building a short stack.
- This year's work was incremental but along tightly focused lines, addressing problems that have arisen in attempts to make NSTF workable for automotive systems. The project now attempts to use best-of-class MEAs to address all issues simultaneously, although efforts to address the durability of transient response will not occur until integration of the more durable Type B interlayer or some other solution. A large amount of data was presented in an unusually clear and comprehensible form.
- The approach is at the level of the complexity it addresses. The integration of anode and cathode 3M catalysts with membrane, GDL, and interlayer is planned adequately; leaching of Ni is not.
- This is a very well-developed approach for testing fuel cell operation under realistic conditions; however, this is something that 3M has been doing for decades. There is no meaningful improvement over previous efforts. It was also really difficult to see any systematic approach—too many parameters are being changed simultaneously (e.g., temperature and the nature of the catalyst) to really develop meaningful insights. The suggestion would be to simply pick the best catalyst and then systematically change the variables to develop a more mechanistic picture of what is happening. For example, the effect of temperature is obviously very important; however, it is difficult to determine what temperature is affecting—kinetics, stability, or something else entirely. It is positive that 3M is using modeling to better understand water management, but the researchers are encouraged to spend more time with real experiments rather than rely too heavily on modeling.
- This project is focused on (1) developing durable, low-cost, high-performance MEAs and (2) improving operational robustness for transportation applications by optimizing and improving 3M's low-platinum-group-metal (PGM) NSTF oxygen reduction reaction (ORR) cathode catalysts. Although this is an MEA-oriented project (not a catalyst project), it would still be best to make the best use of 3M's NSTF catalysts.

However, researchers have relied heavily on the use of interlayers and anode GDLs to overcome fundamental challenges faced by NSTF catalysts, which leads to other critical issues. In particular, the interlayer is of limited implications and would bring additional cost and technical issues.

- The approach has been consistent throughout the project—namely, using the same three methods to address the outstanding issues with NSTF MEAs: (1) small changes in the alloy composition, (2) changes in the GDLs, and (3) addition of an "interlayer." Unfortunately, the approach has not included any major changes to the catalyst layer design or composition, which should also be included because this is the layer that is limiting the performance.
- The approach is focused on 2020 targets on MEA cost, performance, and durability. The weakness of the approach is in incorporation of the interfacial diffusion layer and using the leaching catalyst, as was indicated in last year's review.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The team has done much work, and the number of cells tested by 3M during this project is truly impressive. There has been significant progress in some respects, such as shown in the graphic on slide 17. However, the rejection of the reviewers' comments on slide 17 is puzzling because the limited approaches being used are still falling short of the targets and also have some obvious weaknesses. For example, the addition of an interlayer also adds many concerns, including added complexity and increased PGM (loading); the durability of this layer that includes carbon is also a concern (one of the original major advantages of an NSTF electrode was that it did not include carbon and would therefore be inherently more durable). It seems very likely that the interlayer is degrading because, as shown on slide 10, the load transient performance is substantially degraded after accelerated stress testing, and the interlayer is the component primarily responsible for the improved transient performance, as shown on slide 9.
- Best-of-class MEAs exhibit promising results, with targets for PGM content (0.118 mg/cm^2) and Q/ΔT (1.45 kW/°C) achieved. However, the performances are still 10%–15% below the 2020 DOE targets. The work on improved robustness (GDL and interlayer) would require deeper effort to understand the failures at both electrocatalyst and support cycles (after 10,000 and 10 hours, respectively) and present an MEA of operational robustness. There is little commentary on the work on cold start modeling with the integration of Michigan Technological University's (MTU's) GDL pore network model and Lawrence Berkeley National Laboratory's (LBNL's) MEA model, and the documentation does not clearly explain how this effort is used (e.g., perhaps the project seeks improvements to the model or a back loop to the new GDL/MEA).
- Very good progress and a good deal of work have been accomplished in the last year. Key procedures to improve the MEA performance, integration of the catalysts, and incorporation of an interlayer apparently promise that, in subsequent research efforts, most of the performances will be at 2020 levels. However, several problems with this catalyst are not completely solved: the necessity of using an interlayer with the thickness of another catalytic layer, the dealloying procedure, and the problem with Ni leaching due to the Kirkendall effect. The third point has not been considered at all.
- Progress was made toward addressing the major problems that have arisen in attempts to integrate NSTF into automotive systems, although it is unlikely NSTF stacks will ever be a simple drop-in replacement for stacks made with conventional dispersed catalysts that have shown superior robustness over a range of important operating conditions. The performance of NSTF MEAs in air has gotten closer to that of state-of-the-art MEAs made with conventional supported catalysts, although NSTF still falls short. NSTF now appears to have been made compatible with reinforced 3M advanced membrane materials through modification of the membrane. Modification of anode GDLs has improved operational robustness for cold start, but it still falls short of that of conventional MEAs. Use of interlayers has improved robustness against cold start and transient operation but has introduced a new sensitivity to voltage cycling. The new Type B interlayers may provide at least a partial solution to the durability problem, but it is unlikely that the thin NSTF electrodes will ever fully match the operational robustness of conventional electrodes.
- While there have been some improvements in high-rated power performance and stable temperature range, the operational robustness does not appear sufficient. Furthermore, some of the improved performance

results shown in FY 2015 should be validated in the short stack testing. The modeling efforts by MTU and LBNL look promising.
- Progress has been made toward durability and performance targets owing to the synthesis of more durable and more conductive membranes and more durable interfacial layers. However, this new membrane is still not durable enough, and contamination problems are not solved. The more durable catalyst is still not durable enough.
- Based on the history of 3M in this space, more results were expected that would address intrinsic scientific questions related to this technology. Instead, there is much effort simply to "test" materials to assess their performance. Additionally, much more effort needs to be directed toward understanding durability on the cathode side rather than initial activity of the catalyst.
- The delay in stack testing is concerning. There have been years of fundamental development and studies of NSTF substrates and Pt-Ni dealloying. Stack testing should be predominant in all projects going forward. This approach should be either validated at the stack level or dropped from future DOE funding.

Question 3: Collaboration and coordination with other institutions

This project was rated **3.5** for its collaboration and coordination.

- 3M has effectively coordinated with other organizations to further develop catalyst activity, evaluate the operational robustness of 3M versus conventional electrodes, and evaluate the durability of all aspects of operation. One hopes that the planned stack testing, preferably with best-of-class MEAs with the addition of Type B interlayers, will give NSTF a full chance to demonstrate whether it, in its ultimate modification, can satisfy automotive requirements.
- The broad collaboration looks carefully managed, and collaborators were selected according to the need for their capabilities.
- This is a very good team of people who are trustworthy in the field, and obviously they collaborate very well.
- There is excellent collaboration with activities originating from all involved institutions.
- The project features good collaborations with appropriate partners.
- The effort between 3M, LBNL, and universities is very well coordinated.
- There are excellent collaborations with some groups that utilize the groups' areas of expertise, such as modeling and materials characterization. However, the team still does not have a fundamental understanding of what limits NSTF performance. The unanswered questions include, but are not limited to, the following:
    - How protons are actually transported in an NSTF catalyst layer.
    - How the water saturation level in the catalyst layers is changing with changes in the anode GDLs.
    - How the interlayer works and why it improves the transient performance.
- 3M has led collaboration efforts between strong team members. It is noted that there are good collaborations between MTU and LBNL relating to GDL selection and transient response. However, a greater emphasis should be made on the stack testing, which will be beneficial to all team members. It is important to effectively integrate all key components and have a reliable system integrator, such as General Motors (GM), test at a system level.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.1** for its relevance/potential impact.

- The project is fully in line with DOE research, development, and demonstration objectives for a durable, performant, and cost-efficient polymer electrolyte membrane fuel cell for transport applications. Specifically, the project addresses the following 2020 DOE targets:
    - Performance: 1 W/cm^2.
    - PGM content: 0.125 g/kW.
    - Durability: 5,000 hours with voltage loss <10% V.

- The unique capabilities that have been developed for synthesizing materials, building full cells, and testing their performance are an asset to DOE and will be very important in moving the technology forward.
- The project has a potential to be a game changer for the Hydrogen and Fuel Cell Program (the Program).
- If considerably simplified, this could be an important catalyst technology.
- This project has the potential to have outstanding relevance to the Program since NSTF is a promising catalyst architecture with inherently excellent performance and durability. However, the inherent benefits of NSTF have not been fully realized, and after more than a decade of development, it has not displaced the status quo catalyst of Pt on C (with the possible of exception of use in electrolyzers). What is required is the development of advanced catalyst layers that can realize the true potential of NSTF. Instead, the principal investigator has chosen to use "Band-Aids" to partially mitigate the key issues, and these fixes do not address the fundamental issues or promote the fundamental understanding that is required to successfully convert new catalyst architectures into high-performance and robust MEAs. It is possible that this project has uncovered a new decay mechanism, which is claimed to result from the deposition of ionomer degradation products. However, there is as of yet no convincing evidence that this mechanism is correct.
- NSTF, with its intrinsic durability against voltage cycling, could have a significant impact on the development of widely commercializable fuel cells and electrolyzers. However, a number of shortcomings as compared to conventional MEAs, properly addressed in this project, need to be overcome if NSTF is to become usable in any but a few highly specialized applications. 3M management's blanket policy of not discussing manufacturing costs for any of its products or potential products significantly degrades this project's utility to the advancement of the fuel cell field, as costs cannot be adequately estimated. It would be quite possible that a significant amount of society's resources could be spent pursuing developments with 3M that ultimately would prove too costly for practical application.
- The project is aligned well with DOE goals and objectives. However, operational robustness (under load transients, low-temperature operation, etc.) should be addressed. Furthermore, the use of an interlayer to meet DOE's target may be reconsidered.
- The project addresses a critical challenge facing DOE. However, go/no-go decisions regarding the future of NSTF need to be made so that either (1) the technology can be advanced or (2) resources can be diverted to other efforts (such as alkaline membrane approaches).

Question 5: Proposed future work

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

- The project has identified the remaining barriers (robustness and durability) and has devised plans to overcome them. At some point, component improvement has to cease and a decision on the project's performance has to be made so that testing at the stack level can finally be executed.
- Despite its weaknesses, the proposed work may still resolve many key issues with NSTF catalysts in fuel cells. That said, it appears this project is ending in August 2015, so it is difficult to judge the relevance of the proposed future work without further DOE support.
- It is to be hoped that the planned stack testing, which (one hopes) will incorporate best-of-class MEAs with the addition of the more durable Type B interlayer, will provide a conclusive test of whether NSTF can prove acceptable in automotive applications without very costly, and technically questionable, modifications to balance of plant. If the planned stack testing does not give acceptable activity, durability, high-current performance in air, and operational robustness, difficult modifications of the basic NSTF structure to give a thicker catalyzed layer with adequate porosity may be needed for further progress to be made.
- During its remaining time, the project team should focus on elucidating the mechanism responsible for the decay observed, which may be relevant to the entire fuel cell community. This effort should focus on the degradation of NSTF without the interlayer because the mechanism for interlayer degradation is obvious (the same as other Pt/C electrodes) and will mask the decay of the NSTF layer, which appears to be unique.
- The proposed future work is focused on overcoming barriers related to catalyst and membrane durability.
- The proposed key future work is focused on overcoming the remaining barriers using the NSTF cathodes (with higher mass activity and improved durability) developed outside this project. Considering the short time left for this project, this approach might be the best choice, but the benefit for DOE is not clear. The short stack testing would be most critical.

- The interlayer and Ni leaching problems remain. No definitive resolution is proposed.

Project strengths:

- There are many strengths to this project, which stem from clearly recognizing the importance of connecting fundamental science to real systems. The capabilities developed by 3M to synthesize all parts (e.g., the cathode, anode, and MEA) of the fuel cell are indispensable to making this connection. Finally, the willingness to admit that, even with these great capabilities, there are still many issues (e.g., water management and the durability of the cathode) that must be solved to make this a viable technology demonstrates a clear understanding of what must be done to move the technology forward.
- The project has continued development to take advantage of the durability advantages of the NSTF catalyst layer structure. The project is starting to properly consider the special problems that occur in air at high current density for electrodes with relatively low Pt surface areas (i.e., problems that behave as if a local oxygen transport resistance occurs very close to the active Pt surface). This is one of the most important problems now facing development of fully satisfactory automotive cathode catalysts.
- This new catalyst has resulted in raising interesting questions for the fuel cell technical community, such as how protons transport in NSTF layers with no ionomer. Understanding this may result in the design of new types of catalyst layers with less ionomer than is used in conventional catalyst layers. The ultra-thin catalyst layers, with very low total surface areas, have also been a model system for the impact of contaminants, which may help to reveal new decay mechanisms. However, neither of these key potential benefits has been fully realized to date. This project has also clearly demonstrated that radically new catalyst architectures can also require very different catalyst layer designs (and the development of these new catalyst layers can be challenging but must be included for any novel catalyst architecture).
- Strong team members are making collaborative efforts to meet DOE's targets. It is noted that the efforts to combine modeling and experimental works would be beneficial to better understand the current problems and find better solutions.
- The project has a strong research team, strong resources, and good funding.
- The effort is technically strong and well coordinated.
- The project uses a combination of real-life tests with modeling and advanced diagnostics.
- The PGM content is higher than DOE 2020 targets.

Project weaknesses:

- The NSTF catalyst layer structure itself has insufficient void volume to properly handle liquid water under start-up and load-transient conditions. While the project has made progress in addressing these issues, the overall operational robustness of the MEAs produced to date still appears to be inadequate for automotive utilization without substantial and costly modifications to the balance of plant (e.g., subatmospheric anode pressures). The interlayers that attempt to patch up the cold start and transient issues comprise conventional dispersed catalyst layers, leaving the modified NSTF system susceptible to the same voltage-cycling degradation that plagues dispersed catalyst systems and to which the original NSTF was remarkably resistant. However, it is possible that more corrosion-resistant supports that do not work very well in conventional electrodes could still be effective when used in interlayers. If Type B interlayers prove to be both effective and durable in stacks, this project "weakness" could become a "strength."
- One key weakness is the lack of systematic work. Testing itself is not enough to understand the key limitations and how to overcome them to improve fuel cell performance. It is obvious that there is still a large gap between the catalytic activity observed with classical rotating disk electrode measurements and MEA performance, and 3M may be in a unique position to bridge some of these differences. The team is encouraged to make use of 3M's unique capabilities to really focus on addressing this difference in performance.
- After more than a decade of NSTF MEA development, the key issues have still not been mitigated sufficiently to enable this new catalyst to replace more conventional MEAs, primarily because 3M has not been willing to make any significant changes to the original NSTF catalyst layer structure. The reluctance to make major changes to the catalyst layers is understandable, but it is a major weakness not to include changes to the layer that is limiting these MEAs.

- While 3M's expertise in NSTF catalysts and General Motors' support are the key assets of this project, researchers have relied heavily on the use of interlayers and an anode GDL to overcome fundamental challenges faced by NSTF catalysts. This solution brings with it many other critical issues.
- The project uses an incremental, rather than a transformational, approach, and the modeling does not have predictive capability.
- Weaknesses include the high PGM contents in catalysts and the complex technology.
- The performances and durability are weaknesses.
- Overall, further progress should have been made at this point, given the significant previous investment in this approach.

Recommendations for additions/deletions to project scope:

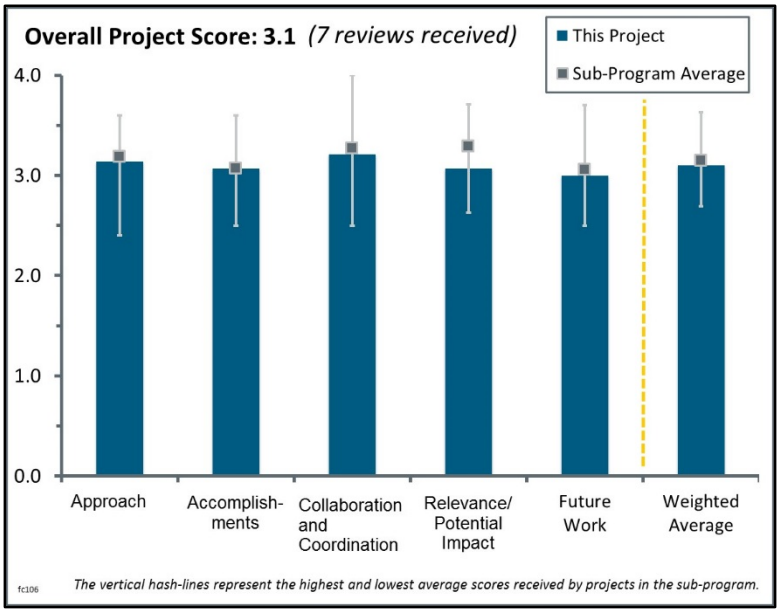
- One hopes that the MEAs to be tested in stacks will incorporate the Type B interlayer in hopes of achieving some level of durability of transient response. Otherwise, the MEAs will have little chance of proving adequate for integration into automotive fuel cell systems. NSTF stacks should be tested with practical anode feedstreams and conditions, (i.e., those representative of anode recirculation or related strategies).
- The project should define a go/no-go threshold for future component improvements so that the project is steered in its overall course.
- With the short time left on this project, a greater emphasis should be placed on the short stack testing of best-of-class MEAs.
- The Ni leakage after dealloying should be addressed.
- It is recommended that DOE hold a review to assess the potential for NSTF/Pt-Ni dealloying to determine whether this continued support is the best investment of federal funding.
- DOE should not fund future NSTF projects unless they include the development of substantially new catalyst layer architectures. DOE needs to recognize that the development of new catalyst layers must be part of any novel catalyst structure.

Project # FC-106: Rationally Designed Catalyst Layers for Polymer Electrolyte Membrane Fuel Cell Performance Optimization

Deborah Myers; Argonne National Laboratory

Brief Summary of Project:

The overall objective of this project is to realize the oxygen reduction reaction (ORR) mass activity benefits of advanced Pt-based cathode electrocatalysts in membrane electrode assemblies (MEAs) and stacks operating at high current densities and on air and at low platinum group metal (PGM) loading. Specific goals are to (1) determine the properties of electrode/catalyst layers that limit the high current density/air performance of electrodes based on advanced Pt-based cathode catalyst; (2) use information from characterization efforts to determine the performance-limiting property of the current d-PtNi electrode; and (3) design the catalyst layer composition and structure and support functionality to mitigate the performance limitations, guided by computational modeling.



Question 1: Approach to performing the work

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

- The proposed approach is to determine the properties of electrode/catalyst layers that limit the high current density/air performance of electrodes based on advanced Pt-based cathode catalyst and, in particular, PtNi (d-PtNi) developed by Johnson-Matthey Fuel Cells Inc. (JMFC) (within the General Motors-led FC-087 catalyst project). This effort is completed by characterization and by designing catalyst layer composition and structure and support functionality to mitigate the performance limitations. Regarding the metrics obtained for this dealloyed PtNi, this approach makes sense.
- The approach is well designed and focused on identification of performance-limiting property of catalyst layers employing dealloyed PtNi catalyst with low loading.
- The overall approach looks good, especially because this is an electrode project and not a catalyst project. Use of the JMFC catalyst is fine. The approach of understanding performance and inks is good, but there needs to be more understanding gained (aggregation from the diagnostics). There should be more transport-related diagnostics and in-cell characterization for an electrode optimization project.
- This is important work to understand better the role of ionomer in low-Pt-loaded catalyst. There is a lot of speculation regarding the oxygen diffusion rates through ionomer used in low-Pt-loaded catalyst. Some comment on this mechanism and how the work in this project is affected by the oxygen limitations (or not) would be helpful.
- This project is focused on the development of MEAs with advanced Pt-based catalysts for high current densities on air at low PGM loading. The Pt catalysts used in this project are d-PtNi catalysts developed by JMFC during a previous U.S. Department of Energy (DOE) project (FC-087). ANL's efforts include (1) determining the limiting factors of d-PtNi at the high current density on air and (2) developing better catalyst layer composition and structure and support functionality to address the critical issues.
- The approach is fairly general, but it is based on the relevance of the project. Simply put, the project team will use all characterization methods at its disposal to figure out why performance is low at high current density for electrodes made from dealloyed PtNi. Then, the project team will attempt to improve the

electrode. The collaboration slides point to JMFC as a supplier of inks and catalyst coated membranes (CCMs), but the approach slides would be improved if they clarified which organization is making the inks and CCMs. Perhaps a flow chart of tasks would be useful. It may be useful to understand more about the cell fixtures used. The operating conditions are different (which takes a "snapshot" of the inlet of a full-scale cell), so perhaps whether the flow field is serpentine or straight is not as crucial, but it may matter. The ionomer-to-carbon (I/C) ratio experiments might benefit from looking at low O_2 concentration, similar to the organic versus aqueous comparison.

- The project features a well-designed approach of taking a high-performing but commercially viable catalyst and developing an understanding around the electrode design requirements. A significant concern is the continued leaching of the Ni and the impact this has on the ionomer and associated performance parameters. This raises the question of whether a self-contaminating system is ultimately a viable approach for very low catalyst loadings. On the other hand, if Pt-metal alloys continue to provide the best path forward in terms of scalable catalysts, then the work is of utmost importance. The extensive use of characterization and linkages to models is a solid approach. However, most of the results on I/C ratio effects are based on only two levels of I/C. More are required to understand trends and to quantify effects. The project is focused only on beginning-of-life or early life effects (up to two weeks of testing). This is appropriate because there is still much to understand and optimize for the initial performance effects. However, additional, previously unidentified effects may be uncovered with durability testing, and it is unlikely that durability will be effectively addressed in this project.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- Significant progress has been made toward DOE targets on MEA performance at 800 mV and 675 mV. Careful analysis of the effect of Nafion® ionomer content and ink composition on the performance of low-loaded catalyst layers is of great significance. Performance-limiting properties have not been identified yet.
- There has been good improvement in performance since the last DOE Hydrogen and Fuel Cells Program Annual Merit Review, but because the project is two-thirds done, there needs to be almost the same level of improvement again by next year to achieve the project targets. Also, the results achieved are under high-stoichiometry, one-dimensional conditions. Actual MEA results will likely be lower. The identification of the effect of Ni^{2+} on the ionomer in terms of O_2 permeability and electrode resistance is an important finding, as well as the fact the Ni oxidation is influenced by the ionomer.
- This project has exceeded go/no-go decision criteria and met several key milestones. In particular, the principal investigators (PIs) have taken good approaches and obtained meaningful data on the effects of ionomer content and the type of organic solvent in ink, as well as successfully demonstrated the effectiveness of the acid-washing process of MEAs. However, the development of the catalyst support surface functionality to improve low relative humidity (RH) performance does not look promising and may need to be reconsidered for the future work.
- This project seems on track to meet the stated DOE goals.
- The team has accomplished a significant amount of work, but the presentation is quite difficult to follow. It should be better structured to emphasis the main highlights. All the go/no-go points and milestones have almost been achieved. Regarding the results, some questions remain, such as what the impact of the treatment (thermal treatment and acid leaching) is on the carbon support, and whether the effect of the catalyst (Pt particles) activity or the effect of the carbon wettability change is observed. The carbon support could be graphitized during thermal treatment and oxidized during acid leaching. Indeed, on slide 8, it is written, "Decreased the performance of the d-PtNi/C cathode in the mass transport region on air at 100% RH (relative humidity)," which is typically observed for hydrophilic support. It is unclear whether the stability of the catalyst has been studied in a single-cell test. Not much progress has been made with respect to the durability of the dealloyed catalyst.
- There are many questions that could be asked about the organic solvent versus aqueous solvent polarization comparison. Some of the results are subtle enough that the subtle increase in loading with an organic solvent could be responsible. It is difficult to make a generic statement that organic solvents are superior without an experimental design that adjusts mixing times, temperatures, levels of agitation, and other

parameters for the particular solvent used. The organic solvent was shown to enhance particle breakup while also increasing gas permeability of the resulting layer, given the same solvent mix and I/C ratio. However, if the two solvent types require different recipes and mixing to achieve particle breakup and high layer permeability, then perhaps a wider parameter range needs to be investigated. Ni loss data need to be reported in association with the methods used for hot pressing and CCM fabrication. The limiting current trends reported for Pt/C, annealed Pt/C, and dealloyed PtNi/C could all be related to parameters other than the particle sizes of Pt and PtNi. Despite similar Pt weight percentages, perhaps there were variations in catalyst layer thickness. It may be helpful to break down pressure-dependent mass transport losses from non-pressure-dependent losses, or to even go further and use methods similar to Nonoyama et al. (2011) to distinguish Knudsen diffusion from resistances related to ionomer film thickness. For what the project is attempting to accomplish, the throughput of data is high, and the data are of high enough quality that conclusions can be drawn based on the processing context in which the project lives.

- The work on inks was interesting, but it needs some further clarification; modeling would help. In addition, it is unclear whether there is any beam damage that may be altering the profiles with drying. The use of RDE studies for determining permeability of the ionomer is questionable because the environment is different. The use of equivalent circuit models, especially with the catalyst layer, is questionable, particularly because the electrochemical impedance spectroscopy can also incorporate Ni movement into the conductivity. It is unclear whether the low surface enhancement factors correspond to what was seen in particle sizes in the inks. The improvement and metrics are noteworthy, although some more guidance to developers on the key issues would be better. More cell diagnostics are needed to provide understanding as well as visualization of the catalyst layer, where the only presented work came from outside of this project. The free ionomer may be providing the conductivity in the layer, so it should not just be disregarded.

Question 3: Collaboration and coordination with other institutions

This project was rated **3.2** for its collaboration and coordination.

- The project features very close collaboration and well-coordinated effort between Argonne National Laboratory (ANL), JMFC, United Technologies Research Center (UTRC), and universities.
- While it is clear that collaboration is occurring, the presentation needs to more clearly explain the roles of the collaborators. If JMFC is preparing an ink, a catalyst layer, or a CCM, this should be spelled out. It appears UTRC is performing tests, but this also needs to be clarified. For purposes of reviewing, it is important to know whether a catalyst layer is prepared by a commercial supplier or by a national laboratory. The Indiana University – Purdue University Indianapolis (IUPUI) collaboration provides interesting results. It would be interesting to see confirmation of the functionalization of the carbon, as well as whether directly functionalizing the carbon yields similar results to those parameter spaces in the rest of the project where I/C contact is enhanced. Collaboration with the University of Texas (UT) was used to show that ionomer tends to associate itself with Pt or PtNi in preference to carbon. It may be interesting to see whether this same trend remains for the IUPUI materials.
- The teaming arrangement is good with national laboratory facilities, a group with stack experience, and a material supplier. Interactions with automotive original equipment manufacturers would be good. It is not clear why the work detailed on slide 13 is done outside of this project, why it is presented as part of the project, and why UT did not contribute, as it seems to be its role. The role of the academic partners seems much more limited in the presentation than as stated in the text.
- The collaborations among several industry, academic, and national laboratory partners are very impressive. It is unclear whether the catalyst dealloying was done at ANL or at JMFC.
- While good collaboration efforts have been made to meet DOE's technical targets, it was disappointing to learn that it took such a long time for the team to adjust UTRC's fuel cell testing fixture for the MEA developed in this project. It appears that more cell testing works under the right conditions (e.g., cell pressure) should be done in the coming year(s).
- The coordination appears to be correct, but it may be improved by using all the characterization techniques and resources offered in FC-020 (K. More).

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.1** for its relevance/potential impact.

- The project is relevant to the objectives of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan. Achieving high mass activity of advanced Pt electrocatalysts at high current density is of great importance to the commercialization of these materials if they are also durable.
- PtNi catalysts are an important new development. Finding optimized electrode constructions is very relevant for these systems.
- This project is highly relevant and has good potential to meet DOE's targets and goals. A greater emphasis should be placed on cell testing and durability studies for this project to be successfully implemented with high impacts in the end.
- The performance of a catalyst layer or entire MEA begins with processing. The processing affects the structure (or the fluidity thereof during operation), which then affects properties (e.g., pore size distribution, gas diffusivity, and electrical or thermal resistances), which in turn affect performance. Because of this, there are two possible risks to the relevance of a DOE-funded project that attempts to integrate a highly active catalyst into an MEA: (1) higher-volume ink processing will differ substantially from the lower-volume ink processing demonstrated in the project, and (2) there are engineering changes that could be made to enhance processing for a given parameter, despite a poor result within the time frame of the project. In other words, the project could make an attempt to state as scientific fact a result that could be addressed with engineering variables (e.g., solvent, mixing times, and temperatures at different steps). The team does attempt to address a very relevant concern, which is the facilitation of high power density using an alloy catalyst that demonstrates high activity for oxygen reduction.
- The work is very relevant, but the targets are not sufficient for longer-term automotive performance. The increased understanding around the Ni dissolution effects as a function of MEA conditions and the resulting impacts on ionomer are an important step toward achieving better results. There is a concern that even if the Ni is acid washed, the Ni will continue to leach out, and the integrated catalyst system may in the end not be durable enough to maintain performance at the high current density conditions. Because much of the work involves the effect of the Ni interactions with the ionomer, this may reduce the relevance of the work.
- The work in the project, especially the inks, is good, although there needs to be more focus on how this impacts overall cell performance. The project addresses the need to understand how catalyst layers form, but not necessarily the functionality and key structures of the catalyst layer. The functionalization is quite interesting and should be continued with more focus on what is occurring. Perhaps there is a way to do the functionalization before making the catalyst.
- Although the project aligns very well with the Hydrogen and Fuel Cells Program, its potential impact is diminished because it employs the leaching PtNi catalyst.

Question 5: Proposed future work

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

- A few things could be added to the next steps: detection of Ni in the membrane (perhaps even in the anode layer) and data interpretation for performance curves from 40° to 95°C. Many of the items mentioned in the next steps are worthwhile, including nanoscale computed tomography and the use of results to assist with modeling. Proton conductivity measurements for the catalyst layer are also worthwhile. Given the troubles with Ni, it may be wise for the project to consider the dealloyed PtCo that also was investigated in the General Motors (GM)/JMFC project. This project may be in a good position to clarify the leaching tendencies of Co versus Ni from a dealloyed catalyst. The team should include some metrics that correspond to CCM quality, such as the uniformity of Pt loading and the uniformity of catalyst layer thickness. Experimental bandwidth could be expanded to include the effect of more graphitized carbons

than Ketjen black, as well as the effects of engineering parameters involved in ink processing and application.
- The future work described is aligned with the proposed work of the project. The evaluation of the carbon support impact on the treatment (e.g., temperature and acid leaching) should be done, and durability evaluations should be started.
- Focusing on the drop in performance at high current for the d-PtNi/C system makes sense. If significant amounts of Ni dissolve in the membrane, then one would expect reduced high current performance. No durability studies were proposed. If Ni leaching is significant, one would expect the voltage decay to be significant over time.
- The proposed future work is focused on identifying other reasons for mass-transport limitations at high current densities. Porosimetry analysis needs to be complemented by analysis of hydrophobic and hydrophilic pores in the catalyst layer.
- The proposed future work is adequate, although ambitious, for the remaining time period. The focus should be on understanding and the remaining questions instead of just more diagnostics—there is not enough time to bring together the findings. More structural data and long-term stability would be good.
- The surface functionalization of carbon supports should be reconsidered. The gain (i.e., proton conductivity) is significantly less than the loss (i.e., decreased ORR mass activity by 55%) observed thus far.

Project strengths:

- ANL is capable of collecting an extraordinary amount of data. It has maintained a systematic approach to the study, maintaining certain control factors to be constant to focus on particular trends (e.g., particle breakup with more ionomer or with different solvents). ANL has collaborated with an MEA supplier (JMFC) and a stack developer (UTRC). It has used characterization techniques that are particularly for the study of inks (e.g., light scattering). ANL has access to numerous collaborators within the national laboratory system. The project began by using a catalyst that has already been shown to be stable in another publicly funded project.
- The project team is a strength, with ANL and JMFC heavily engaged in catalyst preparation, ink preparation, and post-mortem characterization; UTRC in cell testing; and UT and IUPUI in characterization. The project covers catalyst materials and fabrication, catalyst layer characterization, and modeling and diagnostics.
- Significant characterization is conducted to understand the effect of various variables on the electrode structure. This characterization will be linked to performance using models to arrive at an improved understanding. The assembled team is very strong.
- Overall, the good approaches taken and the strong team members are the strengths of this project. It seems the team activities have been well managed by the PI.
- This project addresses critical needs in terms of catalyst layer fabrication. The overall progress has been good, and the project features a relatively strong team.
- The project features very thorough characterization of ink properties and the impact on fuel cell performance. The modeling supports the experimental work very well.
- A strength of the project is how it combines advanced diagnostic and characterization tools with MEA testing capabilities.

Project weaknesses:

- The project is operating in an area that is essentially an engineering area; a vast number of parameters are available to make something work. This is different from many other DOE projects, where conclusions can be drawn from fundamental science and, therefore, more of a “pass” or “fail” rating can be achieved. From this project, a conclusion may be that the dealloyed PtNi catalyst simply cannot be processed in a certain way into a CCM. An unfortunate conclusion would be if the results generated a false negative to say that dealloyed PtNi cannot be incorporated at all. More attention should be focused on processing parameters that may enable success of dealloyed PtNi at high current densities. There may also be other ink constituents (beside solvent, ionomer, and catalyst powder) that would help with particle breakup and the resulting catalyst layer porosity. The project could use more focus on manufacturing and quality metrics.

Some questions may be asked regarding the production scale being represented in this project and the possible effects of non-uniformities and defects on the results.

- It is not clear whether the modeling is just describing data or providing guidance for fabrication and inks. The project contains a lot of data and diagnostics, but there needs to be further analysis of all of the data.
- It is unclear whether Ni-based catalysts are stable enough for long-term durability in fuel cells. The future work listed a variety of characterization techniques but did not relate those clearly to the remaining gaps in meeting performance targets.
- Employing the leaching catalyst adds complexity to the project. The additional acid-pretreatment step will add cost to the system.
- The focus is on the d-PtNi/C catalyst. Although it seems to be a good choice because of the commercial relevance (coming from a GM project, synthesized by JMFC), the findings around the Ni dissolution may dominate other effects.
- The MEAs containing d-PtNi catalysts have been so sensitive to humidity, and surface functionalization of carbon supports would not be the right solution.
- The lack of durability evaluation of the d-PtNi is a weakness to be overcome.

Recommendations for additions/deletions to project scope:

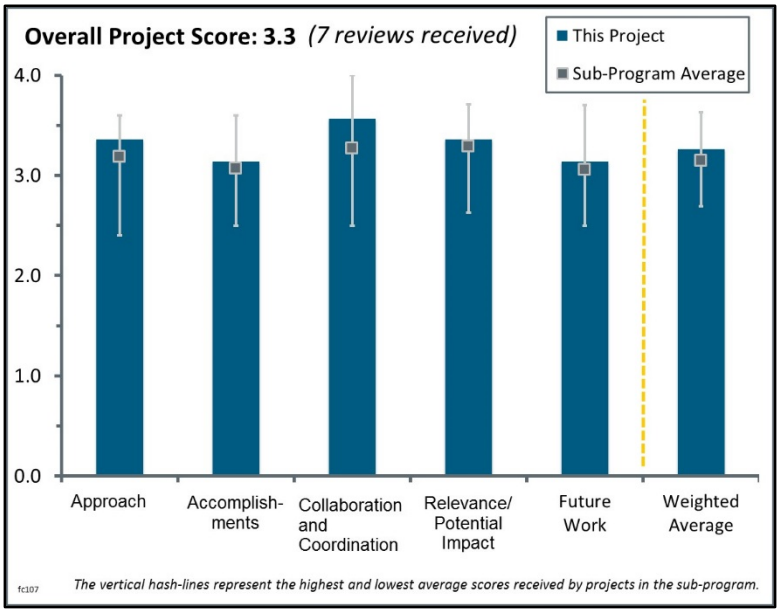
- At this time, there are no recommended deletions. Recommended additions include the following:
    - Greater emphasis on the effect of carbon supports, and on the effect of processing parameters in making inks and catalyst layers.
    - Characterization of ionomer location with carbon functionalization.
    - More attention to catalyst layer, catalyst ink, and CCM quality.
    - Comparison with JMFC dealloyed PtCo.
    - Differentiation of mass transport resistances (e.g., Knudsen, ionomer film).
    - Greater interpretation of results collected from low to high temperatures. Analysis of Ni content in membranes should be reported.
- The project team should provide more visualization of the catalyst layer and also cell-level studies. ANL needs to bring together all of the improvements and understanding for best-of-class MEAs. There are a lot of variations shown, but it is not clear whether they are additive.
- The researchers should increase their focus on the Ni stability in the final fuel cell under normal or accelerated operating conditions. To evaluate the effect of dissolved Ni on performance, it might be useful to study the degree of dealloying versus performance at high current.
- The team should perform experiments to evaluate the carbon support impact on the treatment (e.g., temperature and acid leaching). ANL should start durability evaluations.
- The MEAs containing d-PtNi catalysts have shown poor performance at low RH, and more efforts should be made to improve mass transport at low RH by improving the cathode catalyst layer structure.

Project # FC-107: Non-Precious-Metal Fuel Cell Cathodes: Catalyst Development and Electrode Structure Design

Piotr Zelenay; Los Alamos National Laboratory

Brief Summary of Project:

The overall objective of this project is to advance non-platinum-group-metal (PGM) cathode technology through the development of new materials and implementation of novel electrode concepts, together resulting in (1) high oxygen reduction reaction (ORR) activity, viable for practical automotive systems; (2) improved catalyst durability; (3) high ionic/electronic conductivity within the catalyst layer; and (4) adequate oxygen mass transport and effective removal of the product water.



Question 1: Approach to performing the work

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

- A very comprehensive approach was outlined.
- The project team understands the key issues in developing a non-PGM catalyst: increasing the site density, understanding which sites are responsible for oxygen reduction, using a carbon species that is resistant to corrosion, and developing the tools (e.g., models and fabrication techniques) to minimize the catalyst layer thickness and the associated mass transport resistances. The project scope is compact and avoids activities that add less value, such as building a stack. Ultimately, the catalyst must not include Fe to be compatible with membranes and the ionomer. The approach allows for Fe-free catalysts to be made and tested. However, an enhanced approach would include either greater emphasis on this or greater emphasis on using Fe to understand the mechanism of reaction better. The approach will identify reactive sites, but it leaves out mechanistic studies.
- The project team is incorporating a multi-pronged approach to achieve real progress in the use of non-precious metal (NPM) catalyst. Activity is addressed through modeling and other studies to understand active sites. Durability is measured and characterized, and change in active sites is explored. However, durability cycling in N_2 has low relevance to actual systems. The performance in the membrane electrode assembly (MEA) is addressed through electrode studies and modeling. The use of the Nafion® mapping is interesting, and the linkage to the models is a good approach.
- The project utilizes an effective combination of synthesis, characterization, and modeling. This has led to significant improvements in performance. However, it is unclear whether the modeling implicitly suggests that CM-PANI-Fe-C-based catalysts are limited.
- The project has established defined approaches to increase the activity and number of active sites, and to understand better the nature of active sites in the non-PGM catalyst based on CM-PANI-Co-C and CM-PANI-Fe-C systems. It is not clear how many heat treatment and leaching steps are involved for the catalyst synthesis.
- Given the complexity of understanding performance for non-PGM fuel cell materials, the principal investigators (PIs) have made impressive efforts to utilize all available modern tools to better understand these systems. One important component of this work is to combine experimental results with computational modeling; however, the power of any calculations to model not only the activity of the active centers, but also their stability, is doubtful.

- The focus on non-Fe-based catalyst is a solid approach. The team needs to quantify the amount of metal leached from the catalyst layer to the membrane.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The project team has made significant progress since the last DOE Hydrogen and Fuel Cells Program Annual Merit Review in terms of physical characterization and modeling work to identify the catalytic active sites for ORR and electrochemical characterization to study the catalyst performance.
- The project team has made steady, incremental progress in improving the catalyst and identifying catalytic sites.
- There is good progress against the targets, and all project performance measures and milestones are either met (some exceeded) or remain on schedule to be met.
    - The project has adopted the updated CWG targets, which are more challenging than the original project targets. The June 2015 durability and selectivity targets were achieved and exceeded, and no Fe was detected in the membrane, which is important for membrane durability. However, the durability target is on N_2, and there was a ~200 mV loss due to cycling in air. There needs to be further work to understand and address this issue. There is good progress on understanding active sites, using a combination of measurements, modeling, and experimental approaches.
    - There is good progress to date on the catalyst layer (CL) activities. There is much more to be done, but the project is heading in the right direction. The Nafion® mapping is interesting, and the approach to use the morphological property distribution inputs from nano-X-ray tomography (nano-XRT) imaging and transport property simulations should provide good insight. The model is validated for the shape of the curves and trends for Nafion® content and water content, but absolute performance values are not predicted. Model prediction for no liquid water indicates a path forward for improvements.
    - The Microstructured Electrode Scaffold (MES) also provides valuable information to probe the ORR and conductivity within the CL as a function of relative humidity (RH). The thinner CL and resulting significantly improved performance data shown is on oxygen. If there are O_2 mass transport limitations, then they are very severe. This result should be further explored with the model. The polarization curves on air should be shown as well.
    - There is quite a mix of conditions shown, such as data on both O_2 and air, but not consistently, and a variety of membrane thicknesses. The team should provide increased clarity to enable comparison of data.
- The team has made good progress, but it needs to focus more on high current density performance improvement under H_2/air testing.
- Progress toward DOE goals is good; however, reference was made to previous goals as well as new goals outlined by the Catalyst Working Group (CWG). It was not always clear to what extent these goals were being reached. This should be more clearly articulated in future presentations, via tables.
- Modest progress has been made so far, especially toward improving the stability of materials. The nature of the active sites, however, is nothing new. These types of active centers have been proposed many times. To make non-PGM materials attractive relative to standard materials, the PIs should consider pursuing more challenging directions. The key criticism is the manner in which PIs are addressing the nature of the active sites. As they are very well aware, determining the nature of the active sites is very difficult now using available techniques. As a result, attempting to develop a reaction mechanism based on the nature of the active sites is very dangerous with the current information presented. In situ spectroscopic techniques are required to even begin to make any such claims.
- Co-based, Fe-free catalysts showed low performance, beginning with low open circuit potential (<0.9 V). Peroxide generation was 10%.
- X-ray absorption spectroscopy (XAS) was performed with Fe-only (no Co) catalysts. Results appear to confirm that the Fe-N_x sites are those that are active for oxygen reduction. Although performance improvements appeared to come from the addition of Co, there was no XAS shown to indicate why that would happen.

- Peroxide generation between 1% and 2% is still too high.
- It would be good for the catalyst layer thickness to be reported on some of the polarization curves.
- The team appears to have validated the model predictions versus the experimental results; it is unclear whether the model was used to improve the performance of CM-PANI-Fe-C. LANL appeared to have investigated numerous catalyst layer thicknesses using the same catalyst loading. However, it is unclear which thicknesses were associated with the performance improvement shown, and whether the model would have predicted the improvement.
- It was unclear what the cycles were for the XAS durability study. The cycling may be 0.3–0.7 V according to one slide, but it is hard to say.

Question 3: Collaboration and coordination with other institutions

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

- The project has excellent collaboration among national laboratories, academic institutions, and industries. So far, each collaborator has performed its specified tasks in support of the project objectives.
- The project lead has put together an outstanding team of PIs who are providing a wealth of valuable information required to move the field forward.
- The collaborators appear to be well coordinated, and the presenter made clear their respective roles.
- This is a large project team, with what appears to be good coordination between groups.
- Collaboration is needed with the University of Waterloo on Fe-free catalysts. Hopefully, collaboration can be used to develop better Fe-free catalysts. The nitrogen oxide dissociation observed by the University of Rochester helped to confirm what was also seen by XAS: that the active site is Fe-N_x. The nanoscale computer tomography performed by Carnegie Mellon appears to have greatly assisted the effort to produce a catalyst layer model, so as to predict proton conduction and oxygen diffusion. Oak Ridge National Laboratory showed that there are still disconnects in the catalyst layer between ionomer and the active sites. The project team has done well to add both a supplier and a developer—IRD Fuel Cells and General Motors (GM), respectively. IRD appears to have added to the project by enabling better fuel cell performance, although some of the background on this is not in the presentation. The collaboration with GM does not appear to be represented in the presentation.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.4** for its relevance/potential impact.

- Because the project follows the adjusted targets for non-PGM catalysts, it maintains relevance. These targets are based on the premise that stack size should not be increased to accommodate the removal of Pt. An increase of stack size drives issues with cost, packaging, and robustness at low ambient temperature operation. The removal of Pt would be extremely helpful in allowing commercial fuel cell vehicles to achieve cost targets.
- The project aims at the development of non-PGM cathode catalysts, which are important for the development of PEM fuel cells (PEMFCs) for automotive applications.
- The potential impact of this work is immense; however, the challenges are equally large. If the PIs would be able to develop active and stable non-PGM material, this would be outstanding. Many past effortsts have failed to introduce materials that do not have Pt as the active site. DOE should definitely support more research in this field; however, the progress should be much faster to be able to determine whether working with non-PGM materials is the right direction.
- It is still difficult to determine whether NPM catalysts will have a prolonged role in automotive fuel cells. The work is relevant because the goal to minimize Pt from the PEMFC is worthwhile. However, a more relevant goal would be to target a maximum total PGM per vehicle and stack cost, rather than just the elimination of PGM. The relevance of durability target V cycling in N_2 is low. The cycling in air resulted in much higher degradation and is of much higher relevance.

- The cost analysis shows this catalyst is economically viable, but the results do not definitively suggest the performance will ever be high enough.

Question 5: Proposed future work

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

- A strength of this proposal is its careful and systematic approach. It seems clear this clear approach will be applied to the next steps of scale-up and MEA integration.
- The team has proposed reasonable future work for the remaining project period. It would be good if the team performs durability studies of the most promising non-PGM catalyst developed in this project.
- The team has laid out the important future work to be addressed. However, the durability issue on air requires further attention.
- It would be useful to see more information on how the project team plans to improve Fe-free catalyst activity, improve open circuit voltage, and decrease peroxide generation. It would be useful to see more reporting on the model validation. The model was generated to fit data, but this likely involved some parameter adjustments. The question then remains whether the model can accept a nanoscale computed tomography image of a GM or IRD catalyst layer, and then still predict the resulting performance. Active site determination is mentioned in the future work. However, there should also be emphasis on determining the oxygen reduction mechanism. A number of the future work items are reasonable: improvement of Fe-free catalyst activity, determination of active sites, nanoscale computed tomography on new catalyst layers, and further work to study degradation.
- The research team needs to understand the impact of catalyst layer ionomer content and membrane thickness or the use of state-of-the-art membrane on the H_2/air performance.
- There was not a great deal of details provided, so this is more difficult to assess. The team should provide fewer proposed work items and describe them in greater detail.
- There was not a clear path in the proposed work toward developing non-PGM materials for real-world applications.

Project strengths:

- The lead PI is a world-leading expert in this field, and there is a real opportunity to make a contribution. The unique synthesis capabilities of Los Alamos National Laboratory (LANL) and the ability of the other partners to characterize materials under realistic conditions are impressive. In particular, the use of spectroscopy and transmission electron microscopy (TEM) imaging is incredibly important because the combination of these techniques is really the only way to define the materials' properties and limitations, and ultimately to determine what contribution non-PGM materials will make to the fuel cells community.
- Led by LANL, the project has access to considerable collaboration and analytical capability. The project is led by investigators with extensive history in non-PGM oxygen reduction catalysis. The project has tasks that address most of the barriers to non-PGM catalysis, including mass transport and proton conduction resistances, lack of active sites or active site density, and the need for developing Fe-free catalysts. The project has incorporated collaborators that include a supplier, a developer, and a university with a unique ability to image catalyst layers.
- The project features a large, multi-talented team that is addressing the work from various angles—density functional theory (DFT) and electrode modeling, novel characterization methods, detailed analyses, understanding of active sites, and industrial partners.
- The PI has very good collaboration with national laboratories, universities, and industrial partners. The team has a thorough understanding of the problems associated with the non-PGM catalyst development.
- This project integrates efforts from several different partners effectively. There appears to be close, open collaboration between the partners.
- The creative and capable team is a strength of this project.

Project weaknesses:

- LANL needs to identify a strategy to understand the oxygen reduction mechanism for all catalysts involved: Fe-based, FeCo-based, and Co-based. The generation of hydrogen peroxide is still too high for most catalysts. High current density is still an issue, and it may be difficult to address even with improvements in the catalyst layer. Power density is still only near 0.5 W/cm^2, at best. There appears to be an acknowledgement that durability of the Fe-N_x site is a problem, but it is difficult to see whether there is a strategy to address this issue.
- LANL should more clearly articulate progress toward DOE targets. There was reference to the original DOE targets as well as to updated targets based on the CWG. It was not always clear in the presentation which targets were being used as a reference point. A summary table of these targets versus progress/ milestones would be useful in the future.
- As is, there is no true relation between the number of active sites, the nature of these sites, and the stability and reactivity of these sites. Furthermore, there is no clear direction toward other classes of materials if the materials currently being studied do not work.
- It would have been better if the team had presented some durability results for the CM-PANI-Co-C and CM-PANI-Fe-C catalysts.
- The inconsistency in the data and conditions was an area of weakness. The scope may be too broad for the team to effectively accomplish everything.
- The path to scale up these catalysts to 100 g batch sizes is unclear.

Recommendations for additions/deletions to project scope:

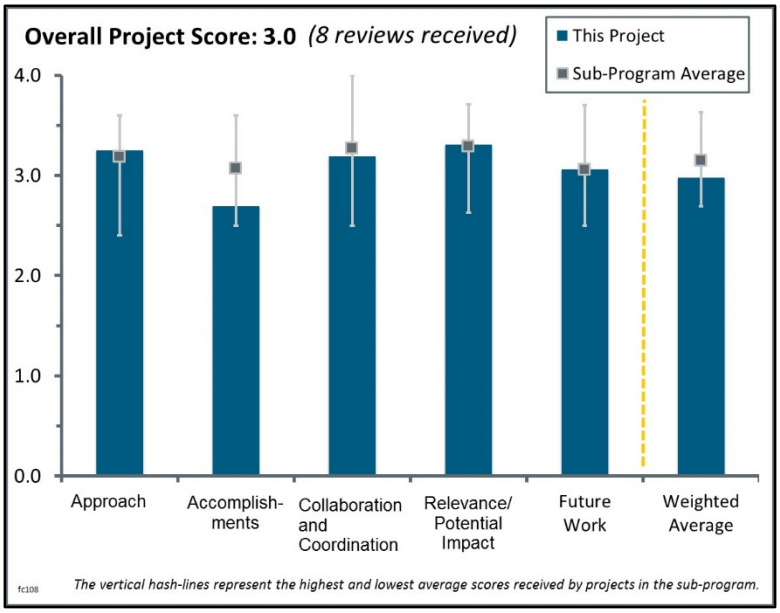
- The project should place greater emphasis on the mechanism for both oxygen reduction and the degradation of the Fe-N_x active site. It would be a problem if the project overachieved in developing the very best catalyst layer for a catalyst that was not active, that was not durable, and that contained a membrane-degradation catalyst (Fe). LANL should add a plan to improve Fe-free catalyst activity.
- The team could use the models to run a scenario analysis and determine what an ideal CL would look like, and what optimum performance could be achieved with current catalyst parameters. Further, it is unclear how this would vary with catalyst activity and active site concentration, within expected achievable parameters. This would provide an assessment of possible maximum performance achievable.
- Future work should include a discussion about the future of these materials. The science itself is absolutely fascinating, and the cost is so low that it is difficult to resist going in this direction, but the question is how fast one can get there.
- It would be interesting to see the correlation between the amount of nitrogen in the catalyst and the catalyst activity/durability.

Project # FC-108: Advanced Ionomers and Membrane Electrode Assemblies for Alkaline Membrane Fuel Cells

Bryan Pivovar; National Renewable Energy Laboratory

Brief Summary of Project:

The goals of this project are to synthesize novel perfluoro (PF) anion exchange membranes (AEMs) with high temperature stability and high water permeability and to employ those high-performance PF AEM materials in electrodes and as membranes in alkaline membrane fuel cells (AMFCs).



Question 1: Approach to performing the work

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

- The approach of pursuing PF AEMs is also a good choice to maximize the possibility of developing AEMs that ultimately have the desired properties. The emphasis on fundamental understanding of the chemistry and the composition and properties of the resulting membranes is also excellent.
- The approach for this project is well founded and seems to be leading to excellent results.
- The approach is interesting and worth exploring. The reasoning—that the morphology generated through the acidity of perfluorosulfonic acid (PFSA), which has been found beneficial, should also prove beneficial in AEMs—is solid. The synthetic pathways chosen by the researchers were straightforward and reasonable.
- The approach to utilize PF polymer electrolytes has merit because the PF electrolytes are chemically robust, and from polymer electrolyte membrane (PEM) ionomer experience, the PF polymer backbone appears to aid in obtaining phase segregation and an appropriate microstructure for ion conduction. Plans to attach the cation through an electron-rich segment such as an aryl group or aryl amide group should reduce the electron withdrawing effect of the PF polymer. The cation groups being employed have decent stability, but they are not the most stable.
- The great majority of AEMs are based on hydrocarbon polymers. The approach of this project consists of developing AEMs with a PF backbone. The PF backbone, while electron withdrawing, should provide higher stability to the final AEM. The challenge here is to synthesize membranes with chemically stable cations and—at the same time—reasonable OH- conductivity.
- The overall approach to this project is good; it is based on the principle of functionalizing a perfluorinated polymer backbone with quaternary ammonium group and a non-alkyl spacer to prevent the Hoffman degradation reaction. While the chemistry is sound for making materials, the project is struggling with making membranes with practical hydroxide ion conductivity. The conductivities are currently more than an order of magnitude too low for practical use, and there is no clear path forward for improving these conductivities. The perfluorinated polymer backbone approach has proven to improve the water transport properties of the membrane, which was one of the stated goals of this project.
- The presentation clearly highlighted the status of the work and the challenges involved. This is indeed a challenging effort, and the scope of the work may have been too ambitious from the start. Future efforts should separate the synthetic chemistry work from the fuel cell stack testing work (which is where the delay is occurring). There were three distinct areas of work mentioned, and all three could readily constitute stand-alone tasks, given the complex nature of the problems: (1) synthesis of polymers/ionomers for alkaline membrane electrode assemblies (MEAs), (2) stack-level testing, and (3) modeling of carbonate formation. This task should really only be focused on item 1, and it is recommended the U.S. Department

of Energy (DOE) Fuel Cell Technologies Office (FCTO) solicit separate proposals to cover areas 2 and 3 in the future. With regard to item 1, the project is taking the correct approach.
- A coherent, focused approach in this project is hard to discern given the several different synthesis approaches which have shown mixed success.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The accomplishments and progress appear to be impressive and seemed well received with the audience.
- Progress for the amide linkage chemistry has been slowed by the choice to initially characterize the chloride form, which did not allow the researchers to see the formation of the much less conducting zwitterion in base. Adding an additional reaction step may allow the researchers to bypass this by methylating the sulfonamide link under basic conditions. Work with small molecules suggests this should fully methylate the sulfonamide position; however, reactions with the polymer suggest incomplete reaction. The alternate synthetic strategy to utilize Grignard reagents to attach the cationic moeity has not been any more productive. Researchers have encountered difficulties finding a suitable solvent that will dissolve, or at a minimum swell, the polymer enough and also be compatible with the Grignard reagent. Semi-fluorinated hydrofluoro ether (HFE) fluids appear to have some promise, and small molecule Grignard reactions work in these solvents. The researchers do not yet have results with the polymer and molecules of interest. The synthetic approaches have been unsuccessful, so MEA preparation, testing, and characterization have not been occurring. The project is behind schedule and will likely need to extend beyond the scheduled end date to prepare and test the AEM in MEAs.
- Significant progress has been made in all fields of work. While the progress toward the development of a PF AEM polymer seems to be satisfactory, bringing these polymers into a membrane, and consequently into a non-platinum-group metal (PGM) MEA that shows high fuel cell performance (>350 mW/cm^2), remains a huge challenge. The reported fuel cell performances for the synthesized MEAs, compared to the CellEra baseline, are still too far from the proposed objectives.
- Much work has been done, and several approaches have been tried, with less-than-complete success. No chemistry to date seems to make the grade, although slide 4 claims victory through 12/31/14. Future milestones are TBD. The team is learning a lot, although the results are disappointing. It would be helpful for the team to provide a summary list of accomplishments.
- The group has unfortunately suffered a lot of setbacks at the synthesis step; this is not terribly surprising because these perfluorinated polymers are very difficult to work with in terms of reactivity and solvent compatibility. The lead's strength, just by resume and reputation, is really in MEA development and characterization, and not as much in polymer synthesis; however, the researchers do have collaborations with very strong synthetic polymer chemists. Their determination to keep finding new pathways is to their credit. In the final analysis, however, there is very little here that can be used either in terms of actually leading to a commercially viable product or deeper insight into AEMs.
- The team has completed 60% of the project work, with 85% of the scheduled project duration complete. This is due to the team setting overly ambitious project goals initially. If the project were to receive a no-cost extension, it may be able to fulfill the stack test work.
- There was very little progress toward reaching the project goals this year. While the fiscal year 2015 milestones have apparently been met, the milestones should have included a hydroxide ion conductivity target rather than, or in addition to, the water permeability target. The project is still struggling with making membranes, and the demonstrated conductivities are too low for these membranes to be practical. In addition, the fuel cell performance, only shown in the backup slides, is very poor compared to the commercial hydrocarbon membranes.
- It is somewhat disappointing that the project is not further along than it is at this point (e.g., cell testing), but it is clear some critical things are being learned that should enable faster progress in the future. The addition of cell modeling work is greatly appreciated, especially in the current absence of cell data. The use of some literature data on carbonate equilibrium to validate the model is also good. However, cell data will be required to validate the complete cell model.

Question 3: Collaboration and coordination with other institutions

This project was rated **3.2** for its collaboration and coordination.

- The principal investigator (PI) obviously has an excellent working relationship with his collaborators, and 3M's contributions to the project are particularly worthwhile because it appears these contributions have enabled the team to make significantly larger and more uniform membranes. The addition of the modeling work is also valuable.
- A fantastic team has been assembled here. 3M and the Colorado School of Mines (CSM) have the best synthesis experts in this area, and CellEra will likely be first to market with these materials.
- The collaboration on this project is excellent and includes a very good mix of academia, national laboratories, and industry.
- The project seems to have excellent collaboration between the different institutions and is well coordinated.
- The team is well rounded and competent, and there are established working relationships.
- The collaboration between 3M, CSM, and the National Renewable Energy Laboratory (NREL) appears to be working OK. The inclusion of CellEra is a positive.
- While NREL has assembled an outstanding, highly qualified team for this project, it is unclear what all the project partners have contributed to the project. While the PI did call this comment out in his "response to reviewers' comments" and said it was addressed through a more detailed description of the team members' roles, the detailed description still showed overlap between the organizations' roles and no clear indication of what project progress is attributable to what organization. This could have been resolved by identifying the source of the data and materials on each accomplishment slide.
- The project received good support from 3M, which is positive. Strong vendor interactions will be required for project success. CellEra could not yet be engaged in a meaningful way because of the synthetic challenges. In the future, the project team should engage additional partners that have strong synthetic organic backgrounds to assist with the CSM effort.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.3** for its relevance/potential impact.

- The development of AEMs has a large potential impact because they would be an enabling technology for alkaline fuel cells that could effectively utilize inexpensive non-PGM cathode catalysts. These fuel cells could have large cost savings compared to current PEM technology. The proposed work attempts to address performance issues with current AEMs.
- This is a good example of the type of high-risk, but potentially high-reward, research that should be included in DOE's portfolio. It is unlikely that AEMs will receive much industry interest/investment unless some of the major risks have been reduced. Yet, if successful, AEM fuel cells could be game-changing, primarily because they offer the potential for lower catalyst costs.
- The project is targeting the main challenges in the development of fuel cells—elimination of the expensive PGM catalysts currently needed in PEM fuel cells..
- AMFCs continue to be an important technical challenge DOE should be tackling as a parallel path to reduce fuel cell operating costs.
- Development of AMFCs aligns with the FCTO's mission.
- AEMs still have a number of challenges, but this project is addressing the biggest issues.
- The consensus seems to be that a new AEM is important, but it is too early to determine whether this is the right approach.
- With only a few months left in the project, there is quite a bit to do. The project needs to overcome many hurdles to demonstrate the polymer materials developed in this project will have any impact on the performance or durability of AEM fuel cells.

Question 5: Proposed future work

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

- The proposed future work seems in line with the objectives and has the potential for great impact.
- The proposed plan appears to be appropriate, considering the current status of the project.
- The proposed future work is logical and appropriate to pursue, although it is not certain this work will solve the problems encountered with the polymer chemistry, especially for the Grignard approach. It may be necessary to start with lower equivalent weight (EW) material and then do crosslinking, or to try other synthetic strategies. Membrane characterization should be performed in the hydroxide form before MEA fabrication and testing are performed.
- According to slide 22, the project has to be focused on improved polymer synthesis as the critical enabling element toward AEMs in energy applications. It is, however, important to identify, specify, and justify the different directions to avoid possible trial/error approaches. At this stage, it looks like that by avoiding one problem (i.e., zwitterionic amide linked polymers), other problems will appear (e.g.., piperazine being a potential crosslinking agent and hygroscopic).
- The future work is perfectly appropriate; it would just be better if the researchers were further along in the project. The use of new functionalities and the chemistries to achieve those functionalities are exactly what the researchers should be investigating.
- The planned future work is rational. Completion in the remaining time seems unlikely.
- The prospects for success of the future work have been dimmed somewhat by the synthetic challenges—MEA preparation and testing may not be possible. The team should drop the CO_2 modeling effort because it is a distraction. CO_2 modeling should be funded as a separate effort next year.
- The proposed future work is a good portion of the entire project's scope, and there are only a few months left in the project.

Project strengths:

- Strengths include the following: (1) the project team's expertise in different polymer synthesis, (2) the project team's expertise in membrane fabrication, (3) the project team's ability to identify the problems associated with the loss in performance (fuel cells), (4) that partners are full participants and well coordinated, and (5) that partners are aware of the high-risk/high-return spectrum when working with AEMs.
- The approaches toward functionalization of a perfluorinated polymer backbone are strong. The project features a potentially strong team, although it is not apparent in what way the team members are contributing to this project.
- The project features a very strong team with good collaborations. There is a sound rationale to the approach, and the project is on a path that should be explored.
- The team's focus on trying to fully understand the synthesis and the resulting membranes is an area of strength.
- The partnership with 3M brings particular strength to this project.
- The team and its creativity are an area of strength.
- The diverse project team seems very strong.

Project weaknesses:

- There is a lack of progress and lack of foresight on how membranes would be fabricated from these materials. There is too much ground to cover in the remaining few months of this project.
- The project has been hampered by the researchers' inability to get the desired reactions to go to completion with the polymer systems chosen.
- The project has experienced difficulties with regard to identifying the problems before assembling the polymers into an MEA configuration.
- The project is too ambitious and should be focused solely on the synthetic challenges.
- Progress on the synthesis is very slow.
- To date, there has been a lack of relevant cell data.

Recommendations for additions/deletions to project scope:

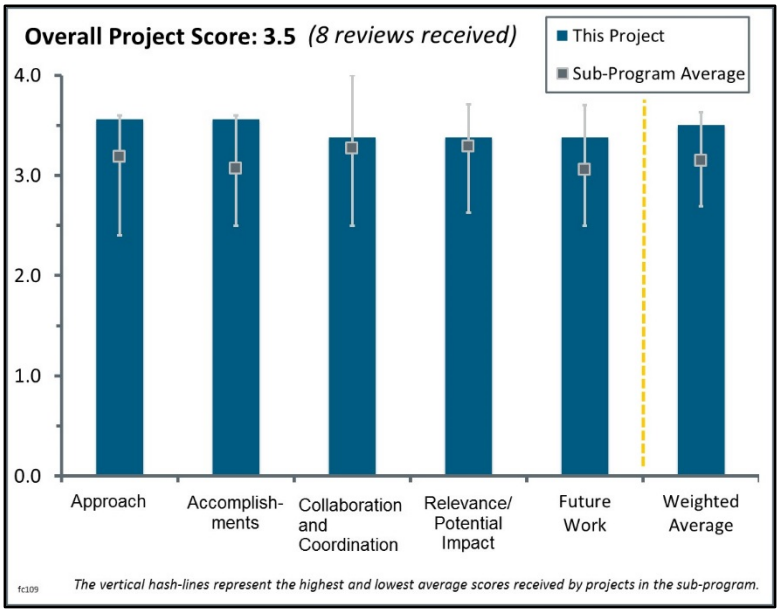
- The cell model should include the impact on ionic conductivity due to the equilibrated concentration of carbonate species (e.g., as shown on slide 17) in both the membrane and the ionomer in the electrodes. In addition, the cell model should include the possibility of free ions in the liquid phases of the cell that are therefore subject to migration and diffusion during cell operation. The presence of concentration gradients of carbonate species may be an issue and may result in localized precipitation of carbonates under certain conditions. If the researchers think the presence of free ions is unlikely, they should demonstrate (in beaker tests, at a minimum) that water in equilibrium with these membranes remains neutral.
- Currently, 3M is providing starting material, but the project may want to find someone who can conduct the synthesis from the start or provide a polymer that is easier to modify. This is likely out of 3M's scope or interest, however, and it is almost impossible to find someone to do tetrafluoroethylene (TFE)-related synthesis.
- The challenges associated with the fabrication of stable ionomers to be used in the catalyst layers have to be considered. Membrane characterizations have to be more focused to its hydroxyl form.
- The carbonate modeling and MEA preparation/testing should be moved to separate tasks.

Project # FC-109: New Fuel Cell Membranes with Improved Durability and Performance

Michael Yandrasits; 3M

Brief Summary of Project:

The overall objective of this project is to meet all of the U.S. Department of Energy (DOE) Fuel Cell Technologies Office (FCTO) Multi-Year Research, Development, and Demonstration Plan membrane targets for performance, durability, and cost simultaneously with a single membrane. Tasks include ionomer development, nanofiber development, ionomer and membrane testing, membrane electrode assembly (MEA) fabrication and fuel cell testing, and stack testing.



Question 1: Approach to performing the work

This project was rated **3.6** for its approach.

- The work is aligned with DOE goals and addresses the barriers of cost and durability. This project addresses DOE membrane targets and is designed to meet all DOE membrane targets simultaneously with a single membrane. The project combines and attempts to improve upon previously effective approaches to reduce swelling and improve performance. The project takes a very systematic approach to developing membranes by looking at how variables such as the number of acid groups per side chain and fiber fraction affect conductivity and swelling.
- 3M is pursuing perfluorinated polyimide acids (PFIAs) as membrane electrolyte ionomers to improve the performance and durability of polymer electrolyte fuel cells. This approach is promising and has the potential to overcome performance and durability issues faced by conventional perfluorsulfonic acid (PFSA) ionomers. The issues with swelling of these low equivalent weight (EW) materials are being addressed by fiber reinforcement, either through traditional incorporation of a weave of an inert support or by electrospinning fibers of the PFIA and support polymers.
- This project is following a well-thought-out plan of synthesis, membrane preparation, membrane testing, MEA preparation, and MEA testing that thoroughly tests out a promising direction of membrane chemistry. The project investigates both new base ionomers for the membrane and new forms of membrane reinforcement in a drive to satisfy all DOE goals simultaneously.
- Up to this point, the approach to achieving the milestones appears solid. The efforts of the different institutions appear well integrated.
- The use of fibers to improve membrane properties is excellent, and results show partial success at this time. The ionomer should really be studied in parallel because it is the key component of the catalyst layer. There are questions about whether a soluble ionomer is available and how it performs compared to Nafion. This should be an essential part of the study, but it appears the principal investigator wants to obtain separate funding to address this issue in the future.
- This project seeks to develop new PEMs perfluoro ionene chain extended (PFICE) with improved conductivity and mechanical properties. Mixed fiber mats are a novel approach—they significantly reduce membrane in-plane swelling while retaining good conductivity (at 80 vol% PFSA). The membranes have been tested for a series of properties, including ion conductivity, swell and water solubility, and mechanical properties. It would be great if some membrane transport properties (e.g., water uptake, diffusivity, and gas permeability) were shown.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The 3M-led team has shown excellent progress this year in increasing membrane conductivity at low relative humidity (RH) (i.e., a fivefold increase in conductivity achieved versus state-of-the-art PFSA at 50% RH). The 3M PFIA membranes now have conductivity exceeding 0.1 Siemens (S)/cm at 50% RH, which is a key achievement toward improving fuel cell performance at low RH. 3M has demonstrated that the PFIA membranes have very low solubility in water—an important consideration when reducing membrane EW. The PFIA membranes do show considerable swelling, which needs to be addressed. 3M is planning on addressing this issue by utilizing nanofibers of PFIA interwoven/fused with inert or PFSA polymers.
- The project has achieved significant accomplishments. Two major breakthroughs were made. For the PFICE-4 membrane, a conductivity of 0.1 S/cm was achieved at 80°C and 40% RH, which is a fivefold improvement. For the dual-fiber PFSA/polyamide-imide (PAI) membrane, membrane lateral swelling was reduced to 5% as ionomer content dropped to 80 vol%, while decent conductivity of 0.08 S/cm (at 100% RH) was maintained. Good correlations—membrane water solubility versus EW and membrane linear swell versus EW—have been generated. The swell predication model allows for evaluating nanofiber materials easily and effectively. Pressure ramping to burst mode is well applied to evaluate membrane failure.
- 3M has made a PFIA membrane that showed performance in a single cell better than its supported 750 EW 3M membrane at 95°C and 26% RH (inlet) and could achieve greater than 0.5 V at 1.5 A/cm^2 at 95°C and 50% RH. This same membrane met the open circuit voltage (OCV) hold target and mechanical stability (RH cycling) target. 3M improved conductivity by approximately a factor of five compared to NR112. This effectively moves the RH at which one gets a conductivity of 0.1 S/cm from 80% RH to 40% RH at 80°C. In the past year, development of PFICE-4 membranes has progressed, and the researchers have achieved a conductivity of >0.1 S/cm at 80°C and 40% RH. The researchers have developed a model that can predict membrane swelling based on the fiber modulus and fiber fraction. They have identified two paths to a membrane that should meet the targets. Data and testing at high temperatures (120°–95°C) are lacking. The DOE target table targets a maximum operating temperature of 120°C. It is not clear how the dual-fiber electrospun membranes compare to other nanofiber-supported membranes, and whether there is an advantage to the dual-fiber electrospinning approach.
- This project has made good progress on synthesizing advanced ionomers, forming them into membranes, and testing the chemical and physical properties of the membranes. Fabrication of 50 cm^2 MEAs and testing of performance and durability are about to start. The project has achieved ionomers of low EW that are also insoluble in water—an improvement over PFSA. It is not clear that currently available (2015) membranes are unable to meet fuel cell requirements, so this area of research need not have the same priority as other, more pressing issues in fuel cell development. It is good to still have two paths available toward meeting Milestone 8.
- Good progress has been achieved over the past year, and development appears to be on track.
- The progress to date appears to be excellent. Ionomer development has progressed, with a number of samples showing promising performance. Data on the OCV hold test indicates that the PFIA membrane passed the test; however, this conclusion is questionable. The OCV for the PFIA material decreased by ~100 mV within <100 hours, and is then compared against other material where the OCV is stable at ~0.93 V. Comparing durability at OCV of 0.82 versus OCV of 0.93 is not a fair comparison. The researchers failed to show crossover data, and the OCV is only a portion of the story. This evaluation needs to be more fairly presented and made; it is possible that the PFIA membrane really failed this test within 50 hours, because the OCV decreases by >100 mV.
- Milestones have been met, albeit with different materials used to meet different milestones.
- Success has been shown in obtaining a membrane that performs better than Nafion at low RH. It is not clear why conductivity measurements have not been shown for 100% RH and liquid water.

Question 3: Collaboration and coordination with other institutions

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

- The project has a great team working on membrane synthesis and characterization. 3M has been well recognized for developing fuel cell membranes. General Motors (GM) is very good at membrane characterizations and tests. Vanderbilt has great expertise on electron-spun nanofiber fabrication.
- The collaborations with GM and Vanderbilt appear to be going well. The electrospinning work is progressing, and testing at GM has been progressing and is valuable to the project.
- The collaborations appear well utilized. The expertise of the different institutions is leveraged well, with no apparent hindrance to synergistic goals.
- The project features effective collaboration between the partners, although the true test of the strength of the interactions will be shown by the level of thoroughness of the upcoming testing in stacks. Project leaders have been helpful with advice and materials to help keep other DOE-funded membrane projects moving forward.
- The interactions with both GM and Vanderbilt seem productive.
- The project features strong contributions from collaborators.
- 3M has assembled a highly qualified team to address the issues. While Vanderbilt's role is clearly defined and necessary as an alternative approach to forming supported membranes to reduce issues with swelling, GM's unique role is a bit less clear—it seems 3M can and does perform many, if not all, of the measurements that GM is performing. GM's role, beyond testing, appears to be in additive development, but this was not covered this fiscal year.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.4** for its relevance/potential impact.

- The project is very relevant because it is aligned with DOE goals and would have a large impact on system cost. A membrane that can meet DOE targets and operate at low RH and higher temperatures would be extremely beneficial for the FCTO because it would allow one to eliminate the membrane humidifier and reduce the size of the coolant system, leading to substantial cost reductions in the balance of plant.
- This approach is promising and has the potential to overcome performance and durability issues faced by conventional PFSA polymers, especially the sensitivity of the conductivity of conventional PFSA to low RH.
- The ability to operate at higher temperatures and lower RHs is a substantial benefit to fuel cell system developers because it increases the operating envelope, facilitates heat removal, simplifies control, and decreases the size of the humidifiers. This project can positively impact the viability of commercial fuel cells.
- Higher-conductivity, more stable membranes are useful. A benefit analysis of these materials versus the current state of the art would be useful; the project appears to address the membrane conductivity, but it is hard to know whether there is a cost-benefit.
- Although current membranes are satisfactory, having a membrane that works at low RH is a good idea. Operating a PEM fuel cell at higher temperatures is not a good idea because the catalyst layer will fall apart and degrade at higher temperatures, even if the membrane survives.
- The potential impact is high, given that the project is on track to develop membranes that meet the 2020 DOE milestones. However, the use of different materials to satisfy the stability versus performance metrics is a potential shortcoming. The true validation of the potential impact will not be clear until Milestones 10–12, when the membrane will be evaluated in an operating fuel cell.
- Membranes with improved conductivity and acceptable mechanical properties could improve fuel cell performance. This project helps keep one of the few laboratories that can synthesize perfluoro ionomers active in the development of advanced membranes at a time when other formerly active enterprises are either inactive or getting out of the business. Membranes have improved so much in the past decade that it

is not clear that further membrane development is essential. This project seems unlikely to significantly reduce membrane costs.
- The improved conductivity at low RH (40%) and improved mechanical property are significant accomplishments. The objective of this project is to meet membrane performance, durability, and cost targets simultaneously in a single membrane. The cost of nanofibers is a concern. In addition, the research is focused on 95°C and 50% RH. It should be stated why this condition is highlighted; for standard conditions (80°C and 100% RH), there are many good existing membrane candidates.

Question 5: Proposed future work

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

- The planned future work addresses the appropriate barriers and is focused on meeting DOE performance and durability targets.
- The future plans seem well thought-out and likely to yield positive results, assuming all partners remain willing to carry out their planned roles.
- Efforts to refine membrane design and conduct stack testing are on track and logical progressions for the project.
- The 3M-led team has an excellent grasp of the remaining issues and how to address them. The development of stabilizing additives and how this is incorporated in the future of the project is not clear.
- The future work plan is detailed and reasonable. However, it should include membrane chemical stability testing and membrane transport properties (e.g., water uptake, diffusivity, and gas permeability); the latter would impact MEA performance at high current density.
- It would be good to see more MEA test data by the next DOE Hydrogen and Fuel Cells Program Annual Merit Review (although data may be limited during the second quarter in 2016). MEA testing should not be limited to stack testing but occur on single cells, and thus not be limited to material scale-up. It is unclear whether the nanofiber development is going to be used or not. If it will not be used in the membrane materials, it is unclear why it should continue.
- The future work should include characterization of the ionomer and its use in the cathode catalyst layer. The impact of the permeability of oxygen and hydrogen on the performance can be clearly understood by using these materials in the catalyst layer.
- It is unclear how it will be determined whether the PFIA- or PFICE-based membrane will be selected for pursuing Milestones 9 and 10. The former exhibits desirable mechanical properties, but the latter exhibits the greatest potential for high conductivity. There is not a clear path forward for how to obtain both of these salient metrics in a single membrane. It is also unclear what the path forward is for improving the mechanical properties of the nanofibers while retaining high conductivity.

Project strengths:

- The project has a high-quality team with the capability to do manufacturing and good knowledge of the current membrane state of the art and requirements in real-world applications. The concepts for improving membrane conductivity and limiting swelling have been proven. The plan to make and test a commercially relevant membrane, which includes the improved ionomer, an improved support, and a chemical stabilization package, is a strength.
- This project provides a highly reasoned development pathway to improve membrane performance while maintaining or improving mechanical properties. This project keeps one of the few laboratories that can still handle the synthesis of perfluoropolymers active in the development of improved membranes, while most of the rest of this industry is moribund.
- For the PFICE-4 membrane, a conductivity of 0.1 S/cm was achieved at 80°C and 40% RH, which was a fivefold improvement. For the dual-fiber PFSA/PAI membrane, membrane lateral swelling was reduced to 5% as ionomer content dropped to 80 vol%, while decent conductivity of 0.08 S/cm (at 100 % RH) was maintained. Comprehensive mechanical tests of the membrane have been performed. Having a great team with complementary experience will ensure the project makes good progress. The combination of experimental data with modeling is a plus.

- The project technology appears to have a clear, tangible benefit for fuel cells that is a distinct improvement over current technologies and can readily be incorporated into existing platforms.
- Material synthesis and testing efforts are progressing well.
- The project features an excellent approach and execution.

Project weaknesses:

- The difference between the mechanical properties in the machine and transverse directions has not yet been overcome, and no clear ideas on how to deal with this appear to have been given in the presentation. 3M management's blanket policy of not discussing the manufacturing costs of present or potential products decreases the value of this project in guiding DOE's development efforts and could lead to inefficient use of society's resources if technically promising developments were to turn out to have costs that are too high.
- Transport properties such as water uptake, diffusivity, and gas permeability are desirable.
- There are questions about the OCV stability of the membrane or the MEA, and why the OCV decay was relatively rapid down to ~0.82 V.
- A weakness is the lack of study of the ionomer by itself and in the catalyst layer.

Recommendations for additions/deletions to project scope:

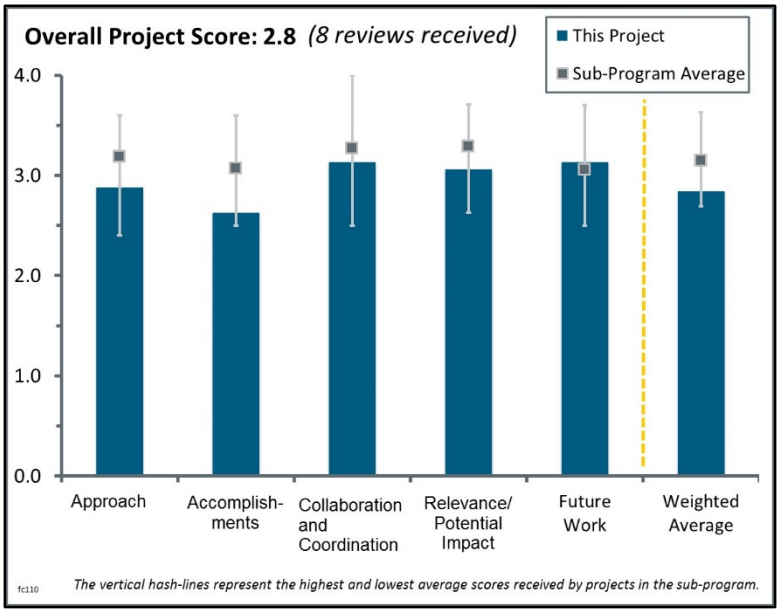
- There are no recommendations for additions/deletions to the project scope—the current scope is very appropriate.
- It would be good to see more single-cell MEA testing. The chemical stability needs to be reevaluated and compared more directly against other current membranes. While it is recognized the researchers used the standard OCV test, comparing one membrane's durability against another with OCVs that differ by ~100 mV is not a fair comparison. The researchers need to present gas crossover data, not just OCV data. Efforts to understand why the OCV decayed so rapidly (within 50 hours) should be added to the project scope if the cause is not just gas crossover. If the rapid decay is just due to increased gas crossover, then this membrane really failed Milestone 4, and the chemical stability needs improvement.
- Although MEA development and testing are tasks within the project, it is not clear whether the catalyst layers are conventional or 3M. If possible, the effort should be extended to ascertain the effect of using imide ionomers in conventional catalyst layers because there could be potential performance benefits, especially at low RH.
- The team should study the ionomer's properties independently and the ionomer's application in the catalyst layer.

Project # FC-110: Advanced Hybrid Membranes for Next-Generation Polymer Electrolyte Membrane Fuel Cell Automotive Applications

Andrew Herring; Colorado School of Mines

Brief Summary of Project:

The overall objective of this project is to demonstrate a low-cost hybrid inorganic/polymer from super-acidic inorganic functionalized monomers with (1) area specific resistance (ASR) <0.02 Ω cm^2 at operating temperature of an automotive fuel cell stack (95–120°C) at low inlet relative humidity (RH) <50%, and (2) a 50 cm^2 membrane electrode assembly (MEA) with desired mechanical properties and durability. The current-year objective is to optimize the two best candidate systems for low ASR, mechanical properties, oxidative stability/durability, and incorporation of electrodes, ultimately eliminating the lowest-performing system.



Question 1: Approach to performing the work

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

- This project follows one of the few really novel approaches to improved membranes and has finally generated a material that should be testable in fuel cells (heteropoly acid [HPA]-FC-2178).
- The multi-directional approaches taken by the team for the completion of all tasks are adequate. All the analytical techniques had been thought through appropriately. The study on materials synthesis based on functionalized super-acidic inorganic moieties is the reasonable approach. However, stabilization of HPA in a polymer matrix seems to be a great challenge.
- The membrane looks promising, and much progress has been made in incorporating a film into a fuel cell. As with many new membranes, materials durability in an operating fuel cell has been a big challenge in the past. Thus it is recommended that an approach be developed and demonstrated that gives the U.S. Department of Energy some indication of membrane material lifetime—in particular, the chemical stability in an operating fuel cell, even if fuel cell performance is low.
- Using HPA functionalized membrane materials is an approach that is not being utilized with other developers, and it could potentially provide lower-cost, performing high-temperature membranes. Regarding the limited samples made to date that are free-standing—primarily thin films on other substrates such as Kapton—it appears that these materials are going to require reinforcement for mechanical strength. There appear to be some fabrication issues that have not been addressed to date and that do not appear in project plans. Regarding the MEA, catalyst layer integration is going to be required, and there are limited plans.
- A well-integrated approach was proposed for this difficult problem that balances risk of failure and beneficial outcomes with three different membrane chemistries. Water transport properties (water diffusion coefficient, electroosmotic drag) will not be measured, but these are likely important to understand membrane conductivity in a low-RH environment.
- The approach taken by the principal investigator (PI) this year is described as the plan would indicate, and it seems that System II with the use of Dyneon has shown itself to be interesting. However, characterization data of the membranes were insufficient to judge whether other barriers such as permeation and durability can also be achieved by the same ionomer.

- The project is addressing performance first, which is a reasonable approach as long as sufficient work has been done to confirm a potential path to meeting durability and cost, but it is not completely clear whether this has been done. Specific details of risks and mitigations are not all clearly addressed, and the approach should be more systematic. There is not enough analysis of relating properties of currently achieved systems with targets, nor is there enough subsequent development of correlations to allow tuning of properties to simultaneously achieve multiple targets. The oxygen and hydrogen permeability data relevance for the low-HPA membrane is not clearly explained. How this will relate to the increased HPA system was not clear, and therefore the permeability data were either irrelevant, or worse, misleading, if these low permeabilities cannot be maintained.
- Professor Herring has shown in his past projects that incorporation of HPAs into polymer electrolyte membranes (PEMs) is a promising approach to meeting fuel cell performance requirements in dry conditions. Unfortunately, nearly two years into the project, there is no conductivity of fuel cell performance data at conditions drier than 95% RH. Professor Herring says that he is at the stage of the project at which he can "tune" the membrane to meet performance and mechanical properties, but no detailed approach was provided for how the mechanical durability requirements will be met.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The project is on track. The membrane picture (slide 12) is, unfortunately, misleading and does not reflect the team capabilities. Clear films without black areas were produced with good thickness control over the film area (doctor blade). Circular features reflect the Teflon frame used to handle the film. A patent was filed for the third-generation FC-2178 chemistry.
- This group's persistence and synthetic skill finally seem to be paying off in a novel material ready to be tested in fuel cells. The new process for functionalizing FC-2178 seems now to provide a pathway for testing the HPA-derivatized membrane concept in fuel cells. Progress in the calculational-chemistry-inspired shift to a low-viscosity but high-temperature solvent for the trifluoroethyl vinyl ether (TFVE) reaction mechanism is encouraging. It is highly disappointing that even with the improved preparation method, the HPA/TFVE is still too brittle to be formed into films to be tested.
- It is good that the team has settled on a system, hybrid HPA-3M FC-2178, to move forward to try to meet the targets. It is a bit disconcerting that after nearly two years, the project is still struggling to make films that are good enough to collect fuel cell data. Last year, the team was asked to collect conductivity data at lower RH, but that has still not been done. Also, no mechanical or swelling data were shown. Professor Herring did say that swelling was not a concern, but it would be nice to see the data. Durability has not been explicitly addressed. It would also be good to see some sort of cost assessment now that the project has narrowed the research down to one approach.
- There is insufficient detail to estimate the probability of meeting all project targets. The membrane systems have improved significantly and are meeting or exceeding some of the targets at some conditions. However, the conductivity at lower temperature and for drier conditions has not been established. There is also insufficient information on swelling, mechanical properties, cost, and durability. It was claimed that significant progress was made in process development: higher yields, good control, much-improved materials, overcoming solvent isolation after every step, and solutions to other criticisms of the old method of manufacturing. Films of sufficient surface area and uniformity are available for partner testing. However, no details were provided, as this is considered proprietary information, and team members are applying for patents.
- Good progress has been made, but some key points still preclude a complete solution to the problem. It is not clear how DOE is to know that this membrane approach has the potential to bring membrane costs down to $\$20/m^2$. It is also unclear how this membrane is better than other membranes in terms of conductivity; the chart on slide 11 shows 0.2 Siemens/cm at 60°C, which is similar to Nafion. Simply making the membrane thinner to meet the ASR target is not suggestive of an improved material.

- On the positive (pros) side, the casted films on Kapton based on the FC-2178 material seem to show promise in attaining the DOE goal of ASR <0.02 Ω cm^2. Here are the cons:
    - Permeation data were not normalized to thickness; therefore, one cannot tell whether this was achieved by the same membrane that achieved the low ASR. When asked about the thickness of the membrane for which the crossover data were measured, the PI could not answer.
    - The PI claims that the "thin films with target ASRs at low RH [are] now system ready" (slide 19, even though this is under the Remaining Challenges and Barriers sections). However, it is not clear whether the films were tested at low RH (the presentation includes no data at low RH). Therefore, it cannot be concluded that the films will have a low ASR at low RH.
    - The PI did not make clear how durability (both chemical and mechanical) is being addressed.
- Progress to date seems slower than anticipated or desired; despite being approximately 60% through the project, the team is still trying to make films that allow testing. Few free-standing films appear to have been produced to date, and those that have been produced are clearly extremely brittle. The project needs to make significant progress in developing usable films. The team will probably need to incorporate mechanical reinforcements and methods to allow incorporation of electrode layers.
- The improvement of the "System II Membrane," the hybrid HPA-3M FC-2178, does not seem to be a feasible path. The dehydrofluorinated membrane shown in slide 12 does not look to be very uniform, and the ion-conductive domain seems to be non-uniformly distributed. The team should determine whether HPA is precipitating out in the polymer matrix. Also, the presence of $-CH_2-$ groups in the proximity of $-CF_2-$ and $-CF_3$ groups may be labile, and the team should conduct Fenton's test to establish the stability of the $-CH_2-$.

Question 3: Collaboration and coordination with other institutions

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

- The team consists of a good mix of national laboratory and industrial partners. Collaboration with 3M, Nissan, and the National Renewable Energy Laboratory (NREL) is very advantageous to the team.
- Materials are just now becoming ready for outside partners to start testing. This project has taken good advantage of 3M's willingness to provide starting materials and advice about how to handle them.
- The partner team is sufficient. Collaborations with Nissan on testing of the materials and with 3M on process/cost input are important.
- There seems to be a solid partnership with 3M. The project is at the stage in which Nissan should start to contribute with MEA preparation and fuel cell testing. NREL's contributions to date are unclear.
- The project has good collaborators with a materials supplier (3M) and an automotive original equipment manufacturer (Nissan), among others for MEA testing. However, very few data were shown from the MEA testing from the collaborators.
- The PI did show slides about Nissan and its participation for the measurement of gas crossover. It is not clear what NREL's role has been until this point. The future work indicates that NREL will become involved for the remainder of the year. It is clear that 3M supplied products for the synthesis, but it is not clear what 3M has done in addition to that. However, the future work indicates that 3M will do characterization and testing and help in the scale-up of System II.
- Some of the work is seemingly duplicated at NREL and Nissan (MEA evaluation). From that standpoint, Nissan is likely the most competent organization to complete these tests. As a result, NREL workload and tasks should be reevaluated.
- It is unclear what, if anything, NREL and Nissan have done in the project to date. Future work is listed, but there does not seem to be any evidence of actual work done to date by the partners. The project is supposed to have third-party testing and have MEAs made, but there seems to have been no interaction to date. The interactions with 3M seem to have been primarily phone discussions.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.1** for its relevance/potential impact.

- The project is relevant to the objectives of the Multi-Year Research, Development, and Demonstration Plan. The activities are aligned to DOE's goal to address the commercial barriers such as performance, cost, and durability. The focus of the project is to demonstrate a low-cost hybrid inorganic/polymer from super-acidic inorganic functionalized monomers.
- Durable, low-cost membranes with low ASR and low gas crossover are essential to automotive fuel cell commercialization.
- A low-cost, durable, conductive membrane is essential to the development of commercially relevant fuel cell systems.
- This project has an innovative path compared to many other projects, with a potentially high payback in terms of higher conductivity at high temperatures and low RH operation.
- A membrane able to meet targets will be highly relevant. However, the membrane's ability to work in the fuel cell may require the appropriate matching ionomer in the catalyst layer. The project team recognizes this and has some activities to address this, but it is not clear whether this can be accomplished within the scope of the project.
- Improved membranes could improve the performance of fuel cells and widen the range of acceptable operating conditions. This project is still a long way from MEA tests demonstrating a real cumulative advantage for their materials. It is not clear that presently available commercial membranes (significantly improved over the past decade) are not adequate for major fuel cell applications, so this area does not require as much research attention as some others.
- The project tries to address the key issues, but so far, after 18 months, the results are not so encouraging. The ASR at high RHs was proven, but permeation and chemical/durability tests were not performed.
- No data or potential was seen for a game-changing reduction in membrane cost or game-changing improvement in membrane performance. The only data that showed significant improvement over Nafion were the gas transport data shown on slides 14 and 15. However, the PI did not know the important factor of sample thickness, so it is not possible to see whether the membrane's material properties are actually improved.

Question 5: Proposed future work

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

- The PI lists all the work that is key to the success of this project as future work.
- Most of the future work path looks appropriate. The project team should consider other design considerations, including mechanical reinforcement, which is lacking from the plans.
- In general, the future work plan looks good. The U.S. DRIVE Partnership's Fuel Cell Technical Team recommended durability testing, such as open circuit voltage (OCV) and RH cycling. ASR measurements should be done if not already planned. The project should include a more thorough cost assessment.
- Along with all the work that the team has proposed in the presentation (slide 23), the team should also work toward getting a uniform membrane and make sure that the HPA is coordinated to the polymer backbone and has not precipitated out into the polymer matrix.
- The proposed future work is logical. However, it does not appear that risks have been adequately assessed and mitigation pathways established. The ability of the membrane to work in an MEA is being assessed but is a very large question at the moment.
- The proposed work is reasonable.
- This project really needs to move forward into testing in MEAs. It is not clear how much faith the project still has in the TFVE/HPA system; it is not clear whether lower molecular weight and/or blending with other polymers can make this system tractable. Given the progress with HPA-FC-2178, it seems uncertain that the group will still put into effort into this. It would be good to get two materials out of this project.

MEA work may require increased commitment from partners in the development of electrodes that can work with the new membranes.
- There should be more focus on demonstrating chemical stability in operating fuel cells or some other ex situ accelerated test.

Project strengths:

- The membrane system front runner is showing very good conductivity and appears to have suitable properties to meet targets. The team has the appropriate partners to accomplish the project.
- The project has demonstrated significant synthetic skill and wisdom in backing away from dead-end approaches. The team is finally back to moving forward with the improved method for functionalization of FC-2178.
- The team is composed of respectable research organizations with adequate expertise. Overall, the team is equipped with the necessary knowledge base, resources, and industry/academia/national laboratory mix that is required for the success of this project.
- Exploring new materials has the potential for game-changing results. The project has a good source (3M) for a large quantity of materials for development work.
- This is really the only DOE project focusing on potentially game-changing PEM concepts. It is certainly high-risk, but there is also a possible high reward.
- The project takes an innovative path not being explored by other developers in the Fuel Cells sub-program.
- The development of three different membrane chemistries reduces the risk of failure.

Project weaknesses:

- Progress appears slower than desired. The number of free-standing films produced to date seems low. The materials produced appear extremely brittle, and the project has not addressed how to make the membranes mechanically viable longer-term, e.g., incorporating reinforcing materials. There was a gap between this project and the prior project, and thus it has taken some time to get back up to speed; nonetheless, this project is far enough through the project path that the gap no longer seems a viable reason for the low number of samples produced. Also, the project seems to have a low integration of project partners to date.
- The following are project weaknesses:
    - Progress is slow. Although the project is at 50% completion, it is not able to prove the ASR.
    - Permeation data are not clearly shown (measurements were taken with different thicknesses). In this case, the data should have been normalized to account for thickness.
    - ASR measurements have been taken only at high RHs.
    - No chemical durability tests, such as OCV hold/cycling, have been performed.
    - No mechanical durability tests have been performed to date, and the project is already at 50% completion.
- The inability to make reproducibly good quality films is a weakness. There are no data for dry conditions, which is where the biggest potential benefit of this concept truly lies.
- The project is thus far unable to convert the promising membrane material into an operating fuel cell. In-fuel cell membrane resistance was 200 times higher than the ex situ membrane resistance tests indicated.
- Membrane water transport properties are not considered but are deemed important for operation in a low-RH environment. MEA evaluations will be carried out at both NREL and Nissan, seemingly duplicating work.
- More focus is needed to prepare polymer HPA composite in which the HPA is bound to the polymer groups.
- Project accomplishments are slow, and the approach as presented lacks rigor on establishing understanding and relationships between material composition and membrane properties.
- This project has shown very slow progress prior to this year but decent progress this year. It is not clear that the motivation to move forward into MEA testing has been sufficiently strong.

Recommendations for additions/deletions to project scope:

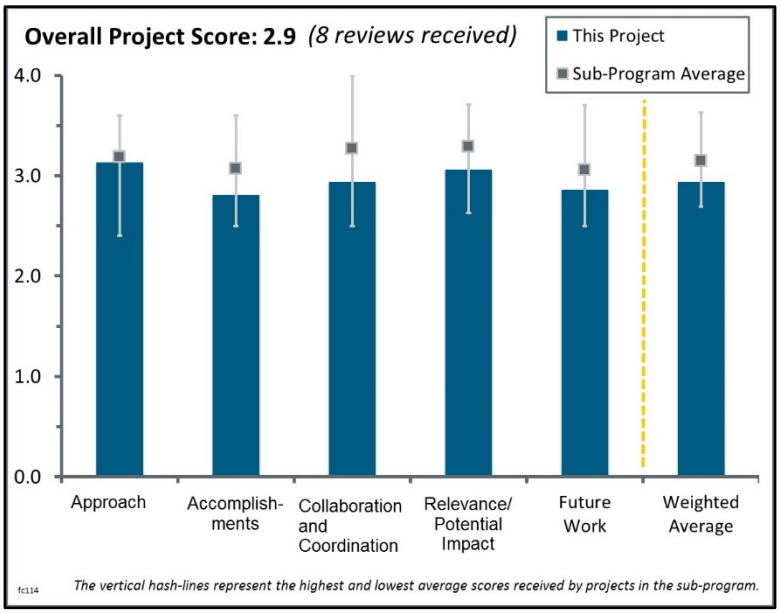
- The team should show some cost indications and compare them to perfluorosulfonic acid (PFSA), even if the team cannot supply actual cost estimates. There is also insufficient information on swelling, mechanical properties, low-RH performance, and durability. It is recommended that the project provide more calculations and predictions on properties. All critical properties should be assessed for risk, with mitigating factors and plans clearly laid out.
- If new ionomers for the electrodes are needed to make the project membranes work, the project should vigorously address electrode development (perhaps through increased commitment of partner resources) rather than declaring electrode development to be the subject of another project.
- More work should be done on making a larger quantity of membranes that can be shared with the MEA manufacturing/testing collaborators. More work on fuel cell tests would serve this project well.
- The project should start doing low-RH ASR and mechanical testing (tensile, swelling, etc.) as soon as possible.
- The project should take measurements of ASR at lower RHs, for example, 50% and 30%.
- Membrane water transport properties should be measured for the down-selected chemistry (2016). MEA evaluation tests at NREL should be eliminated.
- The project should consider adding a reinforcement material so the free-standing films can be better used and evaluated.

Project # FC-114: High-Throughput Synthesis, Oxygen Reduction Reaction Activity Modeling, and Testing of Non-Platinum-Group-Metal Polymer Electrolyte Membrane Fuel Cell Cathode Catalysts

Deborah Myers; Argonne National Laboratory

Brief Summary of Project:

The overall objective of this project is to develop tools for the rapid synthesis, fabrication, characterization, activity and durability screening, and performance optimization and testing of fuel cell cathode catalysts and catalyst layers in membrane electrode assemblies (MEAs). The low platinum objective is to realize the oxygen reduction reaction (ORR) activity benefits of the Argonne National Laboratory (ANL) nanosegregated platinum cathode electrocatalysts, demonstrated in aqueous tests, in MEAs at high current densities, and in air.



Question 1: Approach to performing the work

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

- The concept and intention of combinatorial testing and high-throughput fabrication is reasonable, but it would be interesting to see whether this project will result in new catalyst formulation or optimum MEAs. The methods are currently being developed and validated.
- The high-throughput/combinatorial approach may assist in catalyst development because the catalyst particles are sized nanoscale to approximately submicronscale, but this approach has challenges in its ability to optimize the MEA and its related fuel cell performance because the small-spot MEA may lead to an inaccurate performance measurement because of edge effect and inappropriate processing during MEA fabrication and fuel cell fabrication. The team should calibrate their measurements by comparing them with regular fuel cell results to validate their method and show that the trend is consistent with the regular method.
- The approach is to optimize the fuel cell performance of a particular cathode catalyst by making many variations to the catalyst and testing the fuel cell performance of the catalyst in a rapid screening tool intended to simulate actual fuel cell conditions. The underlying approach is to use the same MEA manufacturing and processing procedure for making many catalyst layers with variations to the catalyst material, including the comparison to the Tanaka Pt/C control. The performance shown, even for the control catalyst, is quite low because rapid testing conditions do not exactly mimic an operating fuel cell. Therefore, the formulation that works well in the rapid screening may not work well in an actual operating fuel cell. Furthermore, there is no reason to believe that an MEA manufacturing and processing procedure that is good for the Tanaka Pt/C control is equally good for a novel catalyst material. It would have been better to see an approach that uses a single large batch of promising catalyst material to make real single-cell fuel cells using different manufacturing processing conditions, ingredients, and methods to hone in on the best processing method for a novel catalyst.
- When trying to screen multiple MEAs, the performance, including that of baseline materials, is miserable. It is unclear why. It is also unclear why the researchers would test 25 samples before actually trying to use one of them. If they did try a single sample and performance was terrible, then it is unclear why they moved forward. It is unclear whether they tested blanks (i.e., just carbon fiber paper) to determine resistance, and whether they used pressure paper to ensure even distributions. It is very difficult to understand what problem this project is trying to answer. Perhaps the researchers have far too many

catalysts to screen; if this is the case, it is unclear how many they have to screen. There are also questions about how many man-hours it takes to process each sample. The input, the backlog, and the needed throughput need to be identified. Those questions need to be asked before the project is started to give it direction and measure its success. The same criticism is applicable to measurement, as well as to generation of catalysts; there seems to be little thought about what throughput is needed. Only then can one determine whether this project is needed and has real metrics to measure its success.

- This project aims to use a high-throughput/combinatorial approach to optimize a low-Pt polymer electrolyte membrane fuel cell (PEMFC) cathode and a non-platinum-group-metal (PGM) PEMFC cathode. The low-Pt cathode is based on ANL's segregated Pt alloy catalyst, and the non-PGM cathode is based on Los Alamos National Laboratory's (LANL's) polyaniline (PANI)-Fe_xCo_y catalyst. The high-throughput approaches the project has developed may help to down-select recipes for catalyst and electrode compositions in an effective way. However, because the low-Pt catalyst and non-PGM catalyst belong to two distinctive systems, it does not sound reasonable to combine these two systems into a project. In fact, each catalyst system should take substantial amounts of effort to make optimized electrodes. The focus of the project is thereby distracted.
- The objectives of the project include development of tools for rapid catalyst synthesis, characterization, activity and durability screening, and performance optimization in MEAs. However, the results presented during the U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program Annual Merit Review (AMR) meeting do not represent the major barriers—namely, electrode performance, cost, and durability.
- The segmented gas diffusion electrode deposition method does not seem to work in this method; it is unclear whether a spray/catalyst coated membrane (CCM) approach with masking has been considered. The goals of this work are quite unclear; it is unclear whether the goals are to develop high-throughput methods or MEA/materials. Clarification of the goals would really be helpful for reviewers.
- The objectives are very ambitious for a one-year project, and the approach is similarly broad and complex.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- Considering the number of tasks, two types of catalysts, and different methodologies (e.g., synthesis, fabrication, characterization, testing, and modeling), the accomplishments are impressive. Important achievements include the use of a high-throughput method for synthesis and characterization of non-PGM catalysts, and the use of powder-ionomer inks and fabrication of combinatorial MEAs.
- The accomplishments of this project are reasonably good. The developed robotic platform and four channel flow double electrodes (CFDEs) appear to be very powerful tools and enable one to screen multiple catalysts simultaneously in a very effective way. A high-throughput method has also been developed for synthesis and deposition of powder-ionomer solvent inks and fabrication of combinatorial MEAs. A density functional theory (DFT) model has been developed to screen non-PGM ORR activity and stability, which is a complementary approach to experiments.
- The team has done a good deal of work—especially on catalyst development—that is conclusive.
- The project is relatively new, and much has been accomplished within a short period of time in both the low-Pt and non-PGM projects. On slide 7, it was not clear what the different pol curves were, because the pol curves labeled as nanoframe-1 (NF-1) to NF-4 all have the same ionomer/carbon (I/C) ratio. It is unclear how one determines that there is a trend in performance and what kind of trend, as indicated in the second point on the slide. It is unclear whether the researchers carried out a test with standard electrodes on all 25 segments to ensure that all electrodes show the same performance. It is important to do this in order to ensure that the trend seen with different nanoframe electrocatalysts is related to the different catalysts, rather than the trend coming from the different segments. On slide 8, scanning transmission electron microscopy–energy-dispersive x-ray spectroscopy (STEM-EDS) was used to characterize catalyst inks with different I/C ratios. The highlights on this plot show that Pt and Ni composition is maintained. However, one would expect that the purpose was to show different I/C ratios using STEM-EDS. On slide 9, it is good to see that a model is being developed to analyze alternating current impedance data. H_2/N_2 impedance is a powerful technique to determine the catalyst layer effect, and it will be good to see more details on this approach and the results. The team demonstrated that a robotic system can be used to prepare different salt

precursors and can produce same-phase composition as a large single batch synthesized at LANL. It is unclear whether there are any issues with contaminants in the process of preparing LANL catalysts. For the CFDE setup, it is unclear how many working electrodes (WEs) and counter electrodes (CEs) there are for the 4 channel 4 WEs, how large the CE area was, and what type of RE was used. On slide 17, it is not clear how one ensures that the catalyst loadings for all electrodes are the same. When comparing the ORR activity using CFDEs and with large-batch rotating disk electrode (RDE) data, the current does not match. It is not clear whether surface area, catalyst loading, etc. were all kept the same. It is not clear how one compares the trend if the current is not normalized the same way.

- As of April 30, 2015, the team claimed it has met two of the three milestones. However, the fuel cell performance in combinatorial fuel cells for the Pt_3Ni nanoframe catalyst (slide 7) and the CFDE results of high-temperature PANI-Fe, Co-C catalyst (slides 16 and 17) are not comparable to the ones generally obtained in a single fuel cell and rotating ring disk electrode experiments. Considering the fact the project is ending in September 2015 and that only 64% of the proposed work has been completed, the team may not have sufficient time to get any meaningful results to substantiate the objectives and approaches of this project.
- The milestones were very low-risk, and they were not well accomplished. The “stretch” goal, which was the one project element with risk, was not well addressed. The researchers did generate a number of samples and a number of catalysts. However, if the goal of this project is to increase throughput, then metrics for comparison are needed. It is unclear how many catalysts can currently be generated and characterized per month per person, and by how much this process improves that figure. The mere fact that the new operation works does not say very much.
- Even if the rapid screening method selects a catalyst that is better than the control, then that manufacturing method needs to be scaled up to a process that could be used to make actual fuel cell MEAs.
- Statistical correlation of the large dataset is a major aspect of understanding screening work; however, it has not been done.

Question 3: Collaboration and coordination with other institutions

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

- There is good coordination and collaboration between ANL and LANL, as well as among experimentalists, characterization efforts, and modeling efforts. It was also nice that the project got the combinatorial fuel cell test fixture and Arraystat from Nuvant Systems for free. The project features good collaboration and leveraging of the available and DOE-funded capability.
- The principal investigator (PI) has good collaboration with Oak Ridge National Laboratory (ORNL), LANL, and an electrochemical systems manufacturer to meet the project objectives.
- The project features excellent collaboration with experts in particular areas.
- There is extensive collaboration between national laboratories.
- The collaboration with LANL is good. The participation of a major fuel cell company (e.g., Ford, General Motors, or Ballard Power) would be very meaningful.
- This is an ANL/LANL/ORNL collaboration, but it seems to be missing the obvious inclusion of industry experts in ultra-low-loading CCM construction and high-throughput methods.
- Collaboration with an MEA manufacturing partner would benefit this project.
- There is not much input from other national laboratories. There is no industrial collaboration, either from those that might use these techniques or those that might market them.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.1** for its relevance/potential impact.

- The project is very relevant to DOE fuel cell catalyst objectives and targets. In particular, ANL’s segregated Pt alloy catalyst and LANL’s PANI-Fe_xCo_y catalyst represent the two most promising catalysts, PGM and non-PGM, respectively. However, previous efforts were mostly on RDE and small-range MEA

fabrication. The proposed high-throughput approaches may help to down-select catalyst compositions and electrode conditions so as to further promote the application of these advanced catalysts at more practical levels.
- The project's efforts are directly related to DOE research, development, and demonstration objectives.
- High throughput is a good approach to screen catalysts and catalyst layers in MEAs. However, the approach should be guided by theory and/or modeling because there can be many different combinations one can try. Otherwise, it will be a trial-and-error exercise.
- The results will affect future efforts in this area.
- The results presented during the 2015 AMR meeting are not promising in terms of meeting the project goals of developing tools for the rapid synthesis, fabrication, characterization, activity and durability screening, and performance optimization and testing of fuel cell cathode catalysts. From the presentation, it is not clear how the durability of the low-PGM and non-PGM catalysts can be evaluated by the end of September 2015.
- Both electrode construction of these highly active PtNi catalysts and optimization of the non-PGMs are aligned with DOE goals. It is not clear, however, that the high throughput has significant value to either goal, nor is it clear that the methods have demonstrated potential to do so. In a very complex experimental variable space, such simple and not fully defined efforts do not seem to have a high value/effort proposition.
- The project seems like it has a long way to go to provide any useful feedback to the manufacture of fuel cells with new catalysts that operate well.
- It is very doubtful anyone outside of this group will use the techniques developed here.

Question 5: Proposed future work

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

- Considering the timeline of the project, the future plan is reasonable.
- The list of future work seems reasonable.
- This project's significance is in its ability to increase/improve the throughput of catalyst generation and characterization. This has to be kept in the context of what is currently being done. To justify the project's cost, $1 million, one needs to know, if a laboratory had been given $1 million to hire new scientists, how many catalysts could have been generated and characterized without this project. In other words, if this task is 100% successful, one needs to know when it will pay off. If it currently takes $10,000 to generate and characterize a catalyst, and with these tools it will only take $1,000, then it will pay for itself after only 110 catalysts. It is unclear whether the change is that dramatic and whether there will be that many new catalysts. These are the questions this project has to answer.
- The team has proposed performing a number of studies, including CFDE studies, electrode fabrication methodology, fuel cell testing, and physical characterization studies. The proposed studies need at least a year to get any promising results.
- The proposed future plan is very comprehensive. However, the project will be ending in September 2015. It is not very clear whether most of these proposed work plans can be implemented.
- The proposed future work is quite substantial, considering the limited time (three months) left in this project.
- The future work involves finishing incomplete tasks.

Project strengths:

- The project features collaboration with an electrochemical system manufacturer and with two national laboratories that have expertise in developing low-PGM and non-PGM catalysts. The project team takes a very good theoretical approach to understanding the ORR active sites of a non-PGM catalyst.
- High-throughput approaches are very powerful and effective for catalyst composition down-select and electrode optimization. In addition, a DFT model has been developed to screen non-PGM ORR activity and stability, which is a complementary approach to the experiment-based approaches.
- The project features a very strong team with a long history of doing this work. If this work needs to be done, this is undoubtedly the right team.

- Project strengths include the technical strength of the groups and the PI, as well as the communications.
- The project has a strong team and resources related to catalyst research.
- This project features a very strong team and excellent collaboration.

Project weaknesses:

- One weakness is the utility and proof of high-throughput methods. Another is the selection of M-OH bond strength as a criterion for optimization. This criterion has led to an active site structure that is inconsistent with experimental evidence for the active site, hence bringing into question the criterion. Comparison to ex-silico information is needed. A third weakness is that statistical correlation of the large dataset is not addressed.
- The combination of two totally different systems into one project has diminished the project focus. In addition, the entire work structure appears to be disorganized, likely owing to the combination of two different catalyst systems.
- The project has very limited time to fulfill the proposed objectives. A two-year project, rather than a one-year project, would have been beneficial.
- The project features too many tasks and diverse methods to accomplish them.
- A relationship between the rapid scanning screening method used in this project and actual operating fuel cells would benefit this project.
- There is a complete lack of proper metrics.
- The MEA fabrication is the weakness.

Recommendations for additions/deletions to project scope:

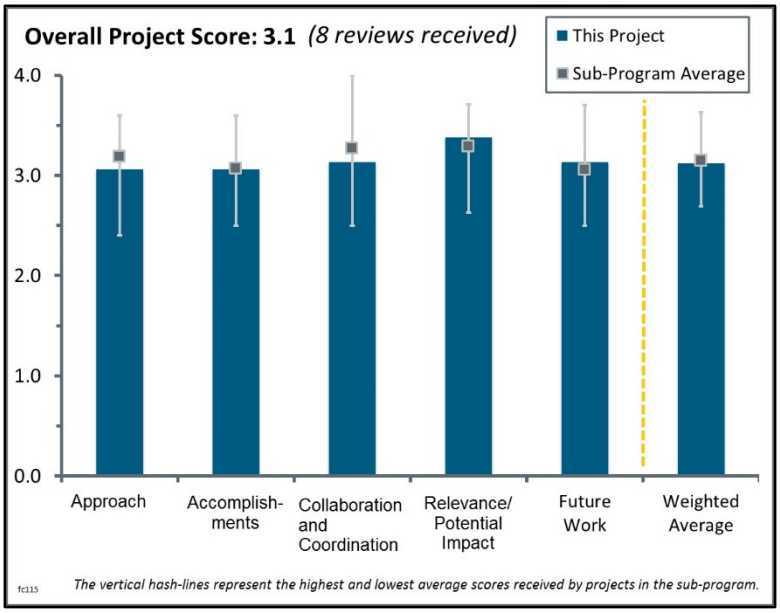
- No specific additions/deletions are needed.
- The team should include industry experts in ultra-low-loading CCM construction, catalyst synthesis, and high-throughput methods.
- If possible, the project should be focused on either low-Pt cathode or non-PGM cathode because there are only a few months left.
- Focusing on a fewer number of tasks would be beneficial for the outcome of the project.
- Adding an MEA manufacturing team member would help this project.

Project # FC-115: Affordable, High-Performance, Intermediate-Temperature Solid Oxide Fuel Cells

Bryan Blackburn; Redox Fuel Cells, Inc.

Brief Summary of Project:

The overall objective of this project is to improve the performance of intermediate-temperature (≤600°C) solid oxide fuel cell (SOFC) technology operating on reformed natural gas through the (1) development of an optimized bilayer electrolyte with increased open circuit potential and thus greater fuel efficiency, (2) optimization of compositions and microstructure for the electrodes to improve performance and impurity tolerance, (3) use of a custom multiphysics model and advanced materials to optimize the performance of bilayer stack designs, and (4) demonstration of a ~1 kW stack for intermediate-temperature operation under combined heat and power (CHP) conditions.



Question 1: Approach to performing the work

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

- The team's approach toward developing intermediate-temperature fuel cells relies on the successful development of Bi_2O_3 (bismuth oxide)/CeO_2 (cerium oxide) ceramic membranes. The materials compatibility will be challenging (i.e., matching thermal expansion, chemical reactivity, and low-cost manufacturing). The approach is one of few potentially viable methods to develop intermediate-temperature solid oxide fuel cells (SOFCs) with good performance.
- The approach includes modeling and experimentation for SOFC bilayer structures. The bilayer approach is intended to provide stability and low electronic conductivity. The team needs to discuss and analyze the interface.
- The approach logically uses scale-up in a stepwise manner to achieve progress.
- The project is currently designed to meet the barriers identified and is structured appropriately.
- The approach seems reasonable, but the absence of planned durability testing leaves a fairly big hole.
- The project needs to speak to total life of cells at high power density and low temperature. Anything can work at high power for a short time—life is a critical factor and should be evaluated as soon as possible. The team should evaluate co-diffusion of gadolinium-doped ceria (GDC) and Bi_2O_3 as a potential issue (or benefit). The effect would be a broadening of the GDC/Bi_2O_3 interface that could affect overall performance and life. A broadened interface may make the bilayer more robust to system upsets that could result in a lower oxygen partial pressure. It could also be detrimental—it is not clear whether co-diffusion will occur or what the impact may be. The researchers should see Oak Ridge National Laboratory's microscopy project (FC-020) as possible support for assessing the interface performance.
- Approaches to performing the work and results obtained to date were not given in the slides or discussed in detail in the presentation. For example, the statement "Leverage 25 years of SOFC [research and development]...." (slide 16) does not convey any information on the approaches proposed in this project to address the critical barriers of the technology.

- In addition to electrolyte resistance and electrode activity, other challenges should be addressed for intermediate-temperature SOFCs. The mechanical strength of the ceria-based cells and long-term operation stability are among those challenges that need to be addressed.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The team has made good progress for GDC/erbium stabilized Bi_2O_3 (ESB) bilayer electrolyte thickness optimization:
    - Achieved Milestone 1.1: ESB/GDC Bilayer Button Cell open circuit potential of ≥0.9 V at ≤600°C.
    - Is close to meeting Milestone 2.1: area-specific resistance ≤0.2 Ω-cm^2 and ~1 W/cm^2 at ≤600°C.
- Single-cell testing has commenced. Preliminary results look promising. It will be good to see the durability data.
- The project has demonstrated the high power density of the cell at 600°C, which is a promising achievement.
- The results to date indicate that the team has made progress toward meeting DOE goals. Presenting anticipated results does not add value to the accomplishments. It will be interesting to see the sulfur stability results. Capturing any performance decrease and the ability to recover is important.
- The project is nine months old. Reasonable progress has been made in terms of performance and modeling. Durability (especially of the interface) does not seem to have been addressed. The open circuit voltage seems low. Cost is not addressed.
- It is early in the project, so judging accomplishments is difficult, but the progress to date seems good.
- Transitioning to a larger format is good; however, there is much that still needs to be learned at the button cell level. Claims of operation on conventional fuels are not currently supported by data. Direct use of gasoline and/or diesel is unlikely because of cracking/carbon deposition at the target temperatures (e.g., ~600°C). Sulfur content of those fuels is also going to be a problem. The presentation should not be a sales document, and unsupported claims are not appropriate in this context.
- Well-defined metrics are needed for this project. For example, the success criteria for the 1 kW stack demonstration should be defined. Numerous statements in the slides (e.g., on slides 8–11 and 13–14) are so general that it is difficult to comment on accomplishments and progress.

Question 3: Collaboration and coordination with other institutions

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

- All aspects of the project are covered by knowledgeable organizations.
- The project includes the following collaborations: University of Maryland, cell research and development; Trans-Tech Inc., cell manufacturing; MTech, commercialization; and Redox Power Systems, cell/stack development and testing.
- The team works closely with the University of Maryland.
- Collaboration exists; however, it is difficult to evaluate the collaboration based off of the presentation.
- The mix of collaborators seems appropriate, but it is difficult to tell where responsibilities lie.
- The actual contributions of the collaborators were not clearly explained or defined. Perhaps it is too early in the project for much in the way of contribution from the collaborators. If so, the team should explain this and provide a schedule of when collaborator contributions are expected.
- In addition to collaboration on multiphysics stack modeling, close collaboration with organizations with expertise in multicell stack design and stack operation/testing may be needed for this project.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.4** for its relevance/potential impact.

- Many DOE Hydrogen and Fuel Cells Program fuel cell barriers have been addressed:
    - Durability:
        - Lower operating temperatures for Redox SOFCs than for competing SOFCs
        - Internal reforming with catalysts for enhanced sulfur and coking tolerance
    - Cost reduction:
        - Using no PGM materials
        - Lowering the operating temperature to allow the use of simple stainless steel commercial off-the-shelf compressive gaskets
        - Using fewer cells because of higher power density per cell
        - Lowering the system cost
    - Performance:
        - 10x10 cm anode supported cell demonstrating >1 W/cm^2 at 650°C (using natural gas)
        - Performing laboratory-scale demonstrations with peak power density of 2 W/cm^2 at 650°C
        - Maintaining performance at ~600°C through various cell enhancements, and improving stability
        - Demonstrating the availability of high-quality heat for CHP (2020 Fuel Cell Technologies Office target for 90% CHP)
- The project clearly identifies the DOE goals it is targeting and the progress it has made to date. It has already met or exceeded some of the identified goals.
- The potential impact is very high. Intermediate-temperature fuel cells offer excellent characteristics for stationary CHP.
- This project seems very relevant. If successful, it could have a great impact on SOFC system costs.
- Low-temperature SOFCs are attractive and in line with DOE goals.
- The success of the project will accelerate the deployment of fuel cells for stationary applications.
- The bilayer approach with these materials is very interesting and potentially a good approach to high power density. Caution is necessary on predicting fuel cell lifetimes with this approach. The claims of internal reforming of commodity fuels are questionable until demonstrated for hundreds of hours.

Question 5: Proposed future work

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

- The plans could lead to achieving goals:
    - Use the upgraded model to optimize stack design for a bilayer cell.
    - Consider different operating conditions that might improve durability and reduce degradation (e.g., reduced thermal gradients).
    - Use the cell portion of the model in conjunction with ongoing button cell and 10x10 bilayer thickness optimization.
    - Translate the already achieved performance at the button cell level (i.e., ≥0.9 V and ~1 W/cm^2 at ≤600°C) to the 10x10 cm cells.
    - Finish mapping the stack performance up to the full 1 kWe size and use residential CHP conditions with natural gas to obtain similar performance for the 10x10 cm single-cell and short-stack levels in the ~1 kWe stack size under CHP conditions.
- The project has a clearly identified path forward for future work and go/no-go decision points. The path forward will achieve the DOE goals identified and overcome current barriers.
- Scale-up from single-cell testing at the laboratory scale to short stacks at 1 kW at CHP conditions is planned.
- The future work plan is acceptable.

- The proposed future work seems appropriate and in line with the objectives; however, the addition of durability testing would seem appropriate.
- The researchers should address the bilayer and overall stack degradation rate before investing excessively in system or full-scale stack development. Systems, particularly CHP systems, are expensive to build and test. This is only appropriate after the stack has been shown to be robust and long-lived—or at least shown to be acceptable for at least a couple of hundred hours.
- The proposed future work does not include sufficient details, especially on the planned approach and risk mitigation.
- The team needs to demonstrate the long-term operation stability and mechanical strength of the cells.

Project strengths:

- The project features a novel approach to achieving high-efficiency SOFCs that operate well below 700°C. The team is highly knowledgeable, with many years of experience in SOFC development.
- Strengths include the clear project goals and identified barriers. The go/no-go decision point will help in evaluating the project at the 12-month mark.
- The team is developing an intermediate-temperature electrolyte and electrodes to reduce the operation temperature of SOFCs.
- The ESB/GDC bilayer is very interesting and potentially a breakthrough technology. It is definitely worth additional investigation.
- The project's strength is its potential impact.
- The project focuses on cell scale-up and stack demonstration.

Project weaknesses:

- There is no emphasis on expected lifetime or degradation rates. This is a major concern that seems not to have received any attention. If the cell lifetime is short, it may take a significant investment to find the root cause and advance the technology to increase cell life. Investments in system or full-scale stack development are not appropriate until the lifetime question has been answered.
- The project is focused only on the electrolyte and electrode development, which is not enough for the stack fabrication and demonstration.
- The presentation did not clearly identify which partners contributed to which portions of the accomplishments.
- The project may be too ambitious for the funding levels. A larger project may be required to determine the feasibility of the approach.
- The project lacks well-defined metrics, risk mitigation, and detailed work plans.
- The recognition of (long-term) issues at the bilayer interface is insufficient.
- One weakness is the lack of planned durability testing.

Recommendations for additions/deletions to project scope:

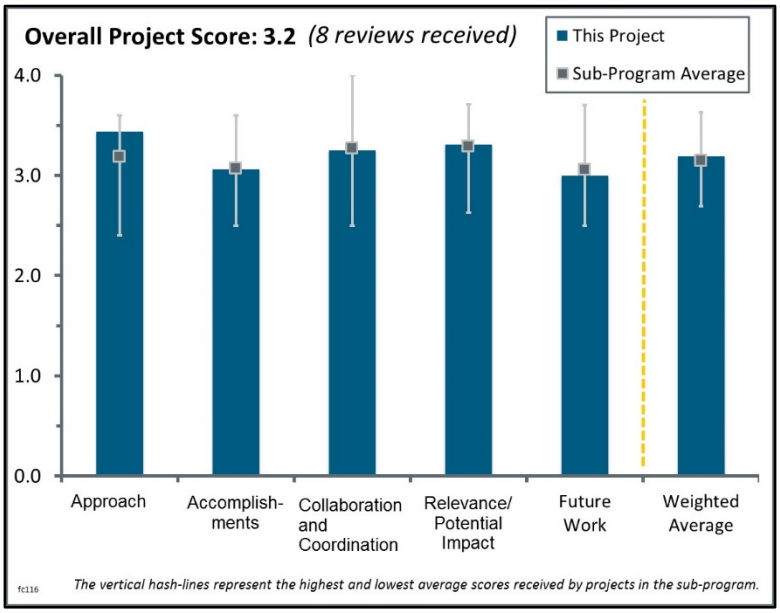
- The researchers should refocus significant effort to demonstrate lifetime under realistic conditions. Domestic natural gas is a good starting point—if that does not work (because of sulfur content or carbon deposition), then none of the other proposed fuels is likely to work. If natural gas exhibits good lifetime, then building a full-scale stack and supporting balance of plant may be justified. Gasoline and diesel are much more difficult—those should be saved for another project.
- The project team should investigate the long-term stability of the bilayer electrolyte and infiltrated electrode catalyst. The team does not need to do direct hydrocarbon oxidization in anode at this stage of technology development.
- Assuming stable performance is achieved with sulfur, the team should analyze the performance loss and the ability to recapture that performance once sulfur is removed. It is unclear whether the system can operate for thousands of hours with sulfur.
- The team should pursue durability testing and study the stability of the approach.
- The researchers should start single-cell durability testing early in the project.

Project # FC-116: Smart Matrix Development for Direct Carbonate Fuel Cells

Chao-yi Yuh; FuelCell Energy, Inc.

Brief Summary of Project:

The overall objective of this project is to develop an innovative, durable, direct fuel cell electrolyte matrix ("Smart" Matrix) to enable >420 kW rated stack power and 10-year (80,000-hour) stack service life. The objectives for the current project year are to (1) develop plans to achieve "Smart" Matrix goals and to enhance degradation mechanistic understanding; (2) develop a high porous matrix structure with controlled pore size; and (3) initiate cell testing of the "Smart" Matrix.



Question 1: Approach to performing the work

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

- The project has clearly defined barriers and effectively conveys how it is going to overcome those barriers. The project is set up well, logical, and feasible.
- The project addresses the critical issue of matrix microstructure degradation for molten carbonate fuel cells (MCFCs) and focuses on evaluation of a new matrix with commercial-sized cells.
- The discussion of the $LiAlO_2$ issues is broad. The work should provide a sound mechanistic understanding.
- There are many thought-out approaches to achieving the objectives.
- The project represents an evolutionary approach toward improvements in a mature technology. The approach seems to be continuing to study the $LiAlO_2$ electrolyte support. The pore size stability tests seemed to be a bit haphazard; however, the project is in its early stages. Very little information was presented about what the "Smart" Matrix technology actually entails. It is not clear that the pore size control is the lifetime limiting factor for MCFCs.
- The project approach is reasonable, except for the goal of achieving 80,000 hours of life in three years. FuelCell Energy (FCE) would need to demonstrate that its accelerated testing is valid out to 80,000 hours, and because it has not achieved this with its current designs, FCE must provide details on accelerated tests. Outstanding questions include how many FCE systems have achieved >43,000 hours and how many have not, and whether FCE did anything special to achieve >43,000 hours, such as derate the power plant.
- The issue with current matrix material is the coarsening at the anode side of the cell, which is more related to the material ($LiAlO_2$) properties in reducing atmosphere, instead of microstructure. Reducing the initial pore size is not likely to achieve a twofold lifetime improvement, as targeted in the project.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The results achieved to date are contributing to overcoming the defined barriers. The project has tightened the control of the pore size diameter in the "Smart" Matrix, which is an important step. Process consistency was demonstrated on the porosity for good reproducibility.
- The team has successfully manufactured a laboratory-scale, high-porosity "Smart" Matrix.
- The progress to date is encouraging and impressive, considering that the project is nine months old.

- Significant progress has been made in the first six months of the project.
- The improved pore structure shows progress. It is not clear whether FCE will be modifying the basicity. More information is needed. The project team did a good job identifying challenges. It is unclear whether all other components are capable of reaching 80,000 hours—that was not identified in the presentation.
- The accomplishments are satisfactory; however, they are not integrated with lifetime improvement models that show that matrix improvements will greatly extend the system life. The MCFC technology is relatively mature, and it will have difficulty competing with polymer electrolyte membrane (PEM) and solid oxide fuel cell (SOFC) improvements.
- Matrix durability is not the bottleneck of current stack life. Reducing the cost in the current stack could have more of an impact in terms of meeting DOE goals.

Question 3: Collaboration and coordination with other institutions

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

- The partners are recognized experts in MCFC technology. The team has a long history of collaboration on MCFC issues.
- FCE has established strong collaborations with the University of Connecticut (UConn) and Illinois Institute of Technology (IIT).
- UConn/IIT has significant prior experience and analytical capability; the project has solid, experienced partners.
- The UConn collaboration is well established.
- The project is leveraging expertise in multiple universities to gain full understanding of the "smart" matrices from multiple different angles. Coordination is required to develop the matrix, as well as to ensure it supports the performance required for the project to be successful.
- Collaboration with an $LiAlO_2$ producer may be needed.
- The presentation did not discuss the partner's progress.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.3** for its relevance/potential impact.

- The project is highly relevant. The project aims to develop an innovative, durable direct fuel cell electrolyte matrix (i.e., "Smart" Matrix) to enable >420 kW rated stack power and 10-year (80,000 hour) stack service life; increase market penetration for stationary fuel cells; enable technology for hydrogen infrastructure and CO_2 capture; and enable domestic clean energy job growth.
- The project has DOE goals in mind and is progressing toward those goals. The project aims to reduce cost and improve efficiencies and operating life. This will contribute to increasing the return on investment on stationary power and combined heat and power systems and help grow the market.
- The project aligns with DOE Hydrogen and Fuel Cells Program goals and objectives.
- FCE MCFCs are an established technology with the potential to meet DOE's technical durability targets.
- This could be a high-impact project if 80,000 hours can be attained.
- The relevance is moderate. The project will result in modest improvements in the established technology.

Question 5: Proposed future work

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

- The future work is focused on solving the matrix problem and has a good mix of material development and endurance testing.
- FCE is pursuing key activities in the future to achieve the following goals:
    - Optimize matrix formulation and processing conditions.
    - Develop stabilized materials based on mechanistic understanding.

- This project has good go/no-go criteria and set points to keep it focused and successful; however, some accelerated test methodologies should be involved in order to more accurately claim success of operating for the very high 80,000-hour target.
- The project is focused on the beginning of life properties of the matrix. However, the team did not allocate enough resources to $LiAlO_2$ coarsening root cause studies or to reducing the cost of the process.
- It is not clear what mitigation approaches will be applied to the results from $LiAlO_2$ coarsening (slide 9).
- The proposed future work is a bit vague. Not much information was presented on how the matrix material will be optimized.
- More details should be given on the approaches for the future work.

Project strengths:

- The project focuses on commercial-sized cells and operating conditions. The project team has extensive experience in MCFC technology.
- FCE is the leader in MCFCs. The development of the project can be smoothly integrated into final products.
- The project team is making incremental improvements toward a commercially demonstrated technology.
- The project is well formed and on target to overcome the identified barriers.
- FCE brings the greatest expertise in MCFCs.
- The project features the world leader in the technology.

Project weaknesses:

- The project does not focus on the key issues of MCFC stack life. Optimizing the initial pore structure is not likely to achieve the technical goal (10 years of operation life). The work plan does not include the efforts for cost reduction.
- The project has a high target of 80,000 hours of life; it is unclear how a 5,000-hour test provides data to show any improvement over the baseline 43,000 hours.
- MCFCs are having a very difficult time competing with natural gas combined cycle power plants. It is difficult to see how this investment will make them substantially more competitive.
- The team will need to discuss durability testing at the next review. Other components that may reach 80,000 hours need to be identified.

Recommendations for additions/deletions to project scope:

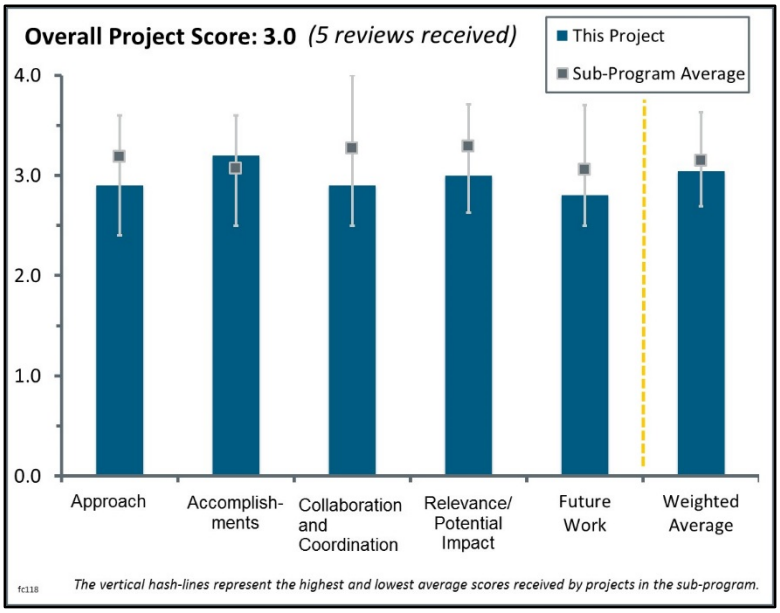
- Perhaps the team should add some validated accelerated test methods or test data from the first 5,000 hours of the baseline system for comparison to the 5,000 hours of the “Smart” Matrix system.
- The project team should include the cost reduction of the fabrication process.

Project # FC-118: Novel Non-Platinum-Group-Metal Catalysts from Rationally Designed Three-Dimensional Precursors

D.J. Liu; Argonne National Laboratory

Brief Summary of Project:

The overall objectives of this project are to (1) design, synthesize, and evaluate highly efficient non-platinum-group-metal (PGM) cathode catalysts using rationally designed three-dimensional (3-D) precursors with significantly improved fuel cell performance; (2) maximize electron, heat, and mass transport by incorporating the catalyst into a porous nano-network structure; and (3) support non-PGM catalyst development through structure-function relationship investigations.



Question 1: Approach to performing the work

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

- Argonne National Laboratory's (ANL's) focus on zeolitic imidazolate framework (ZIF) metal-organic framework (MOF) and membrane electrode assembly (MEA) testing has shown to be fruitful. The team demonstrated significant improvement in fuel cell performance under practical conditions and has produced one of the best, if not the best, non-PGM catalysts. Incorporating organic ligands into the MOF is also promising.
- Studies on benchmarking and understanding the stability of these catalysts are lacking. The transition away from the Fe-based material is very slow. However, considering the short length of the project, this is understandable.
- Incorporating a non-PGM catalyst into a 3-D, porous nanonetwork structure of high-surface-area MOF materials (ZIF) for catalyzing the oxygen reduction reaction (ORR) is apparently a good approach.
- Synthesis of MOFs provides a route for controlled variation of CNFe catalyst precursors, but most of the nitrogen and most of the initial structure are lost during thermal activation, so the final product is not all that different from more conventional non-Pt catalysts prepared by ammonia etching of carbon blacks.
- This project has quite properly moved to MEA testing at an early stage, and it has included a fair amount of data on air, which is the greatest challenge for non-Pt catalysts.
- The project has so far compared its results only to the areal current density targets promulgated in the catalyst working group. These targets correspond to performance that is far too low for automotive application, and they are too low for cost parity with Pt-based catalysts, even for less-demanding applications.
- ANL has yet to report volumetric current densities (A/cm^3), which are the first numbers that should be checked and reported in non-Pt catalyst work, although the field has advanced beyond the need for that number alone.
- This project is repetitive of previous work; the originality and the value proposition are unclear. This is true of the materials set, characterization, and analysis (Dodelet and Jouen did this work 10 years ago, 10.1126/science.1170051, while several groups have made the precursors using the same approach, http://dx.doi.org/10.1038/nprot.2013.100). The relevance of the principal investigator's (PI's) past experience to this project is unclear. The performance reported is for a very short time frame, and testing conditions seem to deviate from U.S. Department of Energy (DOE) protocols and comparable testing conditions reported elsewhere.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

This project was rated **3.2** for its accomplishments and progress.

- ANL has generated a considerable amount of results, particularly related to synthesis and MEA testing. Obviously, the team is capable of making an adequate amount of catalyst in an efficient way. The team has achieved one of the best non-PGM MEA performances. The ORR activity was also close to meeting the DOE target.
- Remarkable area activity has been obtained for the 3-D tetrahedral precursor incorporated in ZIF. The high surface area helps the researchers achieve a high geometric current density; therefore, the per-site activity should be determined to clarify real activity. The durability of the catalyst is not addressed, which should be an immediate task.
- The initial MEA data on O_2 are encouraging, but they still need improvement for practical operation.
- Initial MEA data on air are also encouraging, but ANL still needs extensive work on electrode-layer optimization.
- Much more work is needed on durability.
- Volumetric activities were not reported. Assuming the ~3.6 mg/cm^2 loadings of catalyst have densities similar to those of conventional carbon-black layers, these electrodes would be about 100 μ thick. From slide 9, the volumetric activities would be about 25 A/cm^3 from the 0.8 V data or about 60 A/cm^3 if (improperly, but done in the literature) extrapolated back from 0.9 V to 0.8 V. This is about five times below the state-of-the-art kinetic ORR activity for non-Pt catalysts. However, at this stage in the development of non-Pt cathode catalysts, improvements in durability and high-current-density performance in air are more important than improvements in kinetic activity.
- The "record" power densities claimed here are of limited practical value because they were achieved at such low cell voltages (<~0.65 V) that heat rejection would be excessive and efficiency would be way too low.
- The performances and metrics reported and used are neither relevant nor novel. High and transient performances of non-PGMs are not useful in any real sense. It is unclear why researchers chose to repeat short tests instead of carrying one out for hours or days.
- The synthesis approach is more novel, yet a cursory literature search shows seemingly identical efforts done all over the world, including the FC-086 project (which had yielded a well-performing non-PGM with much better stability).
- The use of Co instead of Fe is to be commended.
- It is unclear how this project contributes to DOE goals related to making technologically relevant non-PGM catalysts. The precursors used are much more costly than those in the Los Alamos National Laboratory (LANL) project, without real performance improvement. Further, because LANL has a more significant effort, it is unclear how this project furthers the technology path.

Question 3: Collaboration and coordination with other institutions

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

- The collaboration is appropriate for the project's size and goals.
- This project team seems to have done a very good job in coordinating the efforts at a diverse group of laboratories, taking advantage of both advanced synthesis and competent MEA preparation and testing, although it is not clear that the calculational work has provided any new information. It would be good to identify where the MEAs were prepared and tested (in the absence of specific information to the contrary, one assumes this was done at the lead laboratory, ANL).
- The industrial partner for the PEMFC MEA work should be named.
- The project's collaborations involve academia, some industrial laboratories with theoretical studies and structural studies.
- While collaborations are listed, they are not clear.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.0** for its relevance/potential impact.

- The non-PGM catalysts provide hope of removing the PGM risk from the technology, although significant challenges still remain, including serious stability and performance issues. ANL's MOF approach is one rational approach to meet the targets.
- This project has been making progress toward the "holy grail" of cathode catalyst work—a non-Pt catalyst with adequate performance over a practical range of current densities and potentials. The results remain far from what would be needed for practical application in fuel cells.
- Proof of durability of both kinetic activity and high-current-density performance in air would be needed before very large resources could productively be expended on this pathway.
- The project seems to be addressing some of the issues needed to get adequate performance at high current density in air out of the thick electrode layers needed for non-Pt catalysts. This problem and durability are the major issues the non-Pt field must address.
- If the catalyst's durability and easy synthesis and scale-up can be verified, this work could have a notable impact.

Question 5: Proposed future work

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

- The issues of stability and one-pot synthesis are the focus in the proposed future work.
- The project team plans to focus on improving activity and durability by using high-throughput screenings of MOF design and organic additives, although the plans were not described in detail.
- New materials development should be focused on a non-Fe system.
- ANL should focus on improving performance at high current density in air; stability in MEAs; and the reproducibility of synthesized catalysts.
- The work with non-Pt catalysts (particularly in making good MEAs for testing of high-current-density performance in air) is sufficiently difficult that it is probably unrealistic to expect that high-throughput pathways will be productive.
- Much greater emphasis must be placed on durability.
- Measurement and demonstration of the stability and durability of these materials is critical.
- The materials set in the proposed work has been extensively studied by Lefre, Dodelet, and Jouen, with poor stability in all cases. It is unclear how this work is different or will lead to stable catalyst performance.

Project strengths:

- Project strengths include the capable team, practicality, and product-oriented culture. The project's organization and management are also strengths.
- The focus on non-Fe catalysts is commendable and should be enhanced.
- The project team has good synthetic skills.
- The project has properly emphasized testing in MEAs, including data on high-current-density performance in air. The poster should have clarified where this MEA testing has been carried out.
- The team achieved facile synthesis of complex precursor materials that allow for a wide range of subtle variations in precursors.

Project weaknesses:

- The goals are nebulously defined, and metrics really need to be more explicit.
- This project is repetitive of previous work; the originality and the value proposition are unclear. This is true of the materials set, characterization, and analysis (Dodelet and Jouen did this work 10 years ago, 10.1126/science.1170051, while several groups have made the precursors using the same approach,

http://dx.doi.org/10.1038/nprot.2013.100). The relevance of the principal investigator's (PI's) past experience to this project is unclear. The performance reported is for a very short time frame, and testing conditions seem to deviate from U.S. Department of Energy (DOE) protocols and comparable testing conditions reported elsewhere.
- The relevance of the "Molecular" design approach to materials pyrolized at high temperatures is not proven.
- Very little, if any, durability data have been reported to date.
- Starting precursors may be needlessly complex for a material whose structure will be extensively altered by activation through pyrolysis.
- Weaknesses include the continuity of funding and the use of Fe in the catalysts.
- Project weaknesses include the small team and a lack of durability studies.

Recommendations for additions/deletions to project scope:

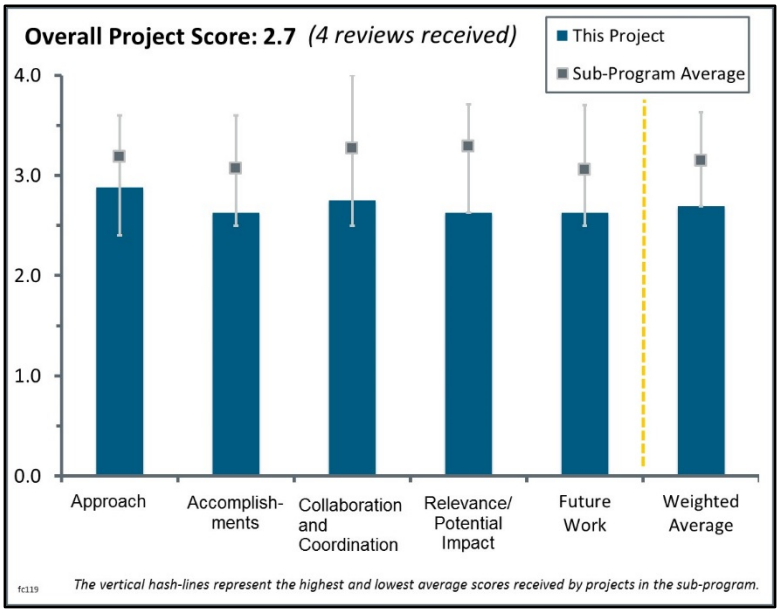
- In MEAs, cathode catalyst layer densities (i.e., thicknesses), porosities, and pore-size distributions should be measured and reported. Durability data should be taken for potential ranges that will give practical efficiencies (i.e., not at 0.4 V RHE, as was done in one oft-quoted paper). It might be good to try to mix project catalysts with conventional carbon-black supports (which give good pore structure) in an attempt to improve mass transport and high-current-density performance in air.
- Reported performance should be of some stability metric.
- The focus on non-Fe catalysts is commendable and should be enhanced.
- The project team should study leaching of Fe and Co, and the effect on the membrane's conductivity.
- The team should pursue a better understating of catalyst poor stability and develop a catalyst without Fe.

Project # FC-119: Platinum-Group-Metal-Free Catalysts for Polymer Electrolyte Membrane Fuel Cells

Hector Colon-Mercado; Savannah River National Laboratory

Brief Summary of Project:

The overall objective of this project is to co-synthesize highly active, low-cost non-precious-metal catalysts for the oxygen reduction reaction (ORR) by doping nitrogen-activated metal complexes into a novel nanocarbon support in a single-step process that is easily scalable and market relevant. A target of the project is to demonstrate performance of non-platinum-group-metal (PGM) catalyst prepared using a chemical vapor deposition (CVD) system to meet 6 A/cm^3 in rotating ring-disk electrode at 0.8 V in acid solutions.



Question 1: Approach to performing the work

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

- Non-PGM catalysts (such as co-pyrolized Fe and/or Co complexes [simple and macrocyclic] with polyacrylonitrile) were developed by Ernest Yeager under Ken Kinoshita of Lawrence Berkeley National Laboratory more than 30 years ago. This paper is similar and advances earlier work by inserting metals in carbon nanotubes and graphenes for better dispersion of metal as well as better conductivity and mass transport. This is a very good approach for advancing the prior state of the art. The team uses a good synthetic approach: CVD and multi-walled carbon nanotubes (CNTs) with NH_3 and Fe, which is better than high-surface-area (HSA) graphite with NH_3 and Fe; it also compared using Fe versus no Fe and found using Fe to be better, but more activity is still needed compared to Pt.
- Fabricating non-PGM using the CVD process has been suggested and deserves investigation. However, its benefit on the performance and stability is not clear. The main benefit is claimed to be its potential lower processing cost. However, non-PGM cost is quite low compared to PGM catalyst; therefore, cost should not be an important motivation. In addition, the high cost of metallic precursors used in the CVD process can cause higher cost than traditional synthesis methods. Evaluation of catalyst relies on rotating disk electrode (RDE) measurement.
- The budget for this work is very modest, so it is difficult to believe that the principal investigators (PIs) will be able to embrace all of the available techniques necessary to make the approach compelling. The CVD approach to synthesizing materials leads to materials that are very ill-defined. Transmission electron microscopy (TEM) images of these materials do not provide a complete picture of the materials' structure, much less the nature of their activity. If the PIs truly wish to use RDE measurements, they should become much more familiar with the technique before proceeding.
- A single-step, scalable process was the goal, yet the poster and slides speak a lot about as-made and surface modification. It is unclear whether this means that the process is no longer single-step. The use of Fe remains undesirable in light of the current membrane durability issues.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The processing of new materials is well controlled, scalable, and done at a low cost. The resulting catalyst materials are well characterized. The performance of these catalysts is consistent with state-of-the-art non-PGM fuel cell cathode catalysts, but still far lower in activity than supported HSA Pt.
- The accomplishments are reasonably good. It is obvious that with the CVD method, the PIs can make a "new" class of materials. The biggest issue is the method they have developed to assess the activity of these materials. With rotating ring-disk experiments (RRDE), one can do many things (e.g., loading, number of cycles, and potential window) to achieve any activity one desires. The issue of the durability of these materials is not addressed at all.
- The RRDE activity targets in activity per volume (A/cm^3) were already revised for the bigger projects of Mukerjee and Zelenay; it is unclear why they were not in this project. Allegedly higher active surface area is obtained with heat treatment, but the cyclic voltammograms (CVs) really only show an increase in carbon area, coupled with an increase in ORR activity that is not quite of the same magnitude. No explanation is given of how the area increased (only "unzipping"), or why the activity is not proportional to this increase. Furthermore, the selectivity toward water is smaller with the higher area, which is contrary to the durability target. The mechanism of ORR activity increase in the N-doped CNTs, going from 800°C to 850°C, is unclear. The Fe content does not seem to increase, and neither does the area (in this case, the activity increase is proportionally larger than the area increase).
- The progress of the project appears to be very slow. Some results (which were quite low) using the traditional synthesis method were shown. Only a small amount of results using the proposed method were shown.

Question 3: Collaboration and coordination with other institutions

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

- It is positive that the PIs have included a variety of partners (e.g., big companies and start-ups). Combining the various expertise possessed by the partners may enable the PIs to do something truly new.
- Most work is being done at Savannah River National Laboratory (SRNL), but with good input from end users such as Ballard and carbon nanomaterials and fuel cell catalyst development companies such as NanoTech Labs and Greenway Energy, respectively.
- The collaboration is appropriate for the project's size and goals.
- This seems to be mainly a single-institution project, with slight guidance and materials supplied by the industry partners. It is not obvious whether there is actual cooperation in development and planning.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **2.6** for its relevance/potential impact.

- Three things are impeding the deployment of fuel cells: cost, cost, and cost. Most of the real cost is in the fuel cell catalysts. Success in this kind of work is vital for making low-cost, effective fuel cells, and this type of work is to be strongly encouraged.
- Non-PGM catalysts provide hope of removing PGM risk from the technology, although significant challenges remain, including serious stability and performance issues. The project goals appear to focus on potential cost reduction, which should be a lower priority for this type of catalyst.
- Non-PGM catalysts are viable but still too far from applications for the DOE Hydrogen and Fuel Cells Program (the Program) to commit funding for them. This is especially true in this project, where most of the research should be considered "academic." This would be better if it were funded by the DOE Basic Energy Sciences (BES) program.

- At this stage, the relevance of the project is very modest. The PIs should be much more creative in order to make this project more relevant to DOE objectives and directions.

Question 5: Proposed future work

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

- The materials development part is about as good as it gets. With the understanding that most catalysts work is trial and error, one suggestion is for the researchers to team with a theorist to assess working catalysts and give a less empirical and more fundamentally rational plan for future syntheses.
- Most of the future work proposed is in basic research. This is good, because non-PGM materials are in this state at the moment. However, it should not be DOE that funds this work. It would be a lot more useful to generate encompassing research toward active sites versus total area versus water-transport properties. There must be more benefits than merely cost for non-PGM catalysts to make the leap toward application.
- Unfortunately, the future work is based primarily on trying to improve the morphology of these materials, rather than focusing on a specific problem—the activity of the active centers. It is very difficult to synthesize promising materials without understanding the role of components in the material that may contribute to activity and stability.
- The progress has been very slow. The project still remains in an infant stage of development. The PIs should reevaluate what they want to accomplish in the remainder of this project.

Project strengths:

- The project features a good team, with SRNL serving as the lead and conducting catalyst characterization. Most work is being done at SRNL, but with good input from end users such as Ballard, nanocarbon provider Nanotech Labs, and catalyst synthesizer Greenway Energy. Perhaps a theorist could be added.
- Evaluation of cheaper non-PGM catalysts is valuable in assessing the feasibility of application in the future. Basic research in defining core properties and determining essential components is important.
- The biggest strength of this project is the use of CVD to try to develop a new class of non-PGM materials.
- The industrial partners are a strength of the project.

Project weaknesses:

- Similar to all non-PGM catalyst development, the work seems empirical but focuses on the catalyst support rather than the catalyst itself. More fundamental inputs or rational explanations of success might accelerate progress.
- There is a lack of a clear-cut approach to how to characterize these materials and evaluate a true activity-stability relationship.
- Weaknesses include the project goals, approaches, and planning.
- This is too fundamental for the Program to fund; at the moment, this research belongs in BES.

Recommendations for additions/deletions to project scope:

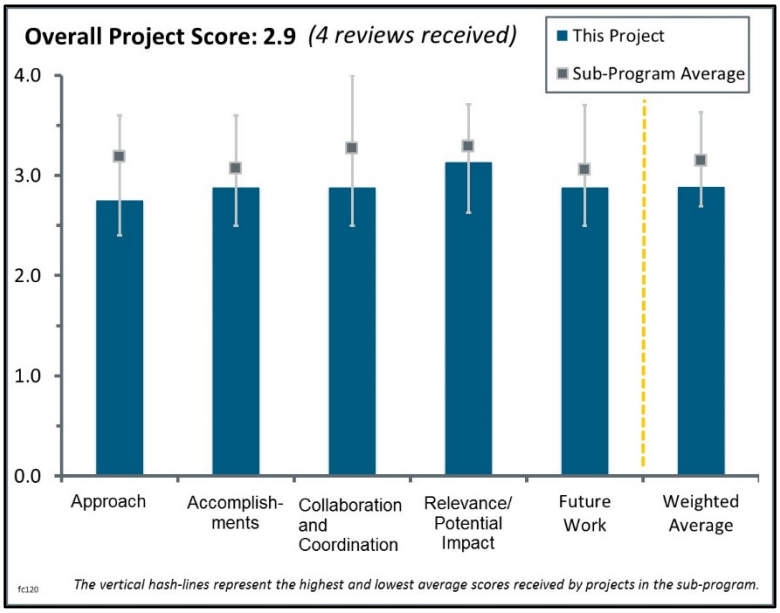
- With the project ending in three months, there is little to change in the scope. The team should scrap most of the proposed future work to really double down on the most essential aspects of the catalysts in order to improve those aspects. It may be useful to make sufficient catalyst for at least a few membrane electrode assemblies (MEAs) to check for basic properties in the MEA environment. This would assess possible trouble in integration in fuel cells (e.g., water transport, gas diffusion, and efficiency). This project should not be extended.
- The PIs should try to establish collaboration with a group(s) that has experience with much more comprehensive characterization of catalyst materials, as well as with how to evaluate the true activity of these catalysts.
- The team should think about getting input from a theorist for non-PGM oxygen reduction catalysts and expanding to non-PGM anode catalysts for hydrogen oxidation and oxygen reduction.

Project # FC-120: High-Performance and Durable Low-Platinum-Group-Metal Cathode Catalysts

Yong Wang; Pacific Northwest National Laboratory

Brief Summary of Project:

The overall objective of this project is to improve the stability of catalysts by enhancing metal/support interactions through engineered graphene for enhanced diffusion properties and by improving carbon support durability with an indium tin oxide (ITO) coating. Electrochemical evaluation using a rotating disk electrode (RDE) test station will demonstrate both the oxygen reduction reaction activity and stability, while membrane electrode assembly (MEA) testing will demonstrate the durability of the graphene catalyst.



Question 1: Approach to performing the work

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

- Moving from two-dimensional (2-D) to three-dimensional (3-D) engineering of graphene to use as a support for catalysts seems like a good idea. ITO is very stable in a polymer electrolyte membrane fuel cell acid environment, but ITO has very low conductivity. It is harder to deposit Pt selectively on a graphene support than on one where the graphene is coated with ITO.
- The approach seems excellent.
- The overall approach is reasonable, but in total, it is much too broad and ambitious for a single-year project. Little background is provided as to why ITO would promote durability improvements. It is unclear whether the 3-D graphene would result in improved transport.
- This relatively small project deposits ITO nanoparticles on graphene and uses the ITO to anchor Pt nanoparticles in order to improve the corrosion resistance of the support. ITO has been shown by several groups not to be stable in MEA operation. It is unclear why the team anticipates a different outcome from this project.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The team engineered a successful 3-D graphene and dispersed ITO well on this 3-D graphene. The researchers showed improvement in durability for the ITO-added support, but tests were done using an RDE. MEA tests are very necessary to verify these results. Other groups have shown that ITO forms hydroxylate species in MEA, so performance and durability may not be the same as seen in an RDE. Also, the presentation did not show clearly what weight percentage Pt was deposited on the support.
- The progress has been good, considering the project's size and duration. The team needs to include comparisons with commercial Pt (similar Pt particle size) on graphitized carbon to provide a reference against the state of the art.
- The results, to date, are good. However, it is disappointing that no MEA testing results are included by this time.

- Some initial results were shown using RDE. The team has not presented any MEA results or conducted any stability tests yet.

Question 3: Collaboration and coordination with other institutions

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

- There is very good collaboration between two laboratories. Pacific Northwest National Laboratory is taking a solution/wet chemistry approach to synthesize the catalyst, and Los Alamos National Laboratory is taking a vapor deposition approach. It will be nice to see the comparison between the two methods.
- The collaboration is appropriate for the project's size and goals.
- The collaboration is appropriate, considering the project's size.
- The two national laboratory partners seem to be collaborating well, but it is disappointing there are no other partners. For example, a non-cost industry partner would be an excellent addition to weigh in on the viability of this approach. Ideally, partners would include an MEA supplier and/or an automotive original equipment manufacturer that could advise on the viability and costs of the manufacturing process.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.1** for its relevance/potential impact.

- The project is very relevant to the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan. With some thorough investigation, this study could be very useful.
- Corrosion-resistant support can reduce the cost of a fuel cell system by about a few hundred dollars per system. This approach could be useful if it meets DOE targets at a modest increase in catalyst cost.
- The project is directly relevant to DOE goals.
- This project is certainly focused on addressing a key target—catalyst cost and durability; however, at this point, it is very difficult to judge the impact this project may have because no MEA testing or durability data is reported.

Question 5: Proposed future work

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

- The proposed future work is appropriate.
- The plan is appropriate; hopefully it can all be completed in the time remaining.
- Demonstrating the same improvement in MEA that is observed in RDE is a very important task. However, the presentation did not mention any efforts to understand why ITO is enhancing the stability. It is not clear if Pt that is deposited on ITO is useful, considering that ITO has very low conductivity.
- The team should evaluate the Pt-support interaction in more depth to understand the role of ITO.

Project strengths:

- Project strengths include the expertise and capabilities of both the national laboratories to carry out this research, that the team successfully engineering 3-D graphene and ITO integration, and that the project is taking two different approaches to deposit Pt.

Project weaknesses:

- Project weaknesses include that there has not been a thorough investigation in MEA yet, and the lack of understanding about whether Pt deposited on ITO is useful.

Recommendations for additions/deletions to project scope:

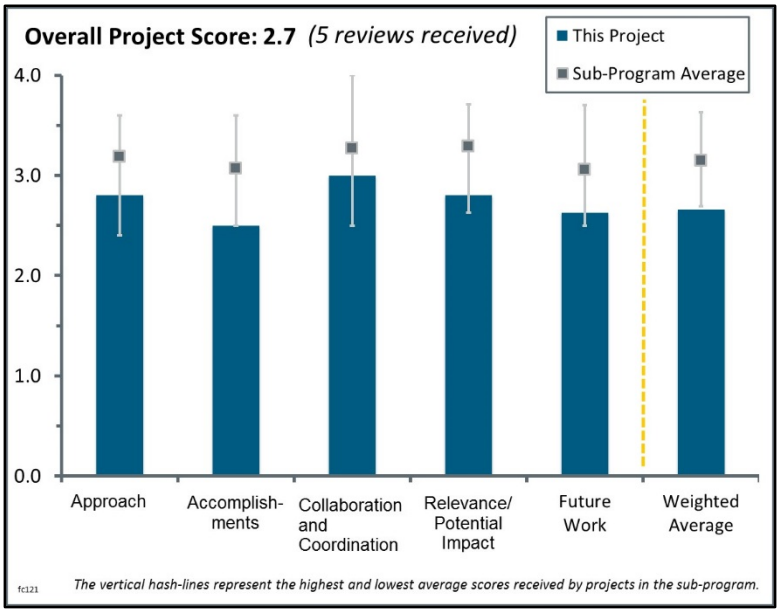
- As proposed in the future work, the team should carry out MEA testing; it should deposit low Pt weight percentage on support, then study whether similar results can be obtained.
- The team should conduct MEA testing and benchmarking against Pt/graphitized carbon.

Project # FC-121: Magnetic Annealing of Pt-Alloy Nanostructured Thin-Film Catalysts for Enhanced Activity

David Cullen; Oak Ridge National Laboratory

Brief Summary of Project:

The overall objective of this project is to explore the potential of high magnetic field annealing to produce highly active surface structures in platinum-alloy oxygen reduction reaction (ORR) catalysts. The impact of magnetic annealing on Pt_3Ni_7 nanostructured thin-film (NSTF) model structures will be characterized by rotating disk electrode (RDE), x-ray photoelectron spectroscopy (XPS), x-ray diffraction (XRD), and scanning transmission electron microscopy (STEM) testing.



Question 1: Approach to performing the work

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

- This is a very preliminary project; the viability and the relevance of the approach are unclear, but the execution of the stated work is excellent. At the same time, some of the most logical preliminary control and characterization work is lacking. Simple, well-proven, and inexpensive techniques should be pursued before more costly ones. Also, RDE tests are really not sufficient; at least membrane electrode assembly (MEA) tests are needed.
- The approach is generally effective; it uses a limited scope of parameters that are varied in a logical progression to assess the effects of these parameters on catalyst morphology and activity. It appears that the only barrier explicitly addressed to this point is activity, while durability and cost have not yet been addressed. However, the desired performance metrics or goals are not clearly indicated. While the project was unsuccessful in reproducing the morphology evolution and activity gain seen by van der Vliet (Nature Materials 2012), the researchers were limited to low-temperature annealing because maintaining the structural integrity of the support Kapton film and perylene red whiskers was a prerequisite. This was not required in the previous work of van der Vliet.
- The project was predicated on attempting to realize the benefits of annealing NSTF catalysts (reported in the literature) with an alternative method (i.e., magnetic annealing). The potential benefit is to avoid the negative impacts of high-temperature (HT) exposure to the perylene-red support. However, the project team has not been successful in achieving this—the mass activity is reduced, and what is believed to be contamination is observed. The project team needs to isolate what positive (and detrimental) results are due to HT versus magnetic interactions versus normal process development/debug, but it has not done this to date.
- The approach might sound unique, but this reviewer does not believe in the ability of magnetism to make a meaningful difference in the activity of Pt-3d transition metal alloy catalysts. Magnetism is usually considered a secondary effect when designing active catalyst materials. A much more powerful effect is the well-established temperature-induced segregation of 3d transition metals in Pt-based alloys.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- To this point, the only barrier addressed is performance—ORR activity to be specific—and every attempt to modify the morphology and grain structure through high magnetic field annealing resulted in a decrease in activity (both in terms of mass and specific activity), as compared to the as-grown NSTF catalysts. In addition, comparing the samples annealed in the same gas but with and without a magnetic field shows negligible differences in the activity. Annealing in hydrogen in a 9T magnetic field shows a marked increase in the grain size; however, this also likely occurred during annealing in hydrogen at 0T. Because of the limited annealing temperatures, the ideal (111)-like, smooth Pt-segregated surfaces shown by van der Vliet et al. (2012) were not obtained. To be fair, the NSTF alloy composition used here is much richer in Ni than that used by van der Vliet et al., which likely has some impact on the annealing process.
- The observed ECA, specific activity, and mass activity are lower than the catalyst as received. XPS suggests that carbon contamination is a contributor. STEM may show some interesting results, but the project team needs to isolate what is due to the magnetic interaction and what is due to temperature annealing. However, more importantly, the project has not demonstrated the benefit of this approach.
- The systems studied are not close to being characterized well enough for the principal investigators (PIs) to be able to provide any true insight into the role of magnetism in enhancing activity.
- There seems to be a lot of characterization, but not much analysis of the results.

Question 3: Collaboration and coordination with other institutions

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

- The partners worked well together.
- The project team is coordinating with the right parties, including both 3M (developer of NSTF) and the original investigator who reported the annealing benefits. In addition, the PI and the National Renewable Energy Laboratory have the expertise to address the characterization. Perhaps more assistance is required in determining the root cause(s) of the contamination. However, it appears the team has the expertise to do so.
- This is a simple project, but apparently it features good division of labor.
- The group of collaborators listed is very good, but the lack of a critical approach toward evaluating the role of magnetism is surprising.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **2.8** for its relevance/potential impact.

- The concept, if proved out, could contribute positively to the programmatic goals, if the following conditions are met: (a) it results in improved catalyst performance, (b) it can avoid negative effects such as support contamination, (c) the process is amenable to scale-up and cost-effective manufacture (the presentation did indicate the concept can be used with rolled goods, which is positive), and (d) it can be shown to be cost effective (the economics of high magnetic field annealing are unclear). However, the researchers have not gotten past (a) or (b) at this time.
- Creating a dominant surface orientation tailored toward the low energy (111) structure of Pt alloys is known to greatly enhance the activity of nanoscale catalytic ORR materials. If this ideal orientation can be translated to the NSTF catalyst architecture, it could greatly enhance the ORR activity of these catalysts, which already exhibit high specific activities for nanoscale catalysts. The use of high-transition-metal-content NSTF is somewhat defeating the purpose because even if grain size is increased and surface structure is smoothed to a relatively low index vicinal (111) surface, post-annealing de-alloying will quickly roughen the structure, nullifying any improvements associated with this annealing procedure. As it turns out, the nanoporous geometry is highly desirable because it greatly improves the electrochemical

surface area of NSTF, limiting the amount of buried precious metal and consequently improving the mass activity. For this approach, however, the homogeneity of the initial alloy, rather than the surface morphology or compositional profile, is the most important aspect of the initial alloy. In addition to surface structure, improving the alloy homogeneity should be a goal of this project because the as-grown alloys are very inhomogeneous, even when using co-sputtering or sputtering from an alloy target.
- While the execution and relevance are unclear, the concept of annealing catalyst has been proven before, demonstrating there is at least potential.
- This magnetic annealing approach is not the best direction to pursue. At the moment, it is very difficult to recommend this approach as the future for making a new generation of catalysts.

Question 5: Proposed future work

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

- The project focuses on RDE evaluation. The approach to incorporate in full-size MEAs would be added value.
- The future efforts are focused on adjusting the annealing conditions and alloy composition, conducting durability tests, and performing more modeling efforts. It is somewhat useless to continue with durability and post-mortem characterization efforts if no activity improvements can be obtained beyond that of the as-grown material. The future efforts should be focused on improving structural ordering and the effect of the magnetic field by using alloys with higher magnetic moments, improving homogenization of alloys at limited annealing temperatures, and limiting catalyst deactivation due to support evaporation and re-deposition on the catalyst surface.
- The proposed future work is very broad and clearly would not all be addressed during the project's remaining time. During this remaining time, the project team should focus on the following:
    - Better process control (i.e., identifying and correcting the cause of contamination).
    - Identifying/isolating the impact of magnetic annealing versus the temperature-annealing effect. The researchers should try magnetic annealing at lower temperatures and with a greater range of magnetic fields (including higher Tesla, if feasible). It would be good to know the actual effects of magnetic annealing without the (believed to be) detrimental temperature effects.
- The relevance of RDE on this effort is unclear, and the future work plan seems to be too general; it mirrors the work to date and does not seem to reflect any conclusions from the work done so far.
- The authors propose to use Pt-Co, which is known to have paramagnetic properties and might seem like a good direction to pursue. However, how successful this direction will be is questionable.

Project strengths:

- This project addresses the need to optimize the surface structure of the NSTF catalyst architecture because it is inherently rough because of the deposition process. This structural reorientation as a consequence of annealing is limited to relatively low temperatures because of the presence of a polymer and molecular solid support. The use of magnetic field-induced annealing could have a significant effect on the reorientation because of the magnetic-field-induced changes of structural ordering in these magnetic thin-film materials. The integration of high-throughput materials synthesis, annealing, catalytic testing, and extremely high-quality microscopy analysis is very efficient in identifying improvements and quickly informing/guiding the project progression.
- The project's strengths include its strong technical skills and its promising preliminary results that show magnetic annealing effects.
- The biggest strength of this proposal is the group of collaborators that has been assembled.
- The diagnostics and characterization capability of the PIs are strengths of this project.

Project weaknesses:

- The modeling efforts are useless because significant computational results in combination with well-defined experimental results have clearly identified the active sites for ORR. The magnetic moment of Pt_3Ni_7 appears to be too small for magnetic field annealing to have any significant impact. Perhaps the

usefulness of this approach is limited to a delicate balance between the temperature required to induce surface diffusion to drive preferential orientation and the curie temperature of the alloys (i.e., demagnetization).

- The amount of work seems very limited, and conclusions are therefore difficult to draw. There seems to be a lot of characterization, but not much analysis of the results. The economics of magnet operations are unclear.
- Weaknesses include the control of the process (i.e., contamination) and the experimental approach (the team should take a more systematic approach to identifying magnetic field effects).
- Unfortunately, the magnetic annealing approach is not the right direction to pursue.

Recommendations for additions/deletions to project scope:

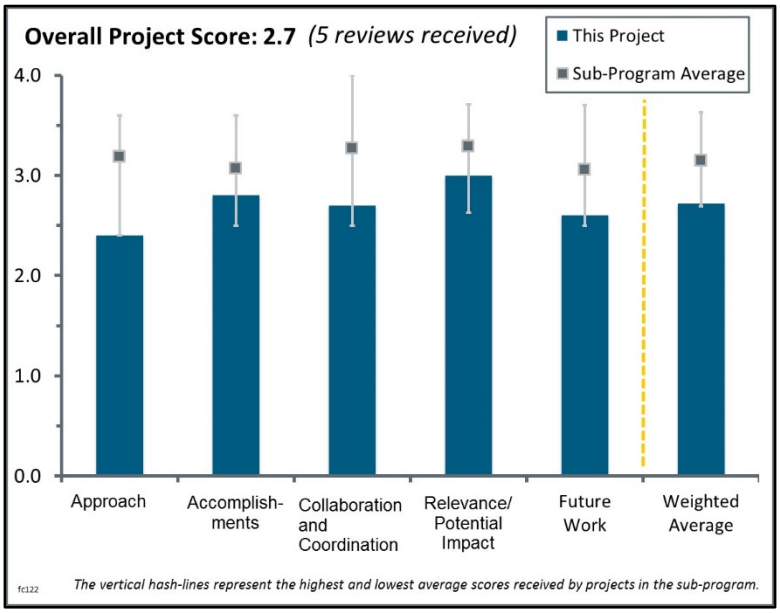
- The researchers should use alloys that are not as rich in the less noble component because they are very susceptible to de-alloying and porosity evolution, which would be counterproductive to the goals of this project. They should try using some alloys with larger magnetic moments, such as Pt_3Fe. These may be more greatly affected by the magnetic field. If they are continuing with high-transition-metal-content alloys, they should also focus on alloy homogenization. The researchers should limit modeling efforts because the ideal ORR active site has already been thoroughly studied in previous computation studies. They should also identify strategies of mitigating carbon layer formation on the precious metal catalyst surface due to support decomposition during the annealing process. Alternatively, they should develop techniques for facile removal and “activation” of the catalyst post annealing.
- The team should focus on good preliminary work instead of sophisticated analysis—for example, XRD refinement to augment aberration-corrected transmission electron microscopy (ACTEM) and thermogravimetric analysis (TGA) instead of a superconducting quantum interference device (SQUID).
- The team should focus on addressing the problems. It should deemphasize modeling until benefits are shown. In addition, durability should be contingent on successful mass activity results.

Project # FC-122: High-Conductivity, Durable, Anion-Conducting Membranes

Tom Zawodzinski; Oak Ridge National Laboratory

Brief Summary of Project:

The overall objective of this project is the development of highly conducting, durable anion exchange membranes (AEMs) and their demonstration in fuel cells. To address this goal, Oak Ridge National Laboratory will leverage its experience in preparing AEMs for zinc-air batteries with high conductivity; new findings regarding the durability of these membranes and how degradation pathways can be controlled; and its capabilities for developing and coating electrodes.



Question 1: Approach to performing the work

This project was rated **2.4** for its approach.

- The high-throughput approach may be beneficial for membrane synthesis, but details regarding what systems are being investigated are not provided. The principles or theory guiding the high-throughput effort have only been vaguely described, and not with enough detail. Without some description of the guiding principles or hypotheses about what will improve the properties of interest, there are simply too many possible combinations to investigate, and high throughput becomes searching for a needle in a haystack. It is not possible to determine whether the approach is applicable to alkaline exchange membranes if the basic chemistry of the polymer systems being investigated is not disclosed. The stability of the polymer backbones, the stability of the cationic functional group, the potential for high conductivity from the functional group, the likely mechanical properties, or even whether these systems have already been investigated or developed cannot be judged with the information provided. The project takes a two-pronged approach to help reduce risk: traditional AEMs and KOH-soaked cross-linked loaded membranes (CLAMs). It is not clear what the benefits of the CLAM approach are, and how the approach will improve on current alkaline fuel cells (AFCs). This approach does not appear to provide the same benefits as conventional polymeric AEMs. It is not clear that this approach will address carbonate formation in AFCs; it seems likely KOH/H_2O uptake in a polymer medium will still result in potassium carbonate formation and decreased conductivity with time. It is also not clear how this approach will prevent loss of KOH under some operating conditions.
- The general approach of tailoring a cross-linked AEM with different cross-linkers, cationic groups, and polymer backbones is reasonable, but none of the chemistry has been disclosed, so it is difficult to provide a thorough assessment of the approach. It is a concern that the KOH would not remain in the membrane during extended fuel cell testing, which would be a durability issue.
- The approach of the polymer synthesis is not clearly given, so it was hard to judge the merit of the approach. The approach has been explained in very generic terms, most likely for intellectual property (IP) protection. However, the team has just mentioned the key words, such as “proto”-functionalized backbone, cross-linking agents, additives, and blending polymers, so it is not possible to understand the actual approach taken.
- The approach seems reasonable in concept, but it is difficult to evaluate without discussing the specific materials. The need to protect IP is understandable, but one cannot give a rating higher than “satisfactory” based on what is publicly discussed to date. The U.S. Department of Energy (DOE) should not view this as a “neutral” rating at this time.

- While the conductivity of the AEMs is high, the area-specific resistance is also high due to the thick membranes. The team needs to study the impact of membrane thickness on performance and durability.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The project has been in place for less than a year and is 50% complete. In this time, the researchers have prepared more than 40 distinct compositions. In this short time, they have achieved some impressive conductivity results for their CLAMs. The reported initial conductivity of 0.8 Siemens (S)/cm is higher than recently reported values for the maximum obtainable with KOH solutions of 0.65 S/cm. It is unclear how much KOH is in these membranes and under what conditions the conductivity was obtained, but one would not expect the conductivity to exceed that of a KOH solution. Mechanical properties were not investigated and stability toward electrolyte loss has not been investigated. If the CLAMs are saturated with KOH, conductivity will be dominated by the KOH, and one would expect the conductivity to be the KOH conductivity X the volume fraction of the KOH phase. The important aspects will then be the stability of the KOH-CLAM system, and whether the CLAM immobilizes the KOH. No results were shown for tests investigating the stability and ability to immobilize the KOH. No results were shown for more traditional AEMs. The poster claims "Over 40 distinct compositions studied." For a high-throughput approach, 40 distinct compositions is not very high throughput.
- The team has met the conductivity and resistivity goals for the membranes. The team has claimed that the CLAMs meet targets for conductivity (>100 mS) at lowered relative humidity (50%), but no data was presented to substantiate the claim. Disclosure of some performance data could have helped reviewers understand the merit of these AEMs.
- Several conductivity values were reported that are very impressive. It will be good to learn more about these materials. The reviewer was concerned that the CLAM approach may not be viable. The permanence of the KOH solution is questionable. The stability of these materials needs more attention.
- Preliminary conductivity and area specific resistance (ASR) targets have been met, although an ASR of 0.1 ohm-cm^2 is not a reasonable goal. For automotive applications, an ASR around 0.02 ohm-cm^2 is needed. Also, it is not clear why "no cathode precious metal" is relevant for an ASR target.

Question 3: Collaboration and coordination with other institutions

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

- The team consists of a good mix of national laboratory, academic, and industrial partners. The collaboration with Eastman, Los Alamos National Laboratory, and the National Renewable Energy Laboratory is advantageous to the team.
- The principal investigator (PI) mentions working with industry to enable scale-up. There is no apparent coordination with other AEM projects. There is no mention of collaboration or interactions with the major players in the AFC or alkaline electrolyzer areas.
- Even though this is a small project of short duration, it would be nice to see more substantial collaborations. It is good to see technology transfer from related zinc/air research.
- There is no evidence of collaboration, because this is an early stage of the project.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.0** for its relevance/potential impact.

- The project is relevant to the objectives of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan. The activities are aligned to DOE's goal to address the commercial

barriers such as performance, cost, and durability. The focus of the project is to develop and demonstrate chemically stable hydroxide-conducting AEMs.
- This project is one of several AEM projects that appear to be in the exploratory stage. This is important work in defining viable chemistry for this type of membrane/MEA.
- Polymer alkaline electrolyte membrane work is relevant to both hydrogen production and fuel cells. It is not clear what the expected advantages of the CLAM are, compared to existing alkaline electrolytes, or what targets or shortcomings they address.
- It is unclear how this project addresses high-level DOE Hydrogen and Fuel Cells Program targets. Even if the researchers are successful in meeting their objective of 0.1 ohm-cm^2 ASR, the membrane electrode assemblies (MEAs) will still not meet power density requirements for automotive applications. Also, without any disclosure of the chemistry or synthesis processes, there is no way to assess the potential cost implications.
- The overall performance of an MEA with AEMs is unclear.

Question 5: Proposed future work

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

- The future work is aligned with the project proposal and the remaining time left for the project.
- The details of the durability testing and how the researchers will achieve their deliverable on determining the relationship between AEM fuel cell durability and accelerated protocols is not provided. So far, the focus seems to be on conductivity loss, which is certainly important. Tailoring mechanical properties is also important, but there is no information provided on the status of the mechanical properties or what properties are required for sufficient durability. Although not included in the presentation, the PI did say the researchers would do thermal stability testing, which is crucial for AEMs.
- The future work appears focused on traditional AEM membranes using the high-throughput approach. This is scheduled as a one-year project; the future work appears to be more than can be done in the remaining three months with the budget provided.
- The future work includes a lot of very difficult tasks: catalyst-coated membrane making, degradation mechanisms, catalyst making, modeling, etc. One fears that the size and time frame of this project will not allow completion of these tasks. One wonders what tasks will realistically be completed by the end of the project.
- The future work should consider using membrane reinforcement to reduce the thickness to less than 25 µ, as well as reducing ASR to <20 milliohm-cm^2.

Project strengths:

- The team is composed of respectable research organizations with adequate expertise. Overall, the team is equipped with the knowledge base, resources, and industry/academia/national laboratory mix required for the project to be successful.
- This project is screening a large range of membrane chemistries and designs, even though the details have not been disclosed, pending IP protection. The researchers have achieved reasonable conductivity with thick membranes.
- This project appears to have made several AEMs with excellent conductivity. The presentation described a very broad number of types of synthetic compounds. This may allow evaluation of a wide range of membranes and selection of an optimum option for performance and durability.
- The project features a solid fuel cell/membrane background.

Project weaknesses:

- The lack of disclosure of specific chemistry, while understandable, makes a full evaluation difficult. The number of chemistry options seems very broad and may need to be more focused in future work. The durability of the cationic functional groups is a key concern for all AEMs at this point. Without specific knowledge of this functional group or the additive groups, this question remains.

- The ASR target may be too high. There is no plan laid out for durability testing. The lack of disclosure of chemistries makes a thorough review impossible. It would be nice to gain fundamental learning about which options provide best conductivity, stability, and durability to provide other developers with guidance in their work.
- The project does not disclose enough about the chemistry to be able to determine whether the ideas have merit. The project utilizes a high-throughput synthesis, but it does not have high-throughput analysis or testing in place yet. This will bottleneck the approach at the testing stage.
- The team has spent significant time on membrane development. It could have conducted some AFC tests to demonstrate the merit of some of these membranes.
- The presentation material lacks the necessary details for one to clearly understand the project's progress. The project also lacks partners to help make good membranes from this polymer.

Recommendations for additions/deletions to project scope:

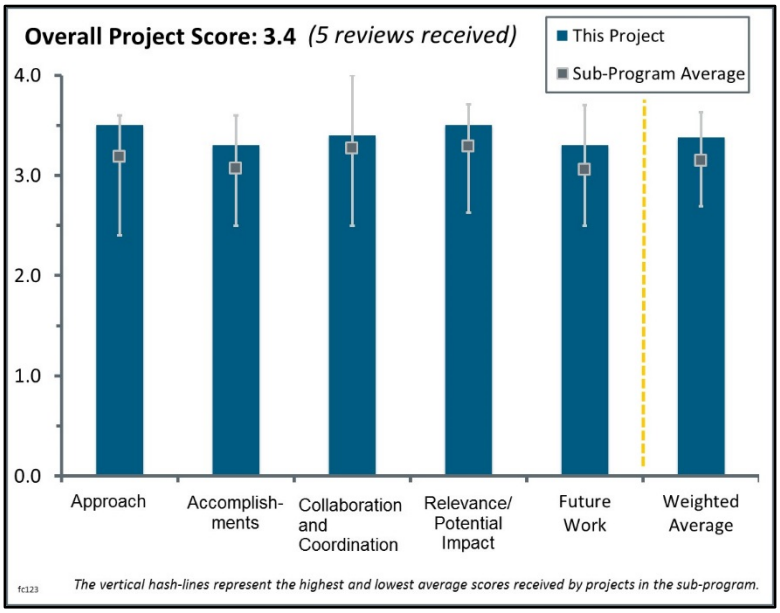
- The team should focus on making thinner membranes (<30 microns) so that the AEMs can hopefully approach automotive ASR targets. It should also start testing for thermal stability. It should test to see whether AEMs lose KOH, and therefore conductivity, during extended fuel cell operation in wet conditions.
- The future work seems much greater than the timing and funding for this project would allow. While all the tasks are relevant, the team should identify a few achievable milestones to be completed by the end of the project.
- The researchers should conduct durability testing to determine whether CLAMs retain the KOH and their high conductivity.
- The project team should add a membrane original equipment manufacturer.

Project # FC-123: Advanced Hydroxide-Conducting Membranes

Yu Seung Kim; Los Alamos National Laboratory

Brief Summary of Project:

The objectives of this project are to develop (1) chemically stable hydroxide-conducting anion exchange membranes (AEMs), (2) solvent processable perfluorinated ionomers, and (3) modeling approaches to demonstrate high-performance/durable alkaline membrane fuel cells (AMFCs). In fiscal year 2015, the perfluorinated ionomer with alkyl amide linkage was designed to increase amide stability under basic conditions; fuel cell performance measurement using these ionomers is ongoing.



Question 1: Approach to performing the work

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

- The approach is excellent. This is the best "alkaline fuel cell membrane" this reviewer has seen to date. In fact, it is the best AEM this reviewer has seen to date. It is better than neosepta, etc. The approach to stabilize the decomposition of the ammonium site looks excellent and is the vital step. The team achieved good stability by eliminating the ether linkage. There is no added OH^- and there is still good conductivity. This should lead to an OH^- CO_3^{-2} conducting membrane with sufficient stability and lifetime to test whether it is true that there are better, cheaper, and more durable fuel cell catalysts available for oxygen reduction and hydrogen oxidation using polymer electrolyte membrane (PEM) fuel cells based on hydroxide (and/or carbonate) rather than proton conduction.
- The Los Alamos National Laboratory (LANL) approach targets the performance and durability barriers for AEMs. The approach specifically targets durability by improving the chemical durability of the cationic functional group and of the backbone polymer. The cationic functional groups being investigated are more resistant to degradation by SN_2 (substitution nucleophilic second-order) reactions than benzyl trimethyl ammonium groups. Also, replacing the aryl-ether linkage with ether-free backbones will prevent ether cleavage in alkaline solutions. The approach to look at multiple backbone chemistries provides reduced risks as well as the potential to discern the impact of phase segregation and electronic effects on AEM performance. The inclusion of modeling efforts to understand CO_2 interactions and water distribution in an MEA should prove beneficial in optimizing operating conditions for AEM fuel cells and minimizing problems with water management. While investigating multiple systems reduces risks, it also dilutes resources. At some (future) point, a down-selection to the most promising candidate would accelerate progress on that option.
- The project seeks to make stable, mechanically robust AEMs and chemically stable ionomers for AEM fuel cells. Chemical stability is achieved through non-polar backbones and advanced cations. New ionomer dispersions are being developed for electrode integration. A modeling approach is being used to understand transport in fuel cells and operational parameters.
- The approach of using alkyl amide, phenyl guanidinium to enhance the stability of the amide linkage is a good way to increase the stability of AEMs. The modeling approach to understand the performance is also a good approach.
- The rationales for the chemistries and structures are presented on slides 7 and 8. The technical and cost targets and their bases are inadequately discussed. The CO_2 issue is recognized and is being addressed.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The project has made good progress. The project has demonstrated the improved stability of the new membranes compared to the control in ex situ testing, and the team has met its milestone for stability for <10% loss after 500 hours in 0.5 M NaOH at 80°C with two membrane types. The team has demonstrated improved stability of the phenyl alkyl amide linkage compared to the phenyl amide linkage in high pH conditions via Fourier transfer infrared spectroscopy (FTIR)studies. Initial fuel cell performance tests indicate hexamethyl ammonium functionalized poly(phenylene) AEM is the best-performing hydrocarbon system under investigation. The area-specific resistance (ASR) of the "AR"-series membrane had a substantial dependence on current density, while the other membranes' ASR had very little dependence on current density.
- The team has achieved good progress in a short time period by establishing seven different membrane synthesis chemistries and finding the weakness in four of them. The team has also successfully determined the "MRH"-series membrane ionomer chemistry as one of the potential candidates for AEMs.
- The project team stabilized the ammonium group, stabilized mechanical properties, and modeled and tested CO_2 effects. It performed excellent impedance work.
- A series of rigid and flexible polymers have been synthesized. The ASR target for the year has been reached. A fuel cell at 60°C shows some promising stability. There is good integration of ionomer with the membrane for some good, improving fuel cell performance. The modeling is showing that input CO_2 amounts will need to be lower than 50%.
- Progress toward improving conductivity, alkaline stability, and mechanical properties is presented and is respectable, but targets are absent, so it is not clear if the progress is good enough.

Question 3: Collaboration and coordination with other institutions

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

- The team consists of a good mix of national laboratory, academic, and industrial partners. The interaction with Solvay, which manufactures PEM fuel cell membranes, is very encouraging.
- The project's full spectrum of team members covers the necessary expertise and project requirements. Collaboration and cooperation were necessary for the project team to have achieved this level of progress.
- The collaboration and coordination are excellent and extensive. All the bases are covered.
- The collaborations appear to be working well, especially between Sandia National Laboratories and LANL and between LANL and Lawrence Berkeley National Laboratory. Including Solvay gets an industrial supplier involved. Participation by or collaboration with commercial alkaline fuel cell or alkaline electrolyzer companies would be beneficial.
- There is good collaboration between national laboratories and one university. Industry involvement is mostly related to material supply. The project needs more fundamental university input. Including a fuel cell component supplier and an original equipment manufacturer in a relevant end-use area would also be beneficial.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.5** for its relevance/potential impact.

- The project is relevant to the objectives of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan. The activities are aligned with DOE's goal of having chemically stable, hydroxide-conducting AEMs.
- The development of AEM membranes has a large potential impact—this would be an enabling technology for alkaline fuel cells that can effectively utilize inexpensive non-platinum-group-metal cathode catalysts.

These fuel cells could have large cost savings compared to current PEM technology. The proposed work addresses the main problem with current AEM membranes—durability.
- This is needed to give a clear test of the hypothesis that alkaline membranes have something to offer.
- This is hard to evaluate because the Hydrogen and Fuel Cells Program does not yet have well-thought-out goals for AEM fuel cells. Certainly the researchers are addressing durability and performance targets that are relevant to AEM fuel cells; it is not clear whether cost targets will be addressed before non-precious-metal catalysts are integrated into AEM fuel cell MEAs.
- The relevance is clear. The impact is unclear without better definition of technical goals and the status of the project.

Question 5: Proposed future work

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

- The project is scheduled to end soon. The proposed work is appropriate for the time remaining. The work focused on the perfluoro alkyl amide polymer dispersion and AMFC testing should be the priority. Work on incorporating the gas diffusion layer and flow channels into the AMFC model is of lower priority at this time.
- The future work is aligned with the project proposal and the remaining time left for the project.
- In particular, the long-term testing comment is good; this has been missing.
- In the remaining time for this one-year project, the researchers are wise to finish polymer characterization so that papers can be finished. Most of the other tasks seem like appropriate loose-end issues that should be completed so that the project will provide the maximum useful information.
- The project is over on July 31, 2015. Cell testing is the most important aspect.

Project strengths:

- The team is composed of respectable research organizations with adequate expertise. Overall, the team is equipped with the knowledge base, resources, and industry/academia/national laboratory mix that is required for the project to be successful.
- The variety of chemistries studied should provide some valuable clues regarding what polymer structures and properties are best incorporated into AEMs.
- Project strengths include that it is investigating non-polar polymer backbone and developing ionomers.
- The project features a great team and a good approach; more of this is needed.
- This project features wide collaboration.

Project weaknesses:

- The project does not have enough resources to do justice for all of the polymer systems studied. It needs more proof that the novel cations will survive in the ionomers. There is not enough emphasis on the challenge of >80ºC and low relative humidity (RH) operation, where these cells will eventually have to operate.
- The team should focus on conducting more AEM fuel cell testing to determine the merit of its membrane in situ.
- Definition of technical targets is needed.

Recommendations for additions/deletions to project scope:

- It is relatively inexpensive to reduce ambient CO_2 to fairly low values; researchers should consult with industry to determine a feasible CO_2 input level instead of guessing. Researcher should also begin to think about non-precious-metal catalysts; there is no point to AEM fuel cells unless non-precious metals are integrated or more complex fuels are utilized.
- The researchers should do the long-term testing of fuel cells with the best membranes right away. If they are truly stable OH^- conductors, then these membranes may have significant use in “bipolar membranes”—a competitor to reverse osmosis for low-salt water purification. One can remove CO_2 by passing CO_2-free gas and monitoring that water pH goes from 5 to 7.

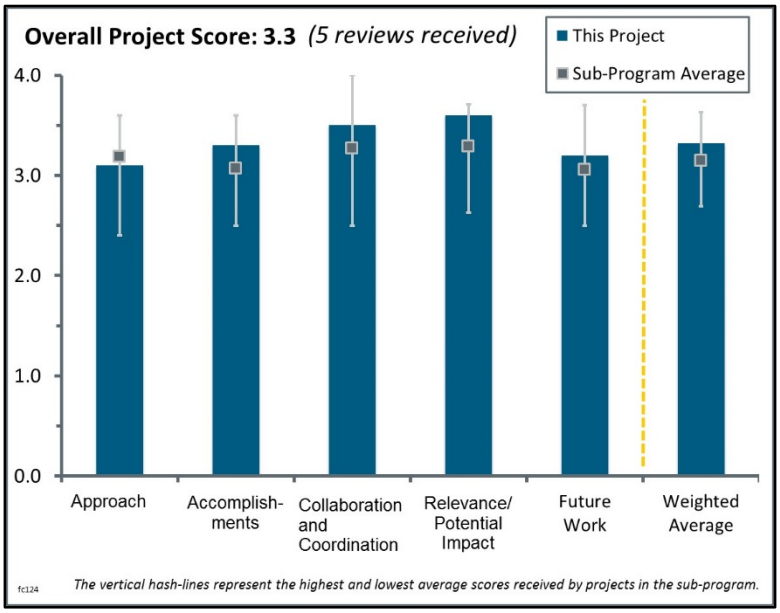
- It would be useful for the team to conduct more mechanical property testing and water uptake and swelling tests. While RH cycling may not be important in electrolyzer applications, in automotive fuel cell applications, mechanical stresses on the MEA due to swelling/shrinking may be an issue.

Project # FC-124: High-Temperature and Low-Humidity Membranes

Cy Fujimoto; Sandia National Laboratories

Brief Summary of Project:

The objectives of this project are to (1) develop a hydrocarbon (HC) membrane that can operate with minimal resistance losses (0.05 ohm-cm^2) at 120°C and humidity ranges between 30% and 50%, (2) fabricate and test membrane electrode assemblies (MEAs) at 120°C and humidity ranges between 30% and 50%, and (3) perform durability testing of MEAs at 120°C and 50% relative humidity (RH).



Question 1: Approach to performing the work

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

- Sandia National Laboratories (SNL) are leveraging their very well-thought-out sulfonated Diels-Alder polyphenylene (SDAPP) chemistry. An unexpected consequence of applying modified SDAPP to a redox flow battery project was the discovery that the acid content of the film could be dramatically increased. If SDAPP could be made to have high proton conductivity at high temperatures (HTs) and relatively low RHs, it would also potentially meet the cost and durability targets. In short, this is an excellent extension of previously funded U.S. Department of Energy (DOE) work.
- The work is focused on the cost barrier and the area-specific resistance (ASR) target. An HT, low-RH proton exchange membrane would decrease demands on the thermal management system and humidity management for the fuel cell, allowing for smaller balance of plant (BOP) and lower system costs. However, the membrane must be able to meet durability targets as well. In particular, mechanical durability during wet-dry cycling must be met, and this project should consider some effort in addressing this target as well. The approach to utilize block copolymers with sulfonic acid substituted polyphenylenes has a high probability of achieving high conductivity at HT and low RH. Previous DOE Office of Energy Efficiency and Renewable Energy–funded work with sulfonic acid substituted polyphenylenes resulted in high proton conductivity, but those materials had poor mechanical properties (they were brittle and had problems with swelling and strength at high RH). Utilizing the Diels-Alder reaction for the polymerization to give a distribution of polymerization at the meta and para positions, combined with the addition of a hydrophobic block, should help overcome the brittleness. The block copolymer approach may aid the swelling and solubility issues observed for the previous polyphenylenes. The project-specific membrane milestones and final targets are unclear. The project ASR milestone of 0.06 ohm-cm^2 is significantly less aggressive than the DOE ASR target of <0.02 ohm-cm^2 at 120°C and water pressure of 40 kPa (~20% RH). The team should aim at meeting all the membrane targets in the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan (MYRDDP) or U.S. DRIVE Partnership Fuel Cell Tech Team Roadmap simultaneously.
- Membrane making is difficult because it involves synthesis and processing. The approach, for better or worse, was restricted to the polymer backbone to improve membrane properties. The good part is that various synthetic approaches were successfully used to develop an aromatic (polyphenylene) polymer backbone for a sulfonic acid membrane with good initial chemical thermal and mechanical stability (high glass-transition temperature [Tg]) and high conductivity. A short-term test is 2,500 hours; a full test is 50,000 hours. One-hundred hours is only an initial characterization. The bad part is that because this is a hydrated sulfonic acid membrane, this fuel cell membrane will have all the issues found in any acid membrane. It requires water (humidification and pressurized gas feeds) for acid ionization ratio and

conductivity, and water affects its performance. This kind of membrane, using water, will suffer Pt oxide formation and therefore low cathode performance (high air cathode overpotential). Therefore, although this polyphenylene fuel cell membrane demonstrates a very good polymer backbone, it is only a first step to another membrane. This specific type of membrane, itself, is not sufficient for commercialization of fuel cells.

- The approach of following the concept disclosed in U.S. Patent 8,110,636 B1 and fine-tuning it to fit the need is good. However, given the peroxide instability of SDAPP, it is very interesting that the team chose to pursue the route to have SDAPP as the ionic domain for the HT membrane. One expects that under an HT of 120°C, the peroxide degradation will be more in SDAPP.
- Hydrocarbon membranes have long suffered from poor conductivity at low RH. This approach appears to address this issue and may be a path to a competitive membrane for perfluorosulfonic acids (PFSAs).

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The project has made good progress in the short amount of time it has been active, and with the limited funds available. Conductivity at 120°C and ~30% RH is almost double that of Nafion. Performance in an MEA (at 80°C, fully humidified) is reasonable, but performance under low RH and or HT has not yet been demonstrated. The conductivity of these membranes appears to have a higher RH dependence than Nafion does at 85°C. This may suggest a larger range of water uptake and swelling, which has negative implications for durability. Minimal results for water uptake or swelling have been provided, but the uptake reported (150%) is quite high and suggests swelling may be an issue. Water uptake/swelling tests—in particular, cyclic swelling tests (using the prescribed mechanical durability protocol)—need to be done.
- The synthetic approach appears to address two issues with previous HC membranes: the rigid rod nature of the polyphenylene backbone and the sulfonic acid density. The 120°C, 30% RH conductivity is impressive. The block copolymer approach is important in overcoming the traditional shortfalls with previous HC membranes.
- One ion-exchange capacity (IEC) of the modified SDAPP has shown a proton conductivity of twice that of Nafion at 120°C and 30% RH, and the ASR is 0.06 ohm-cm^2 for a 30 µ film—a very promising result. Oak Ridge National Laboratory (ORNL) has made MEAs with SDAPP, but it has yet to make MEAs with the modified SDAPP, although this work is about to begin.
- The team has made good progress. However, the team should consider Fenton's test for its new block copolymer membrane (slide 10) to understand the feasibility of SDAPP chemistry for an HT membrane. It is good that the team is exploring the use of this membrane in vanadium flow battery technology, where the peroxide degradation is not an issue.
- By focusing on developing a backbone, the team mostly succeeded in what it set out to do.

Question 3: Collaboration and coordination with other institutions

This project was rated **3.5** for its collaboration and coordination.

- The collaboration among the partners appears to be good. Including the Automotive Fuel Cell Cooperation (AFCC) in the team should keep the project focused on fuel cells and automotive-relevant conditions. It appears some leveraging of work is being done for flow batteries on similar membranes.
- The team consists of a good mix of national laboratory and industrial partners. Working with DOE contractor Sanjiv Malhotra is a good avenue for technology transfer.
- The team was "lean" but effective. The project features synthesis at SNL, MEA fabrication at ORNL, and testing at ORNL and AFCC.
- The project has good collaborations with other national laboratories and with AFCC.
- The collaborations are excellent and involve two national laboratories and a laboratory representing a consortium of original equipment manufacturers. It would be beneficial to add a university partner to dig deeper into the science of these new materials that might help to explain why the higher-IEC material had a lower conductivity than the lower-IEC material.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.6** for its relevance/potential impact.

- The project is relevant to the objectives of the MYRDDP. The activities are aligned with DOE's goal of having HC membranes that can operate with minimal resistance losses at 120°C and humidity ranges between 30%–50%.
- This project is clearly geared toward meeting the performance, cost, and durability targets for membranes in the MYRDDP.
- This is excellent work, but improving a sulfonic acid membrane is evolutionary, not revolutionary, and all the problems of catalysis remain. However, polymer backbone development is one of the core issues in making membranes, and the team achieved excellent development and demonstration of a good polymer backbone that could have a place as a component in a successful fuel cell membrane.
- The project is relevant and addresses DOE area-specific resistance targets for fuel cell membranes and the desire for membranes that can operate at 120°C and under low-RH conditions. The impact could be high if successful, because a membrane that could operate at 120°C at 40 kPa water pressure would allow one to remove the humidifier and significantly reduce the size of the radiator, reducing BOP and system costs. However, the project also needs to address durability concerns with these membranes—in particular, how the membranes handle RH cycling and chemical durability.
- The performance at temperatures above 100°C could be very significant.

Question 5: Proposed future work

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

- Testing these materials at a greater range of operating conditions will be important in order to make a complete comparison to the incumbent PFSA technology. Realistic membrane thicknesses of 15–20 µ will be important for both performance and durability assessments.
- The proposed work at HT and low RH is the right thing to do—adding time testing to evaluate durability seems needed. The workers seem to know what is necessary to reach the goals: innovation on the pendant ionic group seems essential.
- The proposed future work is a logical extension of what has been accomplished so far and should yield enough data to facilitate a decision on whether future—hopefully much larger—funding is allocated for this important work.
- The future work is aligned with the project proposal and the remaining time left for the project.
- The proposed future work still appears to focus on the ASR target. Mentions of durability consider thermal stability, as well as durability at 120°C and 30% RH under constant conditions, but the presentation did not mention the variable cyclic conditions and RH cycles found in automotive applications. Durability under cyclic operating conditions (specifically RH cycling) needs to be brought in. The team should also consider looking at supported membranes, as well as the other DOE membrane targets such as crossover.

Project strengths:

- The team is composed of respectable research organizations with adequate expertise. Overall, the team is equipped with the necessary knowledge base, resources, and industry/national laboratory mix for the success of this project.
- It is good to see work continuing in the HC membrane field. This project is thoughtful and builds on the previous work of Jim McGrath by using block copolymers and on the work of Zawodzinski and Hamrock, who identified the need for closely packed acid groups. The focus on the very difficult operating conditions at 120°C and 30%–50% RH is a good approach.
- The concept is flexible and takes advantage of previous work that showed the potential of polyphenylenes and block copolymer approaches. Initial results are promising and show the researchers can achieve good conductivity at 120°C. The work leverages ongoing work in flow battery membranes.

- The team modified the chemistry of a polymer that has been developed by SNL for a number of years. The project features a very good mix of well-known experts in the field.
- The project features a good team that is focused and has achieved its goals.

Project weaknesses:

- The data to date has been collected on very thick (~100 µ), unsupported membranes. While this is a good place to start, a realistic assessment compared to the state of the art can only be made using <20 µ supported membranes.
- The team should focus more on the stability of the membrane under HT polymer electrolyte membrane fuel cell conditions, which is the primary goal of the project.
- The project seems a little empirical; it is not really science driven. It needs many more resources to properly tackle the ionomer issues expected when constructing state-of-the art MEAs.
- The project lacks swelling and mechanical property measurements, and it is not clear these issues are being given enough consideration.
- The project is too focused; it needs to diversify to pendant ionic conductors.

Recommendations for additions/deletions to project scope:

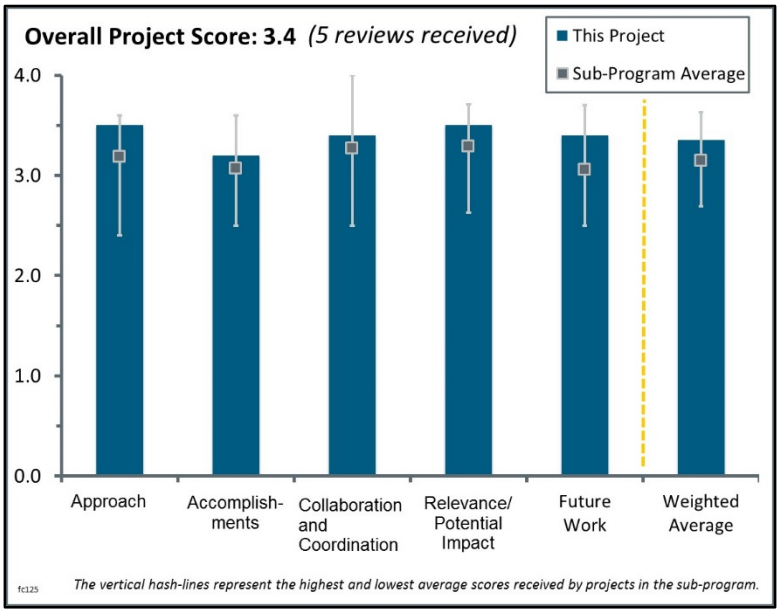
- The project appears to be well organized and has made very good progress. The researchers should continue to look at conductivity under low RH and durability because these are historical weak points for HC membranes.
- The team should conduct longer-term testing (at least 2,500 hours) and diversify to new pendant ionic conductors besides sulfonic acid (e.g., try mixed sulfonic/phosphonic doping in highly doped membranes).
- The researchers should include swelling tests and RH cycling testing, as well as consider including supported membranes.
- The project needs more science; i.e., a full morphological and transport study of the material so that IEC, molecular weight, and block lengths can be optimized based on a scientific approach rather than an empirical design of experiments.

Project # FC-125: Engineered Low-Pt Catalyst Layers

Mahlon Wilson; Los Alamos National Laboratory

Brief Summary of Project:

The objectives of this project are to (1) introduce, model, and develop the materials and techniques for a new approach that focuses on an engineered ionomer topology (EIT) within the catalyst layer; and (2) attain 0.05 mgPt/cm^2 fuel cell performance that demonstrates the potential of the EIT approach.



Question 1: Approach to performing the work

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

- Barriers are addressed and well targeted. Clearly, reducing catalyst Pt content is critical to reduce the cost of the membrane electrode assembly (MEA). Overcoming the shortfall in performance by engineering a catalyst/ionomer interface with better oxygen utilization is a very good approach to achieve this goal. Equally important is developing the model to explain the results, and this project addresses these points very well.
- Conceptually, the EIT approach is very good because thick dispersed electrode layers must be engineered to maximize transport.
- The approach and its outcome are clear. The approach implies that the ionomer film resistance is relatively more important than other resistances. This implicit assumption should be emphasized partly because there is an absence of data on both gas/ionomer film and ionomer/Pt interface resistance values.
- The need for a better understanding of the balance between oxygen diffusion and proton conductivity for the ionomer in low-loading catalyst inks is important. LANL proposes an interesting solution to this problem.
- The proposed approach is to optimize the ionomer topology of the catalyst layer to minimize the shortfall observed with low Pt loadings and to use a two-phase approach of ionomer. This is completed by characterization and modeling. It appears to be feasible, based on the project's progress.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

This project was rated **3.2** for its accomplishments and progress.

- The team has performed well and produced valuable results supported by complementary modeling and characterization. Ionomer characterization improvements shown this year in FC-020 might help further understanding and preparation of this two-phase ionomer approach.
- Performance results exceed the milestone target and are near the General Motors (GM) MEA reference, while EIT catalyst layer improvements are foreseen. Durability evaluations shall be the next step.
- Ionomer coverage of carbon support/catalyst could be considered in the modeling because ionomer distribution might not be uniform.
- Good to very good accomplishments were demonstrated, considering the project time frame and budget. The modeling appears to be rigorous and provides good guidance. Experimental work, while limited, achieved the performance milestones and was comparable to a traditional electrode. Ultimately, improved

performance against the traditional GM electrode is needed to demonstrate the utility of the EIT approach, but this may be beyond the project timeline and budget.
- Clearly, good progress has been made, as demonstrated by the improved performance curves at limiting current.
- The electronic conductivity of the multiwall carbon nanotubes (MWCNTs) is only important if they are not fully covered by an ionomer layer (slide 10). It is likely that the MWCNTs are not fully covered by the ionomer. This consideration should be clearly highlighted.
- It is likely that the ionomer film thickness covering the catalyst particles is uneven (slide 11). Also, it is not possible to assess the ionomer film thickness because it cannot be measured. These considerations should also be added.
- The reproducibility of the results has not yet been determined and could affect the conclusion derived from the polarization curves (slides 13 and 14). Future work should include this consideration within this project or a subsequent project.
- It is regrettable that the oxygen polarization was not measured (slides 13 and 14), because the difference between the oxygen polarization curves and the helium/oxygen polarization curve would provide a direct measurement of the performance loss associated with the ionomer film covering the catalyst.
- The modeling data suggest the proposed approach is viable, but more data ares needed to prove concept.

Question 3: Collaboration and coordination with other institutions

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

- The coordination and collaboration between the partners appear to be excellent. Involving an original equipment manufacturer (GM) is highly appreciated.
- The project work is well distributed among team members, each of which brings relevant strengths to the overall effort.
- Collaborating institutions are well integrated.
- There is good collaboration with other national laboratories, but not with industry.
- It is understood that the project is relatively limited in terms of funding and time frame; however, collaborations with an end user would be useful to establish the true viability of this approach.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.5** for its relevance/potential impact.

- Developing low-Pt catalysts is essential to achieving the DOE Fuel Cell Technologies Office's cost targets. The modeling and initial results demonstrate the substantial promise of the EIT catalyst layer approach to overcome the low-Pt performance shortfall.
- This project has a high impact because it directly targets some of the main cost drivers—namely, high Pt loadings of the catalyst.
- The project is directly relevant to DOE cost targets, by reducing Pt content and maintaining high-rated power.
- A low-loaded catalyst that can overcome the oxygen diffusion limitations of traditional catalyst/ionomer formulations is an important area of research.
- The modeling, in combination with the experimental results, will be important in establishing the viability of this approach.
- The decrease of mass transport performance at low catalyst loadings negatively impacts cost-reduction efforts.

Question 5: Proposed future work

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

- The proposed future work is very well defined and looks good for this project. The durability of these structures will be an important metric in the future work and has been identified as such by the project team.
- The future work on the validation of the EIT concept is the most important. The focus on electrospun ionomers used with thin-film coated catalysts tested in fuel cells will be an important milestone for this project.
- The proposed future work is aligned with the goals of the project..
- The proposed future work is reasonable.
- The project team should focus on generating a wide range of constructions and comparing the resultant experimental fuel cell data to the model.

Project strengths:

- The modeling used in this project is important in identifying a solution to the oxygen diffusion and proton conductivity problem seen in low-loaded catalyst.
- The quality of the project team is a strength of this project.
- The project's strengths include a great approach and a very strong, focused research team.
- The project addresses a clear problem and has focused objectives. The team is well integrated.

Project weaknesses:

- To date, very little fuel cell data have been shared. While the idea appears to be sound, the researchers need to demonstrate that the combination of ultra-thin ionomer-coated catalyst particles with nanofiber ionomers is (1) possible and (2) improves performance over traditional catalyst formulations.
- The reproducibility of the results is currently unknown, and a direct quantification of the performance loss attributable to the ionomer film covering the catalyst is missing.
- The one-year timeline of the project is a weakness. This project should be continued to validate the promising results.

Recommendations for additions/deletions to project scope:

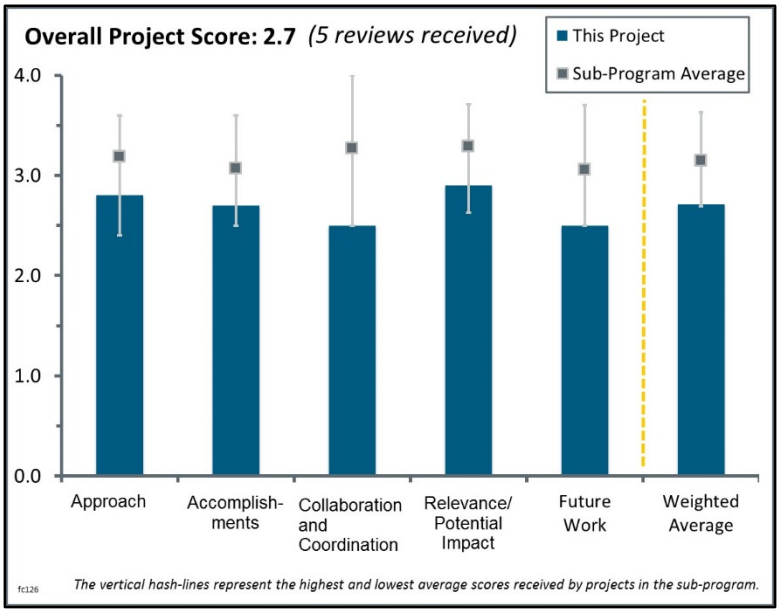
- This project should be continued in light of the promising obtained results and the potential impact.
- There has been a lot of work by Peter Pintauro of Vanderbilt University in both electrospinning of ionomers and the use of catalyst particles in electrospun fibers. A comparison between this approach and those already published would be helpful.
- The research team should define the results' reproducibility and measure oxygen polarization curves to calculate the difference in performance with helium/oxygen polarization curves (i.e., loss due to ionomer film).

Project # FC-126: Semi-Automated Membrane Electrode Assembly Fabrication with Ultra-Low Total Platinum-Group-Metal Loadings

Stoyan Bliznakov; Brookhaven National Laboratory

Brief Summary of Project:

The objectives of this project are to (1) develop a semi-automated system for fast and facile electrodeposition of Pt monolayer shell on non-noble refractory metal core electrocatalysts with ultra-low platinum group metal (PGM) loadings and near 100% Pt utilization, directly on gas diffusion layers (GDLs); and (2) demonstrate the feasibility of the proposed electrodeposition strategy for scaling up and fabricating membrane electrode assemblies (MEAs) with performance exceeding the U.S. Department of Energy (DOE) 2020 technical targets.



Question 1: Approach to performing the work

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

- In a short period of time, successful engineering, development, and deployment of an automated system for the fabrication of electrodeposited catalysts directly on GDLs has been demonstrated. The key technical advantage of the approach is that the catalyst will only be deposited at the optimal areas where a triple-phase boundary exists, ensuring high catalyst utilization percentages (this project successfully demonstrated catalyst utilization near 100%). This project demonstrated a relatively successful scale-up to 50 cm^2; however, the significant loss in peak power density in air is likely due to compositional inhomogeneity and uneven distribution of catalyst on the larger-area GDL. It is unclear whether the ionomer is incorporated into the catalyst layer—this may have something to do with the poor performance at high current density. Typically the microporous layers are impregnated with Teflon, which could limit proton transport to the oxygen reduction reaction (ORR) catalyst.
- WNi is a well-chosen and promising choice of core chemistry to allow the reduction of total PGM loading in core-shell catalysts. Development of the semi-automated processing equipment has allowed facile study of this rather complex catalyst system and expansion of testing in MEAs. Electrodeposition provides tight control of processing parameters but, in its present form, gives a too-thin catalyzed zone, which is the likely cause of the poor air performance in MEAs seen to date.
- Extending the work shown in the area of rotating disk electrode (RDE) to electrode layer is a great idea. Because feasibility is mentioned in the goals, some cost analysis should be included.
- The approach is not well thought-out. Although it may be true the proposed method can enable a low-cost manufacturing process, it is not necessary to demonstrate this with a “semi-automated MEA fabrication” process. If small, high-performance cells can be demonstrated, then MEA suppliers will be interested, and industry is much better suited to develop manufacturing processes. The fact that Brookhaven National Laboratory (BNL) is not well suited to make MEAs is clearly evident by the poor cell performance on air (which is what matters in the real world, not performance on pure oxygen). This type of work should be a collaborative effort between BNL and an MEA supplier. If an MEA supplier is not interested in working on this technology, then BNL should find out the reason(s), and then do experiments that can overcome these objections.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- The development of a semi-automated system for the deposition of complex compositional profiled catalyst materials directly onto GDLs was successfully completed, and durability, activity, and loading targets were all met for small, laboratory-scale MEA sizes. What is left is translating this procedure to larger-scale MEAs, which from the preliminary data, does not appear to be a straightforward process. There is still concern related to the high current density and air performance, especially as the MEA area increases. It is proposed that moving to a dendritic catalyst architecture will help with the high current density and H_2/air performance; however, it is unclear why this would be the case, and the preliminary performance data do not show any significant performance improvement over the primary electrodeposited catalyst structure.
- Rapid progress has been achieved halfway through this one-year project (assuming most of it was not already completed prior to the start of the project). Semi-automation of the production of the complex Pt/Pd/WNi catalyst layers has been achieved. Testing has progressed to MEAs, showing promising performance in O_2 but poor performance in air. ORR kinetic activities are good but not exceptional. The observed problems with performance at high current density in air may be intrinsic to the chosen process of electrodeposition directly onto the GDL, leaving the catalyst in a thin plane right at the surface rather than being distributed through a ~10 µ layer of optimally porous carbon, as in a conventional electrode.
- While the mass activity targets are met, the cell performance on H_2/air at high power densities is well below DOE targets. Slide 11 shows that the team does not understand the DOE targets. Specifically, both the table and plot claim 1 W/cm^2 power density on H_2/O_2. The DOE targets are for H_2/air performance. This is also the case with the voltage at 1.5 A/cm^2. There is also no evidence the team is diagnosing why the H_2/air performance is so poor, much less that it has a plan to address the issue.
- The performance achieved is low and inconsistent.

Question 3: Collaboration and coordination with other institutions

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

- Good pathways for testing of project MEAs by outside laboratories appear to have been established. This project has been active for less than a year, leaving little time to date for collaborators to make contributions, and the presentation did not identify specific contributions of collaborators other than those within BNL.
- There is no evidence of any collaboration with anyone outside of BNL. For example, if General Motors (GM) was actively involved, it would have pointed out the misreporting of the performance targets on slide 11. This team definitely needs to collaborate with an entity that understands mass transport in catalyst layers and can help the team both diagnose and address the core issue. This could be an MEA supplier, an automotive original equipment manufacturer (OEM), or anyone who has experience in fabricating and testing MEAs.
- To this point, the work appears to be contained in-house within BNL. It appears that GM will become involved once the efforts begin for scale-up to larger MEAs.
- Evidence of collaboration is missing.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **2.9** for its relevance/potential impact.

- The current project aligns well with the DOE objectives in that it has the potential to meet the fuel cell performance metrics with a process that could lead to continuous, larger-scale fabrication of MEA materials.

- The project effectively addresses the DOE goal of increasing ORR kinetic activity with reduced PGM loadings and has achieved good, if not exceptional, kinetic mass activities. The project's WNi cores provide a pathway to reduce total PGM in core/shell catalysts. It seems doubtful whether electrodeposition, even if automated, can provide a time- and cost-effective means of generating a cathode catalyst layer for fuel cells; industry tends to prefer chemical rather than electrochemical methods whenever possible. The poor performance of MEAs in air at high current density may be intrinsic to the process of electrodeposition directly onto GDLs, giving an insufficient porous, catalyzed volume to facilitate transport processes.
- It is not clear how this project will have any impact on DOE's key targets because all that is being demonstrated here is that the mass activity and stability of the core/shell catalysts are good, which has already been demonstrated.

Question 5: Proposed future work

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

- The proposed future work correctly identifies the high current density performance and H_2/air performance as areas that need to be addressed; however, no well-defined course of action is identified, other than trying dendritic electrodeposits. The proposed future work should also address strategies for the even, homogeneous deposition of well-distributed catalyst over larger areas to translate the performance of the small-area MEAs to that of the larger-area MEAs.
- Plans to improve MEA performance in air by growing dendritic cores rather than compact particles are directionally correct, but they likely do not go far enough in addressing the fundamental problems at high current density of a catalyzed layer that is too thin. Some of the tricks used in conformal electroplating of rough surfaces might be tried to get deposition of WNi to extend deeper into the microporous layer of the GDL.
- The proposed future work makes no sense in light of the current poor performance on H_2/air. No automotive OEM is going to be interested in testing full-size cells that do not have decent H_2/air performance. Instead, the plan should be to diagnose the cause of the poor mass transport in these MEAs and address it accordingly. Just increasing the oxidant pressure is not a mitigation strategy.
- The breadth of work proposed makes success questionable. Developing GDLs is very much different from other work.
- The strategy for improving MEA performance at high current density in air is not clear. The strategy and approach for using very low PGM and reducing cost using the chosen method are not obvious.

Project strengths:

- The strengths of this project include the well-controlled deposition of highly active catalyst directly onto GDLs, demonstrating nearly a 100% utilization of the Pt. The process shows promise for scale-up to large-area MEAs.
- Codeposited WNi is a promising core material with prospects of decreasing total PGM usage in core/shell catalysts. Electrodeposition provides tight control of metallization. The team developed semi-automated preparation techniques that improve the reproducibility of complex procedures.
- The likely positive results show the promise of the method, but the method needs to be better demonstrated.

Project weaknesses:

- The MEAs perform poorly in air, likely because electrodeposition puts catalyst only onto the topmost surface of the electrode, resulting in an insufficient volume of the (porous) catalyzed layer to support adequate mass transport. The catalyst was deposited directly onto the microporous layer of the GDL, rather than onto a carbon catalyst support layer optimized for mass transport. Even when automated, electrodeposition is likely to be a slow and expensive way of manufacturing a catalyst layer. Pt loadings on MEAs are too low to allow proper testing for optimized performance in air at high current densities and the durability thereof—the most critical remaining factor in the development of fuel cell catalysts and MEAs.

- It is unclear how high current density and hydrogen/air performance are going to be improved. There appear to be no strategies for assessing/qualifying the homogeneous distribution of catalyst with the desired composition and architecture over larger MEA areas. Incorporation of ionomer into the catalyst layer or GDL is not addressed.
- The project is unfocused. If the key work is electrodeposition, that should be the focus.
- The lack of MEA fabrication and testing experience on this team is sorely evident.

Recommendations for additions/deletions to project scope:

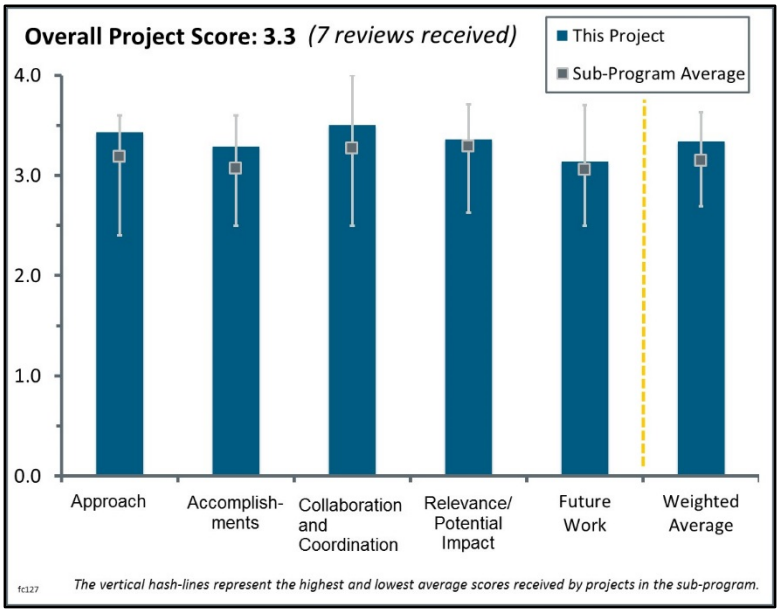
- The researchers should explore chemical, rather than electrochemical, means of depositing the WNi cores. They should deposit the Pt/Pd/WNi catalysts onto proven carbon catalyst-support particles, and then prepare MEAs by conventional methods using the catalyst powders—this is likely to be the most productive pathway toward adequate air performance. If electrodeposition is continued, the researchers should modify procedures to allow deposition of WNi particles into a greater thickness of the catalyst support layer (to improve air performance).
- The team should develop procedures for quantifying the compositional and structural distribution of catalyst on the larger-area GDLs to help improve the transition of activity from small-scale to larger-scale MEAs. It should also determine the effects of ionomer in the electrodeposited catalyst layers and identify strategies for its incorporation into the MEA catalyst layer.
- Clear proof of the concept's viability is needed before the concept is scaled.
- This project should be stopped unless the team is willing and able to get the appropriate collaborators.

Project # FC-127: Durability Improvements through Degradation Mechanism Studies

Rod Borup; Los Alamos National Laboratory

Brief Summary of Project:

The objectives of this project are to (1) identify and quantify degradation mechanisms (discern the impact of electrode structure on durability and performance), (2) develop advanced in situ and ex situ characterization techniques and accelerated stress test (AST) protocols, (3) develop models related to degradation mechanisms, and (4) explore non-system-related mitigation.



Question 1: Approach to performing the work

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

- The approach to performing this work is excellent. Carbon corrosion during the drive cycle is particularly meaningful because previous carbon corrosion studies only used simple voltage cycling. The carbon corrosion comparison for high surface area carbon, Vulcan carbon, and graphitized carbon is also very interesting. In situ experimental observations of Ce dissolution and migration during fuel cell operations have added great values; the high-resolution transmission electron microscopy (TEM) images of catalyst layers with Ce are very impressive. Combined mechanical/chemical AST tests will provide a facile and effective tool to evaluate membrane durability.
- The approach addresses key aspects of interest for the industry related to durability—particularly regarding the catalyst layer and membrane. Targeting the improvements and understanding of ASTs and durability mode quantification is a valuable contribution. Overall, the approach is consistent with existing methods and understanding on analyzing materials from a durability perspective.
- Carbon corrosion work: The approach was very systematic and focused on drive cycles, which has been less explored than start-up/shutdown phenomena. The project situated itself well to understand when carbon loss occurs by using CO_2 analysis, and to understand Pt particle size effects. Ce migration work: The project did well to focus on the "why" questions behind Ce migration. The fact that Ce migrates is understood, but the project went further to address the conditions under which this happens. AST development work: The project is correct to gauge whether protocols are too severe or whether a series of materials fails with a protocol in the same order as it does in the field. However, the project should seek to develop an AST after careful examination of the failure modes observed in the field. This may cast light on what kind of conditions to run, and especially as to whether there should be differing effects on the inlet and outlet of the cell. Perhaps the cell should not be a square serpentine channel cell. Metal bipolar plates work: This is a difficult task to address due to the complexity. Ultimately, there will have to be some interaction between the project team and developers to understand realistic failure modes and to develop an AST that addresses those failure modes.
- The researchers are doing a very good job of maintaining relevancy and performing work that can help over many material and application classes. They need better direction on their Fe work. They seem to be using the membrane electrode assemblies (MEAs) as a probe to measure whether there is any Fe release from the stainless steel plates. It would be better to determine what Fe level the MEAs can tolerate, and then determine loss of Fe into the membrane. General Motors (GM) has done the best work relating Fe to degradation rates, but there is still a lot of room for work here.

- The work presented here is very comprehensive and addresses many of the important degradation mechanisms through an optimal balance between experiment and theory. The combined use of in situ and ex situ techniques provides great clarity and unique insight into the mechanisms of efficiency and performance loss. There is no attempt to develop mitigation strategies for many of these sources of performance degradation; however, that is likely beyond the scope of this work.
- The project is addressing critical carbon corrosion and other durability issues that are barriers to implementing conventional electrodes. It features a nice combination of modeling and experimental work, although it would be advantageous to disseminate learnings to original equipment manufacturers (OEMs) for assessment at full scale.
- The fiscal year 2015 tasks are well aligned with the barriers, but they are very general and not sharply focused.

Question 2: Accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals

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

- Because the work here is more of a fundamental investigation of the various mechanisms of performance degradation associated with the many components that compose the MEA, there are no performance metrics or barriers against which the progress of this project may be directly related. However, the unique insight provided by the results presented here will have a significant impact on the design of MEA components going forward, making an important contribution to the improvement of MEA durability. This work covers a wide breadth of material degradation pathways, containing a significant amount of experimental data, much of which is included into multiscale models that could prove to be very useful to the PEMFC community at large. The accomplishments, to this point in this project, are quite significant.
- Carbon corrosion work: The results the project has achieved align very well with phenomena as they are understood. CO_2 loss was shown for potential down-transients as well as potential up-transients. The carbon corrosion trends with E, V, and EA carbons are consistent with what is known. The particle size increases with more graphitized carbons are also consistent. Ce migration work: The work involving CeZr may be a slight contribution that may assist developers, assuming suppliers have not already surveyed this composition. Ce migration trends were shown with lower pressure as well as in the direction of the cathode, which begins to answer why Ce migrates to where it does. AST development work: The project team has successfully identified protocols that are more accelerated than prior protocols (e.g., combined mechanical/chemical membrane AST and square wave cycle for Pt dissolution). Questions still need to be answered as to how well these ASTs relate to vehicle failure mechanisms, and whether they should ideally be carried out in quad-serpentine 50 cm^2 fixtures. Metal bipolar plates work: The work so far utilized uncoated 316L stainless steel. This is an appropriate prerequisite to understanding what types of stressors will cause higher interfacial contact resistance, as well as higher metal corrosion and fluoride release. The project team also needs to have a way of understanding relevant vehicle stressors.
- Analysis of the water content of the electrode related to the change of the low potential used in the analysis of the E-type catalyst would help identify the effects of the low potential effects. The mechanism effect (pathway) and reactants (i.e., availability of liquid water in the electrode) will be key in understanding the role of the low and high potential distinctions in the AST for carbon corrosion. Confirmation of whether Ce is accumulating as cations or whether Ce is still in a neutral phase is important. The team should consider confirming whether Ce deprotonates the membrane or whether it is simply a neutral molecule in the aqueous phase. Comments on the reaction pathway for CeZr and indications on what the final product of the Zr would be should be considered from both operational and environmental standpoints, considering potential washout effects on a stack or module level for automotive or stationary applications. Spatial maps of membranes via IR analysis or other methods that quantify the local thinning and divots/tearing/pin holes would be ideal to further develop the understanding of the mechanical/chemical AST. The proposed electrocatalyst AST should be considered from a mechanistic perspective; the alteration of upper potential and lower potential hold times are directly affecting the subsurface oxide dissolution, the surface passivation, and bare metal dissolution rates. The design of the potential limits and the dwell times for accelerating the test will be a direct outcome of the previously listed three processes, and means of quantification or analysis would allow targeted ASTs to be deconvoluted for various automotive,

stationary, and other fuel cell applications. In the crack propagation work, yield strengths for the catalyst layers in the simulation should be studied because the expansion of the crack into the lower regions of the catalyst layer could have performance and water accumulation considerations.

- The project has progressed very well in the past year. The durability studies almost cover all the components of a PEMFC, including the catalyst, membrane, and bipolar plates (the gas diffusion layer was previously studied), and some novel and advanced approaches have been adopted. The project has led to some new discoveries and deeper understanding relevant to fuel cell degradation. Some mitigation strategies have been proposed. A few things are noteworthy: (1) the addition of CeZr to the cathode catalyst layer improves membrane durability, (2) the developed E-carbon model is very instrumental for understanding carbon corrosion because it has incorporated parameters such as temperature and relative humidity (RH), and (3) the proposed new AST protocol tests has provided a more effective and accurate means of durability evaluation. There are two questions. Based on the project's discovery that "all types of carbon show carbon corrosion during drive-cycle testing," there is the question of whether adoption of low surface area carbon would meet DOE's ultimate durability target, or whether alternative supports other than carbon have to be developed. The other question is whether the project would investigate the stability of 3M's nanostructured thin film (NSTF) catalyst at drive-cycle testing.
- Regarding slide 9—yes, stable Pt particles on graphitized supports are needed for a durable catalyst, but it is unclear what constitutes an acceptable level of particle growth to be able to achieve durability metrics. Regarding slide 17—the team needs to more clearly define the capabilities of the small-angle x-ray scattering/wide-angle x-ray scattering methods and how the data are interpreted. Regarding slides 21 and 22—the metal bipolar plate corrosion data are interesting, but it is unclear whether they are relevant. With low membrane fluoride release rates, 316L stainless steelshould be durable enough for 5,500 hours. It should be assumed that any metal plate will have some kind of coating to reduce contact resistance, and therefore these experiments should be conducted with a "baseline" coating (e.g., thin gold) and then a representative low-cost material that meets the target for total ohmic loss at beginning of life. Perhaps collaboration with Treadstone is warranted.
- The team should start putting error bars on its experimental data. There are very strong conclusions made based on the trends, but the variability is missing. A lot of work was focused on Ce and concentration changes, but it is unclear whether a lower concentration of Ce has been linked to membrane failure.

Question 3: Collaboration and coordination with other institutions

This project was rated **3.5** for its collaboration and coordination.

- This project has had outstanding collaboration. It is led by a national laboratory, but its collaborators include other national laboratories, universities, and companies (e.g., GM, IRD Fuel Cells, and Ion Power). The industry participation is very meaningful. All the participants have complementary experience.
- This project is a model for how the national laboratories can work with industry. It uses multiple industrial suppliers and multiple site visits. This project represents a really good effort.
- The project features excellent collaboration between experimental MEA testing, data modeling, and post-mortem microscopy analysis. Each partner has contributed greatly to the success of the project to date.
- The project features a strong and diverse group, and now it includes more involvement from automotive OEMs to enhance the applicability of the research outcomes.
- Five different organizations (i.e., IRD Fuel Cells, Ion Power, Gore, 3M, and GM) provided MEAs, but there is no evidence of these organizations participating in the data interpretation or in directing the project. In the particular case of GM, it would be expected that GM would have some greater say in project activities, but that was not evident from the presentation. Collaboration with Lawrence Berkeley National Laboratory (LBNL) is evident through the Ce migration modeling work, as well as in the measurement of membrane crystallinity. The project team likely collaborated with Argonne National Laboratory (ANL) in order to provide data for ANL durability models, but there is no work shown here that indicates the ANL model returned assistance to this project. Like with many projects, Oak Ridge National Laboratory provided TEM assistance. With the possible exception of LBNL, the collaborations more closely resembled provision of service, whether this included materials (e.g., MEAs) or analysis (e.g., TEM). There was little collaboration outside of Los Alamos National Laboratory (LANL)/LBNL in terms of providing the project

direction, and in terms of interpreting data. The project needs much more expansive collaboration with fuel cell stack and system developers.

- The project obtains materials from a variety of industrial vendors, ensuring a broad selection of materials; however, additional MEAs from other vendors should be considered because of the variety of other support materials used (i.e., lower surface area, highly graphitized supports). Evaluation of the AST cycle proposals should be done in collaboration with a cross-section of automotive and stationary fuel cell developers, with some consideration for the link to an accelerated durability test or field return behavior.
- The main collaborators are only national laboratories, and this may limit the materials and fabrication methods being studied. There has been much advancement through developmental research toward mitigation of degradation mechanisms in PEMFCs, and this project has poor linkage. This is likely due to the secretive nature of OEMs and suppliers. However, it is captured in patent applications.

Question 4: Relevance/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan

This project was rated **3.4** for its relevance/potential impact.

- Improving fuel cell lifetime and durability is arguably the most important target of the Hydrogen and Fuel Cells Program (the Program). There are clearly numerous mechanisms by which the lifetime of a PEMFC may be shortened, and the work presented here and in previous years of this project addresses a large number of these mechanisms in great detail, providing significant insight that may be readily used to develop mitigation strategies for each degradation mechanism.
- The impact of this project is significant because this project directly addresses one of two major barriers to fuel cell commercialization—durability. This project aligns well with Program and DOE objectives. It has a great potential to help advance fuel cell durability progress toward DOE and research, development, and demonstration goals. The principal investigator of this project has been well recognized in fuel cell durability/degradation studies and made significant contributions in these studies.
- Durability is one of the primary focal points of fuel cell commercialization, and the work in this project directly addresses both industry-level targets and the long-term targets set out by DOE. Further, the work done in the project is consistent with the state-of-the-art methods for quantification, analysis, and testing of fuel cell materials from the perspective of durability and performance.
- The project team has done a good job of adjusting as materials and challenges change. Carbon corrosion efforts can be scaled down. NSTF does not have it, and automakers seem to be content with current solutions.
- The project team generally performed solid, fundamental work, but it needs to ensure that the problems being studied are relevant at full scale, using material sets that are common in current application hardware.
- This year's project presentation has essentially four sections: (1) carbon corrosion, (2) Ce migration, (3) development of ASTs, and (4) metal bipolar plate corrosion. The study of carbon corrosion on various carbons has been covered internally within OEMs, often in association with particular operating strategies. While it is fair to say that many of this data remains unpublished, the assistance that a DOE-funded study provides to many developers is limited to being a confirmation of phenomena already acknowledged. Furthermore, some patent literature (e.g., US Patent 7,887,963) already points to how OEMs have already studied the role of operating conditions in mitigating carbon corrosion. The study of Ce migration has a similar overlap with OEM work, but it may overlap more strongly with membrane supplier work. Some suppliers have extensive history with Ce cation or Ce oxide additive packages. The development of ASTs may be most relevant to the DOE projects themselves, but there is some value if this project can improve upon some tests that are far too lengthy (e.g., RH cycling). Studies of metal bipolar plate corrosion are relevant and have not been as commonly done by the industry because of the wide variety of substrates, coatings, forming methods, and approaches to joining.

Question 5: Proposed future work

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

- The proposed future work is effectively planned and addresses all of the possible suggestions one could make for the next step.
- The proposed future work is very comprehensive and crucial to further understand fuel cell component degradation and improve fuel cell durability. One example is ionomer/electrode stability and structural changes, which would be meaningful, if successful. The concern is whether the project has sufficient funding to perform the proposed future work.
- The National Renewable Energy Laboratory project on system-borne contaminants approached its topic by first asking a developer what materials would be used in a fuel cell system, and then systematically figuring out which contaminants to test in response. This project could use a parallel approach, where a developer becomes closely related to the project, so that the project has a reference point to use. Instead, the project team presumes that it already knows for certain what degrades during fuel cell system operation. Perhaps literature is used as justification, and in some cases, perhaps an experiment is done to show that degradation is possible. However, neither should be interpreted as an assurance that such degradation is truly limiting to a practical fuel cell system's lifetime. Many of the items listed in the future work slides do appear reasonable for further study. In particular, these include catalyst layer ionomer durability (especially in low Pt loaded layers), alloy catalyst durability, metal bipolar plate durability, and membrane crack propagation. However, each of these activities needs to be addressed from a perspective that has some knowledge of what is really happening in real-world systems. It is not sufficient to say that an experiment says that something might degrade, and then allow that to be the basis for continuing work.
- The work continues to address areas relevant to the industry and fuel cell adoption. The project should include work with system/unit cell modeling efforts to provide either validation data or directed testing to further improve the model mechanisms or predictions.
- The future plans seem reasonable, but the ongoing bipolar plate work plan needs to be revisited.
- There are a lot of open questions around Ce; everyone is using it or another mitigant, and the researchers seemed to have raised more questions than given answers, but this is nowhere in their future plans. Better/faster ASTs are needed, and it is great that they are focusing on this.
- More specific metrics would be helpful in evaluating progress.

Project strengths:

- The full degree of national laboratory analytical capability is available to support the project. Project collaborators have a deep level of experience in fuel cells. The project has provided a good set of AST data on baseline MEAs, which serves as a useful guideline for how materials will respond to ASTs. The project has done a good job of understanding what stressors can possibly accelerate degradation. The project has improved from showing Ce migration to attempting to understand what causes Ce migration. The results of the carbon corrosion study align with other data.
- The project is performing comprehensive degradation and durability studies on the most important components of PEMFCs. Drive-cycle testing was used for durability studies that can better mimic fuel cell vehicle driving conditions. The combination of experimental data with effective modeling can provide more insights and information. The work related to carbon corrosion and membrane durability (e.g., the addition of CeZr to the cathode) has produced significant information. The collaboration with industries (e.g., GM, IRD Fuel Cells and Ionomer Power) is greatly appreciated and valuable.
- The strength of this work lies in the breadth of topics covered with a high degree of detail, providing significant insight into each mechanism of performance degradation. The effective integration of in situ analysis, process, and component modeling and ex situ post-mortem analysis has helped this work to be very successful to this point.
- The project's strengths include its comprehensive analysis; consideration of a variety of state-of-the-art fuel cell materials; and continued focus on the key durability areas of supports, catalysts, and membranes.
- This project has world-class resources that are being utilized in an effective manner. The investigators are inviting the key stakeholders to help steer the deliverables toward relevant experiments and modeling.
- The project features a diverse, collaborative team that is applying innovative experimental methods.

Project weaknesses:

- There are no obvious weaknesses.
- The project often presumes to know what failure modes are limiting the lifetime of fuel cells (particularly in an automotive context) without having enough contact with a developer to really be able to say what is actually responsible. The project needs the ability to cast off any or all committed tasks in order to open itself up to what the true issues may be. Getting insight on realistic failure modes from a stack developer or MEA supplier is something that must be done carefully, given the confidential boundaries that often govern the development of new fuel cell technology. However, with good relationships with developers and suppliers, useful work can still be identified and executed. This is observed in some of the other projects led by national laboratories. This project needs to develop better relationships with developers and suppliers in order focus on the areas that are still problematic for stack durability.
- The project team should pursue more direct collaboration with other fuel cell developers and direct collaboration with model developers on both system and unit cell/MEA levels.
- Development is moving faster than this project. As a result, state-of-the-art materials and operating strategies are not integrated in some cases. The team should focus on patent literature.
- The carbon corrosion data would be more corroborating if delicate TEM images after degradations could be shown. The strategy to mitigate Ce migration from the membrane should be proposed.
- It is not clear that all the components being studied are relevant in actual automotive systems.

Recommendations for additions/deletions to project scope:

- The proposed future work addresses all of the possible suggestions one could make.
- The team should delete the carbon corrosion studies, development of freeze tolerance protocols, and development of any protocols that do not stress materials in a fashion consistent with how materials are stressed in a vehicle. The team should add much deeper collaboration with a developer (or multiple developers, if multiple types of applications are of interest) to understand real-world failure modes first—before durability studies follow. The team should also add a survey of different chemistries used for membrane additive packages. There are other peroxide and radical scavengers beside Ce cations or Ce oxide.
- The team should add further collaboration activities and consider further work on assessing the mechanistic aspect of changes proposed to the AST cycle.
- The team should consider expanding the bipolar plate work to include low-cost coatings to reduce contact resistance.
- It is not clear whether the project has sufficient funding to perform all the proposed future work. If funding is limited, the emphasis should be on how catalyst layer morphology affects durability.
- The researchers should link Ce concentration to a membrane failure.