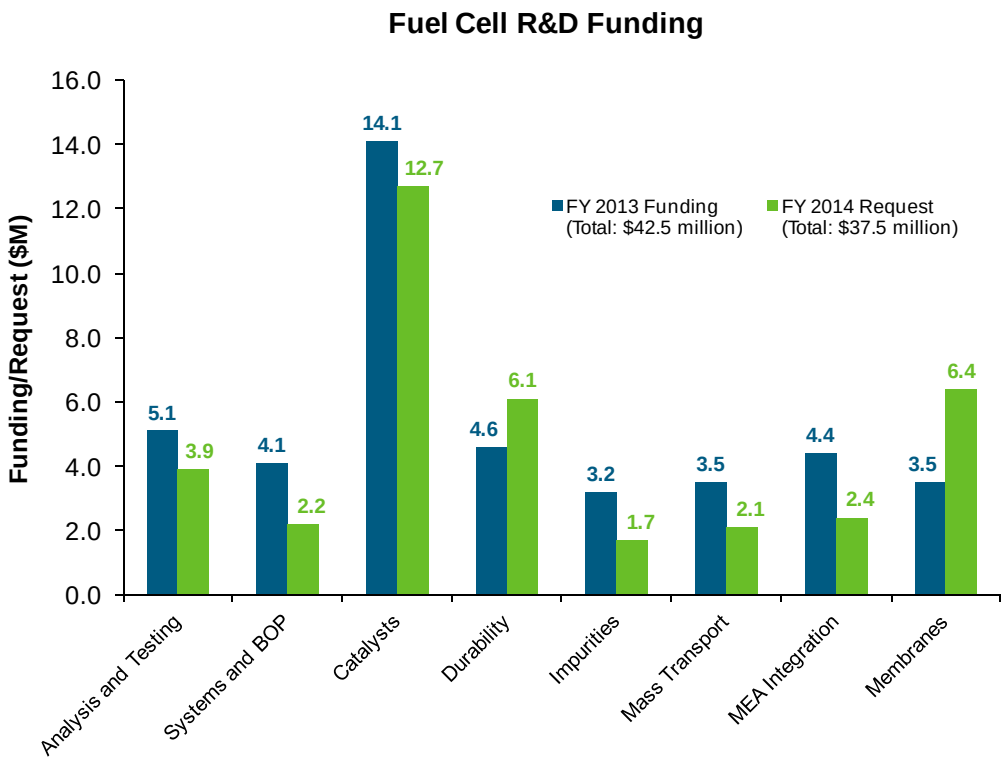
# 2013 — Fuel Cells
# Summary of Annual Merit Review of the Fuel Cells Program

## Summary of Reviewer Comments on the Fuel Cells Program:

Consistent with previous years, reviewers commended the Fuel Cells program for being well managed and clearly focused, with annual progress demonstrated. They noted that key challenges were identified, a strategy was in place to address those challenges, and that the project portfolio was appropriate to address the key challenges. However, reviewers commented that certain areas of the portfolio need to be further strengthened and that fuel cell system cost projections are based on some high-risk components. They also expressed concern about the lag in new starts due to the recent shift in forward funding projects.

## Fuel Cells Funding by Technology:

The Fuel Cells program received $42.5 million in fiscal year (FY) 2013 and $37.5 million is requested for FY 2014. The program continues to focus on reducing costs and improving durability, with an emphasis on fuel cell stack components. Efforts are balanced to achieve a comprehensive approach to fuel cells for near-, mid-, and longer-term applications. Three new projects were initiated in FY 2013 focusing on balance-of-plant (BOP) and catalyst improvements.




## Majority of Reviewer Comments and Recommendations:

At this year's review, 47 projects funded by the Fuel Cells program were presented and 37 were reviewed. Projects were reviewed by between five and nine reviewers, with an average of seven experts reviewing each project. Reviewer scores for these projects ranged from 2.4 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 3.0, respectively. The lowest score of 2.4 for all projects reviewed in 2013 was an improvement over 2012's low score of 1.8 for all projects reviewed.

**Analysis and Testing:** Ten projects were reviewed and received scores between 2.7 and 3.6, with an average score of 3.3. According to the reviewers, the analysis and testing projects benefited from good collaboration, but the reviewers were concerned that the lowest-scoring project lacked focus.

**Systems and BOP:** Five systems and BOP projects were reviewed this year, with scores ranging from 2.4 to 3.5 and an average score of 3.0. Reviewers commended the highest-scoring project for making significant progress toward meeting its goals. The reviewers were concerned that the lowest-scoring project had technical challenges that hindered its success. They suggested adding additional modeling to help identify the best path forward.

**Catalysts:** The scores for the 12 catalyst projects ranged from 2.6 to 3.5, with an average of 3.1. The highest-scoring project was praised by the reviewers for being well organized and for making significant progress toward addressing degradation due to start-up/shutdown events. Reviewers expressed concern that not enough detail was included in the presentation for the lowest-scoring project.

**Durability:** The two durability projects received scores of 3.0 and 3.1. Reviewers praised the durability projects for being sharply focused and for making good progress toward a significant challenge. In some instances, the reviewers recommended studying durability at the molecular level; current studies are at the system level.

**Impurities:** The two impurities projects reviewed received scores of 2.7 and 3.3. Reviewers felt that the highest value of the highest-scoring project was its materials screening effort; however, they noted that the outcomes cannot be fully applied to improve fuel cell durability without better understanding the degradation mechanism. The reviewers had similar concerns for the lowest-scoring project, as well as concerns that impurity concentrations were not aligned with what would be observed under operating conditions.

**Mass Transport:** The three mass transport projects reviewed received scores ranging from 2.7 to 3.3, with an average score of 2.9. Reviewers considered the transport projects to be doing a good job of solving a complex modeling problem. The reviewers recommended performing sensitivity analyses to better understand the role of each variable.

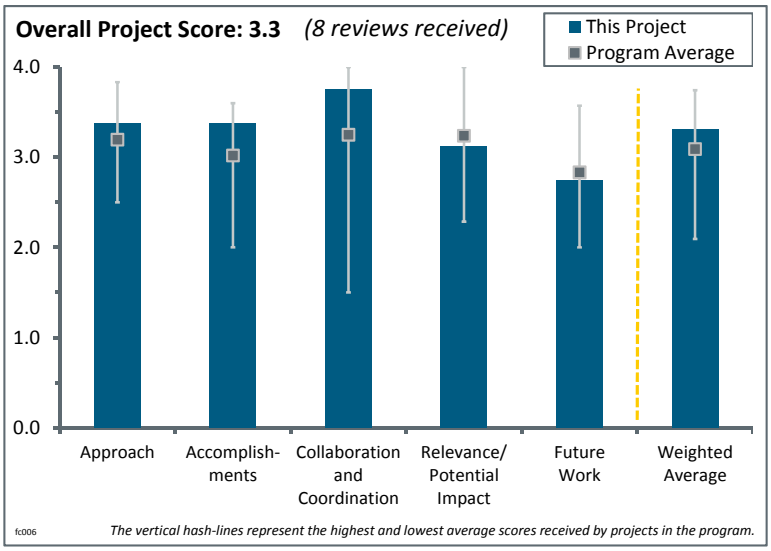
**Membrane Electrode Assembly (MEA) Integration:** One MEA integration project was reviewed, receiving a score of 3.0. Reviewers considered the project to have a good balance of applied research and development. They recommended that the principal investigator address water management and low-temperature operation.

**Membranes:** The two membrane projects reviewed received scores of 2.4 and 3.1. The reviewers considered the highest-rated project to have a good approach and to be clearly focused, but they were interested in better understanding the cost impact and benefit compared to traditional supported membranes. The reviewers consider the approach taken for the lowest-scoring project to be good for increasing MEA area, but they doubt that it will be able to overcome the cost and performance barriers that this project is intended to address.

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

Radoslav Atanasoski; 3M

### Brief Summary of Project:




Developing catalysts that will enable polymer electrolyte membrane (PEM) fuel cell systems to weather the damaging conditions in individual fuel cells during transient periods of fuel starvation makes it possible to satisfy 2015 U.S. Department of Energy (DOE) targets for catalyst performance, platinum group metal (PGM) loading, and durability. Fuel starvation could result in high positive voltages at the cathode during start-up/shutdown (SU/SD) or at the anode during cell reversal. This project develops a catalyst that favors the oxidation of water over the dissolution of platinum (Pt) and carbon (C) at voltages encountered beyond the range of normal fuel cell operation and beyond the thermodynamic stability of water (greater than 1.23 volts).

### Question 1: Approach to performing the work

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

- The project is well designed and focused on overcoming barriers.
- This project's focus and progress to date on a "materials-based solution" to improve catalyst performance and durability will enable the simplification of fuel cell control and operation.
- The approach of this project is unique to currently funded DOE Fuel Cell Technologies Office projects—to develop a material approach to limit the degradation due to SU/SD and cell reversals by modification of the anode (versus the cathode), thus suppressing the oxygen reduction reaction (ORR) activity on the anode catalyst.
- The materials approach to protect the cathode during SU/SD or cell reversal and use relatively low-PGM catalyst loading is a great approach because it is always "on." No monitoring or controls are needed, as in a system approach. The key is that catalyst durability is improved and the cost of the catalyst is kept low in order to achieve DOE cost targets.
- 3M has developed an innovative approach to try to address fuel cell start–stop degradation and fuel starvation. It is a materials-based approach that does not depend on system mitigation. 3M has applied the inclusion of high oxygen evolution reaction (OER) catalysts on the cathode and low ORR catalysts on the anode with nanostructured thin film (NSTF) electrodes. The approach would prove more universally valuable if also applied on non-corrosion-resistant electrodes such as dispersed catalysts on carbon. The 3M approach includes testing at relevant start–stop conditions and $H_2$ starvation conditions, as well as stack-level testing.
- The project features a good balance of fundamental studies and testing under real-world conditions. Utilizing the Automotive Fuel Cell Cooperation (AFCC) to provide gas switching under real-world conditions allowed for the discovery of issues at low OER catalyst loadings that were not observable under simulated conditions. Fundamental studies of Ru-Ir-Perylene interactions should provide insight into differences observed between the laboratory's potential studies and AFCC's gas switching studies.
- The approach is sound; however, using catalysts that enhance the OER rates for the anode reaction is relatively known in the field. The durability of these catalysts in start–stop test conditions has always been

a concern. Further, while reduction in Pt loading for the anode is directionally correct, the impact of fuel cell contamination is a concern and was not evaluated.

- Although this project does address the serious durability issue of start–stop decay, it attempts to do so in a manner that has very limited decay-mitigation potential. In other words, even if the project is technically successful, the decay will still be greater than acceptable, and other mitigations will be required to meet the durability targets. Because these other (system-level) mitigations can enable the targets to be exceeded without this technology, the value of this technology is questionable. This approach may have merit in applications where only minimal mitigation of decay is required. It would appear that the researchers at UTC Power (who first reported on this decay mechanism) agree—UTC patented this approach some time ago, but there is no evidence that UTC attempted to reduce it to practice; please see Bett, et. al., in U.S. Patent 6,855,453 (2005).

### 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.

- Excellent progress has been made toward DOE goals.
- The project developed and demonstrated catalysts with improved resistance to OER and ORR conditions.
- The team has done an excellent job of implementing this secondary strategy and demonstrating the results in a complete stack at AFCC. As expected, the decay rate is still unacceptably high (>100 mV with only 2,000 cycles). Additionally, the team has not provided a good baseline for comparisons; although the team claimed that three stacks at AFCC have been built, the cyclic decay results for only one are presented herein. A "40% to 50%" improvement is claimed, but no data were provided to support this improvement on a stack level.
- In 2013, 3M and the team have done an excellent job of optimizing their electrodes to maximize mitigation effectiveness while minimizing the effect on performance during normal operating conditions. It is hard to truly assess the viability of the mitigation because details of the durability protocols were not provided. The gas switching and original equipment manufacturer (OEM) cycling protocols would be especially important to disclose. There is also no comparative benchmark for the AFCC short stack cycling results. 3M also showed that membrane additives can inhibit OERs at high voltages, yet the significance of this during actual start–stop testing is unclear. It would be nice to know if 3M's OER catalysts are subject to degradation during conventional (0.6–1.0) voltage cycling, which could limit its ultimate effectiveness.
- The project has met all of its initial milestones. 3M has demonstrated stability over 5,000 start-up cycles with <0.1 mg PGM/cm$^2$ on the cathode. 3M has demonstrated considerable durability improvements over current commercial catalysts. The project has progressed to the state that two separate OEMs are evaluating this technology. Testing at AFCC indicates that more work needs to be done but that the technology has the potential to meet automotive durability requirements under real-world conditions.
- Significant accomplishments and progress seem to have been made toward objectives and overcoming the durability barriers. The researchers have demonstrated that relatively low catalyst loading (29 ug/cm$^2$) can inhibit ORR activity on the anode, and the ORR inhibition has been verified in stack testing. They have shown improved cathode protection of 40% to 50% over the baseline OER catalyst during SU/SD and that the OER catalyst did not impact the overall fuel cell performance. They have also shown some work toward the understanding of cell reversal on Pt, Ir, and Ru loss and where they end up, as well as the effect of low catalyst loading on the catalyst structure and morphology. However, how or why this happens is not yet understood. More work toward this would be useful in developing a more durable catalyst. It is unclear how the SU/SD protocol affects the OER catalyst. Understanding the mechanism of this would be useful toward developing more durable catalysts (slide 7). It is unclear what "modified" means and whether the same "modification" is done for all "modified" catalysts. It is unclear why the modified catalyst suppresses ORR on the anode more significantly than the unmodified one at the same catalyst loading of 29 ug/cm$^2$.
- The project has made modest progress towards accomplishing the goals of cost reduction. The researchers have achieved reduction in anode catalyst loading and improved the end voltage for reversal. However, the durability of the same catalyst in simulated start–stop conditions is not meeting expectations. Further, the progress compared to the 2012 DOE Hydrogen and Fuel Cells Program Annual Merit Review seems rather small.

- The researchers developed and tested a low-PGM modified anode with IrRu showing ORR suppression, yet had a hydrogen oxidation reaction (HOR) performance that should not be limiting. Approximately 50% cathode protection was observed (over the baseline). To determine how good this improvement is, the baseline cathode material and carbon material need to be known. It appears to be a relatively high loading of 0.4 $mg/cm^2$. More information on the testing protocols, especially the stack testing protocols, would be helpful to other projects. While anode modifications are shown to be valuable, there still seems to be a relatively large decay over 2,000 SU/SD cycles, which is beyond the commercialization targets. More definitive results on what degradation mechanisms are at play would be useful.

### Question 3: Collaboration and coordination with other institutions

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

- This project features very close collaboration between industry and academic institutions.
- This project seems to be well integrated with its partners. Materials development, materials characterization, and materials testing all seem to be well integrated.
- The assembled team is excellent. It features a university partner that can do catalyst synthesis and basic characterization, national laboratory partners that can do advanced characterization, and industry partners that can do independent evaluation and short stack testing. The coordination and integration of the collaborators in the project are also excellent.
- This project shows effective collaboration between a materials developer (3M), a university, national laboratories, and an OEM. Without the full range of fundamental studies, applied research, and real-world testing employed, the project would not be as advanced as it is.
- 3M has put together an outstanding team with members that all seem to make valuable contributions. The fuel cell testing at AFCC has been essential to validation of the concept on OEM conditions. Dalhausie and Oak Ridge National Laboratory have been essential in verifying that they have in fact made the catalysts they were trying to make and determining how the catalyst structure changes during voltage excursions.
- The project has a reasonable amount of collaborations. The final task of short stack evaluation by the subcontractor has not been completed with the latest iteration of the catalyst to show improvement in "real-world" conditions.
- The addition of AFCC to the team to demonstrate the technology developed on a stack level is excellent. However, the stack results reported by AFCC are far less than expected.
- This project would benefit from adding an additional subcontractor to evaluate membrane electrode assemblies (MEAs) with the improved catalysts; this would accelerate evaluation and commercialization of this work.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The project is very important to the DOE Hydrogen and Fuel Cells Program and has the potential to advance commercialization of PEM fuel cells.
- This is a potential material solution to some degradation effects due to certain transient conditions.
- The project addresses the major durability and performance issues associated with catalyst composition and structure.
- The project aims to develop catalysts that favor oxidation of water over the dissolution of Pt and C during transient periods of fuel starvation. This helps DOE achieve targets for catalyst performance, PGM loading, and durability. Therefore, the project is highly relevant to the Program and has the potential to have a big impact on DOE RD&D objectives. The cost of the catalyst may be an issue.
- Materials-based solutions to start–stop and $H_2$-starvation-driven degradation may prove valuable for automotive fuel cell applications, even though system-based solutions have also proven to be quite effective without adding significant cost. The project's value is limited by researchers' only proving its viability on NSTF electrodes that (a) would experience less damage than conventional electrodes during

voltage excursion and (b) have other issues that may limit their viability, such as water management challenges and low mass activity.
- This project addresses durability during start–stop and cell reversal conditions that have been identified as major contributors to cell degradation. Addressing cell reversal is believed to be critical for OEMs to meet automotive application durability.
- This project appears to have successfully demonstrated a technology that can reduce start–stop decay, but the reduction is not sufficient to meet DOE targets.
- The project is not highly relevant towards cost reduction; while fuel starvation is a known issue, parts of the solutions shown by the project are already being applied in some real-world cases. Further, the frequency and magnitude of fuel starvation can be mitigated with known system strategies. The output of this project will only make modest gains over the state of the art, even though the project is showing significant improvement over the chosen baseline.

### Question 5: Proposed future work

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

- The plans are built on careful analysis of what has been accomplished in the past.
- The future work addresses the needs of the project and will move the technology toward real-world application.
- The proposed future work is relevant to close out the project, even though the impact could be low.
- It would be good to see a comparison of other cathode catalysts, including lower loadings and different carbon support materials, to fully understand the impact of these anode modifications.
- This project's future work plan is well focused, but additional stack testing with a second subcontractor would be appropriate to utilize the benefits of the improved catalysts—assuming that the improved catalysts are commercially viable and the MEAs are ready for manufacture.
- The proposed future work focuses on evaluating the OER catalysts in short stacks for "real-life" automotive applications, improving the durability of the catalyst, and understanding the impact of OER catalysts on other MEA components. The proposed work involves the relevant areas for focus in the last few months of the project. However, it is unclear whether much fundamental work can be done in the short time left. It would be good to see some work toward the fundamental understanding of the mechanism by which SU/SD degrades the OER catalyst.
- The details of the future work are not provided. More stack testing in real-world conditions is all well and good. Hopefully the protocols will be provided. It is not clear how the fundamental engineering studies will be done. These catalysts cannot be "ready for real life" until NSTF is ready for real life. Therefore, some proof of concept of the high-OER cathode/low-ORR anode on carbon-supported catalysts would be very interesting.
- It is not clear that the proposed future work will actually result in any substantial additional improvements; instead, it appears to be just some "tweaking" or optimization of an approach that still requires substantial improvements to be deemed extremely valuable. It is unclear why an all-NSTF MEA is not on the path forward. One would think that a carbon-free MEA with these OER catalysts would provide much greater durability. Presumably, this type of MEA is not being pursued because an all-NSTF MEA presents too many other issues, such as unacceptably high sensitivity to temperature and impurities.

### Project strengths:

- This project features a strong team that has made reasonable progress since the beginning of the project.
- This project benefits from strong technical leadership and the team's ability to exploit unique technology.
- The project has a strong team that can address the fundamentals of stack testing. The results are of high quality and address the barriers.
- This is a sound, creative approach based on electrochemical fundamentals. Testing is done at an OEM at supposedly relevant test conditions. The team is strong.
- Strengths of this project include the collaboration with AFCC and the inclusion of start–stop and cell reversal testing under real-world gas switching conditions.

- This project's strengths include using cutting-edge microscopic and spectroscopic characterization tools to understand the behavior of the IrRu additives, and the team's good grasp of materials (both catalysts and membranes).
- The NSTF catalyst provides a nice architecture to implement this alternative-anode strategy. The principal investigator has done a lot of good work to fundamentally understand what catalysts can provide decay mitigation without having too much negative impact on the cell performance. Another strength is having fuel cell OEMs on the team to assess technology at the stack level.

### Project weaknesses:

- The project is focused on 3M materials only (membranes and NSTF catalyst layers).
- The project needs to define an "optimized candidate" (adjust the Pt loading as needed for the overall benefit) so the industry can take advantage of the accomplishments.
- Weaknesses include the dependency of the concept on NSTF electrodes and the lack of disclosure of protocol details.
- This project is not a high-impact project and only provides a marginal improvement over the state of the art.
- There is no apparent route to developing an MEA that is sufficiently resistant to start–stop decay, which means that other strategies (e.g., system-level mitigations) are still required, and therefore the impact of this project is severely limited.
- The presentation was hard to follow. The information is somewhat cryptic because a lot of data are jammed into a short presentation. The oral presentation did not provide clarity and was difficult to understand. Consequently, the reviewers' work was difficult and time-consuming because they needed to spend a lot of time figuring out the data.

### Recommendations for additions/deletions to project scope:

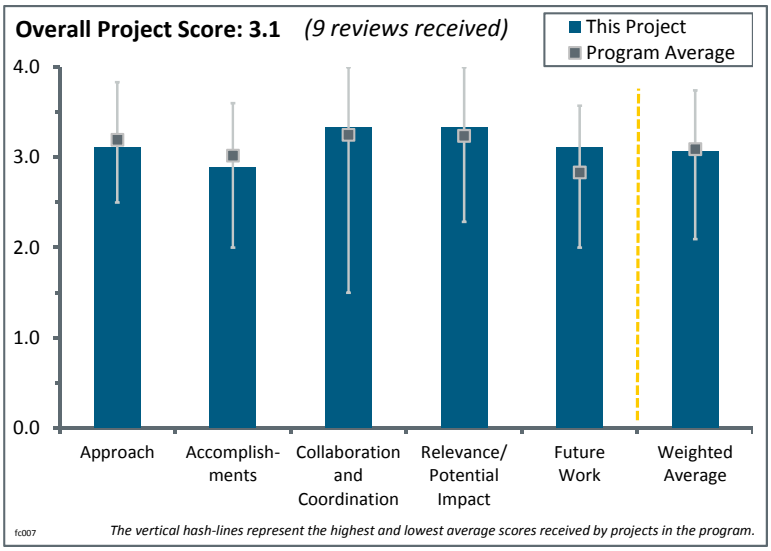
- The researchers should finish the existing scope of the short stack testing.
- The project should continue the focus on catalyst improvements, but a new project should be established devoted to membrane additives.
- The project team should include testing under non-excursion durability conditions and then run the excursion tests. The researchers should also apply the concept to carbon-supported catalysts.
- In situ EXAFS or XANES studies of the gas switching experiments may help elucidate what is happening with the RuIr (if the sensitivity is high enough to detect these low loadings).
- It would be good to see some work done toward understanding the mechanism by which SU/SD degrades the OER catalyst. Furthermore, it is unclear whether the Ru dissolves and crosses over from the anode to the cathode and, if so, how much crosses over. The team should use CO stripping as a method to detect the presence of Ru because this technique is very sensitive. If no Ru is detected on the cathode side, then it is unclear what is happening to the Ru.
- It would be good to see this anode protection applied to the SU/SD protocol with low-loaded cathodes, where the cathode support material can be discussed. In addition, a comparison between high surface area C, Vulcan, and graphitized carbons would be really valuable to see the effect of the anode protection. The start-up events should also be measured at low temperatures (such as 25°C) and with higher relative humidity contents. Related information of the stability of the Ru in the anode catalyst should also be measured—information such as stability to liquid water, which may happen upon SU/SD. Initial characterization of the materials is presented; post-characterization should also be completed.

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

Bryan Pivovar; National Renewable Energy Laboratory

## Brief Summary of Project:

This project aims to synthesize and characterize novel extended thin-film electrocatalyst structures (ETFECS) with increased activity and durability and to incorporate ETFECS with the highest potential to meet U.S. Department of Energy (DOE) targets into membrane electrode assemblies (MEAs) for fuel cell testing of performance and durability. Work is focused on overcoming the cost, performance, and durability barriers for fuel cell commercialization by increasing platinum (Pt) mass activity. Extended surface Pt catalysts were developed for their high mass activity and durability, and these structures were incorporated into robust, high-efficiency MEAs.




## Question 1: Approach to performing the work

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

- The project's approach was both thorough and productive, quantitatively identifying each candidate's strengths and weaknesses.
- It was a good idea to incorporate 2020 targets in the last stages of the project.
- The project is highly focused on the key barrier of catalyst activity and durability. The "all metal" approach of unsupported catalysts is a good solution to the carbon corrosion problem. There is good balance between synthesis and making electrodes/MEAs. It is good to see the team take a "new slant" and pursue relatively low-surface-area materials but high utilization. It was brave to eliminate some less-than-promising catalyst approaches to focus on the best-in-class approaches to date.
- The use of the structures is interesting, and the work with MEAs is good. It is unclear how the MEAs are optimized. The comment on the short stack not being done is disingenuous.
- The extended electrode structures represent a promising approach, but it is not clear that they will deliver the performance that is expected.
- The guiding rationale behind the project—obtaining high-catalytic area-specific activities typical of extended surfaces, while also achieving high mass activities by using thin continuous layers/coatings of the catalytically active metal—is a sound approach and has been demonstrated to be valid by 3M with its nanostructured thin-film (NSTF) catalysts. The unique aspect of this project's approach was to incorporate these extended thin-film catalysts into a thick dispersed electrode architecture to enable mitigation of the issues that arise with the very thin electrodes of the 3M architecture. The project has, however, struggled with obtaining uniform and pinhole-free extended thin films that are thin enough to achieve high mass activities. The researchers should be commended for establishing strict go/no-go decision points for the various catalyst synthesis approaches and for following through on the down-selection of approaches in this last year of the project.
- This project addresses key barriers—the cost and stability of the catalyst—in a manner that is promising and apparently not being pursued significantly enough by others. The thin-film catalyst approach has shown great promise toward meeting the catalyst activity targets and has an inherent stability advantage over conventional dispersed catalysts. Although 3M originally introduced the thin-film catalyst architecture, the company appears to be either unwilling or unable to bring this promising concept to

commercialization. Specifically, the 3M NSTF MEAs are too sensitive to temperature and impurities to be considered commercially viable. This project appears willing to address these issues by exploring alternative thin-film catalysts and MEA compositions.

- The initial design of this project was excellent: grow and characterize extended continuous Pt nanostructures that could harvest the durability and specific activity advantages of 3M's NSTF while allowing flexibility of electrode design that could overcome some of the weaknesses of NSTF, such as cold start and stability during load transient, that arise largely from the thinness of the NSTF electrodes. While the initial design of the project was excellent, difficulties in growing real continuous layers (rather than aggregates of Pt nanoparticles) and in achieving the mass activity target have led the project into down-selects that make the remaining efforts essentially an investigation of Pt blacks, either unsupported (Pt/CoNW) or supported on a corrodible metal (Pt/NiNW), with limited structural control of the active Pt particle layer. Given the frustrating results with the other preparatory methods used, the down-selects were probably appropriate, but they constitute a major deviation from the original aim of the project.
- The use of extensive surfaces for new catalysts holds tremendous potential for value. Higher coordination of Pt has been associated with higher specific activity and better durability. If extended surfaces can be fabricated into high-surface-area catalyst layers with sufficient ability to reject product water, the decrease in precious metal loading could have a significant impact on cost status. A no-go decision was made on synthesis routes that were used in association with more intrinsically stable supports, such as metal oxides and graphitized carbon species. On the surface, the decision appears logical; catalysts that surpassed the mass activity target were studied further, and those that were lower than the target were not studied further. However, those that passed were based on the deposition of Pt onto base metals (Co, Ni, Cu, and Ag) capable of leaching in a fuel cell environment. Other projects from Argonne National Laboratory and General Motors are already studying PtNi and PtCo alloys but in ways that minimize the amount of Ni or Co, which is unlike the approach taken with the PtNi nanowires. The project may have benefited from an approach that first emphasized stability, followed by activity.

### 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 team thoroughly evaluated the ETFECS candidates, providing valuable insight to this class of supported catalysts.
- It is good to finally see some MEA results. The atom probe characterization seems like a good tool.
- The investigators have made very good progress and are incorporating the materials in MEAs. The oxide appears to cause durability issues.
- Great progress has been made toward exceeding the DOE 2020 targets for performance using galvanic displacement (slide 6). The switch from Ag to Co and Ni allows the possibility of spontaneous alloying to improve catalytic activity. The oxide film issue and the differences between Ni and Co are interesting, and the principal investigator has a good approach to understanding and using these issues to benefit the project.
- The researchers have identified catalyst systems that meet DOE activity benchmarks. The oxide states and characterization are important because this feature may lead to numerous routes with respect to activity and stability; however, precise control as an intermediate may be difficult. More electrode structure/incorporation work was expected by this stage in the project.
- This team has made excellent progress in developing new methods to fabricate ETFECS. This provides additional options that may actually be more commercially viable than the 3M synthesis. However, progress in MEA performance is disappointing; additional work is definitely still required in this area, yet the project is 80% complete. It is doubtful that the MEA performance targets can be met unless this project is extended.
- Significant progress has been achieved in obtaining high specific and mass activities in the rotating disk electrode (RDE) environment; however, this project has shown quite well that this is not directly translatable to high activity in the MEA, which is the true test of the potential of the materials to overcome the cost, activity, and durability barriers. The project team should be commended for down-selecting the most promising catalyst synthesis approaches this year and for fabricating and testing the down-

selected catalysts in MEAs. This down-selection should have been done sooner, though, as this year's report has shown that the MEA environment causes degradation and performance issues not observed in the RDE environment. In retrospect, a simple screening method of processing the catalysts as if they were being fabricated into an MEA to look for materials degradation via analytical characterization techniques would have been useful and could be useful to elucidate issues that will arise with the materials stability prior to optimization of the electrode composition and structure for utilization optimization. With so little time left in the project, it will be difficult to tackle all of the issues related to achieving high performance in an MEA, so the project should look for quick screening methods to determine the optimum electrode composition and fabrication procedure for the down-selected catalysts.

- The project has generated some novel catalytic materials with good specific and mass activities that challenge current hypotheses about the origins of high specific activities. NSTF was thought to achieve its high specific activity by taking advantage of the higher specific activity seen for flat Pt surfaces versus nanoparticles, likely due to the suppression of oxygen reduction reaction-slowing OH groups adsorbed on Pt defect sites. This project has generated catalysts with high specific and mass activities—sometimes containing very little alloying element—that have nanoparticle-like, small Pt grain sizes, yet achieve specific activities expected for flat surfaces of Pt or its alloys. These results show that higher specific activities can be achieved with Pt nanoparticles than had been anticipated. If continuing work in this project could explain the origin of these high activities, it could open up new rational pathways for catalyst development that could avoid the concerns about durability that arise whenever a non-noble element must remain present in the catalyst to maintain high activities during operation. The project has led to some further development of the rotating disk electrode technique as a means of screening the activity of catalysts of diverse structures. This is good as long as it does not divert DOE projects from the absolute necessity of testing catalysts in MEAs before decisions about relative merits of different materials are made. In other aspects, the progress of this project is disappointing. Real continuous layers, rather than clusters of nanoparticles, have not been achieved. Some of the structural control of the substrate that was achieved with the crystallographically defined Ag and Cu substrates has been lost in the shift to the less-well-defined commercial Ni and Co nanowires. Only a modest start has been made in learning how to make effective MEA electrodes with catalysts other than the standard Pt/C or Pt alloy/C materials.
- Most techniques for synthesis have not provided tenable catalysts. Even of those that passed the mass activity criteria, PtAg and PtCu were undesirable due to concerns about anode plating. For the PtNi and PtCo nanowires, the highest mass activities were achieved only at Pt weight percentages below 20%. The low Pt weight percentages may lead to thick electrodes and increased mass transport losses. This may have explained some of the low fuel cell performance. Failure analysis has not yet been thoroughly provided from fuel cell testing. It would be interesting to know whether Co or Ni entered into the membrane. The difference in Co and Ni behavior is somewhat puzzling. The Ni nanowires appeared to passivate, while Co leached out. In principle, it would seem that both metals should behave similarly. Some investigation should be made into whether the materials could be further engineered to provide Pt/base metal segregations that are best for activity and stability.

### Question 3: Collaboration and coordination with other institutions

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

- The project has assembled a strong and competent team of collaborators. Suggested additions would include industrial partners with expertise in catalyst ink formulations and application.
- This project features great coordination and management of a large and diverse team.
- The investigators have shown a high degree of collaboration with the partners, demonstrating excellent leveraging of resources.
- This project featured a good team, but everyone's role, input, and productivity were unclear.
- The number and quality of the collaborators were impressive, but it is unclear from the presentation what the contributions of each of the collaborators were to the progress presented.
- The involvement of original equipment manufacturers (OEMs) and catalyst suppliers is a rare accomplishment.
- At least 11 different organizations participated in the project, which was a considerable number. Only 4 organizations (the National Renewable Energy Laboratory, the University of Delaware, Oak Ridge

National Laboratory, and the Colorado School of Mines) appeared to remain involved in the base metal nanowire work that continued after the no-go decisions. The same four organizations were mentioned in the future work section. It was not clear from the slides whether Nissan remained involved.

- This project features a large and diverse team that includes all of the resources necessary to develop these new catalyst architectures. However, it is not evident that the team has the required expertise to fabricate and diagnose MEAs with these catalysts. Insufficient data on MEA performance is provided here, so it is unclear what is being done to understand the root cause of poor MEA performance.
- This project has drawn on the diverse talents of some of the best people available for the different preparation approaches. While the results have often been disappointing, this is not the result of incorrect choices of collaborators or inadequate efforts—the original subject matter of this project was very, very difficult. The project has resulted in a reasonable number of good published papers from multiple institutions. It might have been good to draw 3M more closely into the project to carry NSTF forward as a baseline continuous-layer system for comparison with the novel approaches.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- Durable high-activity catalysts are critical to DOE automotive goals.
- The ETFECS approach is an exciting extension of the NSTF idea without the drawbacks of an organic substrate. The durability and performance possibilities are of great interest.
- The project explored in detail a unique class of catalyst supports, providing an excellent evaluation protocol for future extended surface Pt catalyst candidates.
- This type of catalyst structure appears to have the most promise toward meeting DOE's goals on catalyst loading, stability, and performance. However, much work is still required to develop a high-performance MEA that is not unacceptably sensitive to temperature or impurities (e.g., 3M's NSTF MEAs). This project has enabled new thin-film catalyst options and appears willing to try alternative MEA architectures as well, which is what is required to develop a complete and commercially viable thin-film MEA.
- Cost and durability are the two major barriers to fuel cell commercialization. Developing a catalyst that addresses both cost and durability is the most relevant activity in fuel cell research. This project achieves relevance by attempting to develop an electrocatalyst that can achieve lower cost and better durability by using the principles already validated through research on other extensive catalytic surfaces. The role of extensive surfaces in increasing area-specific activity, while also decreasing the tendency of Pt to dissolve, has already been substantiated. What this project attempts to add is the means to develop such surfaces in the context of a high-surface-area catalyst structure, as well as in the context of a catalyst layer that can provide sufficient rejection of product water.
- The impact could be good, but the project team also needs to understand some of the work that has been done with NSTF and modified electrodes, such as what General Motors has done.
- The work is promising, but there are significant technical challenges to making these materials commercially relevant.
- Unless the significant barriers to achieving performance improvements in an MEA can be surmounted with the relatively little time remaining in the project, the project will have had little impact on the DOE RD&D objectives beyond the more fundamental studies on optimizing activities in RDE measurements.
- The original plan of this project would have had outstanding relevance because it could have helped address some of the shortcomings of the NSTF catalysts that have been developed with substantial DOE support. As the project has evolved toward advanced Pt and Pt alloy blacks, the relevance has been diluted because the potential advantages over standard Pt/C and Pt alloy/C catalysts have been reduced largely to suppression of support corrosion (if decent electrodes, less subject to start-up and transient problems than NSTF, could in fact be made with these materials). Surprisingly good electrodes with moderately low loadings have been made with Pt blacks in the past, but they remained inferior, overall, to the carbon-supported electrodes. In the final months of this project and in the final report, the participants should spend some effort communicating the advantages of their catalysts over advanced versions of conventional-supported Pt nanoparticles because the final catalysts from the project are quite different from those originally proposed.

### Question 5: Proposed future work

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

- The project identified an important issue—the difficulty of converting unique catalyst structures into electrodes—and it should focus on this issue.
- It is time to focus on "best in class" and develop the high-utilization structures that are part of the original approach. Limiting work to galvanic displacement and Pt:Co and/or Pt:Ni makes sense.
- The proposed future work is outstanding because it concentrates on the most important issue of obtaining high performance in the MEA environment through better catalyst synthetic and pre-treatment procedures prior to fabrication and improved MEA fabrication procedures.
- The plan going forward is a good one. There should be a focus on the oxide passivating layer on the Ni; it is unclear if it is a good thing (explaining the durability) or a bad thing (eliminating alloying and reducing performance). It is also unclear if it can be produced on the Co nanowires. There should be a big effort to get the fuel cell performance up. The structure of the catalyst layer needs to be fully addressed. It is possible that existing ink coating technologies are not going to work with these materials. This is a much longer-term activity that should perhaps be considered for a second phase of funding.
- Only a few months remain in the project, and the project team is doing what it can in the remaining time with materials derived from galvanic displacement. It would be good if one of the bullet points described efforts to improve mass activity for higher Pt weight percent samples, particularly with respect to PtCo.
- There appear to be many issues with fabricating MEAs, and it appears that the team has some near-term plans to address some of these fabrication issues; however, it is not clear if there will be sufficient time to overcome even these issues. More importantly, it is not clear if the team understands what the inherent challenges are that must be overcome to construct an MEA with ETFECS that will perform well under all operating conditions.
- The project is nearly over, so the future plans are not as important as they were earlier. The emphasis placed on trying to make electrodes that perform well at both low and high current density with improved versions of the down-selected catalysts is appropriate. More emphasis should be placed on understanding the origin of the very high specific activity achieved from some of these materials, which are aggregates of Pt nanoparticles and often contain very little if any alloying element.
- The future work with MEAs is good. It is unclear how the MEAs will be optimized, especially under different conditions such as saturated humidity, where there might be problems with flooding.
- The investigators have addressed concerns from previous years, but much more progress needs to be made on the materials synthesis.

### Project strengths:

- This project features good planning and direction. The industrial collaborators are a great asset.
- This project features a good concept and diagnostics.
- The project's rationale and underlying premise are valid—that is the underlying strength of the project. The team is also outstanding.
- This project features a novel approach to improving the mass activity of Pt electrocatalysts.
- The project team demonstrated excellent capabilities to screen and down-select ETFECS candidates that meet the required performance goals.
- The "shotgun" approach to catalyst making has worked well, and the result is a good base set of materials to further develop. The team and partners are well positioned to capitalize on their knowledge and analytical base to develop electrode structures and MEAs using these novel "non-carbon" materials.
- The project team has successfully developed new thin-film catalyst options with high performance. This is an excellent team for catalyst development. The team is open to alternative MEA composition and fabrication methods with thin-film catalysts.
- This is a wide-ranging effort to synthesize continuous Pt layers on a range of nonstandard supports. The project has achieved improved reliability of RDE activity testing, although it is not clear whether the full range of surprising results from this technique has been communicated. The team has also achieved down-selects to the materials giving best mass activities in RDE, as well as the exclusion of Ag and Cu systems that can poison the fuel cell anode. A start has been made on MEA testing.

- The original premise is strong—deriving stable, high-specific-activity catalysts from extensive surfaces. The investigators were able to assemble a large number of organizations to assist with the project. Analytical techniques have been able to accurately show material deficiencies, whether such deficiencies are related to electrochemical activity or material structure.

### Project weaknesses:

- The ETFECS technology may be difficult to implement from a manufacturing perspective.
- The project features too much work on RDE studies.
- It is a concern that the industrial collaborators are not as engaged as they might be with regard to scale-up and optimization of the catalyst layer.
- The MEA performance is still unacceptable. It is not clear that the team has the MEA performance diagnostic expertise required to successfully meet performance targets.
- A weakness for this project (and other catalyst evaluation projects) is the inability to convert supported catalyst candidates into catalyst coatings and electrode structures for MEAs. Each candidate has its own unique challenges, and catalyst ink formulations and application techniques can be difficult to resolve.
- Porosity control with all metal structures can be problematic. This problem may be further complicated when the inks are not stable and structures with poor uniformity are created. While mixing corrosion-resistant carbon materials in with the all-metal catalysts is a logical solution to the porosity issue, it seems to be an automatic dilution of the catalysts and thus leads to some loss of activity. If an oxide intermediate is critical, it may be difficult to control uniformly in production.
- The project's weakness has been its difficulty in finding a synthetic approach to obtaining a catalyst with the desired composition and structure. This challenge has consumed most of the effort and duration of the project, when the focus should have been on proving the concept and optimizing the catalyst performance in an MEA by utilizing an existing catalyst or catalyst synthesis procedure (e.g., 3M's NSTF).
- One weakness is the team's inability to grow true continuous Pt layers (this is, in fact, very challenging). Another weakness is the lack of comparisons of all measurements to NSTF, the established continuous-layer catalyst (which may start out as original crystallites but smoothes out closer to a true continuous layer during breaking and operation). Down-selects removed some pathways that may have a better chance to produce real continuous layers—the original goal of the project—than the retained Pt/NiNW and Pt/CoNW paths that produce aggregated Pt blacks.
- The no-go decision against more intrinsically stable supports was perhaps not the right decision. Granted, the activity was higher for materials derived from galvanic displacement, but the galvanic displacement technique allows vulnerability to in situ leaching of base metals. There are also other projects that may be able to address Pt/base metal nanowires while also adding considerable experience with acid leaching and annealing steps. Results yielded high mass activity at a low Pt weight percentage. It will be difficult to transfer the best technology to a practical fuel cell MEA without encountering mass transport issues. Catalysts might be limited to lower current density applications, such as stationary applications or MHEs.

### Recommendations for additions/deletions to project scope:

- The team might want to consider trying to use NSTF and their inclusions in this framework.
- The team should demonstrate the performance of ETFECS in working devices.
- Now that the proof-of-concept stage has been successful, it would be good to see the OEMs and suppliers get more involved in the scale-up and optimization of the catalyst layer/coating processes.
- The project team should add at least one sub-scale cell test and diagnostics partner to the team, preferably one with good modeling capabilities, if this project is continued.
- This project (and other catalyst development projects) might benefit from additional resources within the national laboratories and industrial collaborators that can help with the conversion of promising catalyst candidates into electrode structures.
- The project has already deleted numerous research routes. No further deletions are necessary. The project may want to add some explicit investigation into increasing mass activity at higher Pt weight percentage, or at least make sure that steps related to ridding passivation layers from Ni or annealing are directly related to this topic.

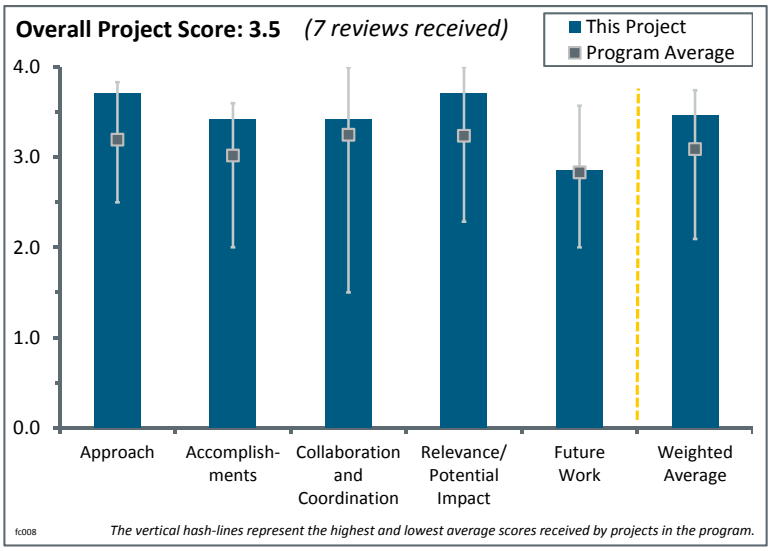
- The project is nearly over, so major revisions are not feasible. To the extent possible, the researchers should try to emphasize the need to engineer electrodes that behave well at high current density on air (under a variety of operating conditions) as well as in the kinetic region under $O_2$. They should take into account the possible role of high specific surface area in reducing local current densities ($A/cm^2$ of Pt), and thus in mitigating possible interfacial mass transport effects.
- The project should transition into electrode making because the initial approach is to build high-utilization structures. The team should develop methods and metrics for porosity characterization with the partners and then compare them to benchmarked traditional carbon structures to understand the difference. The team should also pursue porosity adjustment strategies. Although corrosion-resistant carbon additives could be considered a potential weakness, this avenue should be pursued fully to see if indeed the team can recover some of the activity lost when going to electrodes. It may be interesting to also mix all-metal catalysts of a different morphology to create the desired porosity. The researchers should consider developing a structure–function relationship around ink-making variables, the morphology of all-metal catalysts, and performance.

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

Nenad Markovic; Argonne National Laboratory

### Brief Summary of Project:

The main focus of this project is on developing a fundamental understanding of the oxygen reduction reaction (ORR) on multimetallic systems of PtMN alloys that will lead to the development of highly efficient and durable real-world nanosegregated platinum (Pt)-skin catalysts with low Pt content. The project has established a methodology to form and determine the nanosegregated Pt-skin surfaces for a different class of electrocatalysts, as well as established scalable synthetic protocols to produce larger amounts of materials.




### Question 1: Approach to performing the work

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

- The approach is sharply focused on achieving U.S. Department of Energy (DOE) goals on catalyst activity and durability.
- This approach toward creating nanostructured alloys for enhanced $O_2$ reduction is aimed at ultimately lowering the MEA costs in terms of platinum group metal (PGM) loading. The approach is based on forming core shell materials and incorporating them as nanowires. Using a combination of ligand and strain effects, differences in both mass and specific activities are observed. Some additional benefits of durability are also observed. The approach utilizes a synthetic strategy that combines surfactants, long chain precursors, and metal salts including carbonyl compounds to engender relatively uniform core shell materials to be synthesized. The approach is based on a wealth of prior art from well-known reports, such as those from Bonnemann. While the results are certainly good in terms of ORR activity, scaling up such synthetic approaches is always challenging.
- The general approach is good, although it does seem somewhat more fundamental than what the DOE Office of Energy Efficiency and Renewable Energy (EERE) usually funds. The approach focuses on improving the catalyst activity and durability via fundamental understanding, synthesis and characterization, and MEA fabrication and testing. It leverages the previous accomplishments on nanosegregated Pt alloy catalysts with superior ORR performance.
- To date, the technical work has been very thorough. The challenge now is deciding whether there is something significant in which to invest all of the future efforts in scale-up. A comprehensive test of concept is needed.
- This project is sticking to the original goal of achieving theoretical $Pt_3Ni(111)$ activity and bulk character, and it is taking a systematic approach (Pt multimetallic skin surface and segregated structure).
- The researchers have conducted very careful work in a very disciplined manner, focusing on the fundamental aspect of making high activity and durable catalysts, although it is unclear if these findings would transfer to a real fuel cell.
- This project is focused on the key barrier of maintenance of activity at low loadings of Pt. Whereas some are concerned as to whether this project leads to scalable catalyst layers on MEAs, the strength of this project's approach is in developing the underpinning knowledge of catalyst structure and how it relates to durability and minimizing the cost of the catalyst. In other words, the focus is on developing the appropriate structure–function relationships in multimetallic nanoparticle catalysts, for example, the

"design principles" of Pt on multimetallic surfaces (films, particles). This is a very valid approach that will find its payoff in supporting the global community's ability to design scalable, durable catalysts for MEA fabrication. The community needs a set of breakout ideas, not incremental ideas, and this team's approach has a good chance of providing such a breakout set of design principles for thrifting Pt while maintaining durability. Perhaps this may be one such potential breakout, if the researchers can ferret out what is going on with the "mesostructured" catalysts. The principal investigator's (PI's) capabilities and approach lie in detailing the underpinning fundamentals, not the area of membrane electrode assembly (MEA) fabrication, and the researchers should focus on their strengths to drive the whole community forward. Others more skilled in the fabrication methods will adopt the design principles as they are further developed and if they can be proven with time. This is an excellent approach.

### 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 team has made excellent progress toward DOE goals. All 2015 DOE targets with respect to the catalysts' activities and durability have been met (regarding rotating disk electrode [RDE] characterization) or exceeded.
- The synthesis and characterization of multimetallic core shell nanoparticles and Pt-alloy nanowires are outstanding. The work related to tuning the nanostructured thin-film (NSTF) surface structure is also good. The protocol for durability evaluation was not specified, making it difficult to determine if the durability test used was relevant. It is unclear if the electrochemical areas changed (slide 13). Impressive ORR activities and durability are shown in electrochemistry experiments. The ultimate test is how and whether this performance will be translated to MEAs; this should always be a part of the project. Results from only one set of MEA tests were shown. Much still needs to be done before the project ends. There is slight concern over whether MEA testing (50 $cm^2$) of optimized catalysts and scale-up of catalyst fabrication will be done on time because these tasks are only about 50% complete.
- Good accomplishments have been made in terms of activity enhancements. DOE metrics have been met in terms of the activity rubric. In terms of progress on understanding the nature of such activity enhancement, the progress is incremental. Model systems, for example, do not account for changes with potential. No clear metrics were given in terms of durability towards carbon corrosion. In addition, the durability metrics for catalyst stability were in the range of 0.6 to 0.9/0.95 V. As apparent from the comparison of the cyclic voltammograms, the onset of oxide formation, and hence the associated place exchange with subsurface oxides, is shifted positive for these ternary alloys. As a result, the alloys' higher durability would be predicted when compared to Pt, which has a more negative electrical potential to form such subsurface oxide formation. A true test would be to subject these materials to a higher potential, such as 1.1 V versus RHE. Under those conditions, large changes would be expected for such thin-film-coated nanostructures. Considering the fact that such potential excursions could occur on some cells in a stack, they should be included in such comparisons.
- Argonne National Laboratory (ANL) is making more than adequate progress in a difficult area of performing careful scientific work on difficult-to-characterize multimetallic structures, where the structure-function properties are just being defined—mostly by the team. DOE should be proud of the rate of accomplishments in this project. If there is an area of criticism, it would be the rate at which the milestones relate to the scale-up to MEAs. Because this is not the core technical strength of this team and the researchers appear to be relying heavily on the 3M collaboration, the perhaps slower-than-optimal progress is understandable. But this is a minor complaint. This team has a very respectable rate of publication in high-impact journals. While this is not necessarily an EERE metric, it endows this team with a good deal of credibility that its science is well reviewed and respected.
- There has been quite a stable of catalyst inventions from the work. There is a lack of real assessments of durability and performance.
- The approach to the mesostructured thin-film (MSTF) Pt–Ni catalyst is systematic, and MSTF shows significantly higher performance than NSTF. There are still a couple of remaining questions about (1) the stability of the preferred single crystal alloy surface (crystal orientation and segregation) after the cycle test,

and (2) whether there are any water management problems similar to 3M's NSTF, assuming the ANL team uses a similar whisker substrate to 3M's NSTF.
- The accomplishments over the last year appear to be the work in the areas of the Au sublayer, Pt alloy nanowires, and annealed NSTF. Although some work lacks originality, some valuable scientific achievements were generated. Demonstrated progress was shown using RDEs, but that does not necessarily correlate with MEAs. Syntheses were done in very small quantities; therefore, scale-up work is required prior to the much-needed MEA evaluation.

### Question 3: Collaboration and coordination with other institutions

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

- There is good collaboration between ANL, Brown University, and 3M. The roles of the Jet Propulsion Laboratory (JPL) and the University of Pittsburgh were not very clear.
- Overall, the collaboration and coordination of the project is excellent. It is unclear what JPL's role is in the project.
- This project leverages multiple national laboratories and universities to enhance its capability of analysis. It also uses an industry partner for fabrication of the catalyst.
- Good collaborations seem to be in place for this project. However, Brown University's role in this effort is not clear; perhaps it provided the synthetic approach. In addition, it is unclear if 3M was a materials supplier or whether it provided some other function. A clearer delineation of partners' efforts would help.
- The impact of the collaborations on the overall project is good; clearly Karren More is engaged, and the 3M collaboration appears to be okay. It is not clear what the others (JPL, Brown University, and University of Pittsburgh) are doing to support the project.
- It appears that many of the collaborators have been involved. The participation of General Motors (GM) needs to be strengthened.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The project has the potential to significantly advance the commercialization of fuel cells by using catalysts with ultra-low Pt loading.
- The quantum leap of mass activity improvement is critically necessary to reduce PGM loading.
- The development of high-performing and durable catalysts is relevant and has a high impact on DOE Hydrogen and Fuel Cells Program (the Program) research and development goals and objectives. The impact would be more significant if the high catalyst activity and durability could be translated to MEAs and fuel cells.
- The project addresses fundamental understanding of Pt skin, which is very valuable for designing good durable catalysts. Prior work by the PI has shown important implications for real-life catalysts developed under other projects.
- This project, which is largely focused on developing the underlying design principles of multimetallic nanoscale structures as applied to fuel cell catalysis, is highly relevant to providing the Program with potential non-incremental "breakout" approaches to improving the activity and durability of "thrifted" Pt catalysts. If this project's approach continues to demonstrate progress, these results will be adopted by others in the community that are more skilled in the application of these materials/catalyst approaches to scaled-up MEAs. This is a very relevant approach that can lead to a decrease in the cost of future electrocatalysts.
- This project has the potential to lower the cost of a PEM fuel cell MEA to below $30/kW, providing an important impetus to its commercial success. In this context, the project's meeting and exceeding of the DOE activity metric of 720 $\mu A/cm^2$ at 0.9 V versus RHE (iR-free) and 0.44 A/mg PGM (same conditions) are well received and acknowledged. The potential impact, however, should also reside on some of these nanostructured alloys in sustaining such activities at higher current densities. The PI assumes that such activity enhancement would be scalable; however, most of the ternary materials prepared in this effort

show the expected shift of the onset of oxide formation to higher potentials, and thereby have a relatively lower coverage of oxides at the comparative point of 0.9 V. This would, however, change when Pt oxide coverage changes at lower potentials. A higher current density (lower potential) comparison such as at 0.7 V (iR-free) would be more revealing. The potential impact is also based on the ultimate cost of these catalysts; typically, such nanostructured materials have remained an academic curiosity and have not translated well as commercial materials. Some work on scalability would be an important component in determining the potential impact.
- Until the durability is determined, the relevance/potential impact regarding DOE RD&D goals is open to some question.

### Question 5: Proposed future work

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

- The future work is well balanced between RDE and MEA work, with the focus on MEA testing.
- The proposed future work is adequately addressed.
- All efforts should first be directed at deciding the real status of this catalyst. If this means making a series of laboratory-scale preparations for durability testing, then that is what should be done.
- The proposed future work is reasonable, with scaling up of catalyst synthesis and MEA optimization. It is unclear which of the approaches, or whether all of the catalyst approaches, will be brought to the MEA fabrication and testing.
- At this stage of the project, a higher priority should be placed on demonstrating the activity and durability of the most promising catalysts in MEAs.
- Regarding MSTF, questions still remain and future work should cover (1) the stability of the preferred single crystal alloy surface (crystal orientation and segregation) after a cycle test, and (2) whether there is a water management problem similar to 3M's NSTF, assuming ANL uses a similar whisker substrate to NSTF.
- The proposed future work follows the path of scale-up to address the milestones more geared to application in the last term of the project. No mention was made of what effort was going to be directed at the new and interesting "mesostructured" catalysts. The PI also mentioned in his talk that the influence of lattice mismatch was important to the tuning of electrocatalyst activity, and that the subsurface "core" metal atoms are affecting Pt dissolution, reordering, etc. It was surprising that there was no mention of (a) what studies supported those claims and (b) what modeling was ongoing or proposed to substantiate those claims. This comment may be related to the state of collaboration in this project, which is not well defined, at best.

### Project strengths:

- ANL has excellent material synthesis and analysis capabilities.
- This project's strengths include the team's ability to generate and characterize laboratory-scale catalysts and then show some desirable properties.
- The team has very good synthetic capabilities. The team's knowledge of underlying principles of the ORR electrocatalysis allows it to generate new concepts for catalyst design in a timely manner. New mesoporous thin-film catalysts look very promising both from activity and durability points of view.
- Catalyst synthesis, material characterization, and electrochemical characterization are all strengths of this project.
- This project's strength is its theoretical background to achieve higher mass activity (beyond the DOE target).
- This project details the preparation of nanostructured Pt alloys for higher activity and durability. Both fundamental understanding of the underlying principles leading to higher activity and preparative techniques are addressed in this effort.
- This project features an exacting, detailed approach to get at the underlying "design principles" or structure–function relationships of multimetallic nanoscale catalysts. It also features a good team that is taking a knowledge-based approach to catalyst activity and durability.

### Project weaknesses:

- There is a lack of MEA and fuel cell data at this stage.
- These catalysts will be extremely difficult to scale up. Serious thought should be given to the decision of whether the optimization and scale-up are truly warranted.
- The project is pretty close to the end, but it is still focused on RDE testing (both for activity and durability). Considering that catalyst coated membranes will need to be optimized, the focus should be shifted to fuel cell testing.
- Scalability and the cost of the catalyst have not been adequately addressed to date, though they are on the list of future activities. A simple check on the cost of the precursor materials shows that the end product with lower loading would cost more than the typical Pt/C with conventional loading. This issue must be addressed clearly.
- An approach needs to be developed to transfer the knowledge gained to additional team members that are better suited to address issues such as scale-up of particle syntheses, incorporation into MEAs, etc., without affecting the rate of progress being made on new, more highly active, durable catalysts.

### Recommendations for additions/deletions to project scope:

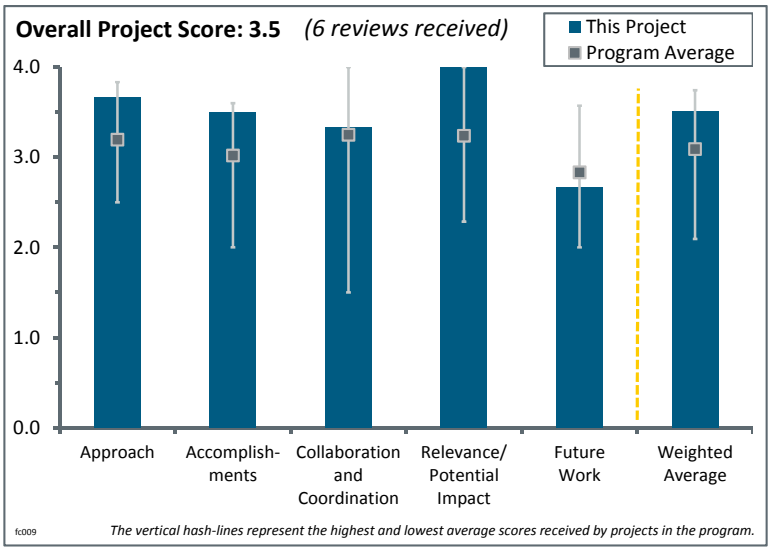
- There should be a stage gate with product durability and performance durability testing on a meaningful scale.
- Fe-containing catalysts may need to be excluded from consideration because of potential Fe leaching into the membrane.
- A clear mass and specific activity comparison with conventional Pt/C should be done between 0.95 and 0.7 V (iR-free). In addition, some OEM testing, such as in GM laboratories, should be a part of this effort.
- The influence of lattice mismatch on catalyst activity and durability effects would appear to be crying out for more theory to help guide the experimental effort and hopefully further increase the rate of progress.

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

Radoslav Adzic; Brookhaven National Laboratory

### Brief Summary of Project:

The primary objective of this project is to synthesize a high-performance Pt monolayer (ML) on stable, inexpensive metal or alloy nanostructure fuel cell electrocatalysts for the oxygen reduction reaction. Another objective is to increase the activity and stability of the Pt ML shell and the stability of the supporting cores while reducing noble metal content.




### Question 1: Approach to performing the work

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

- The approach is sound and strong.
- This project aims at producing a stable catalyst with 100% Pt utilization and thus greatly improved mass and specific activities. The use of core-shell interaction to improve Pt ML properties appears to be quite successful. Employing refractory metal alloys as cores allows for further reduction of platinum group metal (PGM) content.
- The approach to demonstrating novel ways to reduce precious metal loading while maintaining or improving catalyst activity is very good. One or two of the paths will be less likely to succeed in the long term from manufacturability and throughput perspectives—namely the electrodeposition and atomic layer deposition (ALD) techniques. However, it is good to include them for completeness of approach. The other aspect of the approach is the involvement of industry collaborators as independent evaluators of the technology developed; this has been very good.
- The methodology originally developed by the principal investigator (PI) is well established and well aligned with U.S. Department of Energy (DOE) catalyst goals of low-PGM/high-activity catalysts. It was very good to see the move to scale up the results to large-format membrane electrode assemblies (MEAs). There are some concerns about the practicality of using electrodeposition of the catalyst on the gas diffusion layer (GDL) and/or membrane.
- Brookhaven National Laboratory (BNL) is working on very promising catalyst materials, as evidenced by the licensing and scale-up of the core-shell catalysts by N.E. CHEMCAT (NECC). Both the Pt nanospheres and the Pt on PdAu nanowires are promising concepts to get to high mass activity. The electrodeposition directly onto GDLs is also a creative, promising approach to increase Pt utilization, although how much the utilization could theoretically be improved by this method should be considered before investing too much in scale-up of the concept.
- Routes using Pd are subject to the possibility of Pd loss with any loss of Pt ML shell. However, to the project's credit, efforts have been made to reduce low-coordination-number Pt atoms and to investigate stable cores (e.g., refractory metals). The very thin catalyst layers from growth on GDLs could possibly face the same water management challenges experienced by 3M's nanostructured thin-film catalyst. There is also some possibility of degradation from dissolution of either W or Ni. A very interesting part of the approach is the aspect involving Pt hollow nanoparticles, which still need further reduction in the Pt shell thickness.

### 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 presented Pt ML catalyst performance is very impressive. Based on the mass and specific activities given on slide 10, the electrochemically active surface area turns out to be 236 $m^2/g$ of Pt compared to 240 $m^2/g$ of Pt for a true ML catalyst.
- It is truly exceptional that so much data have been gathered on such a wide variety of catalyst options. The team's ability to demonstrate stability to voltage cycling is also very encouraging. The DOE targets have clearly been met for a number of different options, and technologies identified in 2012 have now been scaled up with help from partner organizations. The ability to innovate and then implement is the measure of success for any catalyst development effort.
- This project features outstanding activity and stability of a licensed catalyst. Improvements in mass transport, and noted by the presenter, are needed to truly realize the potential of these materials. "Pt hollows" is a great approach to eliminate carbon corrosion, but it will need substantial electrode development to realize its potential.
- Overall, BNL has made good progress. There is a need for more work with MEAs and understanding the implications of ionomers with the catalysts.
- The activity results on MEAs in $O_2$ are encouraging for all concepts except for the PdAu nanowires. However, these measurements were made at much higher pressures than the prescribed method (350 versus 150 kPa), so it is difficult to know whether these materials actually meet the DOE mass activity target of 0.44 A/mg-PGM. The PI is also encouraged to include the loading of Au as well as PGM in his analysis. Encouraging durability results were shown for ML/$Pd_9Au_1$/GDL MEAs after 2,000 cycles. Tests should be run for the prescribed 30,000 cycles using the DOE-recommended protocols. For most concepts, the $H_2$/air performance is very poor; BNL should work closely with its partners that have MEA development experience to try to optimize these electrodes to minimize mass transport losses.
- Impressive activities have been shown on a per-PGM basis for most catalysts, particularly for monolayers of Pt on hollow Pd (530 A/g PGM), Pt/Pd from ethanol (400 A/g PGM), Pt/Pd nanowires (500 A/g PGM), and Pt/PdAu (500 A/g PGM). Some indication of stability was shown for Pt on hollow Pd, although the true test will be in a fuel cell under realistic load cycling. The Pt monolayers on the PdAu core concept will have to be developed further to improve activity per mass of precious metal. So far, stress tests on catalysts with WNi cores supported on GDLs have not shown degradation in activity, but they have instead shown an increase with 0.6–1.0 V cycles. Failure analysis on these MEAs would be good to see to understand how the durability was maintained. Fuel cell data in air (or activity data in $O_2$) will be needed to evaluate the usefulness of the ALD materials.

### Question 3: Collaboration and coordination with other institutions

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

- BNL has excellent collaborations with universities (for fundamental characterization/calculation) and industry (for material processing and MEA testing).
- It was a great achievement to give a license to NECC. Team members 3M and Johnson-Matthey Fuel Cells, Inc. (JMFC) were important to the electrode structure development.
- This project features good technology transfer and partners, but the project team needs to identify a cell tester.
- The collaboration with partners has been very good. One consideration is the consistency in measurement across the different options of 2,000, 5,000, 10,000, and 15,000 potential cycles—it appears as though there is a need to standardize the approach across the different groups.
- It is unclear what the Massachusetts Institute of Technology did, and the JMFC fibrous support work seems out of place with the rest of the project. It was good to see BNL engaging 3M and General Motors (GM) to get cell and stack data after UTC Power left the project. The fact that the researchers have been able to commercialize the Pt ML/Pd/C catalyst at NECC shows strong industrial collaboration.

- Unlike other projects, this project is not in substantial need of collaboration at the material synthesis level. That said, JMFC has been used for collaboration on more novel catalysts. BNL has recognized that further collaboration is needed for both material scale-up and fuel cell testing. NECC has entered the project for the former and 3M for the latter.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

This project was rated **4.0** for its relevance/potential impact.

- This is a very critical component of the overall Fuel Cells program effort.
- This project addresses DOE automotive targets for catalyst activity and durability.
- The development of stable, high-activity catalysts is one of the most important challenges facing the commercialization of automotive fuel cell systems.
- The ability to reduce precious metal loading and maintain performance and durability is paramount to the successful cost reduction of fuel cells. This project clearly has all of these elements in mind, as well as the equally important technology implementation piece.
- Performance, cost, and durability are major barriers for $H_2$-powered fuel cell vehicles to be commercially viable. This project addresses all of them. The presented results look promising for automotive applications.
- Cost and durability are the major barriers to fuel cell commercialization. The new cathode catalysts provide the possibility of addressing both barriers. The catalysts developed in this project seek to address both cost and durability.

### Question 5: Proposed future work

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

- The proposed future work is in the right direction to a low Pt-loaded catalyst enabling competitive fuel cell vehicles. However, the observed poor air performance needs to be addressed.
- The desire to do further work on electrodeposition is understood; however, perhaps this would be at the expense of other areas in the project, given the timing (i.e., perhaps it would be possible to make more progress in other areas if electrodeposition were dropped). If it is for fundamental understanding, and this is the only way to approach the core design at the moment, then this is acceptable.
- There are still many materials options being developed. BNL should prioritize one or two concepts based on activity and durability testing at DOE conditions and cost projections of the scaled-up process (e.g., it is unclear if electrodeposition is cost-effectively scalable), and then focus on electrode optimization and ultimately scale-up for stack testing of these down-selected concepts. The future work should also include thorough post mortem analysis of voltage-cycled catalysts. The 3M durability results are particularly surprising, and it would be good to know what is happening at the catalyst, particle, and molecular levels. Particle size and composition before and after cycling should be reported. For the electrodeposition work, microscopy should be done to make sure that BNL is actually getting only the proposed structure of accessible Pt.
- Electrodeposition for catalysts has been tried often throughout the years. However, commercialization is rife with difficulties because one needs precise control of the electric field to alleviate non-homogenous depositions, and this issue grows with size. While there are commercial high-speed electrodeposition processes, such as wire galvanization, the goal here is to create impermeable thick films, which is the opposite for fuel cell materials. The plan to focus on electrode structures to optimize utilization of these high-potential catalysts is well aligned with DOE goals.
- Much of the fiscal year (FY) 2013 future work focuses on synthesis optimization of Pd-containing catalysts. However, more interesting topics might include (1) using the new U.S. DRIVE Partnership Roadmap (http://www1.eere.energy.gov/vehiclesandfuels/pdfs/program/fctt_roadmap_june2013.pdf) to substantiate durability of the Pd-containing catalysts, especially the Pt/Pd/W/Ni catalyst; (2) exploring the low-temperature performance of catalysts directly deposited on the GDL in order to explore water

management; and (3) continuing to decrease the shell thickness of hollow Pt nanoparticles (the latter should be moved up from FY 2014).
- There seems to be a lot of work left to do including making enough catalyst and testing it in MEAs within fuel cell stacks, as well as continued optimization, without much time left. It is unclear whether the FY 2014 plans makes sense because the project is set to end this year.

### Project strengths:

- This project features a good approach and progress.
- This project's strengths include its outstanding development of high-activity, stable catalysts and its "top flight" catalyst development team and PI, who have a proven track record endorsed by industry.
- The team has synthesized a wide range of catalyst materials and has moved quickly to MEA testing. Materials have been made that can be scaled up by suppliers.
- BNL has been able to leverage wet chemical deposition techniques to provide a variety of new catalysts. The leadership of the project has enormous depth and experience in the field. The project is able to reach out to suppliers for scale-up of catalyst concepts. The project has shown the ability to reach out to stack and MEA manufacturers for testing support.
- This project features a high "bench strength" with industry partners that can evaluate the technology and provide good input. It also has a solid technology pipeline, starting with the monolayer approach and then going forward with subsequent improvements. The use of multiple manufacturing methods to achieve the desired PGM target is a strength.

### Project weaknesses:

- This project features too much work with $H_2/O_2$; it needs some MEA development.
- This project's weaknesses include the use of electrodeposition for catalysts and electrode development/structures for high utilization.
- This project has inconsistencies in the format of voltage cycling tests. It also uses multiple manufacturing methods to achieve the desired PGM target—this is a weakness.
- This project's weaknesses include a lack of data at DOE-recommended operating conditions, especially activity at 150 kPa. More postmortem analyses would be nice, too. Except for the Pt ML/Pd/C catalysts, the model structures are not supported by HAADF and Pd-EELS analyses. The team needs to focus on optimizing the most promising concepts.
- After developing a catalyst and measuring good beginning-of-life activity, the PI should move quickly toward performing durability measurements in a fuel cell. This should be prioritized over catalyst optimization. The PI should place a greater emphasis on core stability. This emphasis should translate to the scale-up of stable cores and immediate testing to validate the stability of cores. The project materials are often premised on the advantages offered by precious metals, such as Pd and Au.

### Recommendations for additions/deletions to project scope:

- It would be good to see more work on $H_2$/air as well as on the possible impacts of contaminants and water management, transport, and ionomer aspects with these new catalysts.
- The project team should narrow the focus to wet-based methods for catalyst preparation (the team should leave electrodeposition and ALD out unless there is a plan to be more aggressive in each of these areas).
- The project team should test activity and durability using DOE-recommended protocols. It should also conduct beginning-of-life and post mortem transmission electron microscopy analysis on all concepts. In addition, it should engage 3M, GM, and JMFC on electrode design of the most promising concepts to resolve the $H_2$/air performance issues.
- While excellent performance was demonstrated in $H_2/O_2$ measurements, mediocre performance was observed in $H_2$/air operation, which is more relevant to automotive application. The root cause of such poor performance should be investigated in this project, rather than simply stating that it is a mass transport issue.
- Prior to further electrodeposition work, the team should perform a more thorough cost–benefit analysis to compare material throughput rates, cost of equipment, and envisioned configuration, and then compare this

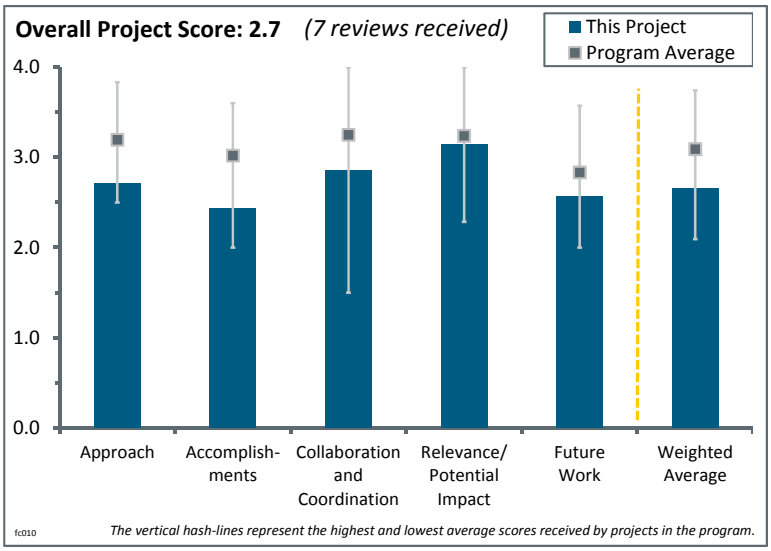
to traditional bulk catalyst synthesis, ink making, and coating. The metrics for uniformity of the electrical field that would be needed should be defined as well as the cost to achieve this. The team should increase the involvement of ink-making and electrode structure development partners.

- The project is fairly late in its timetable, so there are no suggested additions. BNL should increase efforts to validate catalyst durability (especially in a fuel cell) for both prescribed stress tests and durability cycles. BNL should increase the priority for scale-up of Pd hollow nanoparticles, as well as durability testing for them in a fuel cell.

## Project # FC-010: The Science and Engineering of Durable Ultra-Low PGM Catalysts

Mahlon Wilson; Los Alamos National Laboratory

### Brief Summary of Project:




The objectives of this project are (1) development of durable, high-mass-activity platinum group metal (PGM) cathode catalysts, enabling lower-cost fuel cells; (2) elucidation of the fundamental relationships between PGM catalyst shape, particle size, and activity, allowing for better catalyst design; (3) optimization of the cathode electrode layer to maximize the performance of PGM catalysts, improving fuel cell performance and lowering cost; (4) understanding the performance degradation mechanisms of high-mass-activity cathode catalysts to provide insights to better catalyst design; and (5) development and testing of fuel cells using ultra-low-loading, high-activity PGM catalysts. This project will help lower the cost and the precious metal loading of polymer electrolyte membrane (PEM) fuel cells and improve catalyst durability.

### Question 1: Approach to performing the work

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

- The project is focused on key barriers and approaches to each issue. It appears to be sound and is yielding results.
- The project is focused on U.S. Department of Energy (DOE) targets for PEM fuel cell durability and activity.
- This project has included good work in general, although the proportion of performance evaluation and low precious metals studies should be increased. A lot of characterization and modeling has taken place.
- The approach is fair and may be able to overcome the barriers addressed; some of the methods proposed in the approach are already being used to prepare state-of-the-art membrane electrode assemblies (MEAs).
- The approach seems to bundle together as much material as possible. The most important part is the focus on pyrolyzed polypyrrole (PPPy) structures as supports for low-loading catalysts that will work. The rest is largely padding.
- The approach of this project is good, but more material should be down-selected. The approach is well designed and combines the theoretical modeling and experimental work. The ultra-low PGM target for 2017 is 0.125 mg/cm$^2$, so modeling and microstructural simulations should be focused on low or ultra-low PGM loadings. Using ceria as an additive is an interesting concept, but it is not clear how this will help for ultra-low PGM catalyst design. Durability advantages with this approach are shown, but it was not clearly explained how this will help ultra-low PGM design.
- This project consists of several disconnected activities without a clear path to how the various approaches will be combined to address key technical barriers. The modeling work does not seem to tie in with the synthesis work. The ceria work is more focused on membrane stability and has little to do with "Durable Ultralow PGM Catalysts." The extended X-ray absorption fine structure (EXAFS) of the polyol catalyzation does not tie in with the other parts of the project, either. Only the Pt-PPPy nanowire catalysts—which include synthesis, characterization, electrochemical analysis, and fuel cell testing—seem to be a comprehensive study.

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

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

- The project team has made good progress in all areas of focus. Especially notable is the progress in making high-performance MEAs with ultra-low PGM loadings and good performance stability. Because the issue with these types of MEAs is often performance sensitivity with different operating conditions (e.g., 3M nanostructured thin-film MEA performance at 25°C), the team should also report performance at room temperature. If the performance of these nanowire-based MEAs is not particularly temperature-sensitive, then this would be an important result that should be shared with others developing similar catalysts (e.g., 3M and the National Renewable Energy Laboratory [NREL]). Los Alamos National Laboratory's (LANL's) catalyst layer compositions are presumably quite different from 3M's MEAs (i.e., LANL's contain ionomer in the catalyst layer) and may be significantly different from NREL's thin-film catalyst MEAs as well.
- There has been significant progress towards the stated objectives.
- Progress has been adequate. A performance evaluation is a critical need.
- The team is on track to meet DOE targets, but the progress has not been very fast.
- The progress with ceria as an additive is interesting, but ceria's role in designing an ultra-low PGM catalyst is unclear. The Pt-PPPy nanowire catalyst work is very interesting, but performance is below the carbon-based baseline catalyst. Also, durability should be evaluated under the U.S. DRIVE Fuel Cell Tech Team's suggested automotive-relevant accelerated stress tests (ASTs). Many PPPy nanostructures with various templates were generated, but most of them were not tested.
- Pt-PPPy nanowire catalysts need more work to approach DOE performance and durability targets. Activity measurements (either in rotating disk electrode or MEA) are not reported. Thus, it is unclear whether materials development, electrode optimization, or both are required. It is unclear how the density functional theory (DFT) modeling, the catalyst layer microstructural simulations, or the polyol catalyzation will help to meet any DOE goals.
- The progress toward the objective has been miniscule. The principal investigators seem to be trying too many different things and not making a concerted effort in advancing the technology towards the barriers. The claim about ceria addition improving the durability of membranes has been reported in the literature previously.

### Question 3: Collaboration and coordination with other institutions

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

- This project has good collaboration with strong teams between national laboratories, universities, and industries, with clear task assignments among the partners based on their competencies.
- LANL has assembled a strong team with broad expertise from national laboratories, academia, and industry.
- This project features a decent balance between original equipment manufacturers, national laboratories, and universities.
- It appears that many entities are collaborating. There could be some different order imposed based on the criticality of needs.
- There is good collaboration between the industry, national laboratories, and universities. The role of the University of Delaware is not very clear, as it is not mentioned on slide 17.
- The collaboration is reasonable, and the modeling work by Ballard seems to make reasonable progress.
- It would be good to see more interaction with other teams that are developing ultra-low PGM MEAs based on thin-film catalysts (e.g., 3M and NREL). Ideally, this project should have a viable MEA supplier on the team to ensure that the technology developed here can be readily incorporated into commercial MEAs.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- This project is definitely well aligned with the DOE goals, and the focus of the project is on key barriers.
- The project is relevant to DOE goals and the potential impact is high, considering the significant modeling effort.
- The project has the potential to advance progress towards the goals.
- The relevance of the project with the overall DOE goal for ultra-low PGM is very good, and the project features a very good team comprising national laboratories, universities, and industries. One of the objectives is the development of a durable, high-mass-activity PGM cathode catalyst to reduce the cost by utilizing ultra-low PGM loadings; however, no significant improvement in mass activity with ultra-low loadings has been shown so far. Optimization of the cathode electrode layer is limited to catalyst structure, and efforts should be put into understanding the ionomer effect in the catalyst layer, as well as its relation to durability.
- This project, if it is successful at lowering precious metal content with excellent performance, will be highly relevant.
- The stated objectives are relevant.
- Developing durable ultra-low PGM catalysts is one of the most important challenges that must be addressed to enable widespread fuel cell commercialization. Unfortunately, with the way that this project is currently structured, the work from this project is unlikely to promote that.

### Question 5: Proposed future work

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

- The modeling should focus on the nanotube/wire morphology both in DFT and catalyst layer models.
- Performance assessment is key at this stage. Then there should be a stage gate approach to defining the critical aspects required for success. The work plan should be adjusted accordingly.
- Plans need to be more focused on overcoming barriers. The modeling effort is not complemented by experimental model validation.
- It is difficult to understand the actual path forward to achieve the objective with the proposed future work. More experimental work is needed with material similar to what was used in modeling to validate the model. Also, the catalyst structure design should focus on understanding the effect of low loadings on mass transport and durability.
- Only the Pt-PPPy nanowire catalyst future work shows the potential to enable ultra-low PGM durable catalysts, but electrochemical characterization and durability testing should be included. The DFT modeling and catalyst layer microstructural characterization work needs an experimental validation component to provide value. The $CeO_2$ stabilization work is not related to the project goals and should be stopped.
- The proposed future work does not seem relevant towards the barriers addressed. The modeling work seems fine; however, it is unclear how the degradation kinetics will be covered by the model.

### Project strengths:

- This project features excellent bench-scale talents and capabilities.
- The project includes a balance of modeling, synthesis, materials characterization, and fuel cell testing.
- The modeling is a strength of this project.
- This project is focused on key barriers.
- This project features good management and good science.
- This project's strengths include its strong team, very good collaboration among the partners, and very good modeling efforts.

- There is a strong theoretical component in this project that can provide guidance for the design of better low-loaded catalysts and catalyst layers. This project features a very good set of characterization techniques.

### Project weaknesses:

- There are too many non-relevant pathways.
- There is no commercial MEA supplier on the team.
- The value of the approach is not well addressed yet. A rigorous performance test and stage gate review should be undertaken.
- So far, the links between the outcome and the objective of the project are not very clear. An ultra-low PGM catalyst with high mass activity has not yet been demonstrated.
- The modeling is not supported by experimental validation. The performance of the MEAs with Pt–ceria catalysts needs to be compared to the performance of MEAs with a ceria-modified membrane. If ceria additives lead to a gain in durability but also result in performance loss, the point of using them is unclear. If nanowire supports are used, it seems like it would be beneficial to use thin films of Pt instead of Pt nanoparticles.
- The project should focus on the PPPy supports and drop the ceria work, which is in the literature and does not need to be done again. Perhaps LANL is doing too much. There is no link between the different parts of the project. It is unclear when ceria is going to be added to the PPPy-supported catalyst. It is also unclear when DFT is going to be applied as a design tool rather than modeling what is already known.
- The various project work streams are not coordinated to best address technical barriers.

### Recommendations for additions/deletions to project scope:

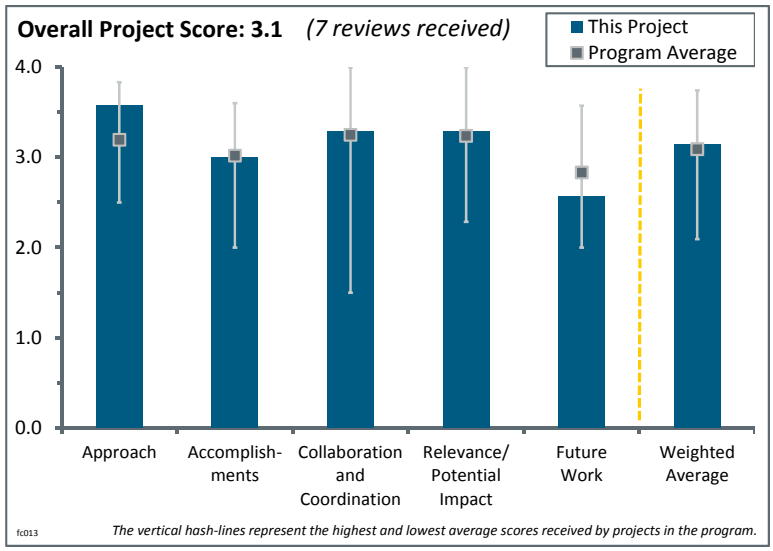
- The Pt–ceria catalysts do not look very promising.
- There should be more interaction with MEA developers.
- The researchers should focus on the PPPy support with experimental work and modeling work. The team should stop the ceria work.
- The team should focus on Pt-PPPy nanowire catalysts. It should measure and report ECSA and mass and specific activity values. It should also conduct DOE-recommended ASTs on promising materials. The team should make sure any modeling and characterization work is directly tied to materials development efforts.

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

Rod Borup; Los Alamos National Laboratory

### Brief Summary of Project:

The objectives of this project are to identify and quantify the components and component interactions and operating conditions leading to degradation in fuel cells, understand electrode structure impact on durability and performance, and develop models relating components and operation to fuel cell durability or degradation. Through this analysis, methods to mitigate degradation of components in fuel cells, increase fuel cell durability, and maintain component costs at a reasonable level are explored.




### Question 1: Approach to performing the work

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

- The project is sharply focused on the key areas of fuel cell degradation mechanisms.
- The approach covers all of the components of the fuel cell and the most degrading modes of operation.
- This is a very broad-based approach that seems to be well managed and productive.
- Understanding the degradation mechanism of catalysts is a prerequisite for addressing the durability problem. This is a well-defined, comprehensive approach addressing the degradation of Pt, carbons, membranes, and bipolar plates.
- Because durability is a key barrier to commercialization, this project is very important, and the approach of obtaining a better understanding of all of the degradation mechanisms is worthwhile. Additionally, the team uses a variety of good experiments to ensure that root causes are fully understood.
- The team members and capabilities are solid. There has been little done related to "real-life" testing or to compare the team's results with others. The fluoride emission rate (FER) has been studied, but only for a short time; this takes hundreds of hours to equilibrate. At low relative humidity, many investigators have shown the FER to increase rapidly with time as end groups are generated. One of the stated goals is to propose mitigation routes and offer solutions—these were not presented. The most important work the researchers have demonstrated is the difference in catalyst dispersion solvents. It would have been good to see a lot more work on this.
- Many of the data are similar to those that developers have already collected. Examples include (1) electrochemical surface area (ECSA) versus carbon loss; (2) evolution of surface oxides versus potential; (3) increased graphitic character of support carbon after cycling, as well as catalyst layer thinning; (4) performance degradation for various carbon types; and (5) fluoride emission rates (FER) from perfluorosulfonic acid (PFSA) membranes under various conditions. The project team needs to ask whether it can go deeper than just these data. If the assertion is made that corroded electrode layers are being compacted, then the porosity should somehow be measured. Changes in surface energies should be studied. It may be more useful for the project to focus on one component and examine it at a deeper level. The project is presently focused on the catalyst, the catalyst support, the catalyst layer, the membrane, and bipolar plates, which is quite broad. The electrode work was able to relate the role of electrode characteristics and processing (Pt%, ionomer/carbon [I/C], solvent type) to types of degradation in an empirical sense. However, most developers already have their own internal findings regarding these types of phenomena. Again, the project needs either to go deeper to discern the meaning in these data or to eliminate the task.

### 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.

- This project team has achieved a much greater understanding of fuel cell degradation.
- The project team has made good, steady progress on all tasks. There is a nice summary of key conclusions to date.
- The project team has made very good progress toward some fundamental, non-overreaching models for the durability of individual fuel cell components.
- This multi-laboratory, multi-team project achieved several useful results and developed models for membrane electrode assembly (MEA) components that help in understanding their degradation, the stability of various carbon supports, types of catalysts, and their loading.
- This project team has made good progress in understanding the electrode and membrane degradation, but it has achieved much less understanding of superimposing factors, specifically how the degradation of one component accelerates the degradation of other components.
- For the tasks that the team set out to do, there is a considerable volume of data. While there may be questions about the approach and which data add value to the community, it is without question that the project has diligently accumulated a voluminous database on all tasks. Interesting EA + E results show decreased thinning of the electrode. The models for catalyst layer degradation, membrane/ionomer degradation, and plate corrosion appear to have received a steady stream of inputs from experimentation and are still in progress. While the data collection has been excellent, the project does need to begin probing the utility of the data to developers. In many ways, a qualitative understanding has been developed of different phenomena, and a quantitative understanding is to be expected later through modeling. But the team needs to be aware of whether results can be extrapolated to more contemporary material sets. For example, there is question as to whether membranes with radical-scavenging or peroxide-scavenging additives would defeat the utility of the membrane model.
- There is far too much emphasis on carbon corrosion. Automotive original equipment manufacturers (OEMs) have low-cost options to avoid carbon corrosion. In addition, a lot of work has been done in this area, and not much light is added here. There have been some interesting results with the types of carbons, but the researchers have not done any work on generating their own supports to help prove their conclusions. The conclusion that decreasing performance is due to catalyst ionomer changes and ECSA, rather than the loss of catalyst surface area under some conditions, is certainly interesting and important. Some very easy experiments could be done to prove this. The cell could be flooded with an $H_2SO_4$ solution to see if the surface area is there but has just lost ionomer contact. Ex situ absorption could also do this.

### Question 3: Collaboration and coordination with other institutions

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

- This project features excellent collaborations in key areas.
- This project features a good team that appears to be well managed and is also well coordinated with other relevant DOE-supported projects.
- Good coordination of such a large number of teams was necessary to obtain the results presented.
- The collaboration and coordination were very good; the project features an apparently well-selected team that is also apparently well coordinated.
- The collaboration effort is very substantial; many qualified parties are involved. It would be nice to see more coordination between Oak Ridge National Laboratory and Los Alamos National Laboratory (LANL) in concluding carbon balance between thinning from scanning electron microscopy (SEM) findings and directly measured $CO_2$.
- In a sense, this is the most collaborative project in the Hydrogen and Fuel Cells Program. Eight organizations are listed, besides those involved in the other durability projects. The collaboration has been strong this past year and in prior years with respect to data gathering. The other national laboratories and University of New Mexico appear to have a strong collaborative network. Collaboration could be improved by contact with suppliers and stack developers to understand where the project could adjust to truly meet

application needs. While it is true that the team is tasked by DOE to carry out particular tasks, the team could benefit from obtaining feedback from developers to find out if its tasks are adding value or not, and then working with DOE to adjust to that feedback. LANL has a fantastic amount of scientific horsepower, but it could go wasted if a project such as this mostly reproduces data that developers already either have or have a notion of from their own findings.
- This project has too many collaborators working on different things with not enough to tie them together. There are 11 institutions at different locations, and only rarely is there coordination between them.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- This project is highly relevant to the commercial success of fuel cell systems.
- This type of project is key to meeting the DOE Hydrogen and Fuel Cells Program's durability targets. LANL has demonstrated that it can continuously deliver good insights.
- Degradation prevention would make some fuel cells applicable tomorrow.
- A project probing individual component degradation could be of extraordinary value to developers. Durability is one of the barriers preventing wider fuel cell commercialization for some significant applications. The project addressed polymer electrolyte membrane fuel cells at low temperature, which is a relevant technology for automotive, material handling equipment, backup power, and other applications.
- Most aspects align with the objectives; however, the real impact to the industry could be less significant.
- Durability models are at the component level; more system-level durability would be more relevant, but this is an important first step.
- A lot of this work is repetitive and has been done without searching for solutions or finding solutions, which is a stated goal. A large effort is being put forth on carbon corrosion, but just in the area of characterization, and a lot of this is already known.

### Question 5: Proposed future work

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

- There is a good plan for future work.
- The project is almost complete. One assumes that a comprehensive final report summarizing the key findings of the project will be issued.
- The proposed future work is good and appropriate in light of the budget remaining. This team apparently has not yet worked much on the thin-film type of catalyst architectures (e.g., 3M's NSTF), which DOE appears to hope is the future for meeting both cost and durability targets. There may be a gap in addressing potential durability issues that arise with these types of MEAs versus the more conventional dispersed catalysts on carbon supports, which has been the primary focus.
- Because contemporary material sets may differ, it may be good to reduce the work on membranes and bipolar plates. The ability of the team to gather data is established. The team would do well in the future work to include consultations with industry to understand if there are deeper directions to investigate with respect to catalyst-related components.
- It is not clear what priorities would be assigned to which aspects of the project, because a lot of modeling remains unfinished for the short time left in the project.
- Most of the emphasis is still on carbon corrosion, with the entire emphasis on characterization. The researchers have done a great job in showing how carbon corrodes the catalyst and where the catalyst corrosion occurs; it is difficult to see what benefit can come from modeling a corroded catalyst.

### Project strengths:

- This project features a strong team and good coordination.
- A strength of this project is its in-depth determination of fuel cell degradation mechanisms and processes.

- Work on the effect of the solvent on catalyst dispersion casting is very interesting and can have an impact on many players; more work on understanding this would be most welcome.
- Discovery of the weak characteristics of the electrode structure under high potentials and mitigation of degradation by mixing different supports into one electrode is worth noting.
- This project features a large, well-coordinated team that is making systematic progress toward understanding fundamental durability issues in fuel cells. Another strength is the good use of LANL's expertise.
- This team has an extraordinary ability to collect a lot of data. It has leveraged the abilities of the national laboratories very well. The team has access to an extraordinary scope of characterization and analytical techniques. The leadership of the project has a deep understanding of degradation phenomena.

### Project weaknesses:

- The project is unlikely to wrap up by September 2013.
- There should be more avenues and routes suggested for the mitigation of degradation mechanisms.
- The membrane degradation work is very superficial; no insight was given into the mechanism or changes with time. There is too much emphasis on carbon corrosion.
- The team needs to become more involved in understanding and blazing trails beyond what developers already know. The project is fairly spread out among a number of topics, rather than finding a particular focus where it can go deep and produce previously unknown results.
- The effect of open circuit voltage (OCV) degradation is described insufficiently. Stating that Pt deposition in the membrane is promoting $H_2$ crossover is not supported by the data. At the same time, potential cycling polarization curves are plotted with the same OCV during aging. It is not clear whether OCV degradation is studied only under OCV conditions or also during catalyst cycle tests, and whether and to what extent Pt loading affects OCV degradation. Mitigation recommendations are very minimal. More substantial recommendations are expected from LANL. Mixing of stable cathode supports into a single electrode is good, though it is not very novel and already utilized in the industry.

### Recommendations for additions/deletions to project scope:

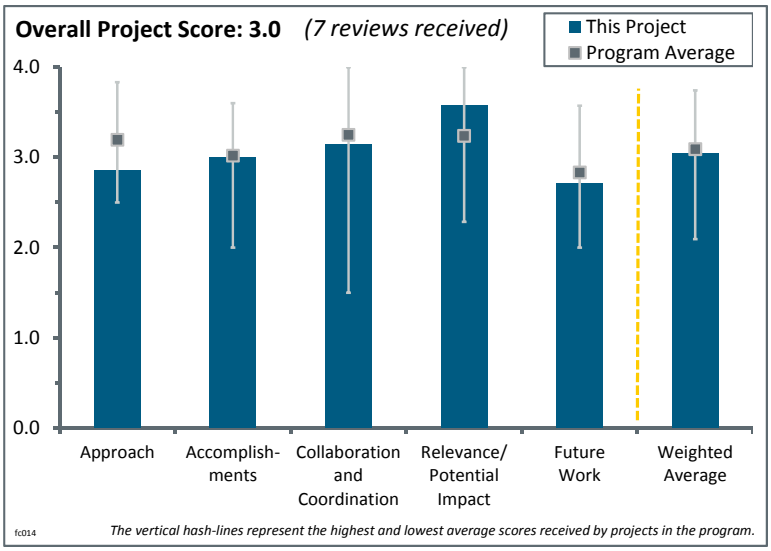
- The project team needs to figure out how to wrap up and publish work by the end of the project—or award an extension.
- The project team should prioritize modeling work of critical components and show at least one superimposing phenomena.
- A large effort to obtain bipolar plates from as many manufacturers and OEMs as possible would be helpful. Modeling transport in a carbon-corroded diffusion medium is of little value; work should focus on mitigating carbon corrosion. The prediction activities (i.e., predicting how long catalysts will last under different shutdown scenarios) should also continue.
- The project team should delete most of the membrane and bipolar plate work if there is no way to extrapolate it from materials under study to materials that will be used in a future commercialized fuel cell system. An automotive stack OEM (e.g., General Motors, Nissan, and Automotive Fuel Cell Cooperation) should be added to the project to provide development context.

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

Scott Blanchet; Nuvera Fuel Cells

### Brief Summary of Project:

The objective of this project is to identify and model polymer electrolyte membrane (PEM) fuel cell durability factors associated with low-platinum membrane electrode assemblies (MEAs) operating at high power density. The key deliverable of the project is a durability model that is validated experimentally over a range of fuel cell stack technologies operating at high power.




### Question 1: Approach to performing the work

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

- The approach is good. The conditions of experimental validation and modeling focus on low Pt loading and high-power-density operation.
- The approaches to collecting data and developing models for describing durability dependence on operating parameters and platinum group metal (PGM) content are well organized and systematic. The results for this activity are outstanding. The initiation of the stack durability tests with automotive cycling appears to be a little behind schedule if the project is going to achieve 5,000 hours. There was no obvious approach to defining the stack cost. The provided presentation did not contain a reference to this barrier. If this barrier will not be addressed until the future, it should be stated; however, under the proposed future work, there was no indication that this work will be done. If it was done in the past, the presentation should say so. Because of the lack of information on stack cost, the approach deserved a rating of "good," rather than "outstanding."
- Both land-channel and open flow field designs are being evaluated. High current density and power operation are being used to address cost. The comparison of decay rates as a function of Pt loading is instructive.
- The project combines the use of modeling and experiments. The use of accelerated stress tests (ASTs) and their linkage to fuel cell or stack data are a valuable part of the approach. The model approach is not entirely clear, because the specific aspects related to phenomena experienced at high currents and the role related to the expected effect on the degradation mechanisms were not fully discussed or linked to the observations seen in the experiments.
- The approach was full of data and charts, but it was hard to interpret the conclusions. Without fully understanding the technology, one would not be able to easily decipher the impact of the plots shown.
- The technical approach is very systematic. Comparing Pt dissolution using ASTs and drive cycle performance in various single cell hardware configurations is very valuable; however, an understanding of shutdown/start-up degradation is necessary to understand performance degradation in the field. Thus, voltage excursions to beyond 1.0 V upper potential limit should be investigated.
- The overall objective of this work—to develop a universal predictor model for catalyst degradation—seems a bit far-fetched, considering the empirical basis from which the entire fuel cell community is presently operating (i.e., it is questionable that one test can be a fair predictor of all of the different materials being used, systems controls, and operational conditions of all fuel cells). While it is a nice thought that one might be able to just sit down in front of a computer, input some parameters, and then see how long a fuel cell is going to last, that seems a bit too simplistic. The use of the General Motors (GM)/Rochester Institute of Technology (RIT) flow field for comparison was also not clearly defined. It is unclear what the GM/RIT

flow field is, but it is well known that GM does not publish what it is really doing, and it is hard to predict why GM worked with RIT on the flow field or its true purpose. The implication is that the Nuvera technology is more stable than the GM technology; this seems like a big step, considering that most companies have many system-level controls in place to prevent degradation, and such conditions would be difficult to implement in the Nuvera (or any) model.

### 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 researchers have made good progress towards their stated goals.
- Nuvera has quantified decay rates as a function of Pt loading. A durability model is nearing completion for dissemination.
- The work is demonstrating the capability to reach the existing cost/durability targets. In particular, there is no acceleration of the degradation mechanisms due to operation at high power density. This conclusion is valuable and useful for the rest of the industry.
- The project has made good progress. Of concern is the large scattering in voltage loss at the high loading in comparison to the low-loading results. Furthermore, the thickness changes with cycling indicate that there is substantial cathode catalyst corrosion, which is quite surprising given that the AST and simulated driving cycle did not exceed an upper potential limit of 1.0 V. Voltage loss breakdown analysis would provide a better understanding of the results.
- The technical progress is steady. The remarkable point of this project is the study for high-power-density operation, as shown in the objective. Surely the conditions of experimental validation and modeling are related to low Pt and high power density, but what researchers study in the project and the description of the outcome should focus more on properties and key factors specific for high-current-density operation, which is more helpful for stack downsizing and thus future costs.
- Excellent progress has been made towards the development of durability models. The information regarding the impact of PGM loading is surprising, but the data appear to be reasonably obtained. The project appears to have a long way to go to get to the 5,000-hour durability target with cycling. If the project ends on September 30, 2013, it is not clear whether the researchers will reach 5,000 operating hours. The third-party data in chart 8 appears to state that the researchers have reached the durability with a short stack; it is unclear if the short stack data meet the objective. Perhaps the presenter said so in the talk. The presentation did not state that the short stack data was acceptable.
- The accomplishments seemed to be the accumulation of the data of the whole 3.5 years, so it was hard to isolate just the past year. If that is incorrect and all of the data presented were just during the last period, then the rating would be higher, but the presenter's talk seemed to cover more than one year.

### Question 3: Collaboration and coordination with other institutions

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

- Collaboration was impressive and well noted.
- This project features a strong team; the collaborative efforts are well coordinated.
- The project demonstrates a strong interaction with both academic institutions and national laboratories.
- The project has a high level of competent collaborators. The modeling activity and the post-test characterization of the Pt in the membrane are both very good. It is unclear if the stack durability testing is a problem at LANL or Nuvera.
- More effective and interactive collaboration can be done (e.g., analysis results from one site can be transferred to another site for modeling).
- The speaker mentioned that the model was available to everyone and was published, but no one can seem to find it. A better collaborative approach would be to truly make the model available for use/criticism/improvement prior to the completion of the project.
- There is no automotive fuel cell systems integrator on the team.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The project addresses stack durability and cost—the top two DOE priorities. A durability model will be disseminated.
- The project supports and advances progress toward DOE Hydrogen and Fuel Cells Program goals and objectives. The data developed to date expand the overall understanding of fuel cell stack durability. A successful stack durability model that is independent of flow field design would be very beneficial.
- Reducing cost and increasing durability of fuel cells are key DOE goals. This project is well aligned with those objectives because it addresses the performance and degradation of low-loaded MEAs and stack architectures that facilitate high-power operation, enabling cell count reduction.
- The project clearly supports the need to improve the durability of PEM fuel cells; in particular, the project's focus on understanding the effect of going to high power is of specific relevance to automotive applications.
- The study on high-power operation is valuable for potential cost reduction of the stack.
- The subject is clearly relevant to lowering the cost of fuel cells; it is just hard to interpret the conclusions.
- It is doubtful that the model developed has any real relevance beyond the Nuvera system, but it is hard to tell.

### Question 5: Proposed future work

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

- The plan and expected achievement should focus on specific factors and properties for high-power-density operation with low Pt. The project is wrapping up. Hopefully the researchers will make their model available.
- The project is coming to an end, and the durability model is well worth completing. It is hard to judge how relevant it will be for all Pt cases because of the complexity of the variables.
- The proposed future work is adequate, considering that the project ends in December.
- The proposed future work addresses the durability model and understanding the degradation of the stacks. The future work does not address stack cost.
- The project ends in 4 to 5 months, so the future work is mostly wrap-up and reporting. Dissemination of the durability model to industry is particularly important, although it is not entirely clear how applicable the model will be to other cell/stack configurations.
- There were deviations in the results of the tests done by each of the participants. Further, there were also issues with the post-analysis test regarding the cathode thickness. Both of these aspects could substantially affect the applicability of the information from the project. Neither of these issues was highlighted in the proposed future work, yet they seem important to address in order to firm up the conclusions and results.

### Project strengths:

- The project has a strong and experienced team.
- This project's strengths include its theme and value.
- This project is a very relevant topic that is of interest to a broad audience. The model could be useful as long as the user understands how to calibrate the variables. The project generates lots of useful, detailed data and an experimental design protocol for catalyst developers.
- The project's greatest strength is the collaboration team. The project successfully builds on the strengths of each of the collaborators. The systematic evaluation of durability with operating parameters is very good work. The comparison of stack architecture is interesting, but it probably needs more explanation. The degradation model information is very good.
- The project has strong interaction between collaborators and a combined model/experimental approach that facilitates both real data and theoretical understanding contributing to the outcome and achievements of the project. The use of different cycles—from voltage, load, and drive cycles—is very valuable in establishing

the linkage between ASTs and the real-world linkage to stacks in the field. The accomplishment of the model verification using an EIS-based parameter determination approach in order to simulate the AST/performance of the various cell configurations is also commendable because it provides insight into key measurement approaches and parameter determinations that can be used to obtain model inputs.

### Project weaknesses:

- Although an enormous amount of data was generated, the conclusions and impacts were not clearly defined.
- No information was given regarding MEA components such as catalyst support. The observed corrosion at an upper potential limit of <1.0 V is a concern.
- The presentation appeared to emphasize the validity of the open flow field design. It is unclear if this is a sub-issue for Nuvera. Hopefully the project was not undertaken just to prove that the design was valid.
- The objective of developing a universal model for stack degradation using contractor technology as the core technology is questionable. While a universal model sounds good, it is probably not realistic. It also seems that a not-for-profit contractor would be a better location for such work.
- There is clearly an unconsidered mechanism or potential carbon corrosion that is affecting the cathode catalyst thickness during the course of the testing. This effect could affect the outcome of the results of the project; in particular, the model does not consider carbon corrosion, only Pt dissolution. No analysis is discussed as to what impact this omission has on the model verification or fitting parameters. The discrepancy in the tests between the RIT cell and the SCOF also should be addressed; there is clearly some local condition that is affecting the results. In particular, given the focus of the project on high-current-density operation, some discussion of liquid water, running water content, the role of liquid water in the degradation mechanisms that are proceeding, and the manner in which the model describes and includes the effect of liquid water should be discussed or considered in light of these discrepancies.

### Recommendations for additions/deletions to project scope:

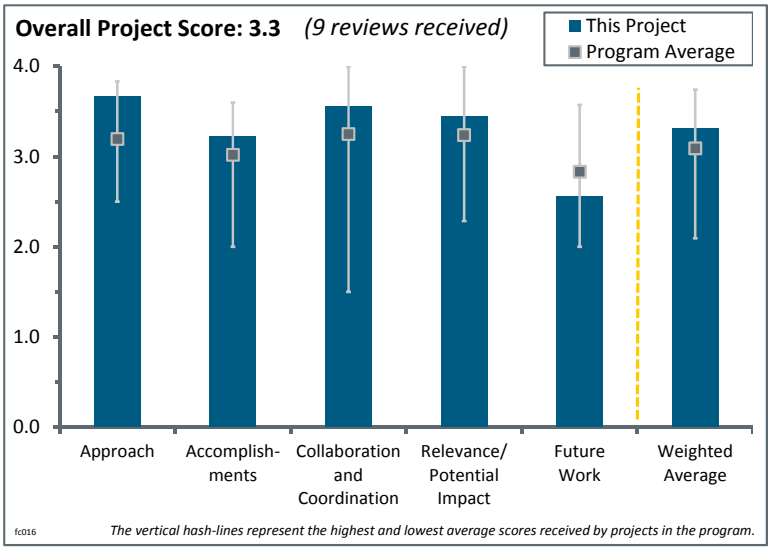
- The project should end as scheduled.
- Resolution is needed regarding when stack cost work will be done.
- The project team should conduct voltage loss breakdown analysis to better understand the impact of catalyst layer changes on performance loss.
- This project can bring potential value and insight for durability and cost/downsizing. Therefore, if the project focuses more on what is more specific for high-power operation with low Pt, it should be accelerated more.
- The project is ending, so there are no recommended changes on this project. However, there is a question regarding whether DOE should pay for-profit companies to develop models that favor their technology.

## Project # FC-016: Accelerated Testing Validation

Rangachary Mukundan; Los Alamos National Laboratory

### Brief Summary of Project:

The objectives of this project are to correlate component lifetimes measured in an accelerated stress test (AST) with real-world behavior of each component; validate existing ASTs for catalyst layers and membranes; and develop new ASTs for gas diffusion layers (GDLs), bipolar plates, and interfaces. The AST is important in that it allows faster evaluation of new materials, provides a standardized test to benchmark existing materials, and accelerates development to meet cost and durability targets.




### Question 1: Approach to performing the work

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

- The approach is very good/outstanding. The key is correlating the AST methods to the bus data.
- Durability is a key barrier, and having good AST protocols is very valuable to the fuel cell community with respect to developing more stable materials.
- The approach to achieving the durability technical target is sound and well designed. Durability is a critical barrier to reaching commercial fuel cell viability. Accelerated stress testing is one way to meet durability targets using a much-reduced time frame. Comparing accelerated test failure mechanisms with actual data from demonstration projects can provide useful and validated test procedures. Identifying specific tests that achieve a targeted failure mechanism is difficult to improve on.
- This work on ASTs is systematic, comprehensive, and sharply focused on overcoming the durability barrier. This kind of work, getting knowledge on degradation out into the public domain, is important so that component manufacturers—which are generally smaller companies, at least compared to car manufacturers—get access to knowledge on degradation mechanisms. It is not enough if the car industry has this knowledge available and holds it confidential. This work is also valid for the stationary sector, in which generally smaller companies are active.
- The principal investigators (PIs) are conducting valuable research on the durability of fuel cells that may contribute to understanding and addressing the degradation issues in fuel cell stacks. The approach integrates modeling and fuel cell stacks in operation. In some ways, this approach is systematic; however, it could be potentially improved if it was capable of pointing out what should be done to prevent decay in performance.
- The project has a well-defined structure focused on achieving the technical barriers. The approach identifies the probable failure modes and systematically evaluates these failure modes under controlled conditions. The approach incorporates expertise from industry and other national laboratories. It is somewhat a concern that the stationary 40,000-hour target does not include purposefully degrading the performance of the stationary system. While this technical target is established by the DOE Hydrogen and Fuel Cells Program (the Program), there was no discussion regarding the conditions at which 40,000 hours would be attained.
- The approach is good. The PIs are using available data from the field to establish the correlation factors for the AST protocols. However, the materials of choice are of old vintage; therefore, the failure rate and correlation factors may not be relevant for newer materials.
- Using the data that are currently available (which are somewhat outdated), the approach of this project will be its legacy. The approach is well organized, and the group has sought the input of key collaborators. The

researchers have adjusted and brought in the right expertise where necessary. The accelerated testing and validation results will serve future work mainly in terms of approach, rather than the actual results.
- The project has focused on ASTs for key components (e.g., the catalyst, catalyst support, GDL, and membrane) and analysis and correlation of test results.

### 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 good progress with respect to refining current ASTs and proposing new ASTs.
- The project team has made excellent progress toward its objectives. Specifically, identifying how many cycles of accelerated testing correlate to hours of operation of various applications is a great step. Updating AST profiles as new data become available, specifically from demonstration vehicles, is also a great step.
- Reasonable progress has been made to understand the failure mechanisms for the various ASTs. Further, the PIs have used industry-recommended protocols and compared these with other protocols. Tests were conducted with drive cycle durability to understand the correlation between field data and ASTs. Overall, a lot of progress has been made.
- The researchers have made good progress on the development of ASTs for the catalyst, membrane, and GDL, but it appears that there are no ASTs for bipolar plates.
- The project team has made strong progress in demonstrating the ASTs.
- The researchers have generated lots of data. It will be interesting to see if they can wrap the whole project up in three months.
- The AST results could be correlated with the U.S. DRIVE cycle. Further acceleration in performance degradation is necessary. Different test procedures have been identified for the different degradation mechanisms. The work went very much into the details of degradation mechanisms. This is necessary, but the focus of AST should be kept. The results are very good.
- The PI presented a fair amount of progress in the last fiscal year. Even though the project is related to validation and testing, the outcome should be a valuable set of plausible solutions that may prevent current state of performance loss. As of now, the PIs are focused mainly on conclusions that are already known.
- The drive cycle testing does not appear to be completely focused. The PI identifies test conditions, but the explanation and discussion for the drive cycle testing is inferior to the other accomplishment discussions. The U.S. drive cycle testing correlates well with the AST for potential cycling; this work needs a better explanation. Carbon corrosion at low potentials is reported to be significant at 0.9 V; it is unclear if the researchers quantified the value. It is also unclear if there is a dependence on the type or manufacturer of the carbon. It is unclear how LBNL confirms the "collapse of cathode structure" and whether the collapse is gradual or occurs at some catastrophic condition. For the voltage loss breakdown analysis, it is unclear what simultaneous fitting of air and HelOx data at different current densities means. It is also unclear whether the work was done or whether this is a statement of future activity.

### Question 3: Collaboration and coordination with other institutions

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

- This project features an excellent selection of team members and data sources.
- The collaboration is outstanding and features a proper division of responsibilities.
- The PIs demonstrated a well-coordinated effort between the participants.
- This project features close collaboration with W.L. Gore, Ballard, and now Ion Power.
- There is good collaboration among the various partners. The segmented cell study to understand the impact of start–stop degradation is useful for the community.
- This project leverages industry partners, demonstration fleet performance information, drive cycles and testing protocols from the U.S. DRIVE Partnership's Fuel Cell Tech Team [see *http://www1.eere.energy.gov/vehiclesandfuels/pdfs/program/fctt_roadmap_june2013.pdf*] and other DOE national laboratories. Coordination with experts in the AST field provides useful data on what parameters the specific end users are looking for and how the experts are currently conducting these tests.

- This project is well coordinated with other DOE-supported projects. There is also good coordination within the project team. It would also be good to include real-world results from other fuel cell original equipment manufacturers (OEMs).
- The collaboration encompasses three national laboratories and four companies, two of which are materials suppliers. All of them are notable players in fuel cells. Moreover, a European university was included—the University of Nancy.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The project fully supports the objectives of the Program.
- This is a very good step in the right direction to validate membrane electrode assembly (MEA) degradation with actual performance data.
- This is an extremely important project that is providing a database that will be valuable for some time. The results generated will significantly advance the progress toward DOE research and development goals and objectives.
- Establishing relevant, validated, and universally accepted AST protocols is critical to meeting durability targets.
- This project is helping to identify AST cycles designed to induce failure of certain components of the fuel cell stack and correlate the number of cycles to a number of hours of normal operation. This correlation will allow rapid testing to obtain data that would normally require thousands of hours. Rapid testing saves time and money for OEMs and users. This project plays a significant role in advancing progress toward meeting DOE durability goals.
- This project has the potential to improve current knowledge about what is causing fluctuations in fuel cell performance over time, and for that reason it is relevant to the Program. It would be important not only to list what is degrading over time but also to propose how degradation can be minimized.
- The project is relevant because it helps to address the barriers listed. The impact of these ASTs on new materials sets may be limited due to correlations to the limited field data. Further, the field data of the older design might be irrelevant in light of improvements in system design to eliminate or reduce some of these failures so that the systems can meet the targets for commercialization.
- In some cases, the ASTs are oversimplified to be of use to fuel cell developers.
- The project targets the major barrier for transportation—durability. The project digs very much into mechanisms of degradation, which is necessary to prevent creating wrong testing procedures in accelerating testing. Nonetheless, an AST project should clearly target an acceleration factor for a whole—even if synthetic—life cycle of a car. The diversity of different acceleration coefficients is physically correct and acceptable. Technically, one acceleration factor can be set for a known (or defined) life cycle of a car by putting the respective mechanism-driven acceptation test procedures together. The impact of the project will be high. Considering the approach, it will be easier to get it into the engineering domain.

### Question 5: Proposed future work

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

- The project is ending in this fiscal year, and the proposed future work is in line with ongoing efforts.
- This team has a plan for future work and builds on the work done in a logical manner. The plan will contribute to further push the durability testing and get closer to meeting the end durability goals. Independent validation of stack-level tests should be considered.
- The plan to update the test protocols (based on the results obtained to date) is recommended. It is unclear if there is a plan for ASTs for bipolar plates.
- Putting the data online sets a great precedent in the Program and will certainly force rigor.
- The proposed future work is appropriate for the funding that remains, but it is not clear how additional work that remains to be done will continue beyond this project.

- The proposed future work is sound; however, it would have been nice to study the failure mode with a newer class of catalysts. The AST related to GDL aging is less important for field failure. The dissemination of information through publications is adequate for failure modes. The models developed under this project should be made available for use by others within the field.
- It seems like the researchers are going to run out of time before they can wrap up their work.
- The focus should be kept on vehicles. The scientific depth in degradation mechanisms is appreciated and fully understood as being necessary as long as the final goal of accelerated testing is also pursued in an engineering way. For certain driving cycles, the different mechanisms of degradation should be quantitatively allocated and combined in a way that the accelerated test represents the particular driving cycle. The existing work is very good, but some engineering perspective should be added to make it really useful.
- The future work is a list of activities to be performed with no discussion of their benefits. Perhaps the researchers assume everyone is so familiar with the technology that no explanation of benefits is needed.

### Project strengths:

- Recommending relevant ASTs for material development is a strength of this project.
- The well-designed testing approaches on key fuel cell components are a strength of this project.
- The technical capability of the team members is extremely high. The results reflect the quality of the team.
- This project features a very good team that is solving real-world problems with MEAs and doing meaningful validation of ASTs versus in-field durability tests.
- This project features a systematic approach in accelerated testing and a strong team that relies on expertise in modeling and fuel cell stack testing.
- Strengths of this project include the very qualified partners and getting ASTs outside of the car manufacturers' domain.
- This project's methods are an area of strength. This project represents the most comprehensive effort for fuel cell ASTs. Its successes and failures will certainly serve to improve the lifetime analyses of those who choose to study the work done.

### Project weaknesses:

- An area of weakness is the impact of system design and material changes on correlation factors.
- Correlation with automotive drive data is recommended to demonstrate the relevancy/validity of the ASTs. The materials are an area of weakness. The materials used in most of the validation work are not state of the art; however, it must be understood that state-of-the-art materials are not generally available with >5,000 hours of service, especially for public disclosure of their performance. The LBNL modeling work seems redundant in light of all of the other projects the group contributes to.
- There is a lack of conclusions about how to prevent or slow down degradation in performance.
- The strongest weakness—the focus on bus data—has been amended. As for future work, there should not be a focus on bus data again.
- The team will not likely have time to finish the project and publish the results.
- The explanations of the accomplishments were in some (many) cases incomplete. The slides were not definitive and did not provide enough information to describe the benefits of some of the achievements. The future work slides did not provide any insight into the benefits of the work. It is recommended that future presentations provide more information on benefits attained and expected benefits of future work.

### Recommendations for additions/deletions to project scope:

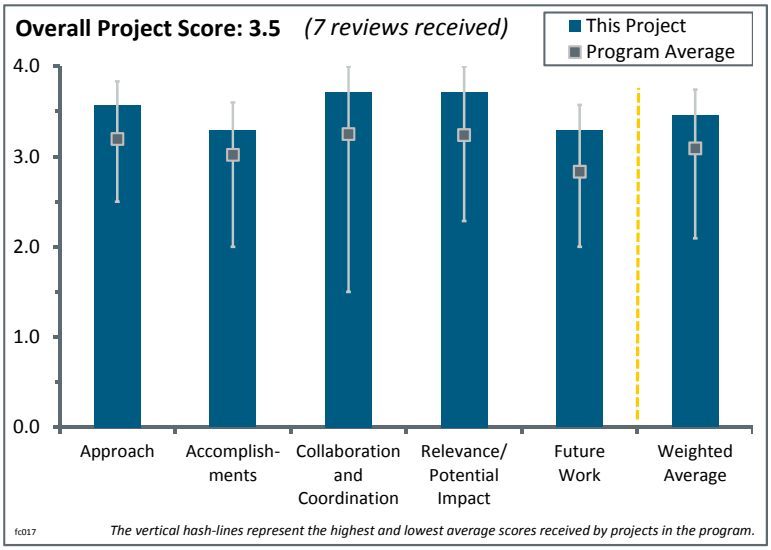
- The project team should study the impact of these ASTs on the state-of-the-art MEAs.
- The team should carefully document not only the successes of the project but also the shortcomings of the ASTs.
- Los Alamos National Laboratory needs some extra time/funding to publish the results.
- The different identified mechanisms and procedures for accelerated testing should be put together to form one test representing the life cycle of a car. Ideally, a routine would be developed that is in a position to identify and quantify the necessary modules to be applied for arbitrary life cycles. This routine should be in a position to emulate a given life cycle and create an appropriate AST program.

## Project # FC-017: Fuel Cells Systems Analysis

Rajesh Ahluwalia; Argonne National Laboratory

### Brief Summary of Project:

This project is focused on developing a validated system model to use for assessing the design-point, part-load, and dynamic performance of automotive and stationary fuel cell systems. The model will also be used to establish metrics for gauging the progress of research and development (R&D) projects and to provide data and specifications for cost estimates on high-volume manufacturing.




### Question 1: Approach to performing the work

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

- This is an excellent contribution to fuel cell technology. The approach is clear and has been well developed over the course of the project.
- This project features excellent continuation of the great work done in this field before.
- The approach to modeling goes down to a level of the science that governs fuel cell performance and behavior.
- The main objective 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. The proposed approach is in agreement with the challenges to overcome, and its annual reorientation does a nice job of reflecting the issues to take into account better.
- These tasks are critically important to the Fuel Cells program. The engineering modeling provides understanding of the many elements that dictate reliability, performance, and cost of all fuel cell hardware. Moreover, these contributions serve to develop a vocabulary that makes it far easier for everyone in the program to communicate effectively. The result is a very clever integration of the research part of the DOE-sponsored work and the commercial part of the program. By using that vocabulary, the entire group of participants has the ability to communicate rapidly and effectively.
- The main technical approach is sound. The modeling is based on experimental validation, as it should be, and several partners contribute with experimental data that are used to validate the model. However, there are two main points for criticism: (1) the fuel cell data is based on 50 $cm^2$ work and should be expanded to stack systems, ideally full-size stack systems, and (2) the work in 2013 concentrated on one particular catalyst type from one particular supplier. While it is important to include nanostructured thin-film (NSTF) catalysts in the work, it would have been better if the work included other electrode layer structures and a comparison of these systems. This would have demonstrated the capability of the model as well as given some insight into a technology comparison of competing catalyst structures.
- The approach taken in this project is very good. The general approach is to develop and validate versatile system design and analysis tools. The collaboration with external organizations for validation of the model is highly appreciated. However, most of the design work is done at high pressure, and considering that cost is a challenge for fuel cell systems, the approach should be focused on low-pressure design and modeling that can reduce the system cost. The project team should look at other fuel cell models developed under other DOE projects while designing the system model under this project. It would be interesting to see system cost at low Pt content for lower volumes, although this may be out of the scope of this 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.

- The sophisticated model is well developed and addresses the details (e.g., mass transfer and Pt-loading issues). The model is validated using real test data and is in very good agreement with the data. The key issue of system cost is also addressed.
- The accomplishments are well documented, but in some cases they represent incremental advancements.
- The project has progressed well this year. Main areas of progress included extending the nanostructured fuel cell stack model operating point, introducing modeled Roots compressors and expanders, validating the humidifier model against data for a full-size unit, and extending and validating the model for $H_2$ ejectors. Unfortunately, only the 3M NSTF membrane electrode assembly (MEA) is investigated, even if its modeling is more detailed.
- The modeling effort and set of results are very good overall; however, the modeling effort aimed at stack-related performance/parameters needs to be verified with an actual stack system. Small-scale single-cell data are not sufficient to verify this model. It is unclear if and how the model is made accessible to the public.
- The progress made so far is significant, but it is not enough to understand the cost savings from different systems. The collaborative work on fuel management (Ford Motor Company), stack model validation (3M), and water management (W.L. Gore, Ford, dPoint) is commendable. The Artificial Neural Network model for cathode mass transport is a good approach, but other models developed under DOE projects related to cathode over-potential analysis need to be considered and cross-validated.
- These activities produce progress in a continuous and rewarding way. Moreover, this progress really covers a rather broad base and is obviously building upon itself—each new calculation uses results of previous ones. Questions are raised, and answers are quickly calculated. Moreover, results are published and made available to many in the community, while protecting the interests of those working to develop fuel cell markets.
- The barriers claimed to be addressed by the model include (a) Cost; (b) Performance; (c) System Thermal and Water Management; (d) Air Management; and (e) Start-Up and Shutdown Time, and Energy/Transient Operation. There is clear evidence that the project addresses barriers (b), (c), and (d). The linkage between this project and barriers (a) and (e) is more tenuous. Although cost results are in the backup slides, it would be good to see some of the major outcomes on cost produced by the model, for instance, a sensitivity study looking at several of the major drivers of cost, Pt loading, current density, and power.

### Question 3: Collaboration and coordination with other institutions

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

- The collaboration is extensive and appears to be very interactive.
- The collaboration is well balanced, and there is an important emphasis on industrial partners.
- The collaboration with external organizations is excellent, specifically with 3M, W.L. Gore, and Ford. Also, the collaboration with the component supplier for the compressor and expander (Root and Eaton) is very good. This project has strong and sufficient partners to accomplish what has been proposed.
- This project features a very good team of collaborating partners. The portfolio consists of a number of research laboratories; companies that create fuel cell system materials and/or balance-of-plant (BOP) components; and national and international working groups such as the tech team, the modeling working group, and the $H_2$ fuels purity-related working group. It is an impressive set of partners.
- The entire thrust of this work is collaboration. The information flow integrates R&D and provides DOE with excellent, timely information, making the management job less difficult. Perhaps this project is the best example of collaboration one could think of. It can be viewed sort of as the model—the one taught to explain what collaboration could be. It is also apparent that these timely results facilitate analysis of proposals for new activities in ways that promote intelligent program management.
- The coordination appears correct, and the quality of the results reflects it. New industrial collaboration is appreciated and enables links between different projects, such as the project with Eaton. Collaboration

between Argonne National Laboratory and the partners appears to be very good, but there is no indication that collaboration between the other partners of the project exists.
- The collaboration looks good; however, it would be good to see system-level validation of the model compared to an actual original equipment manufacturer (OEM) powertrain. That would demonstrate that the model is valid and could be applied to other system topologies. It would be good to have OEMs with more active fuel cell programs than Ford on the list of collaborators.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- This is an excellent contribution to fuel cell technology.
- This project provides valuable modeling tools. It is of high value to industry partners.
- The relevance and impact of this project are very high because it will help OEMs that are designing relevant fuel cell systems according to DOE technical and economical targets.
- This project's relevance to the DOE Hydrogen and Fuel Cells Program is perfect. It is one of the most important studies to address system-level performance and cost barriers. The cost projections are correlated to Strategic Analysis, Inc. (SA) models, which makes more sense. The focus can be increased by featuring more actual stack-relevant system design and modeling than a 50 $cm^2$ cell.
- A comprehensive system model that can guide decision making for DOE as well as industry is a highly desirable tool. However, developing one is a very large challenge because system characteristics are OEM-dependent and the developed tool has to be very versatile to be applicable over a large range of system architectures.
- The extent to which the model is available to, and customizable by, automotive OEMs and other stakeholders is unclear. Making the model open will aid its relevance and usefulness. The model provides a very detailed look into system details as well as physics controlling performance and efficiency. Nothing like it is available in the public domain.
- The project is driven by DOE-derived targets that are somewhat achievable goals. The complicated and thorough analysis done in this project is all about addressing and meeting those targets—the how, when, and if.

### Question 5: Proposed future work

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

- The project team should keep it up. The project is worthwhile and should be funded even further.
- The proposed future work is a natural continuation with convincing additions.
- The future work is to continue these ongoing calculations. There is no question that should happen. The rewards are apparent. It appears that this is money well spent.
- The future work item of most interest is "System analysis with durability considerations on drive cycles." It would be interesting to see this in light of different degrees of hybridization with batteries/µCaps because most fuel cell electric vehicles will be deployed as hybrids.
- The proposed future work is good. The introduction (or at least the presentation, if it is already implemented) of the system control may complete the modeling. The actual durability of a stack will be highly influenced by the control strategy for the BOP components.
- The overall future work to support DOE development efforts at the system and component levels is very good. However, MEAs other than NSTF or de-alloyed NSTF should be considered. It seems that only NSTF was considered because 3M was the only stack validation partner. Other stack validation partners need to be approached, and other MEAs with Pt or a Pt alloy catalyst can increase the value of this project. So far, researchers have not demonstrated enough focus on significant cost reduction, other than the idea of reducing Pt loading in the catalyst. Suggestions for significant cost reductions in other systems would be a great add-on.
- The future work needs to include model verification with one or more stack systems.

### Project strengths:

- This project is very comprehensive and thorough.
- This project is extremely well documented, and the detailed basic physics add up to a robust model.
- This project features a good modeling backbone, as well as good exchange and collaboration with partners.
- This project features a solid approach, very strong partners, and strong work with component suppliers. The team has made excellent progress.
- Certainly it would take a special person to do this, and this principal investigator is unique. Others might grow into such a role, but few could. This principal investigator is a key project strength.
- The project is running well and takes into account major parameters for stack and BOP components. Good links have been established with component suppliers and OEMs. The results are well presented. The collaboration with SA is important because it enables to the team to establish a reciprocal critical review of the outcomes of pure technical and pure economical approaches. The maximum operating pressure proposed (3 atm) is a representative example.

### Project weaknesses:

- No significant cost reduction at the system level has been proposed.
- Even if the presented goal is for either automotive or stationary systems, the presented results only concerned transportation. It is unclear if there are any results for stationary applications.
- These are not weaknesses of the project but of the presentation: (1) the slides contained too many abbreviations known only by insiders, and (2) the slides are far too busy—fewer but clearer ones would be desirable.
- It is hard to criticize what is going on in this project. Even so, there should be attention paid to fuel cell systems of alternative design. The MEA and essential cell fabrication have only been tweaked during the last decade. It would be good to look “outside the box” and see what organizations outside of the DOE family have been up to.
- There was no validation with stacks or real fuel cell systems. All stack performance data points are extrapolated from 50 $cm^2$ cells, which renders the results questionable in their usefulness.
- The project claims to address the barrier of Start-Up and Shutdown Time, Energy/Transient Operation, yet no dynamic model results or runs have been shown. The project lacks collaboration with an OEM on a system-level validation of the model.

### Recommendations for additions/deletions to project scope:

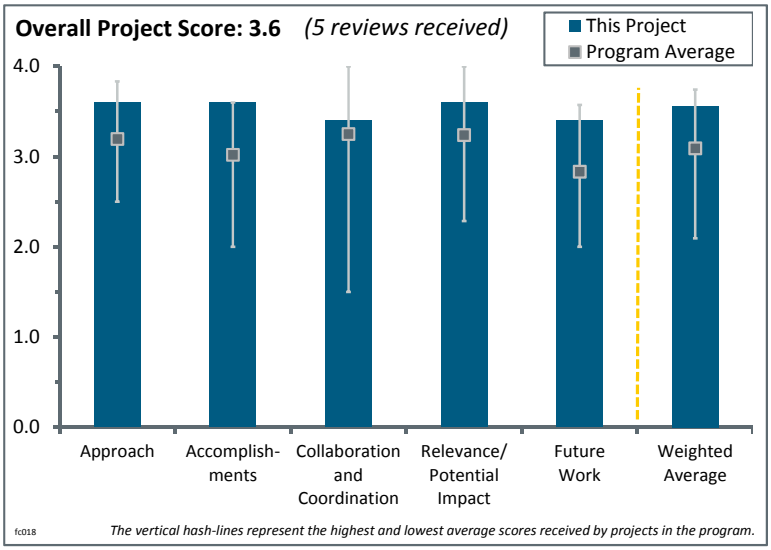
- It seems like all of the bases are covered.
- The project should be extended.
- The project team should pursue validation with full-size stacks, short stacks, and/or systems.
- The researchers should focus on incorporating durability considerations in system analysis.
- This project should produce model dynamic results relative to data from an integrated system. This will likely require tighter collaboration with an automotive OEM to allow comparison of its system to a version of the model tailored to its system.

## Project # FC-018: Fuel Cell Transportation Cost Analysis

Brian James; Strategic Analysis, Inc.

### Brief Summary of Project:

Cost analysis is used to assess the practicality of proposed power systems, determine key cost drivers, and provide insight for the direction of research and development priorities. This project is a continuation of previous years' cost analyses of automotive and business fuel cell systems, and it is exploring subsystems and alternative configurations of those systems. The 2012 analysis looked at a plate frame humidifier, looked at low-cost roll-to-roll membrane electrode assembly (MEA) manufacturing, and improved quality control techniques.




### Question 1: Approach to performing the work

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

- This is a well-established and great process for system evaluation.
- This project features appropriate cost estimation methodologies and well-defined assumptions.
- Independent cost analysis is very important for the U.S. Department of Energy (DOE) to gauge the state of development of fuel cell technology, both within the DOE Hydrogen and Fuel Cells Program (the Program) as well as outside the Program. Knowing the state of development enables DOE to set research targets that reflect a feasible stretch from the current technology. The automotive application has the most aggressive cost target and has received the most attention. However, other applications need similar attention if the Program is to be a success. Strategic Analysis, Inc. (SA) uses well-known cost estimating techniques that have wide industry acceptance. SA accounts for many smaller features and details in its system designs. The results have been vetted by original equipment manufacturers (OEMs). The approach is very good.
- This project features a detailed cost analysis that is identifying key contributors to cost and determining realistic process-based costs. The project also considers realistic production rates for buses. The production rates for MEAs and humidifiers are an order of magnitude higher than the production rates for buses; it is unclear if this is an apples-to-apples comparison. MEAs made by the new process are assumed to have the same performance as ones made by the old process. It is unclear if laboratory data are available to make this assumption.
- The project addresses the barriers of cost and thermal, air, and water management. The bus production rate for cost analysis of buses is set to 1,000 units per annum, which makes much sense for evaluating the market introduction phase. The catalyst loading was increased to a reasonably higher level for the sake of the longevity requirements for buses over vehicles. The procedure and the data used for the cost analysis are apt and up to date (compared to sensitivity analysis and Monte Carlo analysis). The W.L. Gore manufacturing process for vehicles applies very thick catalyst layers, which might not make a big difference in cost. The technology approach does not sound coherent because at the same time very high production speeds of >10 m per minute are assumed, which would result in very long dryers at that high speed. Other than that, the assumptions are sound, and the evaluation is at a cutting-edge level.

### 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 progress to date is very good.
- The project team has done an excellent job of updating work.
- The presenter showed system optimization (slide 28), with selections for catalyst loading, air stoichiometry, and pressure. It is unclear if the model is sufficiently thorough to make conclusions. For example, electrochemistry is affected by air stoichiometry, including water management (high air flows improve voltage but also help remove water droplets in the stacks). The pressure of operation improves current density but induces auxiliary power. Catalyst loading balances performance and more importantly life/robustness. The relationships here can be very interactive. It would be good to know if the researchers' conclusions are in line with OEM views.
- The bus system cost analysis uses the same estimating methodology as the automotive application, which has resulted in cost estimates that align with industry projections. The same level of attention to detail is evident in the bus analysis. The balance of plant (BOP) in these systems remains a significant contributor to the cost estimate and could benefit from additional DOE research attention. The 2013 automotive cost analysis update looked at changes in operating conditions and in humidifier design that did not change the estimate substantially. The automotive analysis is based on the 3M NSTF catalyst system. It would be helpful to prepare the equivalent analysis using a dispersed Pt catalyst, as is the current practice. If the continued analysis of system modifications results in the cost estimate remaining relatively unchanged, then DOE needs to refocus its research efforts in areas likely to lead to more substantial cost reductions to meet the ultimate targets. It is good to see the attention paid to the quality control aspects of the manufacturing process.
- The 2013 data basically confirm the earlier results but on a much more detailed level, and new components were considered. The data seem to be pretty robust and reliable. The cost structure is well understood. The approach to investigate lower production volumes that are relevant for the market introduction is very useful. The investigation of the W.L. Gore process combined a relatively high production speed with very thick catalyst layers. This is not only a materials consumption issue but also creates a problem regarding drying these thick layers at that speed. This process might be looked into closer. This strongly contributes to the DOE goal of cost reduction.

### Question 3: Collaboration and coordination with other institutions

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

- Major national laboratories and major industry players are involved in crucial parts of the analysis.
- There is good collaboration with project partners.
- Formal collaboration occurs between SA, Argonne National Laboratory, and the National Renewable Energy Laboratory. Industry review of assumptions and cost projects is important to the project as a means of ensuring that the results are consistent with industry expectations. Without this interaction, the project would not be successful. SA has reached out to OEMs and the component suppliers to verify assumptions and methodology. Additional interaction with suppliers of BOP components would expand the opportunity for comparison of alternative technology.
- The input and feedback from collaborators are unclear.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The project fully supports the objectives of the Program.
- The impact will be high because the project delivers unbiased data and makes these data publicly available, which is important for economic considerations and even decisions of the supply industry.

- The project is relevant to the Program. It provides an independent assessment of the manufacturing cost of fuel cells. In addition, the method used has been vetted by numerous OEMs and reviewers. While the results are not necessarily spot on, they give DOE a consistent picture of the trends in the manufacturing process from year to year.
- This is an independent validation of costs for fuel cell buses. The project does not present a cost market curve.

### Question 5: Proposed future work

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

- The proposed future work, especially the proposed continued vetting of the assumptions, is very appropriate for this project.
- Continued comparison of the bus system with the automotive system should provide additional insight into the differences between the two systems and an opportunity to take the "best components" and practices from one system and apply them to the other system to explore additional means of cost reduction.
- The future work proposed looks a little weak, considering that just 50% of the project has passed. The proposed work makes sense, though, and needs no changes in terms of the points proposed. It is suggested the project include different production quantities for vehicles as well (e.g., 1,000, 10,000 and 100,000 p.a.) in order to reflect the ramp-up, as was done for buses.
- Determining the sensitivity to annual production volume would be good.

### Project strengths:

- The project features excellent cost estimation and analysis approaches based on well-defined assumptions.
- The principal investigator's team is extremely effective with this analysis. The team should keep doing exactly what they are doing—nothing should be changed.
- Strengths of this project include the creation of publicly available and unbiased cost data, and strong cooperation between a consulting company and national laboratories.
- The project features a good approach in calculating costs for realistic production rates at this time. In addition, the project team has considered ranges of values with upper and lower bounds.
- The collaboration with industry to vet the assumptions and methodology of the analysis is excellent and crucial to the determination of viable cost projections. The analysis is clear and methodical, which provides high confidence in the validity of the cost projects. In addition, the accumulated database also inspires confidence in the results.

### Project weaknesses:

- It is hard to cover all aspects of the bus and automotive analyses in such a short presentation.
- There is a lack of details on the input and feedback of the collaborators and fuel cell system and component developers.
- The individual components are costed at significantly higher production rates than the total system production rate. It is not clear if the new developments mentioned in the study are available for implementation.
- There are no notable weaknesses.

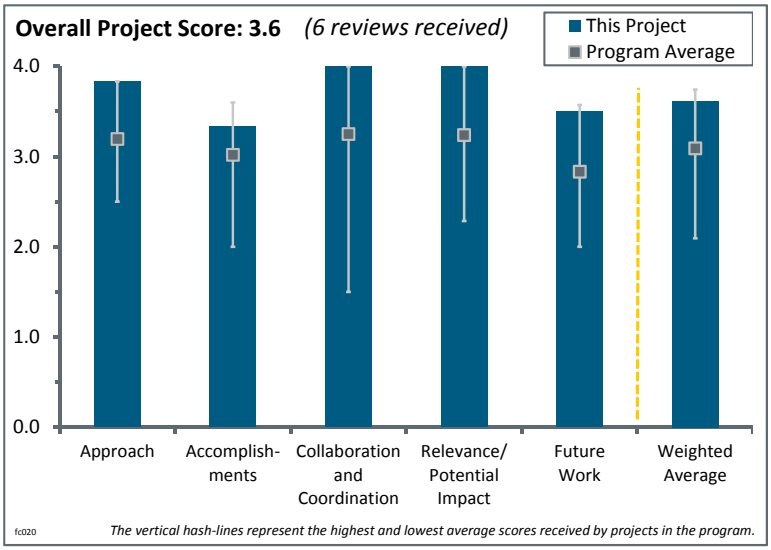
### Recommendations for additions/deletions to project scope:

- The U.S. DRIVE Partnership is continually evaluating research targets in light of the most recent advances in the state of the technology. SA should strive to incorporate the latest information from the U.S. DRIVE Fuel Cell Tech Team into the analysis.
- It is suggested the project include different production quantities for vehicles as well (e.g., 1,000, 10,000 and 100,000 per year) in order to reflect the ramp-up, as was done for buses.
- The project team should consider adding a detailed cost study on high-impact BOP components—the sensors and air loop.

## 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 micrometer-to-angstrom-scale characterization of material constituents composing fuel cells (catalyst, support, membrane); (2) understand fundamental relationships between the material constituents within fuel cell membrane electrode assemblies (MEAs) and correlate the relationship data with stability and performance; (3) integrate microstructural characterization within other U.S. Department of Energy (DOE) projects; and (4) apply advanced analytical and imaging techniques for the evaluation of microstructural and microchemical changes to elucidate microstructure-related degradation mechanisms contributing to fuel cell performance loss.

### Question 1: Approach to performing the work

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

- The project's focus on developing and perfecting the necessary tool and analytical competence needed for evaluating fuel cell materials, coupled with serving an extended group of collaborators, has enabled significant contributions to the DOE Hydrogen and Fuel Cells Program (the Program).
- This is a characterization project that supports the development of others to help understand the development of materials and devices. The principal investigator (PI) showed this is being done and more. The use of microscopy tools by the teams seems to have revealed an important failure mechanism; namely, that compression is a first step leading to local Pt-enriched sites promoting carbon corrosion.
- The approach adopted by this project—to be a center of excellence for nanoscale microscopy—is very valuable to the community. Based on the number of collaborations established, it is clear that the project is well integrated with other efforts.
- The imaging techniques applied are outstanding on a world level. The very inclusive approach makes the basic research contribute in an ideal way to technical development (e.g., the great number of partners, integration of microstructural characterization within other DOE projects, making capabilities available to researchers outside of Oak Ridge National Laboratory [ORNL]). The targeted components such as catalyst nanoparticles, polymers, catalyst support materials, MEAs, etc. are the most important ones for longevity and cost reduction. The research contributes to the most important targets: durability and performance.
- This project applies state-of-the-art electron microscopy to problems of interest in fuel cell design and durability studies. Access to these appropriate tools is only one aspect of the project. Another is that the project develops and maintains a core of experience in issues specific to fuel cells, providing a high level of added value. Having a common electron microscopy resource for many research projects capitalizes on the advantages that specialization offers, "leveraging" the knowledge gained in one research project for the benefit of many. In addition, the collaborative approach demonstrated by this project—for example, using problems suggested by the U.S. DRIVE Fuel Cell Tech Team to guide future work—contributes greatly to its value.

- This project features excellent, highly relevant work. This work provides significant contributions to understanding degradation mechanisms through detailed microstructural characterization. This project provides a focus on the characterization, which is critical to achieving an understanding of degradation mechanisms, which in turn is critical to improving the durability of fuel cell systems. However, the work is focused on the characterization techniques, as it should be, and depends heavily on results from others. This appears to result in some potential disconnects between the interpretation of the structural changes as a function of the degradation mechanism, and hence some potential errors in the hypothesis about mechanisms. It may be better to either limit observations to changes in structure without hypotheses on mechanisms (preferred), or work more closely on the mechanisms. In the former instance, the hypotheses should be made under the durability projects where the mechanisms are being studied. Some examples of potential issues include the following:
    - Validation and comparison to realistic operational conditions is very important; for example, corrosion under $H_2/N_2$ instead of $H_2$/air may affect the structures formed.
    - The amount of carbon corrosion ($CO_2$ production) was not shown, nor was it determined how this correlates with thinning, but conclusions were still drawn regarding this correlation.
    - It was not determined how the volume of carbon converted to $CO_2$ could result in a change in compaction and porosity of the catalyst layer; for example, an estimate based on a packing model may provide a rough comparison.

### 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.

- This project continues to provide the necessary characterizations to better understand failure modes and evaluate new approaches for improved durability and performance.
- Carbon thinning was perfectly proved by imaging. Carbon oxidation and graphite oxide formation were proved and quantified. This is great progress.
- The work to date is meaningful. It would deserve a rating of "outstanding," but there are some delays, although the PI expects things to be back on track in about a month.
- The PI explained that at each Program Annual Merit Review, the project team focuses on a different area of its research and accomplishment (slide 7). This year, the presentation focused on quantifying cathode degradation, especially carbon corrosion. Slides 9–16, together with the PI's oral explanation and the question and answer period at the end of the presentation, demonstrated a significant body of accomplishment in this area. The lack of presentation time precluded a similar discussion on accomplishments concerning measurements of ionomer films (slides 17–21), but there is little doubt that this project is highly productive nonetheless.
- The work provides significant contributions to understanding degradation mechanisms, which are critical to achieving DOE goals. There has been excellent progress on understanding changes in microstructure due to the carbon corrosion mechanism, but it has not been quantitatively linked to the corrosion/$CO_2$ production mechanism.
- The work conducted is very challenging. The ORNL team has made good progress. The results presented for the carbon corrosion effort were very interesting and support the broader fuel cell community. Hopefully in the remaining portions of this work, mechanisms that explain the loss of porosity in the cathode layer will be identified, along with a deeper understanding of the mechanisms by which localized graphite oxide regions are formed, in close proximity to coarsened Pt particles/clusters. The ionomer film studies are unique and could be valuable in determining the differences between traditional supported Pt catalysts and the catalyst-coated nanostructured thin film (NSTF), which apparently behave differently when using 3M versus Nafion ionomers. Hopefully in the next year more studies will be presented detailing the nanoscale microstructural degradation in the $Ni_7Pt_3$-NSTF nanoscale materials, which appear to have been adopted by the fuel cell community even though there is substantial loss of Ni during operation.

### Question 3: Collaboration and coordination with other institutions

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

- A very large number of collaborators are using the facilities for a multitude of characterizations.
- This project's success depends on collaboration in order to have access to material for analysis. The annual report of outstanding accomplishments highlights the synergy developed between ORNL and its partners.
- By virtue of the project team's strongly adhering to the project charter (approach), this is an area where the team has performed extremely well (i.e., by providing critical support to a number of funded efforts).
- This project is inherently collaborative because it provides an expert technical resource to non-ORNL collaborators. The presentation demonstrated an impressive list of university, industry, and government partners (e.g., slides 2 and 4).
- This project features extensive collaboration with a large number of partners. However, the work shown was focused on the efforts at ORNL, so it is difficult to tell the level of collaboration that actually occurs.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

This project was rated **4.0** for its relevance/potential impact.

- The work targets the most important issue in longevity.
- The direct observation of preservation or failure of components is critical, and this team is serving that need.
- The project's investigation and analysis of the effect of carbon structure on carbon corrosion will guide improvements in catalyst development.
- The work is highly relevant, and the results achieved are advancing the supporting knowledge base to understand degradation mechanisms. Ionomer imaging is very important and may lead to the ability to design better catalyst layers to both improve performance and mitigate degradation mechanisms.
- This is a valuable, core project in the Hydrogen and Fuel Cells Program. It provides broad access to electron microscopy and relevant expertise for projects studying fuel cell materials (e.g., the MEA material constituents). Electron microscopy is relevant because it measures and diagnoses the microstructure and nanostructure of the functional materials within a fuel cell. Understanding this microstructure and nanostructure is key to understanding the performance and durability of a fuel cell.
- Given that 34% or more of the fuel cell costs are tied up in the catalysts and membranes, it is crucial that fundamental work such as this is performed. As the community moves to low-platinum-group-metal catalysts in order to meet performance requirements, this work will be key to the discovery and application of these new materials.

### Question 5: Proposed future work

This project was rated **3.5** for its proposed future work.

- The future work proposed is in line with the barriers identified.
- The future work on ionomer imaging is very important. The correlation of the observations with real-world results is critical. There is some concern about the accelerated stress test used and the relevance to real fuel cell operation.
- The electrode structure (catalyst + ionomer) can be significantly different depending on formation (coating, spraying, etc.), and it may be necessary to examine the "as made," "pre-conditioned," and "used" MEAs to ascertain the structure as it exists during actual fuel cell operation.
- The project plans its future work in a logical manner by consulting with the Tech Team and other constituents (e.g., see slide 8.) The future plans described on slide 24 seem reasonable and appropriate, including the studies of ionomer films and the continued development of in situ electrochemical "liquid cell" transmission electron microscopy/scanning transmission electron microscopy (TEM/STEM).
- Besides continuing the very successful work, the researchers plan to correlate the knowledge to experimental data from fuel cell testing. This takes the work to a next, higher stage. As for the observed

compression of the catalyst layers, it is recommended the project put in effort regarding understanding the mechanism of densification in detail. This might open up new options of carbon modification other than the already-identified Pt on low surface area carbon (LSAC).

- The plans for use of the microscopy facilities are well thought out. The researchers should use some simple "sanity checks" to ensure this work is on track. Instead of relying on microscopy to find everything (such as compressive loss of porosity), the team should conduct simple weighing of samples before and after use to see if mass change and thickness change corresponds to densification expected for pore loss. Also, utilizing gas flow through electrodes on gas diffusion layers (GDLs) at constant pressure before and after applying pressure should help determine whether compression is the cause of electrode thinning as opposed to corrosion being the cause of electrode thinning.

### Project strengths:

- This project's strengths are clearly attributed to the ORNL team's continuous efforts to improve its tools and analysis capabilities.
- This project features a skilled team as well as strong and numerous collaborators.
- This project features outstanding experimental proficiency in imaging and an openness to sharply focus on the requirements of fuel cell development. The inclusive approach to the fuel cell community is part of this.
- This project provides core technical competence and develops relevant experience in the field of electron microscopy for a broad range of projects within the Program.
- The team provides crucial characterization capability to the fuel cell community and DOE at large. Without fundamental work such as this, it is unlikely that the barriers facing the fuel cell community will be overcome. Therefore, it is critical that the Program continues to support this work.
- This project provides very detailed work with correlations to structure, for example, the carbon/graphitic structure. The project features an excellent number of collaborators and attempts to draw on the extensive research and industrial community. It also provides significant advances to microstructural characterization capability and features a very accomplished lead researcher. The new focus on ionomer imaging is very important and relevant.

### Project weaknesses:

- The project could use additional staffing in order to make timely progress on the proposed activities for 2013 and beyond. There is a combined need to meet the demand for routine "expert analyses" as well as develop new tools and protocols for ionomer and electrochemical "liquid cell" projects.
- This project features rigid thinking. The researchers need to think "out of the box" a little more. Granted this is hard when one has to focus on handling precision tools, but taking a deep breath and rethinking what one is doing once in a while is a good thing.
- There is a very large number of collaborators, but it is not clear how much is actually done with each of them. The attempts to understand mechanisms based on studying microstructure may be based on inadequate links to the degradation conditions and quantitative analysis of the processes occurring (e.g., carbon corrosion). The corrosion study shown used $H_2/N_2$, which is not a realistic condition, and observations were made without taking that into account.
- There is nothing that could be identified as a "weakness."
- There are no significant weaknesses.
- There are no obvious weaknesses.

### Recommendations for additions/deletions to project scope:

- The project team should include research for understanding in detail the compression effect of the electrodes during operation.
- In addition to the ionomer study, the researchers might include the "interfacial aspects" between the electrode (catalyst + ionomer) and the membrane.
- The researchers should weigh the samples. They should also check the components of the MEA separately (check gas flow through the catalyst layer on the GDL before and after compressing, and check the image by microscopy before and after compressing).

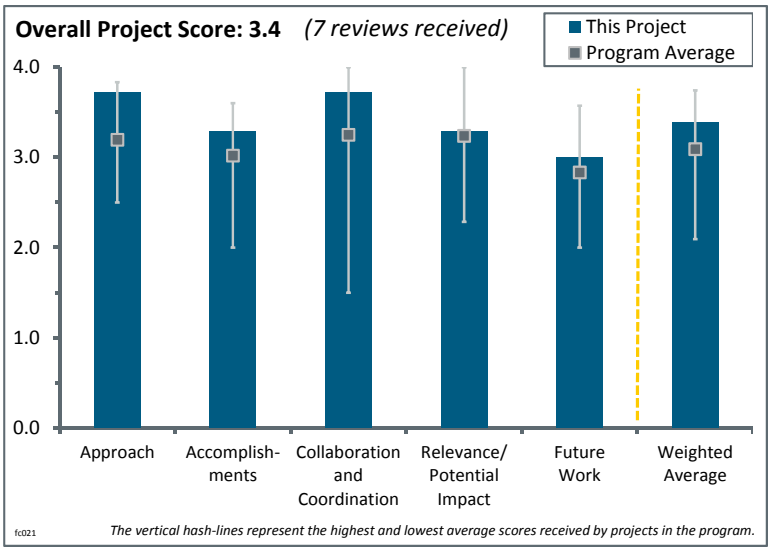
- The researchers should prioritize work to a few areas. They should also increase quantification of correlating data (e.g., correlation of $CO_2$ produced against changes in carbon volumes). It is not clear why thickness changes. It is also unclear how the new carbon volumes, when taking into account porosity changes, compare against $CO_2$ measurements. The researchers should do more comparisons of results to realistic operational conditions. It is unclear why bands are seen in the catalyst layer. They do not seem to be related to the initial high-density Pt regions, which were much more localized. The bands in the catalyst layer may be due to the use of $N_2$, which does not allow the Pt to leave the catalyst layer, which may occur in $H_2$/air conditions. Providing linkages to measures of surface oxidation for the corrosion studies would be worthwhile.
- The scope is fine as is.

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

Muhammad Arif; National Institute of Standards and Technology

### Brief Summary of Project:

Neutron imaging allows for the study of a wide range of fuel-cell-related issues, such as water transport in the flow fields and manifolds, steady-state liquid water distribution anode versus cathode in diffusion media, steady-state liquid water distribution in the membrane, and catalyst degradation induced by liquid water. Objectives of this project include studying water transport in single cells and stacks, enabling the fuel cell community to use state-of-the-art neutron imaging capabilities to study water-transport-related issues, tailoring neutron imaging to the needs of the fuel cell community, and improving spatial resolution to study relevant-length scales such as catalyst layers.




### Question 1: Approach to performing the work

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

- This is a high-tech project to diagnose in situ the water transport in a working fuel cell. This high-resolution work provides a nice tool for fuel cell industries and will also give very good support for future material development. It is excellent work.
- The approach to making microscopic measurements of water transport is very good. No other imaging technique can provide real-time imaging during operation of a fuel cell.
- The project team features a great amount of expertise in neutron imaging of water locations inside of an operating fuel cell.
- The proposed approach is right—to improve the sensitivity of the measurements and the imaging to enhance the resolution and therefore ease identification of the roles in various components in water management. The team is also providing access to the fuel cell community by sharing the results. This approach is of huge importance to contribute to overcoming the performance and durability barriers on the basis of scientific criteria and not only financial criteria.
- The approach is indirectly focused on overcoming U.S. Department of Energy (DOE) barriers on durability and performance by improving water management in the fuel cells.
- The project is focused solely on enabling the visualization of liquid water in a running stack/cell. Recent work in polymer electrolyte membrane fuel cells (PEMFCs) has begun linking the running water content to the acceleration of degradation mechanisms, in addition to the usual linkages made to overall cell performance. As such, the ability to visualize total water content under operation is a highly valuable tool in correlating observed performance and durability effects to cell design. The project focus on increasing the resolution of the test is critical to ensuring that the method has the capability to resolve both water content of the membrane and catalyst layers, which are two key components for both performance and durability.
- The current resolution of the neutron imaging is approximately 10 µm, which may allow determination of the water distribution in the flow field and around the manifold. It is crucial to know if the water content changes at different current densities and if the flow field and manifold design can manage the water. Regarding the water across the membrane electrode assembly (MEA) or along the plane of the MEA, the

water state (localized liquid water or water vapor) and whether the anode is drying out at high current density must be known. It is doubtful that the neutron imaging can determine these issues or help improve the resolution to below 10 µm.

### 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.

- This is very good work. The data sets are good, and it is good to see a real working fuel cell with water transport.
- Significant progress has been made toward improving resolution for water imaging in fuel cells and evaluating the accuracy of water distribution measurements.
- Significant progress has been made on improving the neutron imaging techniques, and completion of the large-area high-resolution detector is very useful for large cell start-up/shutdown studies. Further, the analysis of the error and accuracy is extremely important in interpreting the results and discerning which changes are significant between samples.
- The use of the Gd/Si grating technology, plus the National Aeronautics and Space Administration (NASA) optics (from Chandra), clearly show that resolution on the µm scale is achievable, at least from the optics end.
- The presented results are of high quality and in accordance with the proposed milestones. Achieving the goal of a 10 µm resolution enables users to image the water distribution in the MEA and the channels of bipolar plates. But working to enhance this resolution to 1 µm will enable studies of flooding and liquid-water-related degradation in the catalyst layers of a fuel cell, which is a key point. The quality of the work relies on the use of either the theoretical or practical competences of different partners such as the National Institute of Standards and Technology (NIST), NASA, and the Massachusetts Institute of Technology. Potential artifacts have been investigated so that they could be taken into account for the fuel cell imaging.
- It would be nice if there was more progress on getting down to the 1 µm resolution level. The accuracy of the water measurement analysis was excellent.
- NIST has made efforts to have the neutron imaging applicable to the fuel cell applications. The team should think about what valuable information can be gained by utilizing the technique and what the strengths and weaknesses of the technique are, as well how to provide such valuable results to assist researchers in improving the cell design and performance in a timely manner. This improvement also could give people confidence in the neutron imaging applications in the fuel cell field. Overall, it is a matter of priority determination.

### Question 3: Collaboration and coordination with other institutions

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

- There is close collaboration with NASA and users of the NIST facilities.
- The collaboration is good, and clearly the project supports a wide range of customers.
- The researchers are doing a lot of work with a variety of operational fuel cell systems.
- The achieved results have been possible because of close collaboration between partners. Moreover, the collaboration between NIST and many other partners has been highlighted and appreciated during several presentations at the DOE Hydrogen and Fuel Cells Program Annual Merit Review.
- This project features very good cooperation. It would be better if the principal investigator could explain more about which partner did what job.
- The use of the facility is largely user-driven—in particular, via either open research or closed, paid projects. The process by which projects are chosen and hours allotted could be made clearer, as well as the criteria by which projects in the open research area are selected. Further, pending projects in the open research area should be listed in the presentation to better appreciate the types of work for which the facility is being leveraged.
- A lot of research groups have been involved, although not very deeply.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The project is very important for the Program because it addresses water management issues in operating fuel cells.
- This is the project every fuel cell developer needs. It is a significant work for fuel cell development.
- The location of water inside operating PEMFCs is highly relevant to optimizing the performance of the fuel cells.
- Neutron imaging is a powerful method to help researchers understand a fuel cell or stack under operation by imaging a critical parameter for PEMFCs: the water. Therefore, enabling the visualization of this material has an important impact on the potential ways to improve the performance and durability of PEMFCs.
- Neutron imaging has the potential to make a significant impact in advancing knowledge of water transport at micron-length scales. However, to achieve less than 1 µm resolution, improvements in detector resolution and imaging resolution are required. The team has identified a viable path forward for meeting these goals via collaboration with the Chandra team at NASA.
- Linking the imaging of water to the project objectives is largely left to the reviewer. The project presentation should generally recheck the project impacts against the use of the facility in achieving DOE RD&D goals. This rechecking, in part, could be done via the list of open research projects the technique is used for and some high-level accomplishments and findings. In particular, this rechecking would also help cast the need for increased resolution.
- It helps to publish some papers.

### Question 5: Proposed future work

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

- The future work seems quite reasonable.
- The future work is a continuation of the proposed approach. The project team is encouraged to address the membrane swelling issues depending on the water content.
- The proposed work is focused on improving the resolution of water imaging in order to understand the regularities of water management in the catalyst layers.
- The future plan is correct, in particular the target to improve the resolution to the 1 µm range. This improved resolution will be very useful to better investigate the MEA components individually. The lens optic work has to be continued to improve the resolution while keeping the record time in the minute range.
- The future plan is very good. If the project can detect which layer the water transport goes through, it would be even better. The project so far has obtained two-dimensional images. It would be good to know if there is any way to check three-dimensional pictures, and if there is any connection between the fuel/oxidant consumption versus water transport.
- The future work well identifies the plan to improve resolution using gratings and x-ray focusing techniques. No mention was made of efforts to increase resolution of a 9 x 9 cm MCP detector. This needs to be a focus area for even greater resolution required by the community.
- The development of the grating-based imaging is showing promise; however, it appears there are still significant technical challenges associated with the method that need to be overcome. In particular, the collection time necessary for the grating-based imaging is substantially long, which may in part limit the value of the technique in studying anything other than steady-state water content. The alternative path of using the neutron collecting lens seems to a degree like the optimal path; however, the timing and barriers related to this development are not clear, and largely this amounts to leveraging a technology that “may” reach fruition depending on the success of another organization. Clearly there is significant risk in the proposed path forward.

### Project strengths:

- Neutron techniques are the best way to investigate water issues inside operating fuel cells.
- Applying neutron imaging to water content detection is a strength.
- NIST is bringing a powerful characterization tool to the fuel cell community. As resolution metrics are met, the results will enable a much-improved understanding of water transport.
- NIST features cutting-edge instrumentation for neutron imaging, strong collaboration with NASA, and a combination of modeling and experimental approaches.
- The project provides "eyes" to see the fuel cell water transport in a working mode. Thus, it provides useful information about fuel cell flow channel and manifold design as well as catalyst distributions.
- The main strengths of the project include its (1) effective collaboration between high-quality partners and bringing competences from different horizons (equipment, fuel cell) for a common target; (2) work to improve the measurement quality while increasing the spatial resolution for imaging the water in a fuel cell; and (3) free access for open research, enabling the contribution of any partner on based on scientific criteria.
- The industry use of the facility is strong, and the technique provides much-needed insight into the location and changes in liquid water during operation. The project is clearly focused on the needs of the user community in driving the technique to appropriate resolution for studying water content in membranes and catalyst layers. The project is also clearly leveraging advancements and techniques that are being developed in other organizations/applications, which demonstrates a high degree of collaboration.

### Project weaknesses:

- It is difficult to get the one-micron resolution necessary to address catalyst issues.
- The membrane/catalyst layer and catalyst layer/gas diffusion layer interfaces are not distinguishable.
- It is unclear if the technology can be used for a stack diagnosis.
- There is considerable risk in the proposed path forward for the high-resolution detector. An analysis of the technique in comparison to other approaches (x-ray or nano-CT) should be included to cast the risk of the technique improvement compared to the current state of the art of this and other techniques.
- The imaging resolution limits its applications. Prioritization is needed as to how the imaging helps researchers to improve design and performance by utilizing a valuable result from the neutron imaging.
- Because the record time for a picture can be in the range from some seconds to some hours, the way to ensure the stability of the fuel cell should be made more precise. Actually, in a one-hour range, the water distribution in a fuel cell may evolve, in particular for high-area cells. A presentation of consecutive pictures under stationary conditions would be appreciated. Single-cell investigation is useful, but imaging short stacks would be of added value.

### Recommendations for additions/deletions to project scope:

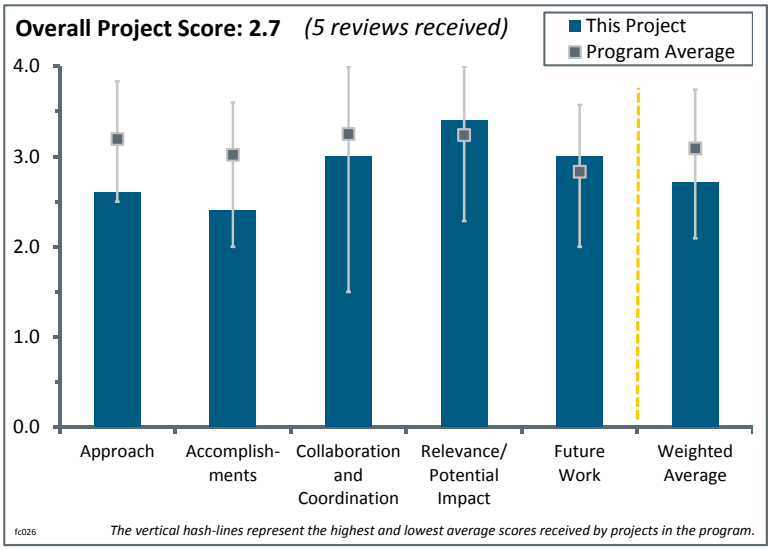
- The project team should move forward toward improving resolution.
- The project team should prioritize the fuel cell stack drying-out investigation.
- Any additional paths that may assist in mitigation of the risk related to the high-resolution detector development would be beneficial.
- The scope is fine.

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

Adam Weber; Lawrence Berkeley National Laboratory

### Brief Summary of Project:

One objective of this project is to gain a fundamental understanding of transport phenomena and water and thermal management at low and subzero temperatures using state-of-the-art materials. Water management with thin-film catalyst layers and water management and key fundamentals in the various fuel cell components are examined. The project seeks to enable the development of optimization strategies to overcome observed operational and material bottlenecks. This project also strives to elucidate the associated degradation mechanisms due to subzero operation and enable mitigation strategies to be developed.




### Question 1: Approach to performing the work

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

- The approach is good and systematic; it involves state-of-the-art materials, studying one of the critical weaknesses of these materials, substantial experimentation, and modeling efforts.
- The project seems to develop individual models to elucidate each single phenomenon. It is unclear if there is any approach to integrate them to develop a membrane electrode assembly (MEA) performance model. It may be effective to clarify which model platform could be proper—the stochastic pore-network model or the deterministic finite element analysis (FEA). Because the nanostructured thin film (NSTF) catalyst layer has a critical water management problem, it is good to add a conventional catalyst later to alternative materials to make the knowledge base more universal with respect to water management in cold temperatures (including subzero temperature start-up). The parallel flow field is widely used by stack original equipment manufacturers and is better (easier) for modeling.
- Although the approach is focused on overcoming U.S. Department of Energy (DOE) barriers on durability and degradation, it looks more like a basic science project.
- The project addresses water management issues. Low-temperature performance is critical to automotive fuel cells, and understanding water condensation and flooding at low temperatures is critical. The project includes NSTF, which has advantages in low platinum group metal (PGM) content and durability but also has water management issues at low temperatures. Solving the NSTF water management issues at low temperatures would be a big step forward. The gas diffusion layer (GDL) effective diffusivity work addresses program needs and is complementary to work from other transport projects. It is not clear what is new in the (bulk) membrane morphology studies. There have been extensive studies of Nafion® morphology, including numerous SAXS studies, TEM studies, AFM studies, conductive AFM studies, etc. It is not clear what is different here and what new information is being obtained. It also is not clear what new information is gained by using the Nafion morphology to model transport through bulk Nafion, or its relevance to low-temperature operation of fuel cells. This portion seems to be a basic sciences study embedded in an applied project, where the basic science study provides little to no added value. Studying the morphology of Nafion thin films on Au and Si provides no useful information for this project. Thin-film Nafion morphology on Pt and C may be useful, but the transport properties are more useful. Direct measurement of the transport properties would be more beneficial than calculated morphologies from a transport-morphological model.

- The overall approach aims at providing inputs from modeling to help industry and national laboratories resolve some of the typical fuel cell performance issues related to 3M, Los Alamos National Laboratory (LANL), and United Technologies Research Center (UTRC). Considering that these diverse sources of MEAs have different electrode structures and natures of catalysts, this effort is expected to provide relative metrics to their operation. In terms of the approach, most of the modeling aspects are not new or innovative; they are well established. Most of the results of this modeling work are, in general, what would be expected. An expected outcome of this effort in terms of modifying the approach would have been the ability to predict results of the various parametric contributions during transient conditions. Such time resolution would be an important step forward in predicting the effect of load variation, humidity, temperature, etc. In addition, modeling inputs such as impedance date are not used; this will be more important as compared to the imaging used to follow water at the interface.

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

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

- The project team has developed a good understanding of the resistivity properties of thin films and narrowing operating windows. The effort should continue into describing areal gradients in order to prevent a voltage drop in the lower-temperature conditions. Critical milestones got delayed or changed in scope. The fundamental understanding of the anode GDL effect on flooding remains uncertain, and specific material factors were not quantified.
- The project team has made significant progress in modeling and understanding some fuel cell transport fundamentals, but it has made only modest progress toward eliminating the water flooding problem at the anode.
- The accomplishments here have been good. The lack of inputs such as impedance data and the lack of the modeling exercise in considering transients are important oversights. It is highly recommended that the researchers incorporate time resolution into these models.
- Good matching model and experiments can be seen; however, modeling information should be disclosed further (such as information on water saturation at low temperatures, what kind of model was developed, etc.). For the anode GDL study, the materials properties should be disclosed to explain why the MRC GDL showed better water management. It is necessary to explain the unmatched PTFE uptake model and experimental data. For membrane water uptake, SANS may be more useful than SAXS.
- The model has been able to reproduce the effect of temperature on NSTF performance. The model predicts that NSTF proton conductivity can have a strong influence on cell performance. Results for a transient MEA-level cold-start model have not been seen yet. A validated transient cold-start model is needed. The project is behind on meeting milestone M3 for the microporous layer (MPL) model in terms of predicting breakthrough pressure and capillary pressure-saturation behavior. The project is behind on milestone M4 for determining the impact of water balance on start-up performance for NSTF and low-loaded traditional MEAs. While milestone M5 has been changed (to adiabatic single-cell tests versus rainbow stack tests), it appears that the researchers are behind schedule for this milestone.

### Question 3: Collaboration and coordination with other institutions

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

- This project features good collaboration between industry and academia.
- The project is pursued with wide collaboration, including with industries.
- It is not clear how all of the studies are being tied together and brought into the final model. Coordination with other projects (3M MEA, LANL durability) is evident.
- The partners carry a significant scope of the experimental work for the project. The design of the experiments on the partner side lacks consistency or was not articulated enough in the presentation in order for reviewers to determine the value of the results obtained for modeling.
- This project features good collaboration; however, the exact role of 3M remains an issue. 3M still remains as the primary source of the materials in this effort. This makes the project heavily tilted toward performing

a systems optimization for a single company. More emphasis should be made on materials from other sources. Despite any proprietary considerations, it is important to link the different MEA design elements to the performance and outputs from the model.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The project is important for advancing DOE RD&D goals.
- Water management and operational robustness are very important for the stack development. High area-specific power density is required to reduce the total active area and enables significant reductions of the stack cost.
- The project addresses water management issues. Low-temperature performance is necessary in automotive fuel cells, and understanding water condensation and flooding at low temperatures is critical. The project includes NSTF, which has advantages in low-PGM content and durability but has water management issues at low temperatures. Solving the NSTF water management issues at low temperatures would be a big step forward.
- If the project demonstrates modeling and component solutions toward tolerating sub-freezing and low-temperature conditions with the respectful operating windows for thin-film electrodes, this may strategically impact the research and development layout of the Hydrogen and Fuel Cells Program.
- This modeling exercise provides important inputs to MEA- and system-level integrators for their respective designs. In this context, the lack of a system integrator approach and clearly defined queries from their perspective represents a glaring gap. Clearly defined inputs from a system integrator, typically an automotive company, would enhance the potential impact of this work.

### Question 5: Proposed future work

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

- The proposed future component characterization work is relevant (excluding membrane morphology work). The planned transient testing and modeling of transients is of high importance.
- MPL is added in the proposed future work.
- The proposed future work is well delineated. However, incorporating transient models and inputs from an automotive company would be further recommended.
- The plans are substantial for each party on the team, which may be overwhelming considering there is less than a year left in the project.
- The proposed future work is still focused on understanding the water management problems; however, at this stage of the project, one would expect to have a solution.

### Project strengths:

- The project team features strong modeling capabilities. There is good collaboration between industry and academia.
- The project features strong modeling capability.
- The approach and collaborations are strong, and the best experts in materials, testing, and modeling are involved in this project.
- Strengths of this project include the GDL characterization work and the neutron imaging studies.
- The project provides important input for system integrators and MEA designers for predicting results of variation in an operating environment. For this project, MEA and other inputs from 3M are considered as the principal materials provider. These are put in the context of more traditional MEA designs from LANL and UTRC. Most of the model outputs are important for this stated objective.

### Project weaknesses:

- More explanation of model development in the project is needed.
- The project is focused on fundamental studies of different MEA components and their assemblies, while less effort is invested in problem solving.
- The correlations between the fundamental properties and component metrics are not well articulated, leaving questions about what component features become governing in tolerating low-temperature operation.

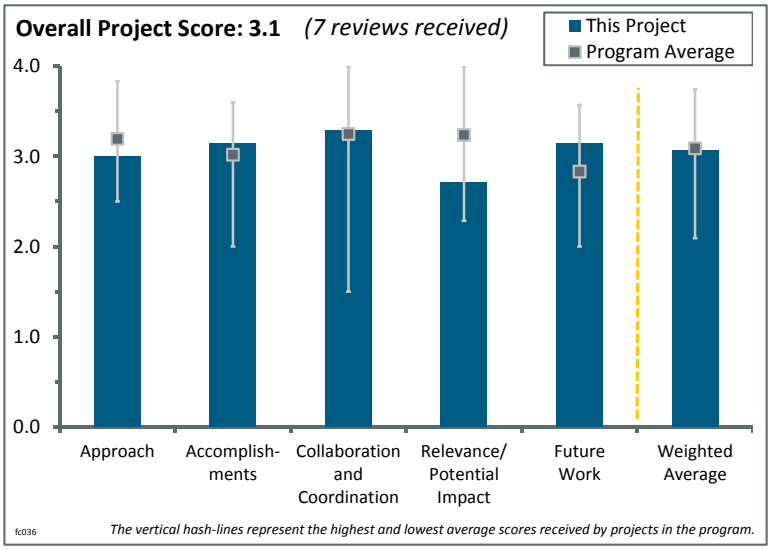
### Recommendations for additions/deletions to project scope:

- The project team should continue the modeling work using degraded membranes to produce narratives on how the whole picture would change with time and how sensitivities would change with time.
- The membrane morphology work should be deleted from the project. It is not clear what new information is to be gained compared to the numerous morphological studies already in the literature. It is unclear what value the Nafion morphology study adds to the project and how any new information from these new studies is critical to low-temperature operation.

## Project # FC-036: Dimensionally Stable High Performance Membranes

Cortney Mittelsteadt; Giner, Inc./Giner Electrochemical Systems, LLC

### Brief Summary of Project:




Three viable pathways were investigated in this project for developing dimensionally stable membranes (DSM™). Inversion casting, which was not pursued further, displayed too many problems with process control and the intrinsic properties of inversion cast films. Ultraviolet (UV) microreplication became a secondary pursuit despite having the lowest ultimate cost, achieving the desired material properties, and having straightforward scaling for the process. The mechanical pathway was the main focus of research because the best materials choices (thermoplastics) are available for this pathway; costs are not prohibitive; and when high (50%) porosity is reached, DSMs™ perform the best.

### Question 1: Approach to performing the work

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

- The team clearly focused on challenging aspects of the problem and screened a variety of options and materials before closing in on the most attractive option to spend the most time on.
- As long as Nafion® or any deformable membrane is used in a fuel cell, this approach will be valuable for meeting durability standards needed for automotive and other applications.
- The approach is clear and well articulated, though single-sided, addressing the mechanical reinforcement of the membrane without ionomer stability.
- DSMs™ are critical to extending the cell performance and life; however, the viable pathways need to be further developed.
- This project features a good approach that seems to address better stabilization but needs to show ionomer incorporation as well as stability and durability of the supports. Its cost impact and benefit compared to traditional supported membranes are not clear. It would be good to see some modeling to figure out the utilization of the catalyst layer, as well as to guide the membrane support design.
- A membrane support structure of 50% porosity may not be sufficient to reach the DOE membrane conductivity target. Fuel cell performance and durability testing using the accelerated stress tests (ASTs) as well as freeze/thaw cycling should be conducted before optimizing the support.

### 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 met all goals and demonstrated meaningful results, such as 10,000 hours of high-performance operation when reinforcing webs are used. The team has optimized web structure and manufacturing process.
- Although there are still issues that need to be overcome, the advances that have been made are quite impressive, and the progress toward the well-defined objectives is what one hopes to see in these types of projects.

- The project has demonstrated good durability of the membrane on the process optimization. Because the project is still in the stage of pathway determination, the team should address the fundamental strength of the selected or prioritized pathway (mechanical).
- Goals for this year included fuel cell qualification of membranes, which was not presented. The team has made good progress in optimizing the UV-microreplication and mechanical DSM™ fabrication methods. Both processes are feasible for high-volume continuous roll-to-roll fabrication.
- The progress has not been that great, but it is good relative to the budget. Good metrics were shown, but too much focus was placed on previous efforts. The research team did nice work on determining the membrane support fabrication materials and methodologies.
- The research team claimed to have achieved the relative humidity (RH) cycling target in ex situ testing, but the team did not provide any data of crossover metrics throughout the types of the supports utilized in the testing. Polarization data from the beginning to the end of the RH cycle test or post-test images would be more credible to show the stability of the ionomer-support interface.

### Question 3: Collaboration and coordination with other institutions

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

- The team is small but effective, as seen by its excellent progress and meeting of goals.
- Solid collaborations are established both at the industrial and university levels.
- This team features strong interaction between project partners.
- This project features a good team, but it is unclear what is going forward in terms of the down-selects and partners. There should be discussion with ionomer suppliers.
- It is clear that collaboration is occurring, but it was not completely clear how the collaboration is coordinated or functions in terms of team meetings and other interactions. Overall, however, the advances are evidence of collaboration.
- Further collaboration may help the project team to accelerate the development in the pathway.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

This project was rated **2.7** for its relevance/potential impact.

- The DSM™ may facilitate the cell and stack design, as well the control of the operation protocol.
- This technology has relevance to Nafion® and any deformable membrane, as seen by the fact that it may just be the final touch needed to make practical the membranes made by another DOE team, Fuel Cell Energy.
- The work could have a “good to outstanding” impact. The potential impact would have been rated “outstanding,” but it seems that there are still quite a few hurdles to overcome before it is clear what level of success and impact will be felt.
- Reducing membrane costs by introducing a dimensionally stable hydrocarbon reinforcement support is on the path toward reducing the cost of the membrane electrode assembly (MEA), and thus the project is relevant to the overall DOE objectives.
- Without addressing ionomer stability, this project partially supports the Hydrogen and Fuel Cells Program and DOE RD&D objectives.
- This could be an important project if the research team can use low equivalent weight (EW) and low-porous matrices, provided the cost is really less expensive and the adhesion and compatibility problems can be solved.

### Question 5: Proposed future work

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

- The future work is highly relevant to proving fuel cell performance.
- The team is on track to meet all of its goals.

- Mechanical focus is the right way to go so that the membrane can be produced on a larger scale and in a cost-effective way.
- The focused plan that is proposed has benefited from the well-structured and disciplined approach. The proposed future work is a logical extension of the work done so far and the interpretation of the data that were outlined.
- The research team needs to show the performance gain before moving on for processing and manufacturing controls to show the benefits and what the costs need to be. The proposed future work looks acceptable.
- The future plan seems adequate.
- Cost analysis should be included in the future work.

### Project strengths:

- This project features good partnering, a good plan, and a logical approach.
- The project has a strong and experienced team.
- The project has yielded good testing results in a practical way.
- The project is well focused. Reasonable progress has been demonstrated in screening the three viable pathways for making the porous support.
- This project features creative thinking about various ways to address a really challenging topic.
- This project features an interesting approach to reinforce membranes. The team has made good progress on making the membrane supports.
- The project explores alternative methods for mechanical reinforcement of the membrane. Establishing new manufacturing technologies in MEA processing is valuable and could be synergistic with other materials developments for the electrochemical power sources.

### Project weaknesses:

- The project needs deeper stress analysis and swelling analysis.
- There are no weaknesses, except it would be nice to know more details of the process for making the web that is put in the membrane to stabilize the dimensions.
- High-volume cost predictions for the engineered supports seem viable for PFSA based membranes but too expensive for hydrocarbon membranes.
- There is question about the overall gain in cost. There is also question about the priority of this project, especially because Giner is already working on this in its existing technologies.
- The timeline and timing for the go/no-go decisions were not specified in the presentation. Judging by the starting time, the project should have passed the end of Year 2. However, the project has not demonstrated whether it passed the go/no-go for Year 2. The mechanical properties of 50% porosity support made by the mechanical deformation method are critical information and should be included in the presentation. One of the objectives of this project is to reduce the cost of the membrane down to <$20/m$^2$. A target should be established for the porous support because it is the focus of the project. As noted by a reviewer last year, the cost target for the support probably should be < $5/m$^2$. The cited cost numbers in the presentation are very high compared to this "target." A more detailed cost analysis is needed to demonstrate the viability of the proposed approaches. More details about the mechanical deformation method are needed to understand the cost/performance aspect.
- This project potentially leads away from the more difficult developments of the higher temperature, conductivity, and stable ionomers, which are durable under fuel cell operating conditions.

### Recommendations for additions/deletions to project scope:

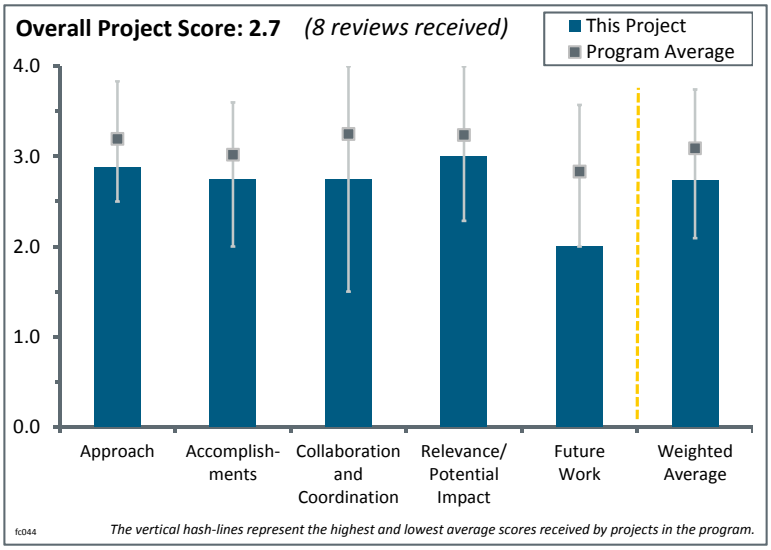
- The researchers should pursue a patent so the Giner team can discuss the work more openly.
- The conclusions need to be backed up with solid data and analysis.
- The team should focus on demonstrating DSM™ performance and durability prior to further optimizing the support structure and fabrication processes.
- If the project continues to focus on solely mechanical durability, the project team should investigate the sensitivity of the resulting DSM™ to different flow fields and GDLs during fuel cell testing.

- One of the objectives of this project is to reduce the cost of the membrane down to <$20/m$^2$. A target should be established for the porous support because it is the focus of the project. As noted by a reviewer last year, the cost target for the support probably should be <$5/m$^2$. The cited cost numbers in the presentation are very high compared to this "target." A more detailed cost analysis is needed to demonstrate the viability of the proposed approaches.
- The project team should look at low EW ionomers and perhaps hydrocarbon ionomers as well to see if there is a more significant gain, but admittedly this is a support project and not a membrane one. The project team needs to think about adhesion of the catalyst layer to the support, especially under stressed conditions of humidity, freeze, etc. The team should also think about looking at accelerated stress tests for both just the support and when a membrane and catalyst layers are incorporated. The team should do some computational modeling to help guide the design of the structure and how it should behave under operation.

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

Eric Brosha; Los Alamos National Laboratory

### Brief Summary of Project:




The objective of this project is to develop a ceramic alternative to carbon material supports for a polymer electrolyte fuel cell cathode. Ceramic cathodes must have enhanced resistance to corrosion and platinum (Pt) coalescence; preserve positive attributes of carbon such as cost, surface area, and conductivity; and be compatible with present membrane electrode assembly (MEA) architecture and preparation methods. Materials properties must possess the required surface area, foster high Pt utilization, exhibit enhanced Pt-support interaction, provide adequate electronic conductivity, and exhibit resistance to corrosion. Materials used in ceramic cathodes should also be amenable to scale-up and have reasonable synthesis costs.

### Question 1: Approach to performing the work

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

- The approach of this project is much improved from last year. It is good to focus on $Mo_2N$ and $Mo_2C$ (excess carbon-free) from various materials and synthesis methodologies. From the technical standpoint, it is also valuable to clarify reasons why other materials and synthesis methodologies were dropped; for example, $Mo_2C$ (polymer-assisted deposition).
- The project is focused on non-carbon supports for Pt-based catalysts. The principal investigators (PIs) had a systematic approach to tackling challenging demands that support materials should meet, such as high conductivity, resistance to degradation, high surface area, and strong attachment to the catalyst particles. Study materials are systematically characterized, and detailed insight into structure function properties was possible to obtain.
- Exploring the possibilities of replacing carbon supports by providing more stability to oxidative degradation ceramic materials is a reasonable topic.
- This project started out with the somewhat vague idea that ceramic supports would be useful. The project ended up with $Mo_2C$, Pt-embedded $Mo_2C$, and $Mo_2N$.
- Los Alamos National Laboratory (LANL) has identified a promising ceramic material based on molybdenum. However, the performance of this material needs further improvement.
- The approach is generally effective (involving many structural and spectroscopic probes), but it could be improved, in particular with regard to electrochemical characterization.
- Carbon supports have been necessary to keep Pt loadings low, but they suffer from corrosion and weak Pt binding. Alternatives with improved performance and durability would be preferred. The development of high-surface-area, electronically conductive, corrosion-resistant ceramics is a reasonable approach. The approach for the project was laid out much better in past U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program Annual Merit Review presentations, and milestones as presented are not clear in terms of time frame or how go/no-go decisions have been made.
- The project focuses on a few materials and relies heavily on the synthesis capability of The Materials Foundry and University of New Mexico (UNM). It is unclear if the team has sufficient resources and expertise. The elimination of all carbon should not be the goal of the project. A noticeable portion of the

effort was dedicated to investigating Pt-support interaction, rather than to developing the support to perform well in a fuel cell.

### 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.

- There is no denying the stability of the $Mo_2N$ supports. The $Mo_2C$ data are not as clear, although it is still better than the incumbent technology (carbon). There is clear progress here.
- The project demonstrates progress toward DOE goals. These materials show promise, but it is not clear that they can be scaled up to produce a cost-effective solution.
- The PIs presented good progress over the last fiscal year and were able to overcome demanding barriers for implementation of novel supports in fuel cells. The high performance has not yet been achieved, but this project serves as a source of valuable data for the future catalyst design.
- The demonstration of the "nanoraft" (NR) $Mo_2C$ materials is a significant step forward and offers some hope in improved performance and durability for the non-carbon supports investigated. These materials appear to have promise and merit further investigation, while the other materials investigated to date had shown little in terms of promise. It is somewhat difficult to fully evaluate the potential of the NR $Mo_2C$ materials because the durability (waterfall) charts presented are based on changes in electrochemically active surface area (ECA), and judging by the limited cyclic voltammograms (CVs) of these materials presented, it is not clear that the surface area or surface area changes reported are appropriate. The high mass activities of these materials are promising, but it is not clear what happens to mass activity after cycling or exactly which cycling protocols have been applied in all cases.
- The focus is narrow, mostly on one support for Pt–$Mo_2C$ and, alternatively, $MoO_3$. The paper by Rossmeisl (*ChemCatChem* 2012) showing that $NbO_2$ is the best support for Pt should be a comparison.
- It is unclear if a clear specification for material selection was set. Few materials investigated have made it into fuel cell evaluation. The results that were shown demonstrated that this is a significant challenge.
- The PIs made minimal progress toward the objectives, especially if the results are compared to the most active Pt-based systems supported on carbon. The stability and conductivity of systems are still far from being realized. Therefore, there has been modest progress in overcoming barriers, but the rate of progress has been slow.
- The fuel cell performance and durability with Pt/$Mo_2C$ is very poor. It is good to distinguish oxygen reduction reaction (ORR) kinetics and $H_2$ crossover problems. It is necessary to investigate whether damage of the electrolyte membrane is critical to use ceramic support for the catalyst layer. Passivation of the $Mo_2C$ surface seems to be critical. It is interesting to see the effectiveness of the lattice mismatch of the $Mo_2C$ support and the catalyst activity of Pt (modeling work).

### Question 3: Collaboration and coordination with other institutions

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

- The PIs have demonstrated a strong team effort.
- Collaboration is very good, as seen by the structural characterization.
- LANL is making very good use of collaborators at multiple institutions.
- Although partners are well coordinated, some improvements are still desirable—especially with the experts for electrocatalysis.
- It would be good to add an industry collaborator to the project and leverage its influence regarding technical approach and fuel cell testing.
- The project has limited collaboration. It should look to expand if the few materials the researchers are currently investigating do not show promise.
- The project consists of modest collaboration, but it is also reasonably small in scope, so this is appropriate. It now contains an industrial partner owing to the spinoff from LANL for the $Mo_2C$ NR synthesis. The use of UNM and Oak Ridge National Laboratory in providing complementary characterization efforts is appropriate.

- There is not a clear path to scale-up. Perhaps it is time to engage a catalyst supplier. Original equipment manufacturer (OEM) fuel cell testing is needed to prove the concept.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The project aligns very well with the R&D goals of the DOE Hydrogen and Fuel Cells Program. The choice of proper high-surface-area support is critical for the future development of catalysts. The knowledge accumulated from this project can certainly serve in future considerations of catalyst supports, considering that the PIs have conducted a systematic structure function evaluation of materials.
- The impact on durability is very clear.
- This project is relevant, but elimination of all carbon is not necessary.
- This project only partially supports the Program.
- The corrosion of catalyst support is a critical issue for fuel cell durability. On the other hand, some mitigation strategies were proposed. If use of an alternative catalyst support would require sacrificing fuel cell performance, the relevancy is relatively lowered.
- The project has the potential to develop more robust and perhaps higher-performing catalysts that could help address cost, performance, and durability of fuel cell systems—the primary remaining technological barriers. The materials developed to date have not yet met DOE electrocatalyst activity targets and have remaining challenges regarding utilization in high-performance fuel cells. The very low fuel cell open circuit voltages and poor performance in MEAs reported on slide 21 are a concern along these lines.
- Finding a good replacement for carbon would have a great impact on fuel cell research and technology.
- Surface passivation, corrosion, and loss of performance are major technical issues that need to be addressed.

### Question 5: Proposed future work

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

- The investigators are making excellent progress in addressing the technical challenges.
- The focus should be made broader, and the Pt-support interaction should be addressed.
- The proposed future work aligns with the project outcomes but does not point clearly to whether ceramic supports are a real alternative for carbon supports. It seems that stability issues may prevent this class of materials from becoming a valuable candidate for high-surface-area supports.
- There is not enough time remaining because this project is supposed to end in September. Therefore, the project team should be focused on some critical areas. In addition to the completion of fuel cell tests (durability), it is important to clarify the cause of the MEA damage, model work for the possibility of performance enhancement, and address the criticality of the surface passivation of $Mo_2C$.
- Few materials are investigated. It is not clear if the team has a good understanding of its materials and the properties needed to obtain good fuel cell performance.
- Owing to the length of the presentation (too many slides) and the presenter's running out of time at the end (in the oral presentation), the future work was not conveyed very well. The approach to future work presented on slide 29 is not compelling or clear; it consists of general statements without guidance to the approach or prioritization.
- The PI should drive toward a conclusion here by making something that works and engaging a supplier and a customer. The project team has something here; it should not waste the opportunity. It is unclear where the value proposition is.

### Project strengths:

- This project features a good idea—a good synthesis that works.
- This project features a novel ceramic support that might provide enhanced properties.
- This project features the capability for various synthesis approaches.

- The project is relatively well organized and has a clear focus. The PIs are good scientists, and synthesis of new ceramic materials is based on understanding of the systems and not just testing of "new materials."
- NR $Mo_2C$ has shown promise in rotating disk electrode performance at low Pt levels. Initial results suggest durability may be favorable, but more work is needed.
- Strengths of this project include its systematic approach, strong team effort, and fundamental insights into the structure-function properties of novel supports.

### Project weaknesses:

- The lack of industry collaboration is a weakness.
- The material is likely not cheap—it is not easy to manufacture and its long-term stability is questionable.
- No suppliers or OEMs are involved. The project team should get focused. The team is not narrowing down to a conclusion. It should take one option to the next phase.
- The selection of new supports is rather narrow. Electrochemical characterization is almost nonexistent, and thus comparisons in activity of ORR on different materials are questionable.
- Weaknesses include the lack of vision for future work and a support of choice that can exhibit outstanding performance.
- Too little is known about the new synthesis route to understand how viable it is for further advances to occur. Significant data were presented, but most of the interesting work revolved only around NR $Mo_2C$. Data that would have been useful were often not presented, while extraneous data with less value were presented.

### Recommendations for additions/deletions to project scope:

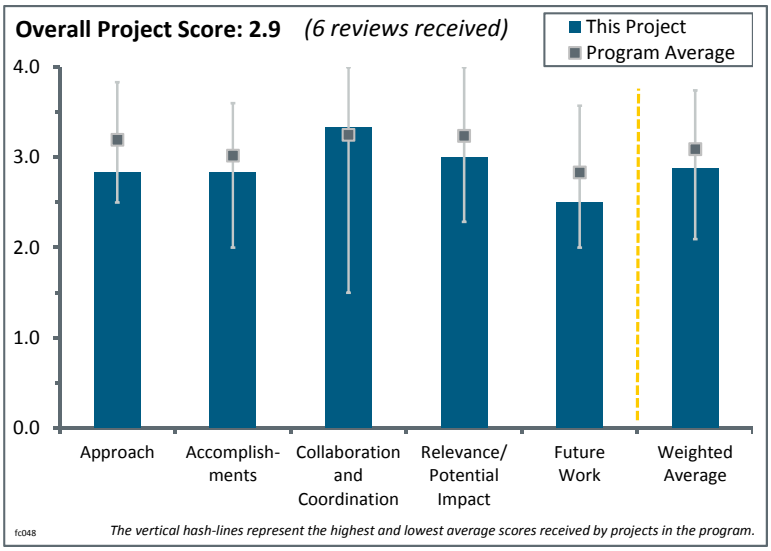
- The project team should focus on scale-up to produce enough quantity of material to perform statistically meaningful tests.
- This project is almost completed, so there is little to be done in the end. The embedded Pt–$Mo_2C$ looks better than the Pt–$Mo_2N$. It is unclear if this is really true, and it is important to know.
- Work should focus specifically on $Mo_2C$ NRs by the new novel synthesis route. These materials show some promise but lack sufficient data to make a strong statement about potential. CVs of these materials should be given, test conditions for cycling should be presented, CO stripping should be used to investigate ECA, and mass activities should be presented before and after cycling. The modeling studies provide no immediate leverage for the project and should be ceased. The fuel cell studies can be de-emphasized until the materials are better characterized and understood.

## Project # FC-048: Effect of System Contaminants on PEMFC Performance and Durability

Huyen Dinh; National Renewable Energy Laboratory

### Brief Summary of Project:

System contaminants have been shown to affect the durability and performance of fuel cell systems, thus prompting research to limit contamination-related losses. As fuel cell stack costs have decreased, the focus has shifted to addressing balance of plant (BOP) materials costs to further decrease overall fuel cell system costs. This project worked to identify poisoning mechanisms and recommend mitigation strategies, develop predictive modeling, and provide guidance on future material selection.




### Question 1: Approach to performing the work

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

- The project provides a very systematic approach to screening a large number of relevant contaminants, understanding key mechanisms, categorizing the contaminants into logical groups, and determining performance impacts and recovery behavior.
- From an engineering standpoint, the objective of this project is valuable for materials screening. From a technical standpoint (this factor is the most important for U.S. Department of Energy [DOE]-funded research and development [R&D] projects), without investigation of performance degradation mechanisms, outcomes cannot be fully applied to improve fuel cell durability. Looking at ORR kinetics and HFR is good in order to distinguish the type of degradation mechanism. It is questionable whether this database can be applicable for types (levels) of fuel cell materials. For in situ analysis, the fuel cell technology (e.g., catalyst and membrane materials) should be disclosed.
- The project is well worthwhile, but it is structurally overwhelming and complicated by too many sources of contaminants. The approach of using model compounds is logical and probably the best approach given the circumstances, but because the actual contaminants and their real-life concentrations are not really determined, the effects seen from model compounds show only potential effects and not real-time effects. Measurement of the macro effects of the functional interaction of the amine or hydroxyl functionality ion exchanging or the organic sorbing on the catalyst is very instructive in determining which mechanism has the most impact; that work is nicely done, but the biggest impact mechanism needs to be studied in actual un-doped cell operation to determine the relationship between model compounds and real operation. Also, no mention was made of mitigation routes.
- The approach has been systematic. It seeks to identify which systems contaminants exist, separate them into fundamental classes (for greater universality of results), and then identify which are most harmful. For such a project, the probability that identified contaminants will not be universal for all developers will always exist. The fact that only one developer has been highly involved in contaminant selection (General Motors [GM]) is a weakness. It does not appear that Ballard or Nuvera contributed highly in that regard.
- The team is quantifying voltage loss into fuel cells but only looking at constant current measurements. It is well known that there is a strong voltage dependence of many of the contaminants, and this dependence should be considered. Also, it is well known that pure organic compounds will oxidize at a Pt cathode in air at >0.6 V or so. A bigger problem is the additives. The team should work harder to study the individual constituents of the materials of interest to the automotive industry and identify the true bad actors.

- The contaminants selected are by-products of other BOP components. It is unclear how it was determined that all contaminants should be tested at 1,280 ppm, for example.

### 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.

- Many materials have been screened to determine their effect on the fuel cell. Some inconsistency exists in the number (55 in slide 6, 62 in slide 7) of materials studied. In addition to quantifying the performance loss, the project tries to identify the type of degradation.
- Lots of data and the big effects are nicely determined for model compounds, but it appears that the translation of the most powerful effects for real-time operation is missing.
- A large amount of data (materials and parametric analysis) enables researchers to identify a tendency of performance degradation. Deeper investigation of the degradation mechanism is expected for some possibly critical material problems, for example, the ORR effectiveness of organic contamination.
- The project had made good progress and already provides a useful tool for fuel cell designers. The website is a very useful tool. Some additional information on the types of mechanisms and recovery behavior would be useful on the website. There has been good characterization of a large number of contaminants to categorize and study in an efficient manner.
- For the contaminants identified and with the techniques identified, the project has generated considerable data and has identified mechanisms (e.g., membrane contamination for 2,6-DAT). It would be useful if some of the results were reproduced on a large cell or stack scale. The study of contaminant mixtures is useful and helped to address some questions with the project. The study did well to include the effect of catalyst loading on the results.

### Question 3: Collaboration and coordination with other institutions

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

- This project features excellent collaboration with very capable partners.
- The collaboration seems excellent; the materials information came from fuel cell original equipment manufacturers (OEMs).
- This project features good collaboration and coordination, particularly with GM. The researchers have also included additional industry partners to ensure that relevant materials are being studied.
- This project has a good-sized group of contributors. It is unclear if all of the experiments were conducted at the National Renewable Energy Laboratory (NREL). It is unclear which partner contributed to which results presented.
- A broadening of developer input would have been better for the project. Ballard and Nuvera appeared to have some input into the testing, but there is no evidence that either provided inputs for contaminant selection. The University of Hawaii has years of experience looking at fuel cell contamination, and it was a good addition to the team. GM made a substantial contribution toward defining the materials set.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The relevance and potential impact are high, and the project objectives of determining the effects of contaminants are needed.
- The project addresses the effect of contaminants that can enter the fuel cell and degrade the performance. Identifying the potential contaminants and their effects is necessary to help maintain the desired performance and durability.
- There is value to developers in knowing about system-borne contaminants. The project risks relevance by going after particular compounds and even particular functional groups that may not be applicable to some

developers' designs. The project did well to enhance its relevance by considering lower Pt loading, as well as the role of water in either washing away or recovering from the effect of impurities.

- From an engineering standpoint, the objective of this project is valuable for materials screening. From a technical standpoint (which is the most important for DOE-funded R&D projects), without investigation of performance degradation mechanisms, outcomes cannot be fully applied to improve fuel cell durability.
- This work is very important for stack/system design. It is a common problem that materials used either in the stack itself or somewhere in the system design can have contamination effects. In a working fuel cell system, these effects can result in performance problems that take considerable efforts to diagnose. A tool that can be used during the design process is extremely relevant and will provide significant improvements to the design process. This tool will become more important as (1) designs become appropriate for commercial cost targets, requiring lowest-cost materials to be used; and (2) increased volumes of fuel cells are introduced into new applications, resulting in new system integrators without past knowledge of acceptable materials. This project is different from most of the funded projects in that it combines research activities with a highly practical and pragmatic approach.
- The research needs to make clearer which particular compounds to avoid and which ones are safe. There is no way to tell from how all of the data are rolled up.

### Question 5: Proposed future work

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

- The proposed future work is appropriate.
- The plan could be a little more specific. It is unclear what "in-depth analysis" is.
- The project has only a short time left, and the researchers mention mitigation strategies, which would be very valuable. Hopefully they can find some that can be demonstrated in the short period of time left.
- This project is ending in September; therefore, there is not enough time to do a new task. It is questionable how valuable the modeling of parametric analysis with operating conditions is, or how it can be valuable to identify degradation mechanisms.
- The project is nearly over. All that appears left is to assemble and disseminate the model, which is appropriate. The only experimental activity that would be interesting to add would be to repeat one of the contamination experiments with a large-size (about 300–400 $cm^2$) cell to see if the results scale.

### Project strengths:

- The team presented a lot of data this year.
- A large number of contaminants have been screened. Their effects have been quantified. The researchers have some insight into how the contaminants are affecting the performance. The research team is good.
- Strengths of this project include the amount of data and analysis, both ex situ and in situ, and the information from fuel cell OEMs.
- Strengths of this project include the extensive list of contaminants considered, the models of contaminants, and the linkage of mechanisms to classes of contaminants. This project is important work for stack/system design; it is addressing a common problem of fuel cell developers. Development of the website is a valuable tool.
- This is much-needed work, and the approach of model compounds was probably the only way to tackle such a complicated problem. The team has produced lots of data and excellent technical hypotheses on how each contaminant might affect operation. There is excellent cooperation among partners.
- Strengths of this project include the team's (1) high-volume data collection capability; (2) collaboration with GM to provide direction on system contaminants; (3) good use of parametric variations to understand the effects of catalyst loading, condensed water, and other conditions that could cause recovery from contamination; and (4) attempt to focus on "fundamental classes" of contaminants instead of particular compounds.

### Project weaknesses:

- One weakness is the depth of the degradation mechanism analysis and mitigation strategy.

- The use of model compounds to determine the biggest negative impact was not followed up by focusing on that mechanism using real-life components. Also, mitigation strategies were not proposed.
- The lack of developer input on contaminants outside of GM is a weakness. The premise of the project could lead to results that are particular for one developer or group of developers.
- Considering the expertise of the researchers from multiple organizations, it would be interesting to hear about options to manage the degradation effect, especially for species that lead to irreversible loss. The level of contaminant exposed to the MEA is likely to be different, depending on the source and release rate.
- The data have not been checked against a larger active area cell. All results are done at 50 $cm^2$. This size is appropriate because a larger cell may mask effects, but some checks should be done. The combined effects of other degradation modes with contamination have not been studied; for example, if ECSA is reduced, it is unclear if the effects will become larger. The effect of catalyst loading is a good addition—the researchers might have considered this as a baseline condition because it would also capture the effects of degradation for current loadings. Correlations of contaminant behavior to cell voltage, or more importantly electrode potential, would provide improved links to mechanisms—only current density is studied.
- The objective to produce a global contaminant model seems unrealistic. It is well known from decades of research on organic and inorganic adsorption on Pt that there are many different pathways to adsorption. Work specific to fuel cells indicates that voltage cycling helps remove many of the contaminants, while some molecules adsorb irreversibly. While it is nice to think that there is some universal process for all adsorbates, it is just not that easy. The work is further complicated by the gas diffusion layer, operating conditions (which vary widely beyond the test conditions probed by NREL), and dose level of the impurity (concentration versus time). It is a very extensive test matrix. The team needs to focus on guiding principles but not a universal model (which does not exist).

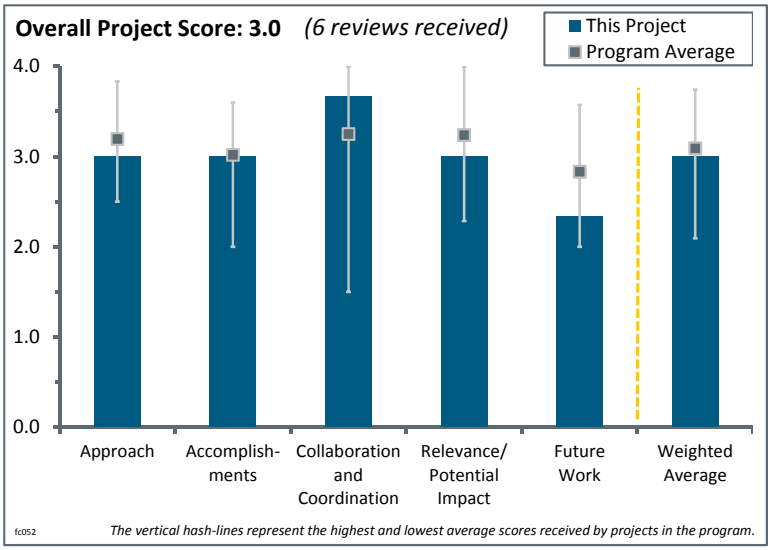
### Recommendations for additions/deletions to project scope:

- Only one addition is suggested: reproduction of small-scale results with a larger cell or stack. It would be interesting to see if the results can be reproduced at that level.
- The researchers should classify the contaminants based on their hazard and propose management options based on the researchers' insights. Identifying the effects at various operating conditions should be a lower priority and paid for by the developer.
- There has been extensive testing on mechanisms and characterization of the compounds, as well as selection of model compounds. Because the website will be the legacy of this project, this work should be better represented on the website. It is understood that the website is a work in progress. Interactions with other degradation modes should be pursued.
- The team should focus more on how to recover the performance of fuel cells, rather than try to develop a universal model to explain voltage loss. It would be more constructive to look at "classes" of contaminants and categorize their introduction to the fuel cell as reversible or non-reversible. The team should also identify additives that are strong bad actors.

## Project # FC-052: Technical Assistance to Developers

Tommy Rockward; Los Alamos National Laboratory

### Brief Summary of Project:




Los Alamos National Laboratory (LANL) provides technical assistance to fuel cell component and system developers and includes testing of materials and participation in the further development and validation of single-cell test protocols. This task also covers technical assistance to durability working groups, the U.S. Council for Automotive Research (USCAR), and the USCAR/U.S. DRIVE Partnership Fuel Cell Technical Team (FCTT). This assistance includes making technical experts available to the U.S. Department of Energy (DOE) and the FCTT as questions arise, conducting focused single-cell testing to support the development of targets and test protocols, and participating in working group and review meetings.

### Question 1: Approach to performing the work

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

- The work appears to be serving a useful and accessible function of sharing technical assistance to developers through fuel cell and materials characterization. The work directly supports DOE because DOE is directing the scope and development of assistance.
- This project takes its direction from DOE in terms of its focus depending on the needs of other projects. LANL supports the other projects by studying a broad array of specific issues as directed by DOE.
- This project was mainly focused on water. The work involved hydration of feed gases through a membrane device, gas diffusion layers (GDLs) of different water permeability, and tests of fuel cell performance impedance as well of imaging of water in fuel cells by neutron scattering. As long as Nafion® or similar water-based membranes are used in fuel cells, this work will be very important. The work here is critical to hydrating a fuel cell and balancing water in a fuel cell and was well done.
- LANL has a wide array of analytical equipment and techniques to thoroughly characterize materials in need of improvement. Understanding material changes and underlying mechanisms is a prerequisite to mitigating degradation.
- The objectives, goal, and overall scheme of how to proceed with the project are not clearly identified. Even though this project is directed by DOE, the scheme should be defined by the project leader.
- One assumes that each task that is reported had a reason or objective and a goal or product that was described to the DOE decision committee by the customer before each task was assigned, costed, and conducted. This process was not defined in the poster, but it needs to be to determine whether the project accurately addressed the issue. Although the tasks are related to fuel cell issues, the degree of relevance to other tasks at LANL was not described.

### 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.

- LANL has successfully addressed several technical issues identified by DOE and other principal investigators (PIs). The apparatus and procedures have been developed as needed.
- The poster exhibits a wide range of characterization results, from Fourier transform infrared spectroscopy and relative humidity degradation characterization of membranes to fuel cell performance of GDLs to water imaging across the membrane. Scanning electron microscopy is also presented for GDL imaging, but this seems to be a more generalized imaging technique rather than a fuel-cell-specific diagnostic or characterization capability.
- The exact need by each outside organization could not be fully explained because of the nature of the particular effort. However, the assistance that the personnel at LANL have to offer each requester seems to be sufficient and willingly provided. How well this was incorporated into the outside agency is difficult to measure. Providing the assistance that was described seems to be adequate for the need presented.
- Most work involved characterization to support others. There is not much innovation. The hydration system was run and tested, the GDLs were tested, and fuel cell impedance and fuel cell performance were tested. Water was imaged.
- What is shown as an accomplishment in the presentation is not distinguished with what is usually done by each collaborator.

### Question 3: Collaboration and coordination with other institutions

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

- The work was well coordinated over a number of industrial and government laboratories. Results were of high quality. Work was complete and done in a timely fashion.
- The list of "clients" is comprehensive and includes automotive original equipment manufacturers, laboratories, universities, materials and equipment suppliers, and technical institutes. Strong relationships have been established in the course of designing the pertinent apparatus and experimental parameters.
- Participation in DOE working groups supports critical areas that DOE has identified as opportunities for a wide range of developers.
- The project appears to span a wide range of collaborators from government, academia, and industry, as well as across multiple Fuel Cell Technologies Office programs. It is assumed that there is close collaboration with DOE stakeholders. Fuel cell training, demonstration, and class material could serve as a useful input or template for more generalized training materials for the college or university level as part of a greater outreach effort in fuel cell education and to help build a talent pool for further research work in the area.
- Lots of collaborators exist. However, it is difficult to understand the big picture of the collaboration (i.e., how to share the various functions/capabilities and combine them to achieve the goal).
- The degree of interaction with other working groups to learn what activities need assistance and how LANL can provide this seems to be quite good. However, the extent of success in portraying the assistance and availability of personnel and equipment to help overcome specified barriers is not described, and no outline is available because this appears to be a rather ad hoc operation.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The issues addressed are relevant to DOE's goals and targets.
- Supporting the efforts of developers with unique equipment and expertise is an important role that national laboratories are very well suited for.

- To the extent that the work can support DOE objectives and DOE working groups, the project is expected to have a consistently positive impact and serve a useful purpose. The relevance is to facilitate technical support and testing of industry, government, and academic partners that are low-cost and reliable.
- This work involved high-quality characterization of fuel cell components pertaining to hydrating and water handling. This is important for DOE in terms of ensuring that performance and durability objectives are met. There has not been much innovation, so progress is not directly impacted by this work, but it has been indirectly impacted because progress depends on the innovation of collaborators being served and the feedback these collaborators get from this work.
- The impact and importance of the assistance are big.
- The tasks seem to be quite well oriented toward assisting fuel cell developmental needs. However, the educational aspect of the reported work seems to fall outside of the intent to assist the advancement of fuel cell technology. Educational efforts should be funded out of a separate fund or category.

### Question 5: Proposed future work

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

- Future work will be defined by DOE in support of the other PIs. It is assumed that the work will be relevant and timely.
- The plan is to maintain these capabilities. Not much was planned to address the deficiencies observed, such as the formation of hydration limiting anhydride in the water permeation membranes of the feed gas hydrator.
- Because of the ad hoc nature of the task and the unknown nature of DOE funding for fuel cell work, it is difficult to describe what needs will arise and how to address them. While it is necessary to make LANL's assistance open to all parties, this procedure precludes developing specific ties for continued work with a given research group or company agency, which is appropriate. It is not clear how the plans for funding should be allocated when the work cannot be planned in advance.
- It would be good to see more in the way of proposed future work beyond "to be defined by DOE." For example, perhaps stakeholders could be surveyed for their input on what is most useful in existing technical support and what could be most useful but is currently lacking.

### Project strengths:

- This project provides great support for other laboratories, academia, and companies.
- Strengths of this project include its potential value and that it is directed by DOE.
- Strengths of this project include its high-quality characterization work and timely response to collaborations regarding the results of characterizations.
- Providing personnel and equipment availability to DOE contractors that need this assistance to perform their work but cannot fund these DOE-funded facilities or need only conditional assistance is an excellent idea. The interaction with working groups to advertise this capability is a very positive feature of this work.
- This project's strengths include its wide range of technical services and characterization techniques; responsiveness to DOE needs and requests; and how it is serving a wide range of partners across industry, government, and academia.

### Project weaknesses:

- The objectives and process are unclear.
- This is a support project, so the most important aspect is to provide timely and high-quality characterizations. Still, more interpretation and recommendations to the collaborators about modifications to the materials seem warranted and were not evident.
- Perhaps it is possible to develop some metrics of success beyond DOE Hydrogen and Fuel Cells Program Annual Merit Review (AMR) feedback and evaluation (e.g., how many sample images, utilization of tools, throughput time). Similarly, perhaps the impact of such technical support or characterization can be quantified or semi-quantified (e.g., "this characterization work led to a 10% increase in performance or developed the basis for a new degradation model or mechanism for this customer"). Also, there should be a

mechanism to specifically call out the new or enhanced capabilities on an annual basis. Otherwise, year-to-year progress is harder to assess.

### Recommendations for additions/deletions to project scope:

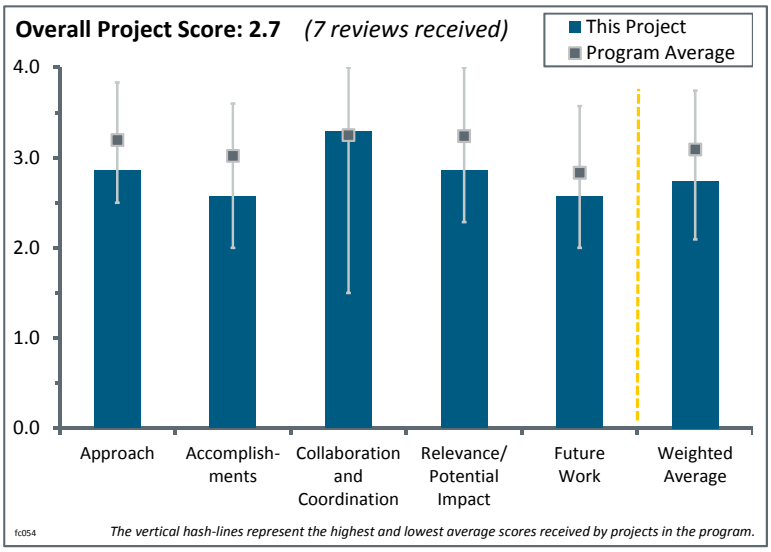
- If the purpose is to provide service to collaborators, this project is fine as is. However, if the purpose is to improve water handling, then more interpretation by government laboratories and adjustment of materials by collaborators (feedback between the team and collaborators) are warranted to stimulate and accelerate innovation.
- Because this is an ad hoc effort and not specifically identified with a specific need in advance of the issues that arise, it is difficult to determine either the workforce or funding that should be allocated. This may also be a problem with respect to how extensive of an effort can be ascribed to a given problem if the funding is to be spread over several unplanned tasks. While this is a valuable "tool" for assisting the advancement of fuel cell research, a significant amount of time is required to establish how effective it is for assisting other researchers. Perhaps including a broad publication of what is available for assistance with any request for proposals would help in identifying what needs could be met by proposers and researchers through including LANL in the proposal at the beginning.
- There are a few suggested additions: (1) perhaps single-cell protocols can be documented either in the poster or in the supporting publications, and (2) perhaps the short course could be the basis or input to a more general short course that can be made available to the community. One could imagine a "train the trainer" session at a future AMR meeting to spread this course content around the United States. There are no recommendations for deletions, but if there was a metric for utilization of the tools/characterization techniques, then maybe some could be downscaled or re-directed if not used, while others could be prioritized for upscaling/expansion.

## Project # FC-054: Transport in PEMFCs

Cortney Mittelsteadt; Giner, Inc./Giner Electrochemical Systems, LLC

### Brief Summary of Project:

The objective of this project is to improve understanding of and the correlation between material properties and model equations. The use of modeling in fuel cell development is widespread, but the fundamentals of modeling are not agreed upon throughout research. This project identifies inconsistencies in design rules for polymer electrolyte membranes (PEMs) and works to standardize the parameters.




### Question 1: Approach to performing the work

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

- The approach to measuring water transport numbers in membranes is novel and good, and it apparently leads to a model that has excellent correlation to the experimentally measured water transport values in actual operating fuel cells.
- The approach is good. By widely varying the transport and structural properties of fuel cell components (mainly the membrane), this project seeks to come up with guiding design principles for fuel cell membrane development and maybe other components.
- This project concentrates on model generation and development of relevant transport numbers.
- The project aimed at understanding the correlation of material parameters to PEMFC transport properties through the development of specific ex situ experiments, a transport model, in situ testing, and verification with in situ data. All in all, this is an ambitious project trying to answer questions that may help to improve/design membrane and gas diffusion layer (GDL) materials with specific properties. It uses a sound approach but aims at high-hanging fruit.
- The approach, it appears, is to develop new membrane materials with different transport numbers. A new electro-osmotic drag coefficient (EODC) was developed, and the degree to which the EODC contributes to performance was estimated using modeling. The model was compared with performance and water balance testing in fuel cell testing to link the component to the fuel cell.
- The approach to the project does not directly address critical barriers, but it does provide support in addressing key barriers of performance, particularly at high current density. The project is not particularly well integrated; the key thrusts of polymer synthesis, transport measurements (development of novel techniques), and modeling overlap but do not critically depend on or leverage each other. The development of a model that cannot be widely circulated has limited value as well.
- Elements of this effort, such as the development of membranes with improved water management, can contribute to overcoming barriers that impede fuel cell commercialization. The modeling and other aspects, such as the segmented cell, are generally similar to past efforts and are less likely to contribute to the development of technologies that overcome barriers.

### 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 progress in membrane design and characterization and transport modeling appears to be reasonable based on additional reading of the published literature in *ECS Transactions*.
- Very good advances have been made with respect to model verification and experimental results, including water balance predictions and cell performance predictions. Some correlations were established between membrane composition and membrane parameters using previously developed experimental techniques. Other correlations included GDL parameters and performance. No data were reported on Tech-Etch. It is unclear what this company's contribution was.
- The accomplishments and progress toward the overall project goals are reasonable based on the lofty challenge that this project is attempting to meet. More effort to validate the model's predictive power based on two-dimensional data is needed.
- The novel polymer electrolytes and novel techniques developed to measure water uptake, diffusivity, and electro-osmotic drag all have value to the community. Some of the structure–property relationships investigated for multi-block copolymers have provided increased insight into the water-related properties of interesting novel materials. The modeling work is showing reasonable agreement with experimental data, but it is limited in the fact that it has not been presented in significant detail, it cannot be openly circulated at the end of the project, and it seems far less advanced than other models that have been developed. The results to date provide little value to the community, although the comparisons between hydrocarbon and perfluorosulfonic acid (PFSA) materials are interesting, and there is significant concern that at the end of the project little specific information will be created that can be built upon by the community.
- This project features interesting measurements and modeling; however, for much of the work there is a lack of in situ validation. A good example is the local distribution of temperature and liquid water cross sections. These have been measured in situ by different methods, primarily in DOE projects by neutron imaging. These modeling cross sections should be validated by actual experimental data—if not from this project, from data collected by other projects. Model validation needs to be undertaken for both the cross-section modeling and the areal "water film thickness" model as opposed to validation of water concentrations by segmented cell.
- Generally it appears that progress has been made, particularly in the membrane arena, but the situation is confused somewhat by presentation or technical backup slides that are very similar or identical to the previous year but still utilize the terms "new" or "achievements" in the title. While EODC measurements in general are contentious, it is not convincing that this technique is superior, only different. Progress toward the technical targets is not addressed, nor is the relevance of much of the work. For example, start-up is listed as both a barrier and a technical target, but there is no obvious tie-in with the work presented.
- It is unclear what the overall objective of this project is. It is unclear what the benefit is of membranes with more or less diffusivity or EODC in operating fuel cell systems. The researchers do show a polarization curve with higher performance due to less water transport and therefore less flooding of the cathode in high relative humidity (RH) conditions; however, this is in an atypical $H_2$/air co-flow orientation of the gas channels. Typical PFSA-based membrane fuel cell systems take advantage of the high water transport and run counter-flow gas channels for air and $H_2$; the researchers did not report on this more typical flow channel orientation. Furthermore, in the more typical low RH operating regime, the low water transport of the new Virginia Tech membrane shows a lower polarization curve, making it unclear if high-water-transport membranes or low-water-transport membranes lead to a better fuel cell system.

### Question 3: Collaboration and coordination with other institutions

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

- This project benefits from a set of strong contributors that provide the majority of the effort.
- The project features good collaborations with university and other industrial partners.
- This project features a good team that involves organizations with complementary skills. The roles of the team members are well defined.

- The project appears to have a good degree of integration.
- This project features collaborators from industry and academia, as well as clear task separation that leverages the strength of these institutions. It also features the exchange of information and materials required to achieve technical progress. The project and collaborations appear to be managed successfully. The contribution from Tech-Etch remains unclear.
- The collaboration with Virginia Tech and the University of South Carolina (USC) appears to be working well. Some automotive industry input may be useful.
- The team has a reasonably broad group of collaborators, including leaders in the areas of polymer synthesis and membrane characterization. The modeling work being performed by the USC team has little other background in this area beyond this project.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- This project relates directly to DOE's needs for understanding and coming up with design strategies for improving mass transfer in next-generation fuel cell components.
- The project's relevance to the overall program is difficult to assess in terms of the value to the fuel cell industry. The fundamental learnings from component studies will no doubt be useful to researchers in the field, which may indirectly result in progress in fuel cells.
- The project indirectly supports a subset of the DOE objectives. The strongest connection is performance, particularly at high current density.
- The understanding and development of membranes that alleviate water management challenges are of significant importance to the field. As the modeling results are similar to those previously observed or modeled, they are unlikely to benefit fuel cell developers.
- No agreement exists regarding the fundamental parameters used in fuel cell modeling. An agreement would be very helpful for meaningful model development with respect to fuel cell performance and water balance. This work thus contributes to developing the capability to engineer/design materials that have specific transport properties for optimized fuel cell performance and efficiency.
- It is unclear how this project will actually be used by industrial developers. There are questions about whether the model is going to be distributed and how the data will be utilized by developers.
- The barriers claimed to be addressed are performance, water transport in the stack, water management, and start-up/shutdown. It is not clear how this work affects any of these barriers. Perhaps a better study would be water transport studies of the diffusion media rather than the membrane; the researchers do recommend that in their future work, but there is not much time left in the project.

### Question 5: Proposed future work

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

- The future work plans seem to be a reasonable continuation of the current activities.
- There is a reasonable amount of future work for the remaining time period. A comparison of nanostructured thin-film (NSTF) layers to conventional catalyst layers would help researchers understand the feasibility of an NSTF catalyst. Optimizing GDL properties for an NSTF-type catalyst may be the only way to overcome the flooding issues of these catalyst layers.
- Now that the membrane water transport is well understood, the study of the water transport in NSTF electrodes (which are known to be challenged by good water management) and the diffusion media (DM) (where there is currently not much good understanding of the water transport mechanisms) is good future work.
- The purpose of the "Emphasis on Saturated Conditions" is unclear; this is clearly the opposite of what the automotive original equipment manufacturers are doing. Examining flooding of thin catalyst layers (such as NSTF) could be highly useful.
- The future work should probably be limited to membranes, diffusion media, and water management, rather than catalyst effects, so that the scope is less broad and more focused on the strengths.

- The future work focuses on studying saturated conditions (whose primary relevance is only during start-up for automotive applications) and electrode effects. In the area of electrode effects, it is unclear how the researchers plan to approach the areas of high current density.
- Investigating flooding of NSTF layers is proposed in the future work. While important, such an effort is a bit of a departure from the current efforts and is quite challenging considering the time remaining on the project.

### Project strengths:

- This project features a good, sound approach that addresses many aspects of transport in PEMFCs.
- This project features good experimental methods to measure fundamental material properties, in addition to a good model with good correlation to measured data.
- This project's strengths include its novel polymer electrolyte synthesis and novel water transport studies.
- The membrane development portion of the effort provides the most potential value to the fuel cell community.
- The approach is the main strength of the project. By widely varying the transport and structural properties of fuel cell components (mainly the membrane), this project seeks to come up with guiding design principles for fuel cell membrane development and maybe other components.
- The progress in membrane design and characterization and transport modeling appears to be reasonable, based on additional reading of the published literature in *ECS Transactions*. The collaboration with Virginia Tech and USC appears to be working well. Some automotive industry input may be of further use.

### Project weaknesses:

- The team needs to better leverage the different project facets (membrane synthesis, water transport, and modeling).
- A poor case is made for the relevance of the various efforts to the "technical targets" and the "barriers to be addressed" listed up front.
- Additional in situ validation techniques could be used. The level of work going on in this project related to hydrocarbon membranes is unclear, as the project is a transport project.
- There is not enough connection made to real fuel cell systems. In addition, it is unclear where the advancement in performance, cost, reliability, and durability comes from for membranes with higher or lower water transport numbers.
- Although the model data seem to predict the overall performance values well, the local performance data shown in the two-dimensional plots of temperature and current look visually discrepant between the model and experimental data. In addition, there are no comparisons to the neutron measurements of water production from www.pemfcdata.org. The model must be able to predict the measured values from neutron radiography as well.

### Recommendations for additions/deletions to project scope:

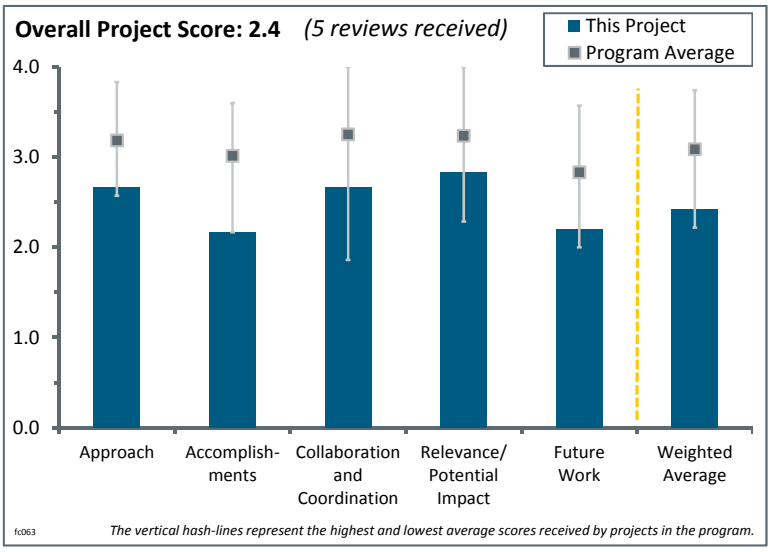
- Start-up is listed in the barriers/technical targets and has been mentioned in the past. Correspondingly, some of the future work should be directly linked to start-up.
- With little membrane work in the current portfolio, the membrane studies bring value. The value of models being developed seems small and does not seem to have a clear path forward; it is recommended that the project cease the modeling work.
- The researchers are using segmented cell distribution measurements as validation of their modeling of water and water flux. These data are at best an "inference." These water concentrations should be verified directly. There are developed techniques to measure the water concentration in situ; for example, the BT-2 beam line at the National Institute of Standards and Technology is funded by DOE/U.S. Department of Transportation as a user facility. The value of the modeling of the hydrocarbon membranes in terms of temperature and liquid water content is unclear, unless a developer is actually exploring using these materials.
- There are no recommendations because the project ends this year.

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

David Mountz; Arkema

### Brief Summary of Project:

The objectives of this project are to (1) develop a membrane technology having low methanol crossover, high conductivity, and increased durability; (2) develop cathode catalysts that can operate with considerably reduced platinum (Pt) loading and improved methanol tolerance; and (3) combine the cathode catalyst and membrane into a membrane electrode assembly (MEA) achieving performance of at least 150 mW/cm$^2$ at 0.4 V and costing less than $0.80/W for the two components.




### Question 1: Approach to performing the work

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

- The project team has been thoughtful in eliminating unpromising avenues and staying focused on avenues with promise to meet U.S. Department of Energy (DOE) targets. The focus on combinations of conductivity/MeOH permeability, and on MEA development and diagnostics, are appropriate and wise.
- This project aims to develop a low-cost MEA that has a better-performing direct methanol fuel cell (DMFC) membrane (lower methanol crossover, high conductivity, and increased durability). It also aims to develop a cathode catalyst that can operate with considerably reduced Pt loading and improved methanol tolerance. These are the right issues to address for DMFCs. However, the membrane technology is not significantly new, and the methanol tolerance catalyst was a no-go.
- The main purpose of avoiding methanol crossover seems to be a difficult task, although some progress has been reported. Generation 2 membranes have the same problems reported in the previous year (leaching of sulfur). The use of cross-linking agents did not help. Other approaches (increasing the polyelectrolyte molecular weight) failed, too. Perhaps some modeling would help.
- The team took the right decision of stopping the cathode development approach, which was not effective. Using a commercial catalyst and GDE for conducting membrane performance and durability assessments is the correct approach. However, the performance and durability test protocols being used by the team are very soft and not adequate for predicting material behavior in true DMFC power system operational conditions. The team should consult with DMFC stack/system builders or scientists from a national laboratory to devise correct performance and durability test protocols for conducting performance and durability diagnostics.
- The project focuses on improving DMFC performance and cost through the use of blend membranes and the development of Pd-based catalysts. The Pd-based catalysts have not shown promise, nor is it clear why it was believed that they would show promise in the heavily investigated area of methanol oxidation. The development of blend membranes offers the potential of decoupling mechanical and transport properties; however, efforts from the team lead over the past several years have not suggested that the proposed approach will result in durable and/or improved performance.
- The targets on slide 4 should be set in units that measure the bulk material properties, not in units that depend on membrane thickness. For example, bulk resistivity should be used for material resistivity, not areal resistance. For perfluorosulfonic acid (PFSA) membranes, the industry standard would become 6.3 Ohm-cm instead of 0.12 Ohm-cm$^2$, and the proposed Generation 1 membrane with a 1.2 mil thickness and an areal resistance of 0.03 Ohm-cm would have a bulk resistivity of 10 Ohm-cm. Thus, the Generation 1 material does not have a lower bulk resistivity as compared to PFSA; the researchers have simply reduced

the membrane thickness to meet the target. This is furthermore evident when the researchers compare the performance of a 2-mil PFSA and a 1.2-mil-thick Generation 1 membrane that has essentially the same areal resistance and thus the same polarization curve. Thus, there has essentially been no advancement in the state of the art. The approach should therefore be more focused on the bulk material properties of a new material that has better bulk material properties as compared to industry standards. A similar approach should be done with methanol permeability.

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

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

- Progress has been made on a new material that seems to behave similarly to industry-standard PFSA.
- Only the membrane work remains from the original goals, and there are still some obstacles to achieve the desired performance.
- Overall the progress is good, but it appears to be stalled. Generation 2 membranes are not panning out; Generation 1 membranes have been optimized and perform fairly well, but this is relatively old work. The MEA work is sensible and necessary, but it is not very creative in and of itself. The diagnostic work on MEAs will be helpful in understanding failure modes.
- PFSA MEAs presently outperform Arkema MEAs, both in terms of performance and durability. The durability milestone had to be lowered and is awaiting a status update. Interfacial resistance is a common major issue when working with a new membrane. The team has not been able to resolve this issue or have a good understanding of it. The team has made good progress in lowering catalyst loading.
- The objectives of membrane development (Task 1) were not accomplished because of the ionomer instability in the membrane under DMFC conditions. The blending of an ionically conductive polyelectrolyte with a PVDF host matrix did not produce a workable membrane because of the high solubility of the polyelectrolyte. The team should have focused more on developing a stable polyelectrolyte to achieve the objectives of Task 1. The evaluation of the MEA was done under an extremely high air stoich, which is inappropriate for the portable DMFC condition. The team should conduct its testing at an air stoich of 2.0, which is closer to that preferred in portable DMFC systems. The team should test its MEA at a higher temperature (80°C) and a lower air stoich (<2.0) to understand the possibility of using its MEA in a DMFC power system. The 2-mil PFSA, which is very stable, also meets the Milestone #5 target. The developmental PVDF/polyelectrolyte membrane does not offer much better performance than the commercial 2-mil PFSA membrane.
- The progress toward DOE goals for portable power has been minimal. The fuel cell performance shows modest performance gains compared to industry-standard values at fixed operating conditions, but it is unclear that this value represents the state of the art, and other published performance values for DMFCs seem to be higher than the presented values. One of the biggest concerns is the low stability/durability of the different membrane approaches employed (one of which was down-selected owing to extremely poor stability). It is not clear that the properties of the blend membranes employed can meet those of state-of-the-art hydrocarbon membranes, even in initial properties performance, and even if they do, they do not appear to be stable. The properties reported—namely areal resistance and permeability—make comparisons between membranes difficult; conductivity and permeability or methanol crossover and resistance would be more appropriate properties for comparison in order to ensure that the thickness effects do not confuse data interpretation.

### Question 3: Collaboration and coordination with other institutions

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

- The lead institution has been proactive in finding the right partners and closing things off with partners that were working in unpromising directions. This is commendable.
- The team consists of good partners, including a university and a catalyst company. However, inclusion or consultation with a national laboratory or DMFC portable power manufacturer could help the team to test and evaluate its membrane/MEA under realistic DMFC operational conditions.

- Most of the work is done at Arkema. There is some collaboration with the Illinois Institute of Technology (IIT), Quantum Sphere, and IRD Fuel Cells, LLC.
- Most of the progress seems to be achieved by Arkema; it is unclear if this is because of the lack of coordination with the partners. The progress is really slow. The team was not able to find an appropriate membrane formulation for the problem of leaching, and the other membrane has a higher permeability than the target (even when the target permeability has been increased from last year).
- A little collaboration exists, but there does not seem to be much synergy or strength in the interactions. IIT no longer contributes to the project, and the Pd catalysis work was tangential to the membrane work. The outsourcing of MEA fabrication and testing does not appear to have much synergistic value and appears to be primarily contract work. Increased interaction with partners that have alternate membrane DMFC testing and MEA fabrication would be beneficial.
- Because this project should be a fundamental material improvement project rather than an MEA project, the project could benefit from university collaborations that help with the fundamentals of the membrane materials and further processing of the membrane materials to improve bulk properties.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- This project aims to increase the performance and durability of DMFCs and reduce cost, which are both relevant to the DOE Hydrogen and Fuel Cells Program (the Program). Its approach to doing this is to develop a better and lower-cost DMFC membrane and cathode catalyst. These are the relevant objectives to pursue with respect to DMFCs and DOE targets.
- The proposed goals are aligned with the Program. However, the slow progress shown so far creates some doubts about the potential impact of this project.
- The project lead is certainly aware of DOE goals and is evaluating project work against them. Progress has been slow, but the team is at least being honest about its findings.
- The project is relevant to the objectives of DOE's *Fuel Cell Technologies Office Multi-Year Research, Development and Demonstration Plan*. The activities are aligned with DOE's goal. This project is focused on low-crossover membrane and MEA for DMFC application, which is very important for the commercialization of DMFC technology. The focus to accomplish the performance and cost objectives is aligned with the fuel cell initiative of DOE. However, the feasibility of the concept is not very clear.
- The project focuses on the cost and performance barriers for portable power. The primary (remaining) focus has been on novel membranes and MEA integration. Issues of catalyst cost—a primary driver for these systems—are no longer being addressed within the project. Meeting system cost goals through only membrane improvements is not likely.
- The research should be focused on a new membrane material that has better conductivity, lower MeOH crossover, and good stability as compared to standard PFSA materials. Pursuing lower-cost membranes compared to PFSA membranes should not be a driver in the membrane development in this project because the amount of precious metals applied in the standard DMFC electrodes is the main cost factor per square meter of the MEAs, not the industry standard of 2-mil PFSA materials. Lower-cost DMFC MEAs should focus on lowering the platinum group metal (PGM) loading of the electrodes.

### Question 5: Proposed future work

This project was rated **2.2** for its proposed future work.

- The project is wrapping up soon; the proposed future goals are sensible and appropriate.
- The future work described is aligned with the proposed work of the project. The team needs to work with realistic testing protocols and avoid using soft testing methods to achieve performance/durability targets. It should use practical test conditions, which are needed to make DMFC systems commercially viable. Using a high air stoich will lead to high parasitic loss and hence loss in the output power of the system.

- The proposed future work looks reasonable, but the major issue of higher resistance does not seem to be addressed. Post-mortem analysis will help, but the researchers need to diagnose the problem in order to improve the durability.
- The proposed approach for the dissolution issue in the Generation 2 membranes is complex and may not lead to the expected results. Thus, the overall success of this project is limited.
- The future work focuses on more crosslinking studies, MEA optimization, and some post-mortem testing; it has little relevance toward eliminating barriers or advancing the Program.
- The project has essentially ended (June 30, 2013).

### Project strengths:

- A strength of this project is having an industry lead with the capability to develop new materials.
- The project team has vast experience with PVDF, which is a potentially good candidate material in ion-conducting membranes.
- The team is well organized and capable of developing DMFC membranes and MEAs. The team is equipped with the necessary resources required for the success of this project.
- Prospects for making very low-cost membranes that also exhibit high performance were good. The approach to testing their ideas is good.
- Membrane development is the strength of this project. However, the researchers have encountered many difficulties.
- The project's strength is in membrane development, but the new membrane is not significantly better than the PFSA membrane.

### Project weaknesses:

- There were poor advances in materials in both the membrane and catalysis areas.
- The Generation 2 membranes and proposed low-PGM catalysts did not work out.
- Durability tests of the MEA are complicated owing to the uncertainties associated with commercial electrodes. The team did not correctly select the appropriate partners or failed in the overall coordination.
- The team could have benefited from consulting with a national laboratory or DMFC company to determine an adequate testing procedure for the DMFC. The team also needs to move quickly to consolidate on the final membrane and MEA structures to facilitate initiation of durability testing.
- The project objectives and approach are good, but the execution and final product were not delivered. The catalyst and MEA development/expertise were lacking.
- Too much time was spent on making MEAs on materials with essentially similar properties to PFSA materials.

### Recommendations for additions/deletions to project scope:

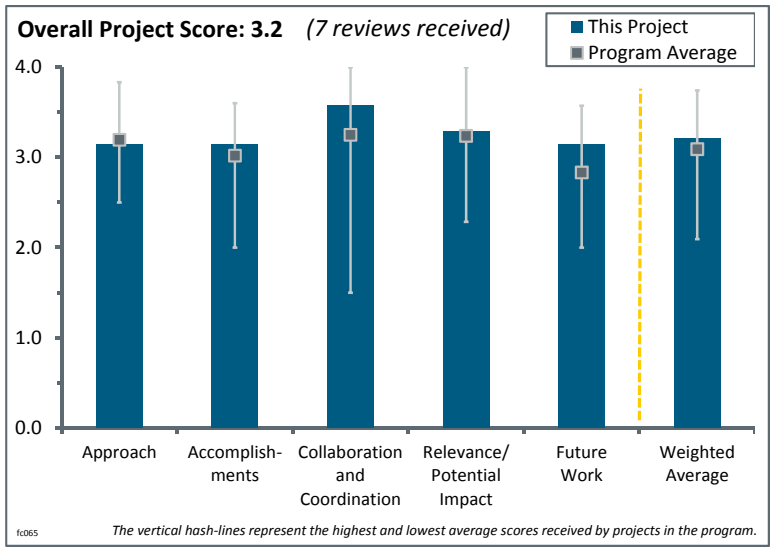
- Because the project is close to completion, there should not be any deletions at this stage.
- The project is ending soon; plans for closeout are appropriate.
- Given the team's strength in membrane development and the time left in the project, the team should focus solely on Tasks 1 and 4. The team should drop Task 3, especially the goal of 150 $mW/cm^2$ at 0.4 V. This goal seems to be a long shot, and presently there is no indication that the goal is achievable. By dropping Task 3, the team will have more time to complete the other two tasks—Task 1 and Task 4.
- DOE has funded for several years the study of blended membranes for improved fuel cell performance. If anything, it appears that the stability of these materials has gotten worse. Further investigation of PVDF blend membranes should be stopped because of the poor properties and advances, despite the reasonable levels of investment to date.

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

Jean St-Pierre; Hawai'i Natural Energy Institute

### Brief Summary of Project:

The objective of this project is to identify and mitigate the airborne contaminants that are adversely impacting automotive fuel cell system performance and durability. The University of Hawai'i (UH) identifies contaminants and determines tolerance limits for filter specifications. The project supports recovery of the fuel cell system after exposure to contaminants by identifying fuel cell stack material, design, operation, and maintenance changes to remove contaminant species and recover performance. The 2012–2013 objectives are to establish degradation mechanisms and quantify performance loss for key contaminants to assist mitigation.




### Question 1: Approach to performing the work

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

- The project is well designed and starts with a list of contaminants, then prioritizes them, and studies their effects at various operating conditions.
- The project's focus on air contaminants is an important task, and the identification of potential contaminants will help fuel cell developers in their selection and the formulation of electrode catalysts.
- The approach is good. The principal investigators (PIs) are using the available data from the field to establish the most common contaminants in the air and the impact of these contaminants on performance and durability.
- The approach is good, straightforward, and based on down selecting from various contaminants. This project addressed most of the concerns from reviewers by studying the effect of down-selected airborne contaminants on voltage loss as a function of operating conditions. This project also addressed the effect of calcium sulfate from a de-icing agent on degradation.
- The approach is good because it used multiple, industrial sources for the selection of contaminants to evaluate. The approach attempts to identify how to recover the fuel cell system from contamination and quantify the performance impact.
- The approach is very reasonable for the evaluation of each contaminant; however, contaminants need to be studied according to their merit. The study has focused extensively on acetylene. This is a relatively exotic source of contamination. Cars will overwhelmingly never see this contaminant. The initial findings show a complete reversibility of contamination and exposure does not cause long-term impacts on the stack (as soon as the contamination is removed, the stack performance comes back). It may be worthwhile to prioritize all the contaminants of interest and to proceed quickly, studying them one by one.
- The combination of experimental determination of impurity effects and modeling is effective. The breakdown of impedance data to extract kinetic, ohmic, and mass transfer resistances is beneficial. This is mostly coordinated with past and present impurity studies to avoid the duplication of efforts. However, it is not clear how cation poisoning work differs from what has been done in the past. The use of low loaded catalysts (0.1 mg Pt/cm$^2$) is a major improvement. The addition of cleaning agents and coolants to the contaminants list is appropriate. The PI needs to work at impurity levels expected in the real world;

acetylene tests performed at 4,030 ppm are not relevant. Even in a welding shop, concentrations should not be this high. Likewise, propane concentrations of 1,000 ppm are quite high and not expected to be relevant.

### 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 demonstrated how to approach the identification and evaluation of air contaminants. The examples of "recovery" are interesting; however, many recovery protocols are well known in the industry.
- Significant progress has been made with respect to the impact of the contaminant on various sources of voltage loss. Further, the current distribution using a segmented cell sheds some light into the propagation of contaminants in a fuel cell system.
- This project has made good progress studying the effects of four contaminants. The performance loss results are supported by interpretations of an underlying mechanism. Tests with lower Pt loading are more relevant considering the direction of the fuel cell developers. The development of a filtering system is mentioned. The mitigation strategy is important.
- Steady and excellent progress has been made so far by identifying and down-selecting various contaminants that cause degradation in the performance of fuel cells. The detailed understanding of the mechanism should help to understand the effect of these various contaminants on different types of catalysts. Novel materials are being evaluated as cathode catalyst materials for fuel cells. These mechanisms would help to mitigate the effect of these contaminants on the newer electrocatalyst materials.
- The project continues to make good progress evaluating potential contaminants. It would be good to see data showing mitigation/restoration for 90% of cell performance for the seven contaminants that this has been completed for, and information on the mitigation/restoration strategies used for the contaminants. The step-change in effect of the $C_2H_2$ contaminant on voltage at a concentration of about 200 ppm needs some explanation and investigation to see if similar results occur at lower concentrations with lower catalyst loading.
- The team has thoroughly examined acetylene. It may be prudent to run parallel evaluation on multiple contaminants as that will provide technology developers with timely feedback regarding potential red flags. This testing should serve as an early warning system to alert manufacturers that certain contaminants need to be addressed in real-car designs. Acetylene appears to be a non-issue and it may have taken efforts away from more important contaminants.
- The team has built up a database of common contaminants with an explanation of the effect on performance and the ability to remove the impurity. It is not evident that the recovery of an acetylene contaminated cathode removed all of the acetylene or if the acetylene was oxidized during removal and left a carbon layer that reduced the active surface area. The researchers did not address this point and did not consider partial oxidation. There was no explanation of why the $H_2$ peroxide rate increased. It is not clear if specification sheets are being developed in this program, but they should be. The authors stated they would "quantify spatial variability of performance loss," but there is nothing indicating this in the accomplishments and progress. It was unclear if the cartoon in slide 15 was the proposed mechanism and if there was some explanation of the drawings.

### Question 3: Collaboration and coordination with other institutions

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

- UH has assembled a good team for collaboration.
- UH has sound collaboration with appropriate partners.
- The collaborations appear to be very well established and working effectively.
- This is a good team of researchers with many organizations contributing to the project. If the partners contributed to the presented experimental results, the credit is not evident from the slides.
- The coordination with other impurity projects is good. It appears to be getting input from most of the fuel cell developers. It is not clear how cation poisoning work differs from what has been done in the past.

- While the project has a reasonable list of collaborators, more stack manufacturers are needed to update the project on the catalysts used in systems that are either in field trials or being readied for commercial release.
- This group should work much more closely with car, bus, and forklift original equipment manufacturers (OEMs). Ballard is a great company to collaborate with; however, real-world contaminant concerns come in where the rubber meets the road in vehicles. OEMs should really be the ones expressing and prioritizing contaminants and methods of exposure.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The effects of contaminants must be determined for widespread commercialization.
- This study supports the performance durability of the fuel cells by looking at the airborne contaminants.
- This type of project is extremely important as it can act as an early warning system for OEMs regarding real-world exposure risks. Based on studies like this, OEMs can decide to ignore, monitor, or design system features to deal with key contaminants.
- This project has developed a useful protocol for evaluating the effects and mitigation of air contaminants on fuel cell operation. It will serve as a valuable model for the fuel cell industry with their internal evaluations in the future.
- Understanding the voltage loss from sources is good. The impact of the project could be higher if they provide ways of translating this data into a meaningful trade-off between the stack and system design like the specification for the air filter.
- The establishment of a contaminant database will benefit all fuel cell parties and provide a reference for future development activity. This codes and standards database is one of the many that will be needed for future fuel cell commercialization.
- Understanding the effect of airborne contaminants and contaminants from other possible sources on fuel cell performance is very critical and the objective of this project works to understand that. Degradation mechanism due to various contaminants would help mitigate the voltage loss at the system or material level.

### Question 5: Proposed future work

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

- The proposed future work is sound.
- The work plan is consistent with the project objectives. The effect of the remaining four species will be studied and mitigation strategies will be developed.
- The plans to perform tests with concentrations closer to atmospheric concentrations and with 0.1 mg Pt/cm$^2$ should be done sooner. The plans to look at contaminant mixtures are relevant and should be expedited.
- The project's future work proposal is well focused and aggressive. One possible improvement would be to "down-select" the possible catalyst systems and focus on those few that are most likely to be introduced in near-term commercial applications.
- The proposed future work is in line with the objective of the project; however, detailed understanding of the degradation mechanism would add lot of value to the project. Also, the effect of contaminants with longer exposure times and different concentrations for low-loading catalysts would provide great information for future fuel cell systems. The effect on non-platinum group metal (PGM) catalysts (if any) and ultralow PGM catalysts would be helpful to anticipate any future challenges for these materials.
- It would be beneficial to streamline such processes and address multiple contaminants in short periods of time. It would also be beneficial to begin to look at interactions of contaminants. Cocktails of contaminants are a traditional means of testing a system; for example, bulk diesel exhaust will be encountered by vehicles on a daily basis. It is unclear if that is acceptable. It would be good if the project could explain this more.

- The spatial variability of performance will be explained (March 2013); that should have been ready for the AMR. It was not clear if there was an explanation of the spatial variability mechanism. The research is addressing low catalyst loading at the end of the project, but it is unclear why this was not an early priority based on known DOE automotive targets.

### Project strengths:

- UH has excellent screening techniques.
- This project has strong collaborations and a well-thought-out approach for down-selecting contaminants.
- The capability of the researchers and the quality of the facilities strongly support the continued development of the contamination database.
- The project's participants have made significant contributions regarding the evaluation and analysis of the effects of air contaminants, including mitigation strategies.
- Tests have been conducted with lower Pt loading. Tests have been conducted to evaluate the effects of both contaminant concentration and exposure time. Performance loss mechanisms are being developed.

### Project weaknesses:

- There are too many contaminants to deal with.
- The contaminants' concentration tolerances need to be specified. A detailed understanding of the mechanism is lacking.
- The project's work scope may need adjustments to focus more on the established catalyst systems and less on the broad and ever-changing list of experimental catalysts that may never make it to market.
- Considering that bromomethane is identified as a "red" contaminant, it should receive a higher evaluation priority. The mitigation strategies for the tested species are yet to be developed. It is not clear how determining the spatial variability will help mitigate the problem.
- The team may need to focus on publishing data in a format easily utilized by industry. It is not clear what the spatial variability mechanism defines. It is unclear if this is this another way of saying the contaminant adsorbs on the surface of the catalyst and blocks sites for electrochemical reaction. It is unclear if this takes into account reactions at the surface.

### Recommendations for additions/deletions to project scope:

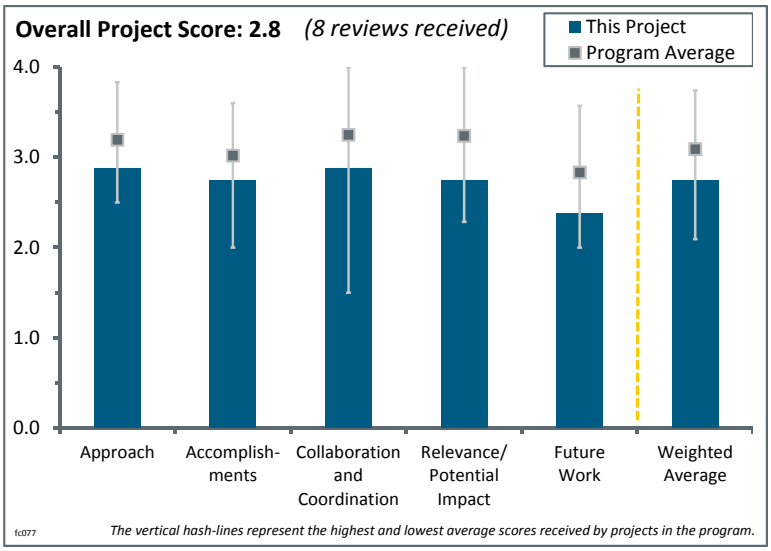
- The project's work scope may need to be re-focused and build on its accomplishments to date by placing more emphasis on near-term commercial systems.
- UH needs to decide what is a reasonable concentration of a given contaminant that might show up in the air stream. This team should classify the hazard of the contaminants based on the mechanism and the reasonable contaminant concentration. It is suggested that this project maintain the focus on the lower Pt loading.

## Project # FC-077: Large-Scale Testing, Demonstration, and Commercialization of Fuel Cell Coolant (SBIR Phase III)

Satish Mohapatra; Dynalene

### Brief Summary of Project:

The objectives of this project include understanding coolant durability requirements by working with different fuel cell types developed by various fuel cell manufacturers and understanding what the cost of coolant should be relative to current automotive coolants. Coolants should maintain low electrical conductivity and other thermo-physical properties for over 5,000 operating hours and should be compatible with a variety of typical fuel cell components.




### Question 1: Approach to performing the work

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

- The approach is effective and innovative.
- The approach is novel and, based on the data presented, effective at reducing ion contamination in fuel cell systems through a variety of means.
- The project's approach to developing an improved coolant for fuel cells is very good. The nano-sized particles for reducing ion concentrations are particularly interesting.
- This is a thorough and comprehensive approach, although experiments may lack consideration of the effects of direct contact of the coolant with the fuel cell system. It is unclear what the contaminating effect of the coolant is when it is in contact with the fuel or oxidant feed streams.
- This activity is not complicated. The project developed a heat transfer fluid and then will test that fluid and see if the results suggest it has economic viability.
- The approach is limited by the fact that long-term testing is really needed to determine its value; however, reliable, short-term tests are not available to extrapolate value.
- The approach lacks clear metrics for achieving project objectives and addresses only a small scope of the potential solutions. There is no evidence of "large-scale testing" as indicated in the project title.
- The use of the fluid overcomes the barriers of other fluids. Low conductivity and corrosion inhibitions appear possible. The particles remain suspended with surfactants. It is unclear if surfactants will degrade. It is not clear why the nano-particles themselves do not contribute to conductivity. The Dynalene fluid has several advantages over current fluids including no de-ionizing filter, lower pressure drop (smaller pumps), higher performance, lower cost, and no clogging.

### 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.

- This project has shown good progress towards meeting the DOE requirements for fuel cell coolants.
- A substantial amount of work has been achieved, but questions remain around colloid stability over time.
- This project has been running through its various phases since 2004. There is a good probability that this project will result in a commercial fuel cell coolant product that is superior to what is presently available.

- This project developed a clear understanding of what type of materials these coolants can be used with. Unfortunately, the materials do not seem to include structural or other plastic materials, which may be the materials of choice for automotive applications due to weight and cost considerations.
- The project addresses a clear need for applicable coolants able to meet the requirements of a PEM) fuel cell. The improvements in system performance, cost, and durability are likely secondary in nature. There are existing solutions in use by many original equipment manufacturers (OEMs), such as DI filter beds. This project will result in the incremental improvement of fuel cell systems, but it is not as groundbreaking as more fundamental work on catalysts, gas diffusion layers (GDLs), and membrane technology.
- The contractor has scaled up 100 L, developed quality assurance/quality control methods, and reduced final fluid cost. Also, the 100-hour test with Protenex is positive and had no increase in conductivity. The fuel cell coolant optimization and scale-up has been completed and Dynalene is capable of producing Dynalene fuel cell coolant in large quantities. It is not certain that conductivity and corrosion tests are conclusive to date; the testing times seem short.
- This project has no clear, relevant quantitative results. The principal investigator referenced about 5,000 hours, but the presentation only included results of 100+ hours. A lot of pictures were shown without a clear explanation of their relevance (e.g., corrosion samples). The presentation creates some doubts about the relevant results and progress. Using nano-particles to avoid corrosion seems to be more of a hypothesis than a real possibility, particularly when it comes to long-term durability and stability of suspension of the coolant liquid.
- The selected fluid, a solution of glycols (this fluid is not described, but one can guess it is propylene glycol and distilled water), is very similar to the most commonly used heat transfer material in the millions of domestic hot water heaters that decorate roofs globally. The engineering requires that fuel cell high-temperature fluids need to also have very low electronic conductivity, unlike the usual heat transfer applications. This requirement results from the fact that fuel cell stacks must be cooled; however, the voltage of the stack increases from plate to plate and the heat transfer fluid addresses each conducting plate. A conducting fluid would short the plates, which is not a good result. This proposal includes ionic scavengers, chemicals that absorb cations, and others that absorb anions. These scavengers are micro particles and are clearly insoluble particles that are entrained in the water-glycol mixture. It appears that progress has been made. The presentation suggests an emphasis on the scavenging, and perhaps that is the only real value.

### Question 3: Collaboration and coordination with other institutions

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

- The collaborations have improved and are good.
- It would be nice to see this coolant tested in more fuel cell systems than just Protonex.
- The collaborators are capable and certainly contribute to the project's progress.
- The contractor collaborates with Protonex, the Naval Research Laboratories, the University of Tennessee – Knoxville, and Lehigh University.
- The group assembled for the given task is satisfactory. There were again only vague statements and having more than 15 fuel cell companies does not really say anything.
- The collaboration could be stronger. It would be good to see high-voltage testing in relevant automotive systems. This could be ongoing, but is proprietary.
- This project used the university collaborator for determining the effects of using the coolant on the fuel cell. The partner from the fuel cell industry is providing a system for long-term tests and other companies will field test the developed coolant. It is unclear if information will be relayed back to project.
- The collaboration appears to be more like a vendor-buyer relationship. The small-scale fuel cell testing is not convincing. Obviously the hot fluid needs to be compatible with a specific set of polymers, metals, coatings, gaskets, pumps, etc. What works in one fuel cell design may not be useful in another.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- As long as there are no quantitative results, the impact is not clear.
- Coolant performance is important in system performance and maintenance.
- The project is certainly relevant to the success of both automotive and stationary fuel cell technology.
- An improved coolant for fuel cell systems is supportive of the Hydrogen and Fuel Cells Program's goals.
- This project is of high interest to the fuel cell community. This interest is also indicated by the amount of companies that test the coolants.
- The relevance and impact are moderated by the fact that coolants exist. This project improves on the performance metrics of the available coolants and so it is an important achievement.
- The project will support the goals of cost reduction and durability. The impact on performance is not clearly demonstrated. It would be useful to see fuel cell voltage degradation rates over time with Dynalene and a DI water control.
- It seems like there are reliable and useful fuel cell stack coolants around. Many fuel cell vehicles have been manufactured and each has a useful thermal management system. Obviously, a lower cost coolant has the advantage; however, if a component has a small cost, it does not save significant money by making that small part cheaper.

### Question 5: Proposed future work

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

- The completion plan is acceptable.
- The project ends at the end of fiscal year 2013.
- To complete only some of the work is not really a good plan; the project needs to achieve its goals.
- The project is about to end; a focus on long-term testing results is critical and will determine overall value.
- More work on the effects of polymer-based materials that are of interest to the automotive industry would create a significant value increase in the project with respect to automotive applications.
- The future seems to be "ship product," which is obviously the goal. There must be some appreciation about the competition, and some need to think about how this work could result in a strategic commercial advantage.
- The future work will validate corrosion inhibitors in a 5,000-hour test, increase anionic particle surface charge to match cationic at 500 μeq/g, and include compatibility and thermal degradation studies at temperatures exceeding 100°C. This project will perform long-term testing of final coolant formulation in three separate fuel cell systems.
- The future work focusing on 100°C testing is important; however, performance should be tested at higher voltages. Given that the fluid uses ionic nano-particles as its primary mechanism for controlling conductivity, it would be good to see how this fluid stands up to voltages around 100 V DC, which may be more common in automotive fuel cell power plants. It would also be good to see full-scale field testing, which is a more accurate determiner of actual fluid performance in the market than the limited lab-scale testing currently underway.

### Project strengths:

- This project is very relevant to fuel cell operations
- The innovative use of nanoparticles for improved coolant is a strength.
- This project is sharply focused on the development of an improved coolant for fuel cell systems.
- This project has a novel approach that can provide modest improvements in fuel cell system cost and possibly durability.
- The company running this project is leveraging its experience with coolants for developing a fuel-cell-specific coolant. This work appears to benefit strongly from the experience of the company.

- This is a commercialization project, which is not a bad thing. Getting a reliable solution for durable coolants certainly has merit.

### Project weaknesses:

- The long-term tests have not yet been completed.
- It is unclear how much collaboration with automotive OEMs has been performed in this project. Using these coolants with light and cheap non-metal materials may be key for automotive applications, but to date they are questionable at best.
- There is a lack of full-scale field testing of the fluid in relevant systems. Also, the presentation claims that the fluid can be non-toxic, but the health effects of many nano-particles are unknown and the claims of non-toxicity should be validated prior to being promulgated further.
- Some real technical questions remain with respect to colloid stability over time. The tests should be run under load as well. The effect of contaminants, such as di- and tri-valent ions, is unknown other than assuming that any corrosion of the metal plates would have delivered contaminating ions. This assumption may be true, but the purposeful addition of ions would have determined boundary limits since almost all colloids are ionic-strength sensitive.
- The necessary metrics for a good heat transfer fluid must include no plugging of the flow path within the radiator and no fouling of the heat exchanger (HEX) surfaces, especially by putting a thermal barrier on the HEX surface. The downside here is all about the wisdom of pumping the micro particles through the fuel cell stack. Small particles can agglomerate and it seems like this approach adds risk to achieving the reliability demanded by the targeted market. Certainly, fouling in a specific stack involves an interaction between that fluid and the stack components. This type of consideration appears to be missing.

### Recommendations for additions/deletions to project scope:

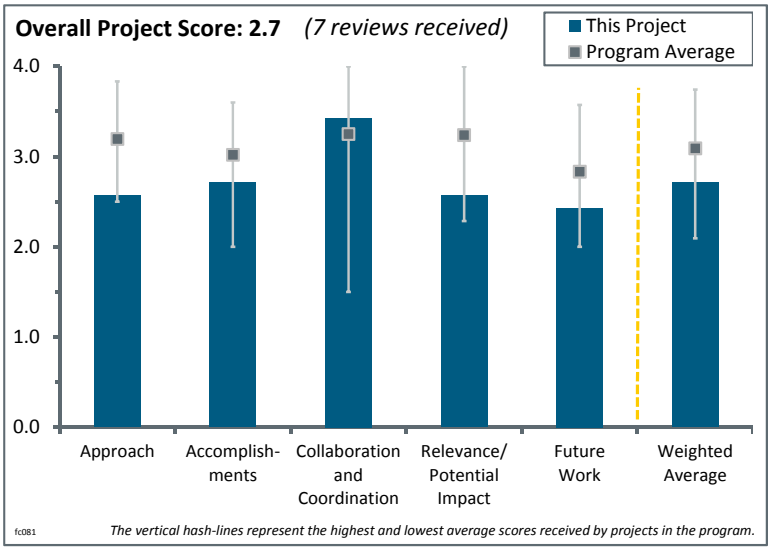
- Three reviewers commented that they had no recommendations as the project is ending.
- This project should continue to work to its conclusion. Temperature has a strong influence over the results.
- Future research should consider full-scale field testing at high voltages in a relevant system.
- The researchers should work to understand the effect of the coolants on structural plastics and other materials that may be used in automotive fuel cell systems.
- This project uses biofuels and glycols synthesized from a biological starting material. Although it is admirable to use these products, they add some uncertainty to the results. Biological products are derived from available feed stocks, and they tend to be variable. This approach is rather unlike using high-purity methane as a starting point for glycols derived from a chemical plant. It is certainly true that the two products are similar. Even so, there are two formulations in sight. There is the chemical glycol with additives and the bio glycol with additives. It makes sense to first test with a glycol formulation that has assured quality and then repeat the test with the new bio-sourced organics. Most likely the tests will give similar results. However, for now there can be some question. There should be some thought given to using a stationary “filter” sort of scavenger hardware, an ion exchange sort of device that remains outside of the stack. Certainly, if that were in-line with the principal flow, there would be a pressure drop. However, the entire heat transfer volume is being pumped around; therefore, all of it will be contacted by a well-placed, impurity-catching gadget. This gadget could be positioned in a side arm, a fraction of the total flow, or it could be placed in the reservoir tank. Such alternatives need to be considered. It is also important to test the concept on a stack designed for automotive applications. This hardware has extreme power density on a volume basis and necessarily has HEX components with small-gap heat exchanger flow paths. If things are going to plug up, these small slots will be the site for that precipitation. So, there will be a need to do coolant testing of prototypical hardware. Tests could be done on a “used” stack—one that is no longer making design voltage but has a complete thermal management system. This project should install an inline heater to heat the coolant and then pump that around for a few weeks. They should then address flows through each of the plates and show that no blockage occurred.

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

Jennifer Kurtz; National Renewable Energy Laboratory

### Brief Summary of Project:

This independent assessment of state-of-the-art fuel cell technology benchmarks the fuel cell durability status using uniform analysis and reporting methods to evaluate proprietary data from leading fuel cell developers. Data from developers is collected annually, analyzed, and compiled into two products: detailed data products, which contain confidential material and are shared with each contributor individually, and composite data products, which list aggregated data without revealing any proprietary information and are shared with the public.




### Question 1: Approach to performing the work

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

- This project seems well organized, well managed, and successful.
- The project offers the fuel cell industry an excellent methodology to evaluate fuel cell stack durability, even at the prototype stages of development.
- The approach to obtaining and analyzing the data is very good; there are considerable efforts pursued to access data and report out on the analysis using a variety of metrics. The areas of concern are identified by the researchers, and an additional effort could look into ways to address these concerns.
- The National Renewable Energy Laboratory's (NREL's) approach in this project is very good and clear. This project is reaching out to as many fuel cell developers across the world as possible to gather real-world data. One objective, studying the difference between lab and field data, is of prime importance to know the readiness of fuel cell technology and challenges. NREL is using already established tools for this data compilation and analysis, which is useful for getting the most out of the funding provided.
- NREL's approach to evaluate the data they are given from developers is reasonable. There are so many unknowns about the data sets and they cover such broad ranges of protocols that the value of the compiled data is small. It would be good to see them focus on gleaning information about other targeted metrics, such as power density loss or quarter power efficiency loss, in addition to the time to 10% voltage loss at a fixed current. Also, the steady-state data for automotive systems is irrelevant.
- The project has been shown to be responsive to the objectives of benchmarking the available technology. However, some of the fundamental characteristics of the fuel cell systems involved are missing from the analysis. Some examples of these characteristics include precious metal loading, membrane thickness, measures to maintain membrane durability (additives, reinforcements, etc.), operating conditions, carbon type, system mitigation strategies, and other characteristics. In terms of providing the U.S. Department of Energy (DOE) with status information, the project appears to work best for providing durability information. However, even this data still leaves its audience with an incomplete feeling. It is not clear what systems have reports on durability and what technologies those numbers represent. The project does some good in showing that there is a sincere difference between laboratory and field data, but questions still remain as far as which technologies might provide the greatest disparities between the laboratory and the field and why.
- The overall approach of this project is flawed. As currently constituted, it is difficult to understand how this work contributes to the barrier of addressing the durability of fuel cell stacks and systems for several

reasons. First, it combines different types of fuel cells (polymer electrolyte membrane [PEMFC] and solid oxide fuel cells [SOFC]) into one homogenous set of data. It is unclear how one can identify the current state of the art when all of the different fuel cells are combined into the same pot. Second, the operating conditions in the lab are much different from those in the field. The lab conditions can be closely controlled while the field operations are not. For example, the effects air contaminants encountered in the field have on the performance of the fuel cells is unclear. The effects will be different for different operational environments. A forklift operating at the factory door where a tractor trailer truck may be idling and emitting sulfur diesel exhaust that is being ingested by the fuel cell will be different from the same brand of forklift operating inside the bowels of the warehouse. It is unclear if the project's results are getting better. There was nothing in the slides that offer any conclusions that the results are getting better over time. This work should identify what areas need to be addressed. The desire to keep the identity of the different fuel cell suppliers secret may be self-defeating when trying to understand the current state of the art. This information needs to be segmented by fuel cell type. The bottom line is that is unclear what meaningful information is being identified in this project. It seems to be more of a modeling exercise rather than a search for specifics. Most (if not all) of the slides address modeling and not the outcomes. This effort is not useful.

### 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 data is starting to provide a view of fuel cell durability for a number of applications.
- If one accepts this reviewer's premise that the basic approach is flawed, it would be impossible to make meaningful accomplishments and progress.
- The project's data handling and statistical analysis of expected durability provides an excellent model for adoption by the fuel cell industry.
- In some ways, this is a "reporter" sort of effort. The daily reports come in, records are made, and the results are distributed on the Internet. It does not cost very much to do and makes for interesting reading.
- Excellent progress has been made so far. NREL or the principal investigators (PIs) have reached out to fuel cell providers outside the United States, which is very important. Sixteen data sets were gathered last year; this is great progress. This project is very transparent with the results and presentations by publishing them on the website, while also protecting the intellectual property of various data providers very well.
- The main objective of this project is to benchmark state-of-the-art fuel cell durability. There is no way to know whether the technology being analyzed is, in fact, state of the art. The project also does not directly track progress towards DOE's durability goals, as protocols are not disclosed. For example, if data for cells/stacks run was analyzed using the DOE automotive durability cycle, it would be much more informative.
- Unfortunately, the presentation was constrained to reporting what data sets exist, as opposed to the meaning that could be extracted from the data sets. For the data to be more meaningful to developers, NREL will have to be more aggressive about having data contributors be more open about material sets, Pt loading, carbon types, membranes, operating conditions, system mitigation, flow-field designs, etc. The volume of data generated was impressive along with the 99 different data products that the analysis conceived. Despite the amount of data, the data still suffers from a lack of details about systems.

### Question 3: Collaboration and coordination with other institutions

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

- The project has worked hard to obtain data from numerous entities.
- NREL has received data from 15 developers; without their collaboration there would be no project.
- The entire effort is based on collaboration, which involves input from the global community. The project team obviously operates a trusted procedure in which results are given, but those who generated those results are not identified. Such collaboration requires a perfect record in keeping identities protected, and that is what NREL is doing.

- The project does not exist without collaboration. Just to have the data that the project has, collaborative relationships had to be established. Some question exists as to whether the collaborations were able to sufficiently leverage the relationships. Since many material sets, cell designs, and operating condition parameters were left unknown, there is some uncertainty about whether the investigators were able to understand as much about the stacks and systems as they should have.
- While many potential collaborators (fuel cell manufacturers) were invited, few have participated, which is somewhat understandable given the competitive and confidential nature of an emerging industry.
- There have been many companies contacted to try to get more data. While the 15 entities that have shared the data represent 98 data sets, this data set is still quite limited. It is unclear if there are cost-effective ways to get more companies involved, either through reassurances or by offering incentives.
- The fact that 68 developers were contacted and the project was able to gather data from 15 developers is commendable. As this data sharing is voluntary, it is very necessary to reach out to as many developers as possible and the PI and NREL have done an excellent job collaborating.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The fuel cell activities supported by DOE clearly have commercialization as the goal. This project maps a part of that success. It seems essential and certainly relevant.
- There is some relevance. The main value of the project is to report public status numbers and to illustrate to DOE how the lab data is often different from vehicle data. For developers specifically, the project is not relevant. Progress towards meeting the program goals would proceed with or without the project.
- This project provides the needed durability benchmark status and progress concerning the emerging commercial applications of the fuel cell industry. The "third-party" expertise and analysis provides the necessary credibility to substantiate manufacturer's claims during the pre-commercial stages of introduction.
- It is very critical to know the readiness of the fuel cell technology and this project is helping a lot by collecting and comparing real-word or field data with lab data, which at times can be misleading to know the current real-world readiness of the technology. This information fits well and helps with DOE's Hydrogen and Fuel Cell Program's long-term goal.
- Fuel cell performance is going to be addressed by the individual fuel cell developers. The market competition is going to drive them to improve performance. DOE needs to analyze the data to see if there is some systemic issue common to all the developers that can be address at an overall programmatic level. The work done in this project does not do that.
- This project may provide some value for the developers who provide the individual data sets, assuming they do not have the capacity to do this analysis themselves, and help to assess the voltage degradation of their technologies. The compiled data is of limited value due to the unknowns and variations in the protocols, materials, and designs. Plus, this project only addresses voltage decay at high power. It may have more impact if it also addressed other failure modes, such as efficiency and power density loss.
- The data is highly relevant as it provides a view on the status of durability over a range of applications. This data is important for DOE to understand how the industry is progressing, and it is valuable for companies to benchmark themselves against others. However, some of the relevance may be lessened due to the inclusion of such a broad range of types of data (e.g., from accelerated to steady state and from short stack to system).

### Question 5: Proposed future work

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

- The future work proposed is to keep counting and reporting.
- The proposed future work is well thought-out and fills the gaps in current analysis. The system-level voltage degradation data for automotive seems to be minimal or missing. If NREL could get this information from original equipment manufacturers, it would really help with understanding the real-world

challenges. The proposed price information addition and other aging parameters for fuel cell durability would be great.

- The future plans, such as compiling results by test protocols and by current density (especially low-current density), are good ideas. Investigating start/stop and soak time is also a good idea. The plan to add price information may be valuable, but it is unclear how NREL will get and verify such information from its data providers.
- This project needs to be fundamentally redesigned to get meaningful results that will support systemic changes that can be supported by the fuel cells program. If this cannot be done, then the project should be discontinued.
- The project's proposed work effort seems to be too broad. Focus should remain on stacks and not expand to single cells. The benchmark data for PEMs, SOFCs, and direct methanol fuel cells (DMFCs) would be useful to the industry for "comparisons," but it may be difficult to get the interest of enough suppliers of SOFC and DMFC stacks to provide data.
- The future work as outlined is reasonable. However, the highest needs should be to 1) increase the number of data sets and 2) increase the comparability of the data. For example, including single-cell data will help the former, but at the expense of the latter. More effort should be put into adding additional companies so they can contribute data and add additional requested parameters to better compare data.
- The future work must attempt to convince developers to provide information about the characteristics of their systems. Without this information, there is little quality information that will come from the project to advance the Program toward its goals. In fact, without further information about system or stack characteristics, the project may serve to misinform the public or government officials in either a negative or positive light about the progress of fuel cell technology.

### Project strengths:

- NREL has outstanding analysis capabilities and access to a wide variety of durability data.
- The project's ability to analyze data and validate durability is excellent. This capability may develop into a "service" for those manufacturers unable to perform these evaluations in-house.
- This project has a strong team and a clear approach. The use of already developed NREL tools for the data compilation and analysis is a strength.
- This project has confidential data input and industry-relevant output, a non-biased view of achieved durability by application, and extensive data analysis.
- The NREL team is doing a good job and the strength is there. It is also apparent that this is a unique effort, and many people are reading the results. However, the NREL team is the core strength.
- This project has a vast array of collaboration. The information gathered has been treated with considerable analytical prowess. The project has been a public source for fuel cell durability information.

### Project weaknesses:

- This project's lack of shared details of data sets makes the compiled analysis of little value.
- This is a not a weakness but a limitation as only data is shared by the developers and not the material information or other details on understanding the mechanisms of degradation.
- The project needs some way of obtaining a material set and an operating condition and cell design context for all of its data products. The project is dependent, by definition, on information generated by others.
- The work is very important, but this reviewer is concerned that the confounding factors seriously impact the quality of the data set. It is understood that the data is what they get and they cannot influence that. It should be considered whether there is a way to adjust data for better comparisons.
- This is a flawed approach. This project is homogenizing the fuel cell performance of many types of fuel cells including large stacks, short stacks, SOFC, and PEMs with no attempt to analyze the data to determine what research and development needs to be done on a systemic basis.
- This is not necessarily a weakness, but more of a barrier to success: the extensive data requirements and confidential nature of the industry protects the material and system-design information and durability performance. Manufacturers will assume the responsibility to provide performance warranties and the expanding fuel cell applications will make comparisons (as currently done) more difficult. As mentioned during the review, "too many bins" (unique applications) will eventually dilute the comparison data.

- The information gathered, as in almost all reporting, is only as credible as the source. There are many reasons for distorting history. Bad data could be changed to "look good." There could be outstanding data, but it is undesirable to alert the competition before the project is ready and those numbers might be obscured. The data set is, in truth, very small, so credibility is an issue. The error bars tell the story, perhaps.

### Recommendations for additions/deletions to project scope:

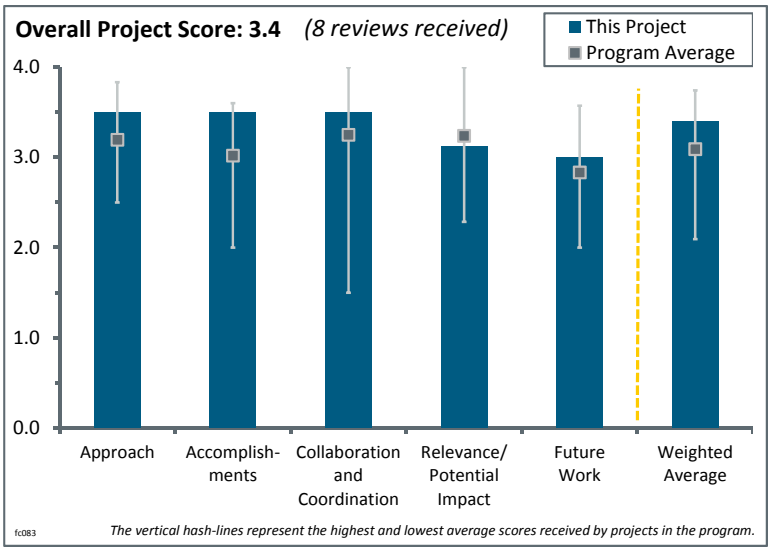
- This project needs to get background information on the stacks and systems it has analyzed.
- The investigators should totally overhaul this project to get meaningful results that address the barriers. If that cannot be accomplished, then the project is of little value.
- The project team should explore how a "service" might be provided to the industry, similar to what is done by others (e.g., Oak Ridge National Laboratory with clients for "characterizations of fuel cell materials"). This project should develop examples and publish protocols for the industry, including the relationship of voltage degradation as a function of operating parameters, etc., which is one of the items in the list of future work.
- This project should not include any steady-state data sets in the compiled automotive data; it should track the time to 10% quarter power efficiency loss and to 10% peak power density loss. This project should separate data sets with and without voltage recovery procedures and do a separate analysis for tests runs with the Fuel Cell Technical Team's recommended durability protocol. For automotive data sets, this project should run an analysis at maximum current points that meet the $Q/\Delta T$ requirement.
- It would make sense to have some large-scale durability tests conducted under DOE sponsorship and then have NREL include numbers derived from actual, well-defined hardware. That is, incorporate some data that is verified and serves as the "reference," perhaps several in each application class. There is no question that good data is verified data. It would also be interesting to collect some details about operational conditions, such as duty cycles, average current density, and operating temperature. This suggestion may not be possible, but it might make sense to try.
- Some data could be adjusted to account for various effects (e.g., model effects of current density on voltage degradation) and then use that to adjust the data. This project should then show data as both collected and adjusted data. The inclusion of the accelerated testing is of significant concern. Many accelerated tests are designed to fail within a few hundred hours or less. It is not clear how much acceleration is included. Following are some possible approaches : (a) when collecting accelerated data, the project should also request an acceleration factor and adjust the data accordingly, possibly with a large error bar; (b) the project should gather information on stressors and cycle times used in the accelerated data in order to adjust the data using known factors, although this would likely be more difficult in terms of data gathering. The inclusion of steady-state data is also a concern on the opposite end of the spectrum, but it at least shows the product capability given benign operating conditions.

# Project # FC-083: Enlarging Potential National Penetration for Stationary Fuel Cells through System Design Optimization

Chris Ainscough; National Renewable Energy Laboratory

## Brief Summary of Project:

The objective of this project is to build an open-source tool that helps combined heat and power (CHP) fuel cell developers, end users, and other stakeholders determine the appropriate sizing to reduce cost; integrate commercial building control and heating, ventilation, and air-conditioning (HVAC) systems to maximize durability; compare performance relative to incumbent technologies; determine optimum system configuration; and evaluate potential market penetration. These activities will help drive economies of scale and cost reduction in full-scale systems.




## Question 1: Approach to performing the work

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

- The approach to the model formulation in this project is excellent.
- To build a flexible, modular software framework that allows the addition of a wide variety of modules is a good approach. The cost and durability challenges are addressed.
- The approach taken in this project appears sound. It leverages existing models and information, works with organizations to access valuable databases and to improve those databases, works with original equipment manufacturers (OEMs), validates the model with existing data, and has a go/no-go step.
- This project incrementally improves upon the work done under previous years, which strengthens the confidence in this project. It directly targets the cost barrier by utilizing open source to allow multiple different users and suppliers to have some idea of the bottom line prior to initiating and allocating funding.
- The project has a solid, realistic, and unbiased approach. It exploits and analyzes a huge data volume and, thus, gets very good results. Analyzing 16 model buildings (even considering their vintages) and 16 climate zones is impressive and provides valuable local data. A web-based tool was realized and is helping local decision makers see if fuel cells will be of benefit. Guidance by a user group strengthens the project.
- The approach is solid and comprehensive. It is detailed enough to work as a planning tool, but lacks thorough benchmarking with competing technologies outside of the CHP sector (e.g., comparison to or in combination with heat pumps and better insulation).
- This project is well-focused on three critical barriers: cost, durability, and performance relative to the incumbent. In the presentation, the researchers make it clear how both of these barriers relate to DOE goals and how the research specifically addresses these barriers. While the approach addresses these barriers, the late and still somewhat limited outreach and coordination with other efforts raises a question about whether and how this work could be more effective.
- The project has included a very comprehensive survey of energy usage in buildings. This inclusion makes the study very realistic and relevant. The idea of the user group is pertinent. However, assuming a constant cost for fuel cell systems over a wide range of rating (100 kW–3 MW) may not be valid. The model is not flexible enough to incorporate technical aspects of individual developers: some can load follow better than others, some cannot load follow, etc.

### 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.

- Progress so far has been very good.
- Excellent progress has been made on model development. There should be more comparisons of the model to real-world operational stationary fuel cell systems.
- The progress within the scope described is very good for its comprehensiveness, detailed character, and applicability.
- The progress of the project and the additional measures taken to make the results even better are worthwhile. The interaction with other projects, particularly regarding cost, is very good.
- There has been good progress on the model and a significant database on data across the country and for many building types, as well as the addition of new control cases.
- This project currently has models for 400 kW phosphoric acid fuel cells (PAFC), 300 kW molten carbonate fuel cells, and the natural gas genset (as a control). It is developing models for polymer electric membranes (PEM) at 1, 5, 25, and 100 kW and for solid oxide fuel cells (SOFC). The movement toward cloud and open source computing is a good direction.
- This provides an excellent reference to past reviewers' comments that have strengthened the program and directly impacted the progress made this funding period. Getting Commercial Buildings Energy Consumption Survey (CBECS) inclusion for industry feedback was a critical success and should help validate model assumptions made thus far. Updating the model to include 15-minute increments that allow for thermal storage requirements is a great step. The means used to predict the storage and power needs of the system for the next day in order to reduce the thermal cycling of the fuel cell are interesting. It is unclear if there are predictive patterns or a connection to the National Weather Service.
- The researchers have made significant progress toward the project's objectives and have addressed most of their stated barriers. In particular, the effort to integrate with commercial building controls and HVAC systems to maximize durability could benefit from added efforts to reach outside of the National Renewable Energy Laboratory (NREL) and make use of resources at other national laboratories, such as Lawrence Berkley National Laboratory (LBNL) and Oak Ridge National Laboratory, and the emerging resources from the DOE Buildings Innovation Hub. Making use of these broader resources will provide the researchers with more opportunities for evaluating the reliability and overall performance of their scenario analysis.

### Question 3: Collaboration and coordination with other institutions

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

- The industry involvement has been broadened. The open source collaboration is an excellent tool.
- This project has very good collaboration with its project partners. User groups needs to be expanded and members need to provide feedback on the model' usefulness.
- The "consortium" is big enough to cover all topics and small enough to be efficient. The introduction of a user group adds value and the collaboration with other projects is well performed.
- This project leverages other DOE-funded work and the establishment of a user group is valuable. This project has a good collaboration with the University of California, Irvine (UCI). However, it is not clear how committed the user group is to the project or the amount and quality of the feedback that they will provide.
- Although there has been an effort to expand collaboration relative to last year's review, this area still needs some improvement. The presentation identified collaborations, but it did not provide clear evidence of whether and how these collaborations have benefited the approach and outcomes.
- Some important stationary fuel cell manufacturers are absent from the collaborations (e.g., Bloom Energy). It does not seem that there are very many commercial building organizations associated with the user group.
- This project has good collaborations with NREL's Electricity, Resources, and Building Systems Integration Center; UCI, which is subcontracted for controls and integration work; LBNL, which has a tie-in with a

separate DOE project (FC098) for manufacturing cost analyses; Strategic Analysis, Inc.; and Battelle. Stationary fuel cell OEMs are providing product data sheets and supporting information.
- This project is leveraging separately funded projects from DOE that have the information needed in this program to provide a much more efficient program and reduce duplicative work and wasted funding; this project makes good use in this area. Forming a user group is a critical step toward validating that the model is able to be customized for proprietary inputs and still provide useful data. A user group is an excellent use of collaboration since both this project and the user can benefit greatly once validation and refinement occur.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The analysis is important. It can have a high impact on implementation since it provides guidance for planning and might also be used to enhance the confidence of investors in stationary fuel cell systems.
- The project addresses some of the program's goals and objectives. However, the project does not address the critical issues of meeting the cost and reliability targets, as these issues are not within the project's scope.
- The project has a strong impact due to the many different cases investigated and thus provides valuable data for decision makers with regard to new buildings or even retrofitting old ones.
- The goals of this project are aligned well with the DOE Hydrogen and Fuel Cells Program (the Program), but the researchers seem to be working in isolation and have not planned ways to test and evaluate model performance through interactions with other researchers, other similar models, and a broader number of data sets.
- This project covers two major potential impact areas: (1) users, who by inputting grid information and building type, can see what type of fuel cell set-up is best for their application and get controls, HVAC, and cost analysis, and (2) developers, who can see how many users could be impacted prior to making investment decisions and committing funds. This is a versatile tool that can impact both sides of the technology used and can help avoid wasting funds.
- This project supports CHP application implementation by providing an analysis tool for fuel cell CHP developers. This analysis is currently a strong barrier for fuel cell developers, requiring access to data they might not be able to source and significant analysis time. This tool should enable a strong understanding of value propositions for fuel cells as a function of geographic location and use type.
- Stationary fuel cell manufacturers already consider the factors that the model summarizes into their business plans. They have relatively little need for a global model that evaluates their potential market penetration. The primary users of this model are companies that are trying to decide if fuel cell stationary power systems make sense for their organization. It is not clear what steps are being taken to "sell" this modeling activity to the user group. There is a need for such an evaluation model for business users who are seeking to determine if stationary fuel cell power makes sense for their commercial buildings.
- This project is highly relevant to deployment success. The tool is relevant and will be used to minimize lifecycle costs, lifetime greenhouse gas (GHG) emissions, or installed capital costs of fuel cell installations; characterize the largest segments of the U.S. building inventory for use in the tool, leveraging the CBECS; characterize building control systems and include in the tool, advanced control strategies for integrating fuel cell systems, and building control systems; validate the model outputs against real-world data from stationary fuel cell installations; and use the tool to determine the set of the most favorable system sizes and types to achieve national GHG emissions and energy-demand reductions.

### Question 5: Proposed future work

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

- This project has a clear scope of future work.
- This project has continuity between existing progress and future goals in most aspects.
- The proposed future work items are sound. Benchmarking to systems other than CHP are recommended.

- The proposed future work is straightforward with meaningful objectives. A decision for continuation should be taken.
- There is an upcoming go/no-go decision point on this project. The project seeks to develop participation in the user group.
- The future work was not discussed as it is contingent upon a go/no-go decision. The future work should include addressing other goals described in slide 3.
- While the investigators have included a validation step for the model, it is not clear how extensive this validation will be. They have not outlined risks associated with this validation. The inclusion of high-temperature PEM would be useful.
- In reference to Verizon, it is unclear if this project can utilize data from installed systems to validate the model as part of future work. As Europe and Japan have been in the CHP game for a long time, this program should set up models of European buildings, utilities, and rates to help validate the model's concepts on actual ongoing CHP systems.

### Project strengths:

- The modeling approach is comprehensive.
- This project has excellent collaboration and has created a user group.
- This project addresses issues that are important for the Program and includes visualization methods for viewing model outputs.
- The major strength is the detailed and comprehensive character of the project. It has the potential to be used as a realistic planning tool.
- The development of an open source tool that is applicable to multiple fuel cell developers is a strength of this project. This project also considers realistic and comprehensive data for electricity usage in distributed generation.
- This project should result in a valuable tool to model the applicability of fuel cell CHP systems for various building types around the country. A validated model will provide an essential tool for fuel cell developers. The use of a user group to validate the model is good. The incorporation and leveraging of significant data sources that may not be available to fuel cell developers provides a strong data set for the model and a significant advantage for use by fuel cell developers.

### Project weaknesses:

- This project needs to strengthen the user group's participation.
- With CHP programs ongoing around the world, this project could validate these models and present the data.
- A small fraction of fuel cell developers have joined the user group. The project does not consider the customers' input. It is not clear what the requirements of the customer are besides cost.
- It is not clear how much validation will occur. Go/no-go criteria were not defined and success depends on use by fuel cell developers. Since the users are not an integral part of the project, there may only be limited evaluation.
- The researchers seem to be working in isolation and have not planned ways to test and evaluate model performance through interactions with other researchers, other similar models, and a broader number of data sets.
- The restriction to CHP is the major issue. It is understood that benchmarking to other competing technologies cannot be done as comprehensively as the CHP analysis. Nonetheless, for certain cases, it should be included to avoid working with a sub-optimum model. Such an analysis should yield a feeling for when alternative technologies have to be considered and analyzed in detail. This analysis is required for the model's use as a comprehensive planning tool and, moreover, to win over potential investors.

### Recommendations for additions/deletions to project scope:

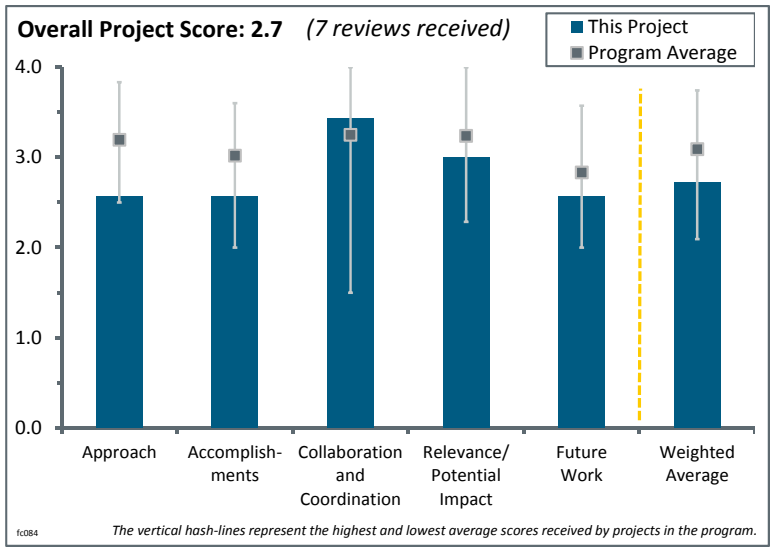
- This project should add users with committed roles, as opposed to a voluntary user group.
- This project should build in more capacity for model performance evaluation.
- This project should consider the relatively higher costs for smaller (e.g., <100 kW) fuel cell systems.

- DOE is funding a cost analysis for large-volume manufactures of automotive PEM stacks. It seems worthwhile to check if there could be synergy with this effort. Looking at different fuel cell technologies (e.g., PAFC, high-temperature PEM, and SOFC) may be difficult, but would be interesting.
- This project should continue the work. It needs to continue working very closely with marketers in fuel cell companies. User groups are very important and the project needs to provide some value for them. Fuel cell operators and building managers may not have the capability in-house.

## Project # FC-084: $WO_3$ and HPA-Based Systems for Durable Platinum Catalysts in PEMFC Cathodes

John Turner; National Renewable Energy Laboratory

### Brief Summary of Project:




The objective of this project is to improve electrocatalyst and membrane electrode assembly (MEA) durability and activity through the use of Pt/$WO_3$ and heteropoly acid (HPA) modification of carbon support to approach automotive proton exchange membrane fuel cell activity and durability targets. Enhanced platinum (Pt) anchoring to the support is expected to suppress losses in Pt electrochemical surface area under load cycling operations and to increase electrocatalytic activity. Lowering corrosion of support materials will increase durability under automotive startup/shutdown operation and suppress Pt agglomeration and electrode degradation. Ultimately, improvements to the electrocatalyst durability and performance will simplify the fuel cell system and lower system costs.

### Question 1: Approach to performing the work

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

- This project demonstrates the performance in working devices.
- As WOx has been unsuccessful and $SnO_2$ becomes the current focus, this project seems to rely on trial-and error.
- The principal investigator and his team are sharply focused on critical U.S. Department of Energy (DOE) goals and barriers and are pursuing sensible, but creative, approaches to their work.
- The stated objective is to use Pt/$WO_3$ and HPA to achieve performance and durability targets. There is no hypothesis as to why this would work and no attempt has been made to combine them. $WO_3$ and HPA were just ideas that might work. In fact, $WO_3$ has been tried before and found to have issues.
- This project is working to develop more durable catalysts with a two-pronged approach: using $WO_3$ as the support and synthesizing catalysts using HPA. Neither of these approaches seems to be aimed at increasing the mass activity of the Pt, which is required to meet DOE's targets. The use of advanced characterization techniques, such as EXAFS is good to provide basic information.
- The approach is quite standard and based on the experimental screening of promising materials. $WO_3$ was dismissed earlier this year (2013) as an option, given the poor performance. The program approach should have been crafted to reach this conclusion in a much faster time frame than three years. This way, the program partners would have had more time to explore and work the other options (e.g., HPA).
- The goal of using a non-carbon support to get around the degradation of traditional carbon supports under start-up/shut-down operations and at high voltages is to be lauded. There is much potential for scientific return in this approach, especially if the team continues the search for oxides and compounds with better conductivity. There are numerous paths to explore, but non-stoichiometric WOx and $SnO_2$-x are marginal pathways. The use of atomic layer deposition (ALD) processing to obtain conformal Pt coatings at low loadings is also commendable. Given that ALD processing is still in its infancy, compared to wet-chemistry or sputter-coating techniques, there is much to be learned from this project. ALD processing shows the promise of cost-effective scaling to large volumes.

### 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 materials show improved durability, but low mass activity.
- The team made good progress towards its objectives. The return of knowledge was worth the effort, even if DOE's performance metrics were not achieved.
- The work on $WO_3$ could have been accelerated to achieve the go/no-go decision earlier in the program. The progress on HPA seems encouraging.
- Refocusing on Pt/$SnO_2$ and Pt/C-HPA is a good move that yielded promising results for durable catalysts with improved activity.
- $SiW_{11}$ does not seem like a hetero poly acid (figure 3 slide 8), as some $O_2$ is missing. All the efforts in $WO_3$ did not produce any benefit, as shown in the literature. The HPA work did show some promise, at least in durability. The Pt/$SnO_2$ is just a distraction; the project should focus better. $SnO_2$ is only conducting in a reducing environment and will not survive startup and shutdown transients.
- This project synthesized, characterized, and measured the performance of a number of materials based on the WOx support. Mass activity measurements indicate that these are not competitive with traditional Pt/C catalysts, let alone more advanced alloyed (or dealloyed) catalysts. The Pt/C-HPA results indicated that some improvement of durability to the accelerated stress test cycling protocols is achieved; however, no initial increase to the mass activity is made, thus the modifications do not help the catalysts make the activity targets.
- The work on HPA-modified carbon appears promising and is being pursued in a sensible way that will yield actionable results and reveal the positive attributes of this approach to improve durability. The team has worked very hard on the other system involving $WO_3$ and Pt and has been careful and thoughtful in their approach. Even though that system ultimately did not make the milestone needed for the go/no-go decision due to insufficient support conductivity, the team is commended for their approach to the work.

### Question 3: Collaboration and coordination with other institutions

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

- The project appears to be very well coordinated with the partners.
- The collaboration on this project was well organized.
- This project has good, productive collaborations with industry, national laboratories, and universities. However, the role of the University of Colorado-Boulder (CU Boulder) is not clear when WOx is no longer included.
- The collaboration between CU Boulder and the Colorado School of Mines (CSM)is clearly evident with the materials being produced and transferred to the National Renewable Energy Laboratory.
- It is good to see catalyst suppliers involved, represented by the Tanaka Kikinzoku Kyogo Group (TKK). Andy Herring (at CSM) knows what he is doing with HPAs. It is early yet for scale-up and commercialization.
- There is clear collaboration among the partners. However, it is not clear if collaboration with TKK (and the use in this program of a commercially available catalyst) is relevant to accomplishing the objectives of the program.
- The collaborations appear excellent on both materials and characterization fronts. The team will soon be moving forward on MEA fabrication and testing, and they are encouraged to take advantage of the many resources available in DOE for making and characterizing MEAs, including both performance and microscopy.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- This is a relatively low-cost approach to improving the effectiveness of Pt catalysts.
- The durability of catalyst supports is currently a critical path item. Sadly, progress here was limited due to distractions from what actually worked.
- This project is working to stabilize catalyst activity from degradation mechanisms. This work should be expanded to increase the mass activity of catalysts so that they meet the DOE mass activity targets.
- The corrosion of catalyst supports and loss of Pt ECSA are very important problems; the project's work is highly relevant to solving problems in these areas.
- It is ideal to have a corrosion-resistant support for catalysts; however, its cost needs to be justified because there is a way to mitigate carbon support corrosion at the system level.
- The goals for this project are well aligned with the Hydrogen and Fuel Cells Program's RD&D objectives; however, the team has not fully addressed how the cost benefits will be obtained solely from using more durable support.
- The pursuit of novel catalytic structures to address durability, cost, and performances is definitely in line with DOE's goals. However, these new structures are far from being at the maturity level needed to turn them into commercially available products. Alternative work, focused on the optimization of the catalyst layer for existing carbon-supported Pt, has been proven to be effective to approach DOE's targets. In particular, the optimization of the water management in the catalyst layer is a big opportunity to accelerate the achievement of the goals.

### Question 5: Proposed future work

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

- The work plan is excellent and will help achieve project goals.
- The proposed future work is feasible with a focus on Pt/$SnO_2$ and Pt/C-HPA.
- The focus should be on HPA and the project should forget everything else. It should take HPA to subscale testing.
- The proposed preparation and characterization of MEAs is sensible. The team probably needs some new ideas on other areas to pursue. Some work aimed at revealing mechanisms by which HPAs improve performance will be helpful.
- The future work does not appear to continue with the Pt/WOx materials and to date there is no data to support that this work is promising. Moving the Pt/C-HPA to a graphitized carbon will probably provide more positive results. However, the value of the HPA is questionable, as the HPA has not improved activity to date and it is already clear that the graphitized carbon is more stable than carbon black.
- The program will sensibly focus on testing the most promising configurations (HPA and Pt/$SnO_2$). The value of testing the Pt/$SnO_2$ is not fully understood. Given that it is a commercial catalyst, the majority of work should be aimed at assessing performance of HPA, which has been the main output of the program up to now.
- The plans build on past progress, with logical down-selects chosen. However, the project ends in April 2014 and the use of carbon-stabilized HPA phases or sub-stoichiometric tin oxide needs to be supported by additional materials development beyond stating that the team will explore wet chemistry Pt deposition of alternative support carbides, etc.

### Project strengths:

- The right experts are involved with HPA.
- There is lots of collaboration with strong academic and industrial partners.
- This project has a strong team, particularly on electrochemical characterization.
- The use of lower cost materials with improved corrosion resistance is a strength of this project.
- This project has done good fundamental work on a promising system combining HPAs with Pt on C.

### Project weaknesses:

- Low mass activity.
- There is no desire to narrow down to a conclusion.
- There is limited value in short- and medium-term plans for fuel cell commercialization.
- With work on $WO_3$ a no-go, the project is relatively narrow. Some new initiatives are probably needed.
- This project really only concentrates on improving the durability of catalysts, not improving the mass activity, and the mass activity of the catalysts are low compared to standard benchmarks. The durability improvements are mostly only marginal.
- This project lacks the theoretical analysis to help the development of corrosion resistive support. For example, the impact of electronic conductivity on catalyst activity should have been predicted by a mathematical model that considers electron conduction.

### Recommendations for additions/deletions to project scope:

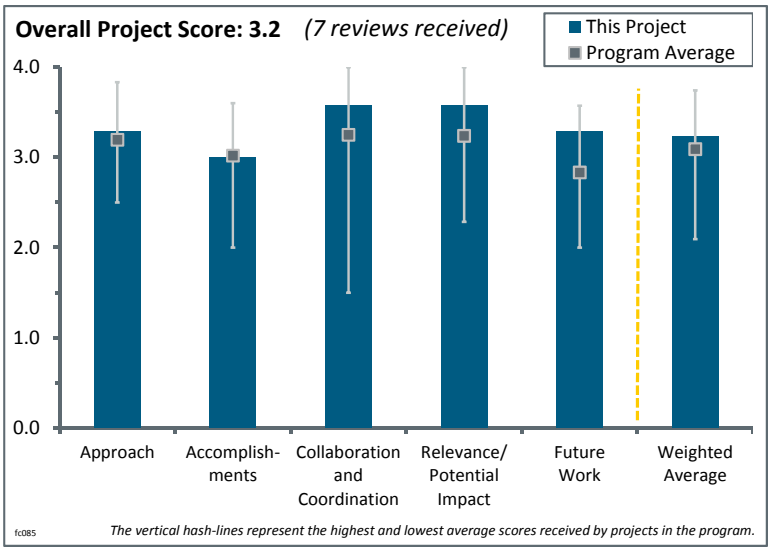
- As WOx has been excluded from future work, this project should include a theoretical analysis to explain why WOx is not as good as $SnO_2$ as a catalyst support.
- The future work should address the question of whether the HPA modification of carbon helps to prevent carbon corrosion and/or Pt sintering. Also, the project should consider the proposed idea of combining $RuO_2$ with $WO_3$.
- There should be a focus on the HPA work and a drive to demonstrate its value for improved durability in subscale fuel cells. This project should engage TKK to scale up this catalyst to allow some original equipment manufacturer (OEM) testing. This project should find an OEM willing to evaluate it in the short stacks.
- It looks like the work with Pt/WOx should be discontinued; it is hard to see any benefit. The project has not really examined the detrimental aspects of the electronic conductivity yet, which is about eight orders of magnitude lower than carbon. This project needs to look at the stability of the Pt/$SnO_2$ in terms of this and past work. These Pt/Sn materials were explored about a decade ago by the Phil Ross group with the conclusion that the activity was not stable. ALD does not seem to be amenable to cost or to make materials with high catalytic activity; this does not seem worth pursuing. More data on the activity of colloidal Pt/C-HPA and how it relates to the benchmark of Pt/C in rotating disk electrodes and DOE activity targets would be beneficial to understand the value of these materials and if modifications can be made to increase the mass activity.

## Project # FC-085: Synthesis and Characterization of Mixed-Conducting Corrosion Resistant Oxide Supports

Vijay Ramani; Illinois Institute of Technology

### Brief Summary of Project:

Research objectives in this project are to (1) develop and optimize non-carbon mixed-conducting materials with high corrosion resistance, high surface area, and high proton and electron conductivity; and (2) concomitantly facilitate the lowering of ionomer loading in the electrode for enhanced performance and durability by virtue of surface proton conductivity of the electrocatalyst support and reduced ruthenium (Ru) content in support materials. This project also will develop a cost model for the optimized support materials.




### Question 1: Approach to performing the work

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

- Exploring mixed conducting oxides as an alternative to conventional carbon supports is a sound idea. In terms of stability, the materials examined are clearly superior to carbon. The $RuO_2$-$TiO_2$ system looks promising, but the non-platinum group metal supports, such as indium-tin-oxide (ITO), are even more interesting and worthy of greater attention.
- This project has some very interesting approaches and has rightfully narrowed work to the most promising the $RuO_2$ supports. However, divergence from U.S. Department of Energy's (DOE's) and U.S. DRIVE Partnership's automotive cycling protocols is not good at all. It is fine if the Illinois Institute of Technology (IIT) team thinks it has found a better way, but it is essential to do the standard test for comparison.
- This project should use DOE protocols for catalyst cycling (0.6 V–1 V at 50 mV/second). The reason these exist is to make sure you can compare material sets being developed by various projects. There is nothing wrong with doing the more severe carbon corrosion test; in fact, the U.S. DRIVE Fuel Cell Tech Team has already adopted this accelerated stress test. The Nissan Technical Center North America (NTCNA) wants to run their own protocols, but it will be useful to DOE to have the same materials evaluated under DOE's protocol. The summary slide included this information, "< 30 mV electrocatalyst support loss after 100 hrs at 1.2 mV"; it should have said, "< 30 mV electrocatalyst support loss after 400 hrs at 1.2 V."
- The support materials chosen are directed towards a more durable support. $SiO_2$ is more durable than carbon black, as are most oxides. The key component is $RuO_2$, which is deposited on the $SiO_2$ to provide electronic conductivity and the ability to deposit small particles of Pt on the modified support. It is unclear if Ru will dissolve and move into the membrane or anode of a membrane electrode assembly (MEA) in a fuel cell. It is also unclear if $RuO_2$ also provides a protonic conduction and if the ionomer can be eliminated. These questions should have been answered by the third year. Ex-situ measurements on electronic and ionic conductivity of $RuO_2$ have not been reported.
- The approach addresses the key obstacles in improving and demonstrating support durability and includes testing under relevant conditions. The much higher density of the mixed metal-oxide (MMO) supports compared to carbon suggests that the Pt loadings should be adjusted to lower Pt loadings to achieve comparable volumetric loadings for Pt/C. Assuming a 50-50 $RuO_2$-$TiO_2$ ratio, the Pt loadings should be reduced by a factor of two to three for $RuO_2$-$TiO_2$ compared to Pt/C (i.e., should be 13%–20% Pt on $RuO_2$-$TiO_2$ to compare to 40% Pt on carbon). $In_2O_3$ has an even higher density, so Pt loadings should be reduced even further for the ITO support. The different electrical and proton conductivities in the MMO supports

may lead to different optimized Pt loadings, but it would seem that starting at the same volumetric loading (rather than mass ratio) would be a good starting point. It is not clear if the Pt loadings used were at 40% because the loadings on carbon are also 40%, or if they were optimized based on electronic and protonic conductivity considerations. Water management in the catalyst layer can have a large impact on performance. The project has not yet looked at the effects of changing the catalyst layer structure (removing ionomer, changing hydrophobic C to hydrophilic oxides) on water management. Tests should be done under conditions where flooding may occur to see if water management issues arise from changing the support material to a less hydrophobic material.

- The overall approach is to develop alternatives to carbon supports in order to mitigate the corrosion problem. This approach is particularly important from the perspective of start-stop operation, lower humidity operation, and conditions that could cause cell reversal, such as fuel starvation, etc. The approach, therefore, is based on developing metal oxide support materials to replace carbon. These metal oxides, besides providing for a more corrosion-resistant support, also provide for greater wettability and proton transport (under dry operating conditions).The attempt to use metal oxide supports to replace carbon is not new and, therefore, not terribly innovative. The choice of materials proposed and followed through in this effort is interesting. Particularly interesting are those that are well known to form underpotential deposition and even bulk deposition within the operating window of fuel cell operation. These include Ru and tin oxides. Both have the potential for dissolution at higher electrical potentials, and their ability to re-deposit on Pt may have debilitating consequences on oxygen reduction reaction (ORR) activity. In addition, any migration to the anode would be even more devastating as they form bulk depositions below 0.3 V versus RHE. It is interesting to note that these MMOs (some with $SiO_2$) are stable when cycled at high potentials.
- This project demonstrates a good knowledge of what properties a non-carbon support requires to yield improved durability electrocatalysts for fuel cell applications while at the same time being compatible with catalyst and MEA preparation. The integration with an original equipment manufacturer (OEM) partner is apparent and at an appropriate level. With their partner, the project has adopted an accelerated testing procedure to probe rates of corrosion. The project has adopted a laudable "fail fast and move on" approach that enables testing a much wider array of materials that may have promise. Perhaps an improvement in their approach would be to do less "front end" characterization of surface areas and crystallite sizes via X-ray diffraction and more materials testing, saving the more detailed characterization for materials that offer demonstrated advantages, or doing post mortems of materials that have unanticipated poor corrosion properties. Sometimes focusing on outliers at both ends of the spectrum can reap rewards to faster rates of progress. While keeping an eye on materials cost is a good thing, one may not want to narrow ones view too early at the expense of developing the learning and knowledge of what structures and compositions lead to the desired properties. In terms of the community and their fixation on surface area, one might want to consider not the surface area/mass, but rather the surface area/volume. It is the surface area contained within the reactor volume that is relevant in most cases. In the case of the MEA, it is the MEA that is the "reactor," and so it is the surface area available in the volume of the support in the catalyst layer that is important, not the number of grams of support in the catalyst layer. Thus, one might want to consider normalizing surface areas per unit mass to surface areas per unit volume by multiplying by the density of the material. Otherwise, one is comparing mostly apples to mostly oranges.

### 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.

- This project has made good progress on the $RuO_2$ supports. They do not normalize for mass activity, they only do ECSA; this omission is a major mistake.
- The divergence in goals for e- and H+ conductivity is confusing. Using a typical telegraph model for a catalyst layer, the electrical conductivity can be much lower until it becomes limiting.
- The milestones and go/no-go criterion outlined in slide 8 were all satisfied in the 2012 review. The dispersion of Pt on ITO looks great, but it is unclear why the electrode was inactive. It will be nice to see if the group can get to the bottom of this in the remaining grant period.
- This project is making good, steady progress toward controlling corrosion/loss of ECA. Hopefully, the work on the Ru cost model did not hold up progress in the more important areas of screening more

materials. Although highlighting the influence of durability on cost is always good to remember, cost modeling is not the principal investigator's (PI's) strength; materials development is. The progress included some very valuable knowledge gained from accelerated testing studies to benchmark a variety of materials. This occurred with good feedback from the OEM partner.

- The technical accomplishment of Pt/$RuO_2$-$TiO_2$ with only a 34% ECA loss compared to a 49% loss when cycled in the voltage range of 0.6–1.0 V, is not spectacular. The same results can be obtained with Pt/graphitized carbon black. The initial surface area of the Pt/$RuO_2$-$TiO_2$ is half that of Pt/HSAC; therefore, since the Pt particles are large, their dissolution is lower. This is not an indication of an improved catalyst. On slide 13, the limiting currents for the RDE measurements vary from sample to sample, which has been explained as due to the dispersion of Pt on the support. This explanation is fundamentally incorrect since both poly-Pt with a very low area and Pt/C with 10–100 times higher area have the same limiting current for a given $O_2$ partial pressure. It is good to see a cost model being presented.
- This project has good MEA results. It is unclear how the proton conductivity of the support (stand-alone) was measured. More details on this need to be provided. It is not clear if the project has run fuel cell drive cycle measurements with the catalysts. At low potentials, it seems the $RuO_2$ could be reduced and alloyed with Pt to decrease the ORR. It seems the project has to take these supports to lower voltages in order to evaluate that effect. It is not clear if any other comparisons have been made with other supports such as Vulcan carbon (e.g., MEA studies). Those supports might be a fairer comparison for durability since the Pt ECSA of these catalysts is far lower than the HSACs and might be more comparable to Vulcan, which will obviously have better durability than the HSAC (slides 7 and 8). If possible, a highly graphitized support should also be included for comparison. There is a good cost model included with simplified assumptions.
- The accomplishments in this effort are good. The durability measured under the DOE durability working group's recommended metrics show a high state of durability. These measurements are under both 100% and 40% relative humidity operating conditions. Even though the surprisingly high durability results are reported with oxides, which typically dissolve into soluble valance states below pH 1, no clear underlying studies are reported on how this feat is accomplished. Some impedance data, together with conductivity measurements as a function of potential, would be interesting and important for future consideration of these materials as mixed-oxide supports. Traditional reports in a direct methanol fuel cell with PtRu and Pt on $RuO_2$ show very different results, wherein Ru dissolves and ultimately kills the cathode and anode electrodes. The absence of this behavior in a slightly different format where $RuO_2$(OHx) is used as a support is most interesting and needs an explanation.
- The stop-start cycling (1.0–1.5 V) results for Pt/$RuO_2$-$TiO_2$ are very encouraging. The lack of negative impact of $RuO_2$-$TiO_2$ support in catalyst dissolution cycling protocol (0.6–1.0 V) is encouraging, but needs to be repeated with samples of similar Pt particle sizes. Previous results compare cycling at 0.6–1.0 V for Pt/C and Pt/MMO were not of comparable Pt particle sizes. Recent efforts to prepare smaller Pt particles on $RuO_2$-$TiO_2$ should lead to a more appropriate comparison. The much higher density of $RuO_2$-$TiO_2$ compared to carbon indicates Pt loadings should be adjusted to lower Pt loadings on the support in order to achieve comparable volumetric loadings for Pt/C. Assuming a 50-50 $RuO_2$-$TiO_2$ ratio, the Pt loadings should be reduced by a factor of two to three (i.e., they should be 13%–20% Pt on $RuO_2$-$TiO_2$ to compare to 40% Pt on C). If the loadings are decreased to give comparable volumetric loading as for Pt/C, it should be easier to achieve the smaller particle sizes desired. The initial cost study shown was based on the initial performance of Pt/$RuO_2$-$TiO_2$ with much higher Pt loading for the $RuO_2$-$TiO_2$ support than for Pt/C. Based on volumetric loading arguments, this loading should be able to be reduced substantially. This loading suggests that $RuO_2$-$TiO_2$ supports will have cost advantages due to increased durability once the Pt loading for this system is optimized. A true cost comparison should be on optimized Pt/$RuO_2$-$TiO_2$ loadings. The initial work with ITO is encouraging, but a synthesis providing higher surface areas while maintaining conductivity is needed. It is not clear what impact the order of magnitude lower electrical conductivity, compared to $RuO_2$-$TiO_2$, will have on performance. If the performance is significantly impacted, the cost differential between ITO and $RuO_2$-$TiO_2$ will be overshadowed by the increased Pt cost due to lower performance.

### Question 3: Collaboration and coordination with other institutions

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

- The collaboration between IIT and NTCNA is working well.
- The collaboration with NTCNA is very good.
- This project has excellent interaction with NTCNA. This interaction is a major strength of this project and keeps the project relevant to automotive applications.
- The collaboration with NTCNA appears to be fruitful although the data between NTCNA and the IIT do not appear to match well.
- This is a small group, which is good. However, they should continue to follow the Fuel Cell Tech Team's recommendations for comparisons.
- The IIT project lead indicates that NTCNA has been an excellent partner. Hopefully, they can get the performance and durability studies of the Pt/ITO system completed in the remaining grant period. It is unclear what is going on with the sulfated tin oxide system.
- The project review resulted in many examples of excellent integration and communication between the PI and the OEM partner in developing materials, properties targets, and accelerated test procedures to make more rapid progress. Sending students to interact with colleagues at the OEM is extremely valuable for the student. Hopefully, this interaction can continue.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- Durable, non-carbon supports have the potential to significantly increase durability.
- They are doing exactly as asked; however, automotive OEMs are saying that they have good strategies to avoid carbon corrosion all together so the need for these supports is questionable.
- The work has clearly demonstrated that $RuO_2$-$TiO_2$ support exhibits superior durability compared to traditional carbon supports.
- Alternative supports for Pt are a critical area, but this is not so easy to solve because the materials that are corrosion resistant are usually electronically insulating and also have a low surface area resulting in low Pt dispersion. Select projects on alternative supports need to be continued.
- The PI provided some cost estimates, which look promising. However, the use of Ru and Sn still seems risky. The impact of this effort is based on the ability for this process to be scaled up and implemented by a commercial entity.
- The development of corrosion-resistant supports will help the DOE program achieve its research and development goals. This project, if successful, can help commercialization of fuel cells by providing an alternate to carbon, which is the only support used today. The authors have provided a good cost study that helps alleviate concerns regarding Ru costs.
- Improving support durability and overall catalyst stability is highly relevant to DOE's goal of advancing progress toward less-costly commercial fuel cell systems. The development of alternative support materials for fuel cell applications that avoid the shortcomings of carbon supports, while providing advantages beyond what carbon offers, is very relevant to DOE's objectives of providing durable, cost-competitive fuel cell systems. This project is well focused on the relevant barriers and is well-integrated with an OEM to maintain the project's focus on acceptable materials for contemplated future use in commercial systems.

### Question 5: Proposed future work

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

- The proposed future work is adequately addressed.
- The emphasis on lowering Pt particle size is good, but the project did not speak to what is limiting them now. IIT should consider alloys following the PtCo and PtCoNi materials on carbon now. Specific activity should be measured.

- The proposed experimental work appears to be sharply focused on what is required for continued progress to meet relevant DOE objectives. This reviewer would recommend that any cost modeling work after the materials down selection should be held to the appropriate level. The strength of the project is in the capabilities directed at the materials development and learning what some of the composition and function relationships are for alternative supports. The substantial cost modeling efforts would distract the project team from their strengths of materials development and the development of accelerated testing protocols for alternative support materials.IIT should study lower loadings, especially when looking at reducing ionomers. IIT should try not to use loadings over the DOE 2015 targets.
- The project should see how the ITO and sulfated tin oxide support work in terms of catalytic performance and durability. It is not clear what lies behind the inability to measure the activity of the nice Pt dispersion on ITO shown in slide 20.
- The future plans address optimizing Pt particle deposition and decreasing Pt particle size. The future plans discuss a down-selection of supports. Water management in the catalyst layer can have a large impact on performance. The project does not appear to have plans to look at the effects of changing the catalyst layer structure (removing ionomer, changing hydrophobic carbon to hydrophilic oxides) on water management. Tests should be done under conditions where flooding may occur to see if water management issues arise from changing the support material to a less hydrophobic material and removing the ionomer.
- In addition to the future work proposed, electronic and ionic conduction measurements as well as surface area measurements should be conducted ex-situ. These measurements should guide the possibility of success in electrochemical measurements. Such measurement may help the selection of materials for further work. For example, it is unclear how one can expect to deposit 2–3 nm Pt particles (100 $m^2/g$) on an ITO support after having a BET surface area of 40 $m^2/g$. It is unclear if Ru is found in the membrane and anode side, and if the ionomer content can be reduced due to the use of $RuO_2$.

### Project strengths:

- This project addresses critical needs for the DOE Hydrogen and Fuel Cells Program. This is a reasonable approach to solving the problem and has good collaboration with industry.
- Nissan provides accelerated testing expertise and knowledge of system requirements. The potential cycling test results were very encouraging.
- This project makes a compelling fiscal argument, the RuOx is viable. RuOx is definitely a real technical possibility.
- This project has good interaction between NTCNA and the IIT. The durability offered by $RuO_2$-$TiO_2$ is impressive. It will be good to hear more about the performance/behavior of other Pt ITO.
- This project has great collaboration with NTCNA and good, promising initial results. There needs to be high activity on these promising supports.
- This project has a strong PI-OEM interaction and communication that has led to a focus on materials requirements and the appropriate accelerated testing protocols to screen against. There is a good focus on benchmarking test protocols and materials against the best currently available materials.

### Project weaknesses:

- This project needs to improve ex-situ diagnostics and measurement of material properties to guide electrochemical work.
- The project should not lose its focus on materials development to cost modeling in the future. The strength is in materials development.
- The project is still looking at too many supports. It is good to look at various things at the beginning of a project, but the team needs to focus as the project gets mature (it is 65% complete at this point).
- There is a need to measure specific mass and surface activity and compare two automotive targets. This project needs to do long-term testing. It is unclear if these catalysts ripen. The project needs to hunt harder for Ru dissolution over the long term.

### Recommendations for additions/deletions to project scope:

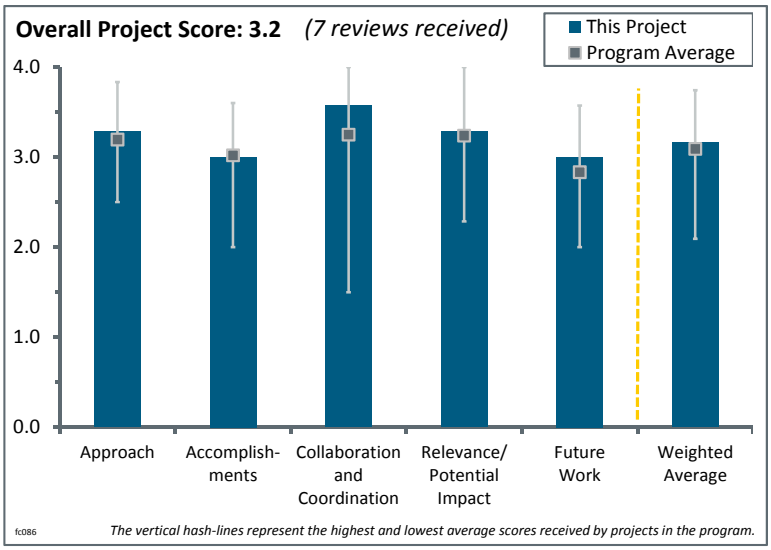
- The scope is appropriate at this time.
- This project should sample the materials to external groups for characterization as well.
- The investigators should see how the Pt/ITO and Pt/sulfated $SnO_2$ supports perform and/or what prevents the effective activity measurements.
- This project should ensure the balance between materials development and cost modeling makes sense. They should consider engaging others in the "alternatives" community to share samples and do some round-robin testing against the best-in-class, carbon-supported catalysts. Perhaps the PI or DOE can consider taking on this initiative.
- It is difficult to evaluate how much of the effort is being spent on developing the ITO support and the other three supports in addition to the $RuO_2$-$TiO_2$ support that has already been developed, and how much is being spent on developing a workable catalyst on the $RuO_2$-$TiO_2$ support that has already shown great promise. The best approach is not sequential, such as the one the PI is following. The research team has the one support and are optimizing the catalyst on that support and now are exploring another support and plan to optimize that too (maybe doing this five times in total). It is better to down select one or maybe two supports and do all the optimization (MEA, durability) work on that one or, at most, two supports.

## Project # FC-086: Development of Novel Non-Platinum Group Metal Electrocatalysts for PEMFC Applications

Sanjeev Mukerjee; Northeastern University

### Brief Summary of Project:

This project will develop new classes of non-platinum group metal (PGM) electrocatalysts to meet or exceed U.S. Department of Energy (DOE) 2017 targets for activity and durability. This will enable decoupling of polymer electrolyte membrane fuel cell (PEMFC) technology from platinum (Pt) resource availability and lower membrane electrode assembly (MEA) costs. The science of electrocatalysis will be extended from its current noble metal catalysts to a wide array of reaction centers that have a greater tolerance to poisons, which typically affect Pt and Pt alloys.




### Question 1: Approach to performing the work

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

- This approach is an exceptionally good ramification of early work on pyrolyzed macrocycles.
- The approach includes computation, catalyst and electrode design, powder preparation, out-of-cell and in-cell testing, and stack testing.
- The approach of this project is valuable and systematic. It covers all relevant aspects that should be considered for the class of catalyst that may revolutionize fuel cell technology.
- This presentation was one of the best. The approach used to develop the non-Pt-based catalysts shows a high degree of understanding of physics, chemistry, and materials science. This project shows that there is strong promise in this area.
- This project is following a multi-pronged approach to developing non-PGM oxygen reduction reaction (ORR) catalysts to significantly reduce the cost of PEMFCs. Three organizations are pursuing three approaches to synthesizing Fe-C-$N_2$ catalysts. The project also shows a balanced approach in determining the active site and ORR mechanism using computational chemistry, XPS, X-ray absorption, and RDE studies. The project originally had other types of catalysts, but these have been down-selected due to low ORR activity.
- The overall approach to performing the research is reasonable, with a good materials development strategy. The collaboration strategy also seems reasonable and appropriate for the project. What is not clear in the approach is the parameters to be reported on the electrocatalyst activity and active sites for ORR. The principal weakness of a poor ORR electrocatalyst is that the loading would have to be very high resulting in high thickness of the electrodes. Although the units of A/$cm^3$ is a good choice to take into account in terms of electrode thickness, the actual electrode thickness, loading, and resistance (using ,EIS, not HFR) should be clearly measured and reported for simplicity and transparency.
- This area is important to explore, but the project's approach is incremental relative to an earlier attempt at co-pyrolysis of CHN precursors with metal ions/complexes. The team's approach to this important area will not lead them to raising the bar in the search for non-PGM electrocatalysts. This pyrolysis approach, while yielding materials that have demonstrated some PGM-like activity and some durability, necessarily leads to uncontrolled catalyst properties, such as structure and composition. That is OK, but it makes it very difficult to make rapid progress in understanding the underlying principles of this class of catalysts, and the researcher's approach should recognize the degree of difficulty and apportion resources appropriately. The

principal investigator's (PI's) approach should be a little more careful not to oversell the ability to control active sites, electronic structure, and metal-support interaction as there is no supporting evidence that any of this is possible at such an early stage in research. For example, there is not any definitive evidence that the activity is metal-centered. The approach to understanding ORR electrocatalysis by spectroscopy and computation is laudable, but it encounters the usual problem in catalysis in that the minority species that go missing by "the usual" approaches may be responsible for all of the catalytic activity. What is seen is not generally what is doing the catalysis; it may be a mere "spectator" along for the ride. The theory and spectroscopy efforts have leapt to assuming there is some M-Nx center(s) responsible for the catalysis. This assumption is not supported by any direct evidence; correlation is not causation. The approach at this early time should focus on very elementary catalysis characterization approaches rather than on the approaches that are more appropriate to more well-understood catalyst systems. There is value in learning what makes these things tick, even if the activity and durability is not yet on par with PGM.

### 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 milestones and go/no-go points are being met. The results approach DOE performance targets in some conditions. There needs to be more operation and testing with air. Durability seems to be an issue with some formulations and operating conditions.
- Substantial progress has been made over the last fiscal year and the PI was able to address challenging critical barriers. Moreover, the project brings an overview of an entire new class of catalysts and it spans from lab to scale-up efforts that engage research labs and industry.
- The materials shown exploit the Fe2+/3+ redox couple. The researchers should focus on modifications of the chemical moieties where the local atomic-scale environment (e.g., near-neighbors, coordination, and short-range ordering might result in shifts in the E1/2 value to higher potentials [reduced over-potential]).
- A lot of excellent work has been carried out, but the main deficiencies of these catalysts have not been reduced. The major issue is their stability, and another issue is their activity. Considering that the cost of production cannot be low and there is still insufficient performance, it appears that approaching PGM catalysts will not be an easy task.
- The progress is incremental with respect to earlier efforts in this area. Progress in performing computations and modeling "active sites" and advanced spectroscopic studies is of dubious value as there is simply not yet even an elementary understanding of how or what these catalysts are functioning as. For example, it is not clear if these are metal-centered or not. Hence, the progress in "mechanistic understanding" is of yet more dubious value.
- In slide 7, the current density has not been reported, so only the relative values of current at 0.8 V are known. In slide 8, the resistance of the catalyst layer and its thickness should both be reported; no activity values are reported or extracted from the curves. In slide 9, millivolts at 200 $A/cm^3$ is reported and the back pressures are arbitrarily different. No attempt has been made to identify the active sites, estimate the number of active sites, or obtain the turnover frequency. A study of the effect of loading might enable one to estimate the values that are critical in understanding how these non-PGM materials work. In slide 11, the DOE Durability Working Group (DWG) protocol for non-PGM has a cycle from 0.2 to 1.1 V. This range is not realistic for an automotive fuel cell. The ranges used by Nissan and by the DWG for Pt-based catalysts are much more reasonable. There is no reason why a non-PGM material should have a special protocol that deviates from a protocol used for Pt-based catalysts and simulates automotive conditions.
- The project has shown excellent progress towards enhancing the ORR activity of the Fe-C-$N_2$ catalysts and characterizing the active site. Whereas last year they were quoting ORR volumetric activities at 0.8 V based on extrapolation from higher potentials, this year they are reporting directly measured ORR activities at 0.8 V which are approaching DOE targets. While the MEA performance is still well below that of Pt/C, the progress demonstrated by this project and in other labs working on the same type of catalyst show that these materials are becoming viable both in terms of activity and durability. This project is behind schedule in meeting its milestone for the in situ measurements for degradation and electrocatalytic pathways for ORR.

### Question 3: Collaboration and coordination with other institutions

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

- The team seems to be operating coherently.
- This project has excellent collaboration.
- The collaboration team for this project is excellent.
- Collaboration between mostly top experts for this topic appears to be very good.
- The team includes an automotive OEM, universities, national laboratories, and materials suppliers.
- Very good team effort is demonstrated throughout all phases of the project that enabled a systematic approach in tackling challenging issues in non-PGM catalysis.
- This project has an excellent breadth of experience and balance between academia, national laboratories, and industry. The Nissan participation is vital to keeping the project relevant to automotives. A new partner has been added to the project this year. Though this partner's work was not presented this year, it is apparent from the supplemental material that they are scaling-up the most promising catalyst materials. A cost estimate from Parajito powders on industrial-scale catalyst synthesis would benefit the project.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The work addresses the issues of Pt cost and availability by replacing Pt as the catalyst.
- The realization of Pt-free materials for the cathode in PEMFC will be a game changer.
- The project, if successful, will significantly advance fuel cells as a power source; however, considerable research will be needed to realize the possibilities of this approach.
- The projects aligns very well with the Hydrogen and Fuel Cells Program and serves as a valuable source and foundation for future considerations of non-PGM catalysts.
- If this project is successful in improving the ORR activity of the non-PGM catalyst even further, the results of this project would be of enormous benefit to the Program as it would eliminate one of the major barriers to commercialization—cost.
- Non-PGM materials need to be studied for the next decade before they can achieve targets comparable to platinum group catalysts. Projects of this nature need to be funded even though the progress might seem slow. Better fundamental electrochemical measurements, along with improved analysis, are partially lacking. The targets should be rigorous and identical to that for platinum-group materials, so that there is relevance for automotive targets. The impact of success in such a project will be disruptive, but it will speed-up automotive PEMFC commercialization.
- The goal of discovering, characterizing, and utilizing non-PGM catalysts is highly relevant to DOE objectives in making progress toward active, durable, cost-competitive electrocatalysts to support future fuel cell vehicles. Where the relevance falls off in spite of the promise for the future, is that the approach taken by this team will not likely produce successful results as quickly as it might. The project's emphasis on modeling and advanced spectroscopies is too early in the development cycle and saps resources from an effort to do some of the very elementary catalyst and materials characterization steps. In this very early stage of catalyst discovery, focus needs to remain on developing some of the elementary systematics of how, why, and what this catalyst might be. This is necessary so that in the future, meaningful experimental input can be given to the theorists and carefully chosen, well-designed experiments can integrate spectroscopy into the experiment—spectroscopy—theory cycle. This should be done; otherwise, the project will be of limited relevancy.

### Question 5: Proposed future work

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

- The proposed future work is logical and in line with the overall project approach and goals.
- This is a good path forward. Again, this project should focus on reducing the over potential and then try and get kinetics.
- Although the future work sounds reasonable, the key task is to meet the DOE activity target at 0.80 V and 0.90 V in A/cm$^3$ with a better understanding of the actual active sites of the catalyst for ORR.
- Spectroscopic mechanistic studies should be compared with the RRDE data, which can clearly determine the sequential mechanism, and with diagnostic criteria involving formal electrode kinetics.
- The project has well-focused future tasks; however, several milestones in this project are only 50%–70% accomplished. Considering that the project is entering the last phase, there is a concern about whether these milestones will be met.
- There is some value in placing these early non-PGM materials into an MEA; but of course, the risk is that these catalysts will be far from optimal, will not perform as well as PGM at this early point, and that this approach will be discarded before the true potential of these catalysts can be shown or one can conclusively identify that there is no path forward. This is a wordy way of saying to stay the course on the materials discovery aspects of this project.
- While the overall thrust of the project— improving catalyst activity and mass transport—is on the right track, it would have been beneficial if there were more details given as to the approach that would be used to achieve these goals. The modeling of catalyst layer transport would presumably provide guidance for the design of catalyst layers with improved mass transport, but this was not presented and was relegated to the backup slides.

### Project strengths:

- This project's scientific approach is a strength.
- This project has a strong research team of top experts on the topic.
- The multiple classes of materials and collaborations to choose from are a strength of this project.
- This project is working on an important topic in catalysis that has implications beyond fuel cell catalysis. The project is potentially very relevant to DOE objectives in thrifting (to zero) PGMs.
- This project has novel systems and non-PGM catalysts, a systematic approach from fundamental atomic scale to real applications, and a strong team effort.
- This project has many approaches to synthesizing Fe-C-$N_2$ non-PGM catalysts. The X-ray characterization of multiple catalysts and correlation of characterization results with activities to determine active site and mechanisms is very good.

### Project weaknesses:

- Pt-free anode materials are a weakness.
- This project has partially achieved their milestones. The volumetric activity to actual fuel cell performance is not well defined and the durability issues are substantial.
- Need a report on electrochemical activity results under conditions that are comparable and with sufficient detail that allows one to determine whether one is approaching the activity of baseline Pt/ C. Fundamental electrochemical measurements with improved analysis is partially lacking. The targets should be rigorous and identical to that of platinum-group materials so that there is relevance for automotive targets. Although the future work sounds reasonable, the key task is to meet the DOE A/cm$^3$ activity target at 0.80 V and 0.90 V with a better understanding of the actual active sites of the catalyst for ORR.
- The approach is not well-aligned with realistic catalyst discovery approaches. There is not enough information at this point to feed to theorists, and there are not enough elementary catalyst characterization studies. There is also not enough information to determine if the metal plays a role in the electrocatalysis, or if it is important (or crucial to know) for the formation of a non-metal active site. This project is not staffed with conventional catalyst characterization types that can approach answers to these rather elementary questions.

- The presentation lacked details on how the proposed improvements were to be made and also details on the various catalyst synthesis methods, making it difficult to evaluate the merits or feasibility of the proposed future work. The roles of Los Alamos National Laboratory and the University of Tennessee-Knoxville (UTK) were not clear in the work that was presented. A discussion of mechanisms was interesting and important, but it could have been greatly abbreviated to allow for highlighting the results from other aspects of the project, for example, the UTK transport modeling work.

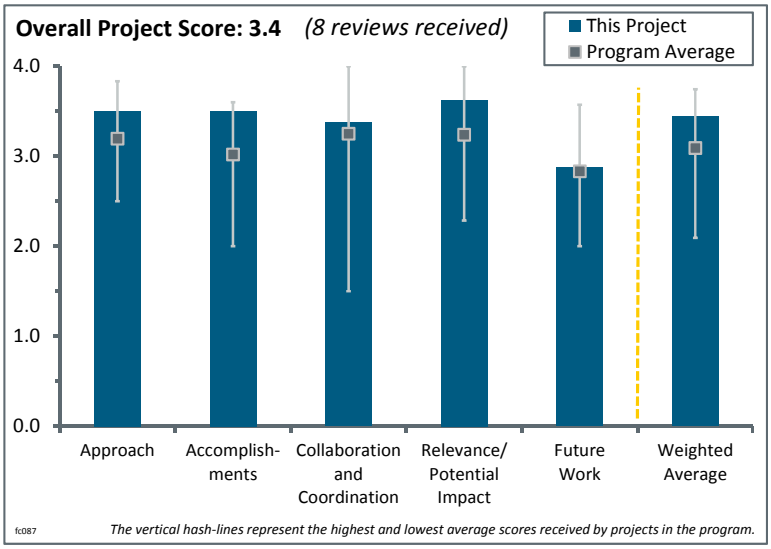
### Recommendations for additions/deletions to project scope:

- Modeling the structure of the active site is not appropriate at this time. A useful modeling effort needs additional elementary information about what the active site properties are. This is not to say to keep the theorists out of the loop, just that performing modeling on a very incompletely defined system is not productive. This project should stay the course on materials discovery; the community needs more potential non-PGM materials, and thus an effort to go look for them.

## Project # FC-087: High-Activity Dealloyed Catalysts

Anusorn Kongkanand; General Motors

### Brief Summary of Project:




The objectives of this project are to overcome cost barriers and increase the durability and performance of fuel cells using dealloyed catalysts. Research should (1) demonstrate reliable oxygen reduction reaction kinetic mass activities that achieve the U.S. Department of Energy (DOE) target in hydrogen/oxygen ($H_2/O_2$) fuel cells using manufacturable synthesis and dealloying procedures, (2) achieve high-current-density performance in $H_2$/air fuel cells that meets DOE heat rejection targets and platinum (Pt)-loading goals, (3) demonstrate the durability of the kinetic mass activity against DOE-specified voltage cycling tests in fuel cells, (4) determine where alloying-element atoms should reside with respect to the catalyst-particle surface for durable activity, (5) demonstrate the durability of high-current-density performance, and (6) scale up to full-active-area fuel cells.

### Question 1: Approach to performing the work

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

- The approach is systematic and focused. Investigating different leaching methodologies is important in optimizing and "stabilizing" the catalyst.
- This is a very well organized program that is following a sound approach based on intensive collaboration with partners and an iterative experimental process. The program team has done a very good job defining the milestones to progressively increase confidence in the findings and is oriented toward identifying a practical solution for commercialization.
- The approach is good. There could be some additional challenges for further scale-up, such as: quality control of the dealloying process, resulting in a higher cost; approximately 40 mV voltage loss due to dissolved Ni can have a significant impact on performance and cost, considering fuel cell electric vehicles (FCEV) total life; and modeling capability to validate demonstrated performance/durability is preferable.
- The focus on large-scale catalysts provided by a catalyst supplier allows this project to have very strong short-term benefits. Shifting the focus to Co and Ni only prevents some of the poisoning concerns from Cu that had been in the project. Reasonable cases are made for the performance of these materials being acceptable after accelerated test protocols. Longer term testing in short stacks under vehicle-relevant conditions would be interesting to confirm that the accelerated stress tests (ASTs) are relevant for the durability concerns of these alloy catalysts.
- The project is working on developing cathode catalysts to meet DOE mass activity targets. The main approach is exploring dealloyed materials based on Pt. The initial precursors include $PtCo_3$, $PtNi_3$, $PtCu_3$, and other combinations that include Ir, Au, and Pd. Some of these appear to be good approaches; others appear to be less valuable and more questionable, including the use of Cu and Ir, especially in view of past comments from General Motors (GM) about using Ir.
- The project is focused on demonstrating catalysts that meet the DOE activity and durability targets and that can be manufactured in large batches. The approach to try to improve the formation of core-shell particles by controlling rates of base-metal dissolution and Pt surface migration is sharply focused on the major barrier of the durability of these materials.

- The group is focused on exploring the role of dealloying in the performance and durability of Pt-Ni and Pt-Co electrocatalysts that will be central to any meaningful progress in the engineering of polymer electrolyte membrane (PEM) fuel cells. The efforts of GM, Johnson Matthey Fuel Cells, Inc. (JMFC), and the Massachusetts Institute of Technology (MIT) are well represented in this report.
- The approach in this project has significant weaknesses and does not bring new insight into catalyst properties. Moreover, it does not consider any novel class or type of catalyst. It is mainly focused on catalyst performance that cannot be correlated to the physical/structural properties of the catalysts due to the lack of consistency in particle size and elemental distribution.

### 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.

- All the 2012 milestones have been met.
- The progress is excellent. The validation for scale-up has progressed gradually. The gas transport measurement is very helpful and effective in addition to the activity in terms of catalyst microstructure.
- This project has made excellent progress toward the project objectives. The further loss of Co and Ni with fuel cell operation is not surprising. It is expected to affect membrane and catalyst layer ionomer, thus voltage loss analysis could shed some light on the loss mechanism. In order to understand the impact of the carbon support on the observed performance loss, it may be valuable to test the equivalent Pt/C catalyst.
- A big improvement was shown in this years' work in terms of improving the initial Pt particle size distribution. The researchers have met initial activity and durability criteria. They are demonstrating that the concept of dealloyed catalysts has promise for improving the state-of-the-art cathode ORR catalysts. The work has been accomplished at a good scale for these catalysts.
- The progress over the past year in this project included materials that met DOE targets in the initial activity and added the ability to meet durability targets when tested under AST conditions. The materials set expanded to include Co as well as Ni. These materials have shown a good capability to meet DOE's targets and currently represent the state-of-the-art for fuel cell catalysts in membrane electrode assembly (MEA) and a significant step forward to commercially competitive systems.
- This project has made substantial increases in mass activity over the past year, from 0.5 A/mg to 0.75 A/mg platinum group metal (PGM). They have made substantial increases in the durability of dealloyed $PtNi_3$. JMFC has made good progress in improving the particle size distribution and has decreased the maximum particle size from greater than 25 nm in the old synthesis to approximately 10 nm. The project has also demonstrated catalysts with mass activity greater than 0.5 A/mg Pt after 30,000 potential cycles. PGM loading (g/kW) has not decreased despite substantial increases in mass activity, suggesting mass transport issues need to be addressed. Ni leaching from catalysts into membrane and catalyst layers still needs to be reduced.
- This project has made significant headway exploring the performance and durability of the Pt-Ni nanoparticle system. This includes studying the influence of variations in the synthesis and dealloying procedures, although the details of the former were not provided. Slide 13 demonstrates completion of milestones 1 and 2. Interestingly, variations in the dealloying process led to a significant range of Pt/Ni net composition ratios (from 0.8 to 1.8), yet the performance and durability milestones were still satisfied after 30,000 cycles. The results indicate that the new alloys are very forgiving in terms of sensitivity to the dealloying treatments, and speak to a certain robustness of well-engineered PtNi catalysts. From a different perspective, the compositional heterogeneity of the dealloyed particles speaks to the net Pt/M ratio as not being the essential parameter that specifies catalyst performance. Going forward, the scientific and technological impact of this work would be greatly enhanced if the size and compositional scatter of the precursor particles could be improved. It is not clear why the project adopted the synthetic path reported by the Argonne National Laboratory group in 2012. The presentation is nominally focused on $PtNi_3$ precursors, yet slides 11 and 12 indicate that the milestone satisfying material is closer to PtNi stoichiometry.
- Most of the conclusions from the project are already known and do not add new insight into the functional properties of these catalysts. The progress made since last year's report is not obvious.

### Question 3: Collaboration and coordination with other institutions

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

- The project represents a well-coordinated team effort that is led by GM.
- The project is well executed; the collaboration between all partners is excellent.
- JMFC appears to be coordinating well with the GM activities.
- The team is well structured with an automotive original equipment manufacturer (OEM) as the prime and a material developer and universities as subcontractors. The roles and responsibilities of the partners are clear and the reviewer thinks the technical results are the outcome of a strong collaboration.
- The project has several partners and defined roles. The inclusion of a leading OEM and a catalyst synthesis company are particularly strong and the efforts of team members appear to be well defined. How the efforts of Northeastern University (NEU) and George Washington University (GW) were leveraged in the rest of the project is unclear.
- The collaborations appear to be transferring knowledge from universities to catalyst supplier and from OEM to catalyst supplier to provide information on what makes active catalysts and feedback on performance and durability, resulting in an improved catalysts produced in large batches.
- The link between the work at GM, JMFC, and MIT is clear. Hopefully more insight will be forthcoming from MIT, NEU, and GW once the "rush of fascinating used MEA-samples" that satisfied milestones one and two have been examined.
- The collaboration is going well, but it seems independent of each pair. More organic and effective collaborations can be carried out among the collaborators.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The work has satisfied the 2017 DOE mass activity and loss targets and is approaching the PGM total loading goals.
- This project, led by the OEM, covers three major challenges that are very important for automotive commercialization.
- The project addresses cost and durability barriers and is directly targeted at meeting cost and durability goals in the DOE *Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan*.
- The project is well aligned with the DOE objectives that aim to reduce cost and improve the durability of PEM fuel cells. It focuses on dealloying $PtCo_3$ and $PtNi_3$ catalysts to improve durability, while maintaining high catalytic activity of the alloy catalyst.
- Catalyst mass activity and catalyst durability are key target metrics that require parallel improvement for long-term commercialization. This project is addressing the Pt particle durability and the mass activity, and thus, cost. However, the project neglects the other portion of catalyst durability, which is the catalyst support.
- The project is well aligned with DOE goals and objectives. It addresses the key fuel cell concern of catalyst cost and durability while maintaining performance. In the past year, the team has been able to meet the DOE targets for electrocatalyst and MEA performance using industrially relevant materials and approaches.
- The adoption of low-Pt loading materials (retaining performances and durability) is crucial to achieving the cost targets allowing broad commercialization of FCEVs. The reviewer wonders whether this work should be combined with the optimization of catalyst-layer properties (e.g., water management) that is perceived as a big driver to be able to fully leverage the low-Pt materials.
- Considering the lack of novelty in this project, it is hard to envision the substantial impact to the Hydrogen and Fuel Cells Program. Despite the unfortunate choice of systems and poor quality of samples, the team is capable of conducting a systematic approach that spans from fundamental insight to real application. For that reason, the set of knowledge accumulated from this project may serve in the future as a valuable set of results that justify utilization of Pt alloy in PEM fuel cells.

## Question 5: Proposed future work

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

- The shift to short stack is good. The model validation of demonstrated performance change/durability is preferable.
- The proposed future work addresses the most important issues. With little time remaining in the program, a down-selection of catalyst systems in order to focus on one system seems appropriate.
- This project is ending in this fiscal year, and the outcome does not seem to serve as a foundation for the future work on dealloyed catalyst.
- The future plans seem adequate, but are possibly a little aggressive for the remaining six months of the project.
- The work performed to date on the project has yielded significant benefits. The future work is focused on minor tweaks to the system as optimized to date, with a focus on fine-tuning dealloying, employing thinner membranes, and modifying syntheses/treatments. It is unclear that these materials can offer much in terms of further improvements. Clear rationales explaining why further improvements in performance or durability are possible, or why the approaches being suggested are likely to succeed, are missing.
- A number of the future work items seem to be from reviewer comments from the 2012 DOE Hydrogen and Fuel Cells Program Annual Merit Review, instead of the comments being addressed in the past year. The future work should include measurements of the catalyst durability, including the support durability. If this project is relying on the synthesis of the catalysts on nonviable supports, this effort could be wasted. It is unclear what they are actually going to do in terms of "Advanced Characterization."
- The future work seems to be in line with the strategy to progressively increase confidence in the technology. It is not clear to the reviewer how the porous catalyst will interact with water and whether the project team has investigated this aspect. In particular, if ice formation could happen in the porous structure or its proximity as a consequence of low-temperature events, the reviewer would invite the researchers to conduct degradation studies to better understand this phenomenon. On a similar note, it would be interesting to understand if the catalytic structure has an impact on the water transport (e.g., if a higher retention of water may yield to a lower dynamic response when passing from low to high current density).
- The NEU/GW side of the team needs to move forward with the "rush of used MEA samples" provided by GM. In addition to characterization of the aged specimen's closer correlation, like that shown in slide 11, the as-formed catalysts are called for. Throughout the presentation, the as-formed materials are referred to as $PtNi_3$ (e.g., slide 12), yet the composition of the as-formed catalysis appears to be a function of the particle size (e.g., slide 11). In this regard, attributing variations in the composition of the dealloyed particle-to-particle size might be misleading without a proper assessment of the variations evident in the precursor. The parting limit for face centered cubic alloys (Erlebacher and Newman) is thought to be near 58.4% of the reactive metal. How this impacts the aging trajectory of the nanoparticle population is an interesting question, with the authors presenting some very useful data in this regard.

## Project strengths:

- This project is led by the OEM.
- The project has a strong and well-coordinated team.
- This project has good collaborations across the project.
- Commercially relevant materials with the ability to meet DOE's targets have been demonstrated in fuel cells and have maintained performance following ASTs.
- This project has a strong team effort. The potential to conduct systematic evaluation of the structure function correlation is positive.
- Excellent progress has occurred in the last year that includes satisfying milestone one and two. The characterization work shown in slides 11 and 12 is most welcome. This program represents the key bridge that connects exploratory laboratory activity with real-world applications, and, as such, is a key pillar in the DOE Hydrogen and Fuel Cells Program portfolio.

### Project weaknesses:

- The ability to further advance these materials beyond what was demonstrated in the project is unclear.
- Poor-quality samples and a lack of novelty in the approach and choice of systems are weaknesses.
- The quality control/cost for further scale-up and the model's capability to validate demonstrated performance change/durability related to material structures are weak.
- It will be nice when the full power of the NEU/GW subcontractors characterization capabilities are brought to bear on the latest generation of catalyst that satisfied milestone one and two. Improvements in the compositional and size dispersion of the precursor catalyst particles would greatly improve the quality and strength of the conclusion and insight that will come from this study.
- The lack of samples due to GM's relocation from New York to Michigan is a weakness. They state that "0.6–1.0V cycling is too aggressive for vehicle operation. Could induce carbon corrosion. More relevant tests than 30,000 cycles at 0.6–1.0 V are probably required." However, they have yet to examine whether the catalysts being developed are being put onto stable substrates. The substrate plays a key role in the particle precipitation, yet they are ignoring this key parameter, and appear to want to go to a "less stringent" testing protocol.

### Recommendations for additions/deletions to project scope:

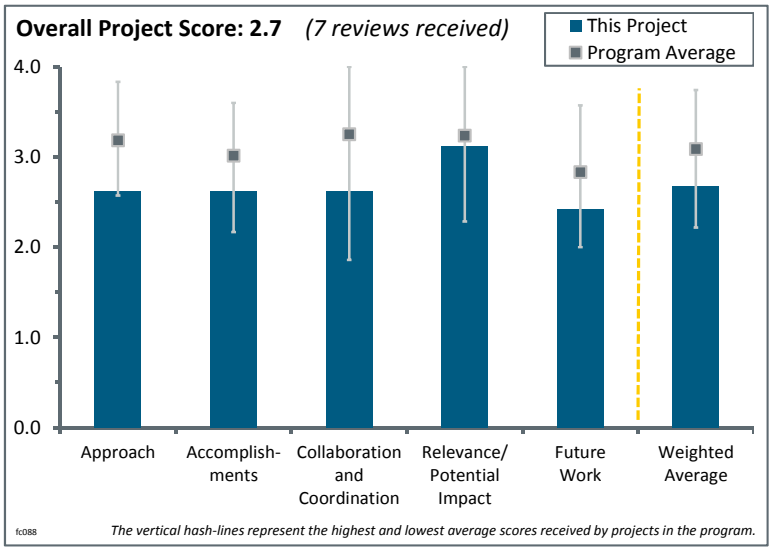
- This project should continue going full steam ahead.
- At this point, much of the need for these systems likely lies in the scale-up, introduction, and long-term testing under relevant operating conditions of these materials.
- This project should compare durability of the dealloyed PtCo/C and PtNi/C catalyst durability with the equivalent Pt/C catalyst.
- Without knowing the stability of the carbon support, the progress in this project is still unclear. It is not clear if high surface area carbon (HSAC) or a more stable graphitized carbon is being used. It is easier to get higher dispersions with the HSAC, but these catalysts are less stable during the higher potential ASTs. It is recommended that the catalyst support and the stability of this support, as well as AST testing of these materials, be conducted and presented. GM should end all activities for Ir as their own company guidance to other developers is that Ir will never work. If this is a reasonable path, it seems odd that there are comments in the past from GM to other projects to never use Ir. The ternary systems work should concentrate on other components that are low cost in contrast to the Au, Ir, and Pd explored to date. The effect of de-alloying different atmospheric conditions should be explored (in air versus $N_2$). The researches should also examine the effect of Ni leaching on the performance and durability of the other components in the system. It would be interesting to understand how this work compares to the similar catalyst develop by Stonehart et al., for phosphoric acid fuel cells in the 1980s.

## Project # FC-088: Development of Ultra-Low Platinum Alloy Cathode Catalyst for PEMFCs

Branko Popov; University of South Carolina

### Brief Summary of Project:

The objective of this project is to develop high-performance, low-cost, durable cathode catalyst and support. The research focused on optimization studies of carbon composite catalyst (CCC) supports, development of a process for the in-house synthesis of carbon nanocages (CNCs), development of an advanced hybrid cathode catalyst (HCC) based on a CCC support with low-platinum (Pt) alloy loading catalyst, development of CNC supported Pt-alloy catalysts, and development of high-volume procedures for the synthesis of a promising catalyst.




### Question 1: Approach to performing the work

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

- The approach has been consistent in terms of trying to meet the targets set out by the U.S. Department of Energy (DOE).
- The basic approach to develop a catalyst-support system with a corrosion-resistant, heat-treated support is valid and seems to result in high activity electrocatalyst systems that are close to the DOE targets.
- The approach based on synthesizing graphitized carbon and then finding adequate Pt deposits apparently worked well. However, there is no information on why it worked and if the Pt-C interaction is determining the activity and stability. Only the degree of graphitization is considered.
- This project seeks to simultaneously develop more durable support materials, some of which have $O_2$ reduction activity, and more active Pt-based catalysts. The experimental approaches taken towards these ambitious goals are reasonable and have produced some positive results. It is not clear that the membrane electrode assembly (MEA) testing is yet state-of-the-art and capable of demonstrating the true potential of these catalysts. The idea of combining some ORR activity of the support (to carry current primarily at low potential/high loads) with low loadings of Pt (to provide current at high potential/low loads), the HCC really needs to be checked by modeling that can adequately handle the mass transport losses in the thick films needed to get useful currents out of the active support. Not enough results from the modeling shown in the reviewer-only slides are given to demonstrate that this issue has been adequately considered. It is essential that the hybrid catalysts be tested under $H_2$/air conditions, and it appears that this work is just starting now.
- In general, the approach has significant weaknesses. First of all, the designing "rules" for the ORR are ill defined and it is still difficult to understand what is unique with PtNi and PtCo preparation methods. It is more like making materials and testing them.
- The principal investigator (PI) claimed in his talk that Pt is better than any alloy. This seems to contradict several other speakers and undermine the very title of this project (alloy). The researchers must provide more proof when making such claims. It is not clear why alloys are being studied in this project. It seems to meet all targets based on DOE protocols. This project needs to evaluate the new U.S. DRIVE Fuel Cell Tech Team accelerated stress tests (ASTs) on carbon corrosion (1–1.5 V cycling instead of 1.2 V hold). This is a new change and should be followed for the rest of the project. The PI did not answer questions satisfactorily. He started talking about $H_2$ crossover when asked about $H_2$ evolution in the cyclic voltammograms (CVs). The electrochemical surface area should be determined from CVs that do not go

down to the $H_2$ evolution region. The approach could be better stated with a slide that shows exactly what gains they hope to achieve by using active supports. It is not clear if the support is actually contributing to better activity or if the activity of Pt overwhelms anything from the support. The support corrosion durability is also being improved by increasing graphitization temperature. All this should be stated simply in one slide with clear criteria for go/no-go decisions.

- The overall project approach and objective are unclear from the presentation. While it appears that the approach is to utilize an ORR active support for platinum group metal (PGM) catalysts to reduce the cost of the catalysts, the loading of Pt on the CCC support is as high as what is utilized in current state-of-the-art materials, and an initial performance benefit was not demonstrated. The Pt-based metal catalysts are achieving high ORR activities, but these are similar to or lower than what has been demonstrated for the same classes of Pt alloy catalysts on traditional carbon supports. It is not clear if the approach of this project is to enhance the ORR activity of non-PGM catalysts by adding Pt, or if it is to enhance the ORR activity of Pt by supporting it on an ORR active support.
- The approach appears incremental, at best. Pyrolysing carbon blacks at a variety of temperatures with or without additives have been a part of heterogeneous catalysis and electrocatalysis for a long time. This project does not move the dial very far, if at all. Giving a new name to an old carbon does not make it new, nor does placing Pt on it make it either "advanced" or "hybrid." The project approach would be improved by bringing some focus to explaining what, if any, the differences are between commercial carbons and the CCC or CNC, why those differences may be important in durability/activity, and how the comparative characterization/electrocatalysis/durability data supports their hypothesis. The electrocatalytic studies are appropriate; the accelerated durability testing parameters could be better defined or illustrated.

### 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 team has made good progress year after year, especially in terms of evaluating the catalyst technologies developed and making some key advances in understanding how to control Pt particle size on the support material. The team has a very short list of candidates that have the capability to meet the DOE targets.
- The PIs made a small amount of progress towards the objectives, especially if the results are compared with the catalytic activity of ORR obtained from the other groups.
- The catalysts' performance and durability are close to DOE's targets for 2017, but they are not yet there. There is no information on the alloy composition after the tests of durability. The Co content in $Pt_1$-$Co_1$ alloys should be known given the devastating effect of $Co_2$+ or $Ni_2$+ on a membrane's conductivity. The project needs techniques that provide more basic structural and molecular-level information.
- High catalyst activities close to DOE's target at the beginning of life have been demonstrated in this project in the MEAs of fuel cells. Although the support is supposed to be very durable after very high-temperature heat treatment, the support still corrodes and results in activity losses of approximately 50% compared to the baseline Pt/C in durability studies, as seen in slide 15. The goal of a more durable support has not been achieved.
- The activities and durability of the Pt/CCC should be compared to state-of-the-art Pt/C rather than comparing the activity of HCC to Pt/C, which includes the effects of both the unique carbon support and the alloying effect. An impressive number of iterations have been synthesized and tested in this project. The most promising at this point are the Yonsei University (YU) catalysts, but these should be compared to their counterparts using traditional high-surface-area carbon (e.g., Ketjen) and graphitized carbon for a valid comparison and to validate the premise of the project.
- The project accomplished a lot of work if work is measured by the sheer number of data panels on the slides. There is far too much data, far too little interpretation in the presentation to result in any clear accomplishments and progress. To deflect criticism that these materials are not novel, effort should be put into demonstrating that the carbons that result from the modification of carbon black is somehow different than, say, the pyrolysis of carbon black. The evolution of products is not clear; they could arise from the methanation of the amorphous fraction of the carbon black. The fate of the surface modification additives is also unclear (e.g., where does the $O_2$, $N_2$, etc., from the additives go?). The project should use a TGA/MS

to at least give some idea as to mass recovery and off gases of the pyrolysis step(s) such that reviewers can ascertain the "novelty" of the support. Essentially, there are no elemental characterizations of the carbon or of the catalyst to allow one to determine the uniqueness, if any, of the compositions.

- Progress since the 2012 review appears to be limited. The 2013 slides show only one catalyst, the YU $Pt_2Ni$/CNC (in both unleached and leached forms), meeting both the initial activity of 0.44 A/mg of Pt and the loss of less than 40% of the mass activity after 30,000 cycles 0.6–1.0 V. The same catalyst was shown meeting these targets in the 2012 slides. The 2012 slides showed a University of South Carolina (USC) $Pt_2Ni$/C at 0.45 A/mg of Pt, but in 2013 this is listed as 0.43 A/mg of Pt (not a significant change), and presumably the result of additional experiments pulling down the number slightly. New and productive work was done on new generations of the modified carbon supports (CCC), including more durability testing. The performance of MEAs at moderate- to high-current density in $H_2$/air testing is still below the targeted values, even before aging and even with iR correction, which is not commonly applied to $H_2$/air tests.
- The ECSA losses reported are artificially low since the ECSA scans are taken down to very low potentials (<0.85V) where there is significant $H_2$ evolution. The PI should repeat these measurements down to a reasonable voltage and then report the percentage of losses. These will be significantly higher than those reported in this talk. Once they do this, they will find that their ECSA percentage losses are a lot more consistent with their mass activity losses, which are far greater than ECSA losses now. In slides 10 and 11, the cycling is done to an upper potential of 0.925V, but in slides 12 and 13 the upper potential is 1.0V. The DOE protocol is 1.0V. It is unclear why there is this discrepancy. More post-characterization data and the particles sizes and electrode layer thicknesses before and after the various durability tests should be presented. These should be shown in addition to the polarization curves. A slide summarizing the behavior of all three catalysts (fresh and after the two AST protocols) would be very useful. It is not very easy to evaluate the progress since the details are missing. While there is lot of polarization data, there is no link to what was done to achieve this improvement. The reasons behind the improved performance of the various catalysts are not clear, nor is the difference between the 2012 and 2013 catalysts (first generation and second generation HCC). The durability of the CCC is not clear. After durability tests, it is not clear if the transition metal is still there. It was also not clear if there are any post characterizations of the carbons used in this study, if they still retain activity, or if they contribute to the activity of the overall catalysts (not just the overall catalyst activity controlled by Pt).

### Question 3: Collaboration and coordination with other institutions

This project was rated **2.6** for its collaboration and coordination.

- The collaboration is strong and this is a strong component of this project.
- This project has significant collaboration with the Korean universities and companies.
- There are many collaborators, some for a short time. The coordination of their efforts should be improved.
- The collaborators from YU appear to have made a very significant contribution in terms of helping the team to reach their milestone for the project.
- This project is rather narrow in its collaborations versus other catalyst development projects, especially in the area of characterization and computational studies. Some of this was apparent in the reviewer-only slides, but the impact of the characterizations and the impact of modeling on catalyst layer design should be indicated.
- It is not apparent what the level of collaboration, communication, or coordination was with the other partners as few references were made to them. This perception may, in part, be due to the overwhelming data content of the presentation.
- The official partners on this project are YU and Hyundai.YU and USC are developing their own catalysts and Hyundai had no funding last year. This is not a good collaborative effort. The PI did not mention the percentage of effort or funding that went to the collaborators. The PI needs to interact better with other players in the field (original equipment manufacturers, laboratories, and other universities) to help scale up and validate these catalysts. The additional interactions on the TEM studies are good.
- The presentation did not make clear whether the activities of the prime- and sub-contractors are coordinated, or if they are essentially independent activities. The presentation did not make clear where the MEA testing was done. In the absence of such clarification, one must assume that it was done at USC. The

level of involvement of the automotive subcontractor was not at all clear, and the line about the interruption and restart of funding leaves the reviewer with unanswered questions. The data on the YU catalyst with good activity seem to be the same as those presented in 2012. The collaboration might be better facilitated if the PI made a conscious effort to listen to, and thoughtfully consider, what others have to say and to achieve greater brevity in his own answers to questions.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The project aligns with cost reduction and activity targets for catalysts.
- The project is focused on PEM fuel cells development.
- The project is developing catalysts that can meet the DOE targets and, as such, is very relevant.
- The project is relevant to obtaining high activities and durability of catalyst systems to lower costs and speed up the commercialization of automotive PEM fuel cells.
- Most project aspects align with the Hydrogen and Fuel Cells Program and DOE RD&D objectives.
- Given the highly incremental nature of this approach, it is unlikely that this project will significantly advance the state of the art in fuel cell electrocatalysis.
- The topical area of developing ultra-low Pt catalysts having appropriate durability is highly relevant to DOE's objectives in developing long-lived, cost-competitive components for fuel cell vehicles. This project is aligned with those goals, but the approach will not significantly advance RD&D toward the goals.
- The potential impact of a synergistic effect between an ORR active support and a PGM catalyst in reducing PGM loading and cost is large. In theory, this project is relevant to DOE's goals if the PGM catalyst loading can be vastly decreased using this concept. However, what was presented this year utilized high Pt loadings on the support and appeared to just reduce Pt loading through alloying, which is the focus of many other DOE projects.
- This project addresses the activity and durability of catalysts, supports, and complete MEAs and, as such, is well positioned to improve the state of the art of PEM fuel cells for a wide range of applications. The modest progress on several fronts and the weaknesses in MEA performance to date, coupled with the inadequate modeling analysis of the prospect of the hybrid catalysts, raise doubts as to whether the project can achieve enough of its very ambitious goals to positively impact the field.

### Question 5: Proposed future work

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

- The proposed work appears to result in small incremental improvements. Similar treatments of the catalyst's components have been already tried.
- It will be important to have the industry-led evaluation of the technology/other proof points from independent groups.
- The proposed future work should focus on reducing the Pt loading on support by increasing the ORR activity of both the alloys and the non-PGM support.
- It is not clear how the go/no-go decision is going to be made. It is unclear if the best of the three tested catalysts can automatically go forward and if this catalyst needs to meet some minimum requirements. There needs to be some comparison made to state-of-the-art catalysts that can be purchased commercially. Currently, all catalysts are being made at USC.
- Future work seems evolutionary with small modifications to materials. The corrosion resistance of the CCC and CNC should be carefully measured in ex-situ measurements and compared to Ketjen Black and Vulcan and graphitized Vulcan to get a quantification of improvement of these new materials. Currents in A/mg and A/cm$^2$ carbon should be plotted to investigate if these carbons truly have better corrosion resistance that is worth the expensive heat treatments. The supplementary slides showed an effect of Pt loading that is highly questionable and reverses the trends of what is expected. This may have to do with the method of catalyst deposition or spraying onto the membrane.

- The future work section appeared to be mostly regenerated from ongoing work. The goals for future materials improvements were stated, but the pathway to reach those goals was poorly explained, if at all. Carbon modification plans included purification, etc., but the underlying justification for doing this, which was perhaps obvious, went unstated as did the methods for obtaining and confirming that the purification process resulted in changes in compositional/structural properties of the supports. The future approach would be improved by detailing what the hypothesis behind the surface modifications and purification is and then addressing the data to be obtained to prove or disprove the hypothesis.
- The proposed work under the no-cost extension is reasonable, though proper modeling of the hybrid systems, using the kinetic data from the support-only ORR data, and adequate inclusions of traditional treatments of mass transport within the catalyst layer should receive greater emphasis. Making good high-activity electrodes that meet DOE activity and kinetic durability targets on corrosion-resistant supports, as done in this project, is very difficult and modest shortfalls versus numeric targets should not necessarily be taken as a major failure. The viability of the work proposed under Phase II (if the go/no-go is passed) is dependent on improved MEA and short-stack testing capability, either at USC or through partners.

### Project strengths:

- This project used great teamwork to get to the milestone.
- This project closely meets the DOE targets for activity.
- This project has plenty of performance data on various novel catalysts.
- This project is focused effort on two to three factors determining catalyst performance.
- Any strengths were difficult to ferret out of the extremely voluminous amount of material presented.
- The PI has a strong background in carbon chemistry and in electrochemistry. The PI is highly experienced with synthesizing materials with unique stability and activity.
- Some progress was made in taking on simultaneous improvement of support stability and catalyst activity/durability. The project's modified carbon supports do appear to be significantly more stable against corrosion (as would be seen in unmitigated start/stop) than are conventional carbon supports. Progress has been made in achieving reasonably high activities with low levels of corrodible, non-noble metal atoms in the electrodes. The development of partially chemical pathways for producing graphitized carbon supports is a strength, compared to the inert thermal annealing at much higher temperatures than is commonly used.

### Project weaknesses:

- Heat-treated CCC and CNC support do not show high corrosion resistance.
- There was no comparison to commercial materials, no clear understanding of the role of the catalyst carbon support and its contribution to HCC activity, and no clear understanding of the durability of the CCC.
- This project would benefit from techniques that can provide relevant basic structural and spectroscopic information. Protocols used to judge the technology may not be aggressive enough and may thereby give a false sense of security in terms of applicability to industry.
- The systems are in general ill defined. The measured activity/stability is subject to debate. The PI is still questioning the existence of Pt-Ni/Co alloys. If it is not an alloy, then it is not clear what it is. More fundamental knowledge is needed to improve the stability and activity of the systems.
- This project has far too much data and too little interpretation and supporting evidence to support the researchers' hypothesis that these materials are "novel." This project appears to be incremental.
- The kinetic activity targets before and after cycling were met, at best, marginally. The MEA performance at moderate and high-current density is below expectations, and catalyst utilization numbers at higher loadings are unusually low. There was a lack of adequate modeling analysis of the extent of advantages at high-current density that can be realistically expected from use of a carbon support with limited ORR activity.

### Recommendations for additions/deletions to project scope:

- One-to-one Pt-to-Ni or Pt-to-Co alloys are not likely to provide useful electrocatalysts.
- This project is unlikely to lend support to moving DOE's RD&D objectives forward and should be considered for redirection or early completion.

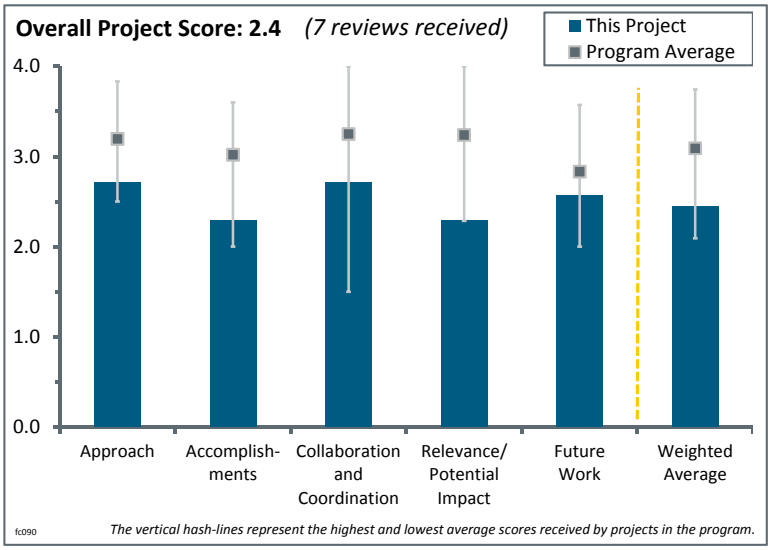
- Only those catalysts that show improvement over state-of-the-art commercial catalysts should go forward to Phase II. Hopefully, these are evaluated somewhere other than at USC, maybe at Hyundai, who is one of their partners.
- The proper modeling analysis—using available kinetic data and standard analyses of mass transport within an electrode layer—of the magnitude of advantages that could be realistically expected at high current density in $H_2$/air for a hybrid catalyst incorporating a low loading of Pt on a carbon-based support. It is not clear if one can get a meaningful augmentation of total performance at high current density from the ORR-active support in a layer sufficiently thin that it does not throttle off the performance through mass-transport limitations.

## Project # FC-090: Corrugated Membrane Fuel Cell Structures

Stephen Grot; Ion Power

### Brief Summary of Project:

The objective of this project is to pack more membrane active area into a given geometric plate area, thereby allowing both power density targets and platinum (Pt) utilization targets to be achieved. The project aims to demonstrate a fuel cell single cell with a two-fold increase in the membrane active area over the geometric area of the cell by corrugating the membrane electrode assembly (MEA) structure. An ultra-low, Pt-loaded, corrugated MEA structure will be incorporated in a single cell that achieves the U.S. Department of Energy (DOE) 2015 Pt loading target while simultaneously achieving power density targets.




### Question 1: Approach to performing the work

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

- This project has a good approach to increasing MEA area.
- The approach of making corrugated, three-layered MEA (five-layered MEA, including the gas diffusion layers [GDL]) is a good approach to condense larger active surface areas within the confined, two-dimensional (2-D) space. By moving from a 2-D, flat MEA geometry to a corrugated MEA geometry a factor-of-four improvement in power density can be realized. The approach clearly addresses the high power density barrier and is well integrated with the DOE goal.
- The approach is interesting, but there are questions regarding how the stack is going to be assembled.
- The creativity and ambition of the project are commendable; however, there are many challenging technical issues with the approach.
- Corrugated MEA may increase the surface area and potentially enhance the performance, though there are a number of challenges to making that happen.
- While this is an interesting approach to increase the volumetric power density of a PEMFC stack through a corrugated cell and flow-field design, it is very technically challenging and requires a completely new flow-field material set and cell support structure.
- The corrugated MEA approach is unlikely to overcome the cost and performance barriers as this project intends. The complex MEA construction and the use of a metal wire screen will likely lead to higher costs. The corrugated structure may have performance issues related to heat and mass transfer, electrical resistance, and durability. The latest performance results in slide 17 are still substantially lower than conventional MEA design.

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

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

- The project has gotten into modeling and structural/thermal/electric analysis, which provides a clear view of the potential challenges and risks. The project has also moved on to a practical way to demonstrate if it

can eventually be made. The modeling can be improved by introducing a swelling change and by a related conductivity change.

- Progress was made on GDLs. The project needs to address membrane swelling in corrugated structures. Based the on figures shown, current densities may be larger in MEAs and GDLs without changes in cathode/anode plates. It is unclear how this will impact MEA durability and whether General Motors (GM) will be able to support the development of improved seals.
- Most of the results in the last year were modeling by one of the subcontractors. It is not clear how much progress was made by the project lead and the other subcontractor. The overall progress is modest.
- Many tasks are behind schedule due to the demanding nature of the design and materials property requirements. There has been good progress in the modeling of structural and thermal stresses and voltage drops.
- Some progress has been made in establishing the feasibility of the approach. It is unfortunate that the cell design is already limited to a maximum current density (1.2 A/cm$^2$) to avoid overheating, as this undermines the original premise of increasing power density.
- It is unclear what the gain is in terms of cost and processing, especially with the higher areas. GM's move from New York to Michigan impacted the project significantly, and there needs to be a plan going forward or the project is delayed. The researchers need to understand how membrane swelling is going to impact durability and performance. There are problems getting the membrane to hold its shape. This project has a good analysis of operating conditions, but this could be a big limitation.
- Very little progress has been made in corrugated MEA development, which is the most important part of the project. The membrane forming tool was made, but the membrane could not be formed into a corrugated pattern. Polymer membrane, such as Nafion®, is known to change its dimension and morphology upon humidity change, and it is not expected that the membrane can be formed into a permanent, rigid, corrugated structure. The team should take the initial approach of making a corrugated GDL, which will support the MEA to remain in a corrugated shape in the cell. It is not clear why the team is taking a Cu membrane approach to meet the cell-compression, mediated, cell-impedance target. The target should be met with a functional, corrugated fuel cell MEA, not a Cu membrane. The team should focus more on making a functional corrugated-structured MEA that can be evaluated in the cell.

### Question 3: Collaboration and coordination with other institutions

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

- The collaboration has led to data comparison and improvement—even though only limited data are available.
- The team consists of good partners, GrafTech and GM, who can address the GDL and testing/validation of the corrugated MEA concept. GM and GrafTech can help develop a workable stack using corrugated MEAs.
- It is evident that this project has many relevant collaborations. The team is well-balanced and has an automotive original equipment manufacturer (OEM) to insure that the project is relevant to automotive applications.
- This project has good collaborations with the OEM that should be able to provide input into feasibility. This project should also look into working with other suppliers of GDLs, including those who work with open field and foam structures. With the removal of GrafTech, it is unclear what is going on with their portion and involvement.
- The researchers need to improve the overall design architecture with GM.
- The suppliers appear to be responsive to the effort; the subcontractors are less so.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

This project was rated **2.3** for its relevance/potential impact.

- The project is relevant to the objectives of the DOE *Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan*.

- The activities are aligned to DOE's goals. This project brings in an unconventional approach of using corrugated MEA to access higher active areas and, hence, harvesting higher power density from a defined stack volume.
- The researchers have to wait until a fuel cell stack using the corrugated MEAs shows promising results.
- Any changes that can realize a two-times increase are good to explore. However, the project has not fully described an end-to-end solution for making a cell. Perhaps this was in earlier presentations.
- New approaches to overcoming the limitations of current conventional hardware are relevant to the field, but the potential impact of this project is low due to the many challenges inherent in the approach.
- This project falls in the high-risk, high-reward category. It could have a large impact on volumetric power density and reduce the cost of the stack attributed to bipolar plates; however, it is not certain if the manufacturing costs, which would appear to be higher for this non-planar structure, will outweigh the cost savings from reduction in a bipolar plate area.
- It is not necessarily fair to compare on the geometric basis instead of membrane basis, and an effort should be made to compare it to a non-corrugated system with the same membrane area. This can be used to determine the specific targets that one could gain with the corrugated structure. Pt utilization was not presented in this talk.

### Question 5: Proposed future work

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

- The reviewer is looking forward to seeing more data come out of the project.
- The actual operation of a convoluted MEA is, of course, critical and justifiably the primary focus of the future work.
- The de-bug work and approach to fixing the sealing issues are not described. The plans that are on the table to solve these problems are not clear.
- The proposed future work was very broad and what was shown in the presentation is the deliverable of the project.
- The project is ending and not much was presented. It is not clear if the researchers will meet the go/no-go and what will happen in the last few months after that.
- The future work described is partly aligned with the proposed work of the project. The team not only needs to focus on sealing issues, but also on the corrugated MEA fabrication. So far, the team has not come up with a fabrication method of a good performing corrugated MEA. There is no clear outcome on the types of GDL, membrane, catalyst materials, and fabrication process that will be used to make such corrugated MEA. The team also needs to plan the performance and durability studies of corrugated MEA under various temperatures and relative humidity (RH) cycling.

### Project strengths:

- The MEA area increase is a strength.
- The novelty and strength of collaboration is a strength.
- This project's strength is providing a high-surface-area MEA.
- This is an interesting way to change volumetric power density.
- The team is well organized and capable of evaluating such a different idea.
- The strength of this project is creativity, starting with the original idea and then formulating solutions, such as the ability to quickly develop an alternative wire mesh-based GDL when the original was inadequate.

### Project weaknesses:

- The complexity of design and a need for new materials sets is a weakness.
- The project needs to address the catalyst accessibility versus high MEA geometric surface area.
- While significant technical challenges should not necessarily be perceived as a weakness, the magnitude and variety of formidable technical challenges are overwhelming in this project.

- This project does not appear to be a game changer, only a way to increase MEA area. The impact on cost needs to be shown, especially on an MEA area basis, and the problem of being able to only run at lower current densities seems to limit the applicability and gains in volumetric power density. There are membrane and formation issues.
- The concept may have some theoretical merits, but the proposed technical approach may also lead to performance issues related to heat and mass transfer, electrical resistance, and durability. The project timeline has passed the 60% mark, yet the results and progress so far are not encouraging. One of the barriers this project is trying to address is cost; yet there is no cost target. It is very likely that the proposed cell structure will incur a higher cost, and a cost target should be established.
- The team does not have a clear plan for fabricating corrugated MEAs. The team should down-select GDL membrane and catalyst coating methods that are appropriate for making corrugated MEAs. The team assumes that, in a corrugated MEA system, it would not incur any additional stress around the corrugation during temperature and RH cycling. The team also needs to consider the heat dissipation mechanism along the wall of the corrugated surface of the GDL/MEA, which is not in contact with the bipolar plate; hence, the effect of coolant will be very little along the walls of the corrugated surface and can give rise to heat-spots resulting in the formation of pin-holes in the membrane. The heat management and, hence, the cooling of the MEA in a stack, is always a challenge and the team should consider heat management challenges for corrugated MEA seriously.

### Recommendations for additions/deletions to project scope:

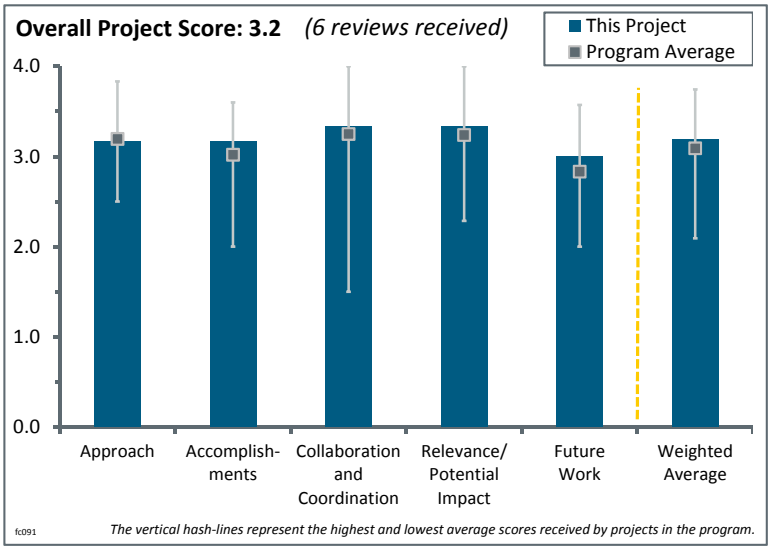
- It is suggested that the project make full cell stacks to figure out all the issues and then lay out the plan to address those issues.
- The whole project approach lacks a solid fundamental and engineering foundation; its lack of progress is not a surprise. It is questionable what we can really achieve by continuing the project.
- While modifying the hardware for obtaining proper compression of the membrane for sealing purposes, the team should get the appropriate MEA materials for making rigid and corrugated MEA that can be compressed in a cell.
- It is late in the project, but it would be nice to know (a) if the proposed design, with its more complicated manufacturing process and new materials set, would be more costly than that of state-of-the-art conventional stacks and (b) if the cost savings due to reduced use of bipolar plate material would be negated by increases in cost in these other areas.
- This project should look at doing a more detailed cost model for the system to show the benefits and the targets needed to reach them. It is not clear what the impact on the stresses on the material and MEA properties are; this should be correlated to work that is ongoing in other DOE projects. Perhaps this project should try reinforced and stiffer membranes. The project needs to think about water management issues and multiphase flow aspects in this structure where one could get water films and channeling where it would be detrimental.
- So far, the project has described a manifold configuration for single cells that is already much thicker than the convoluted structure. Manifolding the anode, cathode, and coolant flows along the two entrance and exit sides of the parallel channels in a stack will be very difficult without likewise thickening the unit cells appreciably, thus negating the benefit of the convoluted MEA. Provided the first successful convoluted cell is accomplished, the project should invest in an effort to develop a stack design that shows how the manifolding can be efficiently accommodated.

## Project # FC-091: Advanced Materials and Concepts for Portable Power Fuel Cells

Piotr Zelenay; Los Alamos National Laboratory

### Brief Summary of Project:

The objectives of this project are to develop advanced materials (for catalysts, membranes, electrode structures, and membrane electrode assemblies [MEAs]) and fuel cell operating concepts capable of fulfilling cost, performance, and durability requirements established by the U.S. Department of Energy (DOE) for portable fuel cell systems. The project will also explore the development of a path to large-scale fabrication of the successful materials.




### Question 1: Approach to performing the work

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

- The approach is very reasoned and sensible. The team includes seven individual thrusts but the principal investigator (PI) holds it together. Despite some apparent isolation of the individual efforts, they are all of high quality and are focused on DOE milestones and barriers.
- The multi-directional approaches taken for the completion of all tasks are adequate. All the technical barriers had been addressed appropriately. The responsibilities for anode, membrane, alternative fuel development, and performance/durability testing were given to the research teams with significant experience and strength in respective areas of research.
- The approach is really a collection of small projects with no cohesion or overall focus. Interesting catalyst work is done, as is interesting alternative work, but there is no connection between the two, for example. This project has a large number of groups each going on their own way with no reason as to why they should be put together in one group. For example, ethanol and dimethyl ether (DME) will provide unique membrane challenges and it is unclear if the membrane group is targeting these. Doing durability measurements for 100 hours is not a good approach, and not much Ru dissolution should be expected. This short test does not prove the stability of a catalyst.
- The oxidation of small organic molecules on Pt-Ru-X (X = Sn, Pd etc.) electrodes has been an "attractive" research direction for decades. Therefore, it is surprising that in this proposal these systems are considered to be advanced catalysts. Although the PIs have mainly focused on activity, very little is presented about the stability of non-noble components (e.g., Sn and $SnO_2$). It is also surprising that Cu-based nanowire (NW) has been used as a "support" for depositing Pt-Ru alloys. Having Cu in fuel cell environments is undesirable because even a trace level of Cu (from Cu underpotential deposition) would have a huge inhibiting effect in the electrocatalysis of small organic molecules. The computational approach the PIs used to find a new generation of catalysts is appreciated; however, the leading role of density functional theory still draws some skepticism. This is simply because, in addition to strain, which may or may not have a significant effect, the defects are usually playing a much bigger role, especially those created on thin metal films. The combination of FTIR with electrochemical experiments is good and is the right direction to go. In general, the approach is good, but could be improved.
- The overall approach in this effort is to develop advanced catalysts and materials concepts for direct oxidation fuel cells designed for portable power. It incorporates (1) advanced anode catalysts, (2) electrode structure, (3) hydrocarbon membranes, and (4) extending to more complex fuels, such as ethanol and dimethyl ether. The PIs identify a project success for direct methanol catalysts on the basis of ultra-thrifted PtRuSn catalysts where 200 mA/mg Pt activity is claimed. This catalyst is not new and the performance

improvements in terms of lower loading, etc., are mostly a function of electrode structure and possibly the higher concentration methanol (2M) and elevated temperatures of 80°C. These systems have been extensively tried in several commercial entities with very limited success in terms of its sustainability. This aspect is manifest in the durability tests where, instead of a full cell cycling test, a DHE-based half cell is used with $H_2O$ (2M MeOH) at the working electrode and $H_2$ on the counter electrode. This does not provide a proper projection to cell operating conditions, wherein cycling and resultant effects at the interface cause severe losses. It is suggested that this approach to a durability test will never provide proper information on the stability of these systems in a working direct methanol fuel cell (DMFC). In addition, the use of Cu NWs is not without its problems, with issues related to its possible poisoning of the interface. No one in the commercial arena uses Cu as an additive in any form, including core materials, such as Cu NWs, due to their deleterious effect on the cathode.

### 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.

- In a short period of time, the PIs have made great progress towards the objectives. This is particularly true for lowering the amount of Pt without paying penalties in catalytic activity.
- Los Alamos National Laboratory has made good progress on increasing the anode activity; this is the most important contribution. The researchers do not properly address whether these materials can be made at a low cost. It is difficult to understand how the membrane work fits in here. The block copolymer work for DMFC has been looked at many times, including by McGrath. It is hard to see what is novel here.
- Significant developments were reported in lowering the catalyst loading at the interface, providing a pathway towards making these systems more viable. However, faulty use of durability tests provides no projection of understanding the sustainability of these systems for practical use. The steady state chronoamperometric (CA) results are a testament to quick poisoning of the catalysts, which are in direct contrast to pseudo steady-state polarization measurements. The PI should explain the differences between the CA results and those from polarization measurements.
- The team reported that the tetramethylbisphenol A (TM)-based, multi-block copolymer demonstrated a reduction in methanol crossover by 54%. However, the 54% decrease in methanol crossover resulted in only a 6% increase in power density. The polarization curve clearly shows that the MEA containing the TM-based, multi-block copolymer membrane suffers from mass transport limitations, which may be responsible for the lower performance increase. The team should focus on enhancing the mass transport gain and maximize the gain due to lower methanol crossover.
- The team achieved the methanol mass-activity target of 200 mA/mg Pt at less than 0.25 V (80°C) with an ultrathrifted advanced anode catalyst (uTAAC), but obtained high DMFC performance with an advanced anode catalyst (AAC). The high mass activity of the uTAAC catalyst is not translating to high polarization performance due to its cathodic loss. Therefore, either of these two catalysts (uTAAC and AAC) may not be suitable to provide high performance at the system level. These two catalysts have individually achieved two different DOE performance goals; however, for constructing a high-performing DMFC power unit, only one anode catalyst with high methanol activity as well as polarization performance is needed. Having two different anode catalysts meeting two different goal targets is not practical.
- The work on "ultrathrifted" Pt/Ru/C catalysts is outstanding, with the PI strongly focused on DOE goals and strongly committed to finding and mitigating potential problems. This is as might be expected for a national lab PI, but even so, he appears to be a good group leader. Other projects are all very good to excellent. In some cases (e.g., catalysts supported on Cu and on Au), there could be more work done to address durability; in other cases, early-stage work has not yet been translated into prototype power sources. There is more to be done, but overall this is a very strong effort with many accomplishments all at the cutting edge of fuel cells utilizing liquid fuels.
- Very good progress was made in anode catalyst research. On the membrane research, it would be very useful to check how the TM membrane/electrode interface changes in a durability test to validate the hypothesis that matching water uptake to Nafion can improve interface durability. Also, it is perhaps more useful to have the membrane swelling ratio data. The milestone for ethanol oxidation catalysts is delayed. The $SnO_2$-modified catalyst made by micro-emulsion synthesis has much lower activity than the one made

by the Cu UPD method. No explanation was given. It is important to understand the cause, which can help further improve the catalyst. On the MEA durability test, the new results showed that catalyst-layer cracking has no major effect. The question still remains unanswered as to why MEA failed faster in a high-methanol concentration operation.

### Question 3: Collaboration and coordination with other institutions

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

- As in the past, the lead PI put together an outstanding team of researchers. The team consists of a good mix of university, national laboratory, and industrial partners. The collaboration with SFC Energy (SFC) and Oorja Protonics is very advantageous to the team since SFC has expertise in portable DMFC systems and Oorja has expertise in high-power DMFC systems. The team can obtain valuable information on the material/performance requirements for portable-power and high-power DMFC systems.
- The PI reports good collaborations among the partners in this effort. Some of the advanced concepts have yet to be incorporated into working fuel cells at SFC, however. All SFC results were with conventional catalysts and loadings. The highlighted advanced catalysts actually operating in an SFC cell will be proof of concept in this proposed effort.
- There are five focus areas and it is not at all clear how they are related or how these groups are working together.
- The individual efforts are all well executed, but there was not much synergy among them. They seem to hang together mostly because they are all funded by the same project.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- Most project aspects align with the Hydrogen and Fuel Cells Program RD&D objectives.
- This team has done excellent work addressing the key issues in DMFC and related liquid-fuel fuel cells for portable applications.
- Each of the project areas is relevant. Improving catalyst activity, developing better membranes, and using ethanol would all greatly help this field. It is not clear if any work was done on electrode structures. The ideas they wanted to pursue are not shown here.
- The potential impact of this effort is yet to be realized. As a practical fuel cell person, the proof of concept is yet to be shown with the new materials—be it the catalysts shown in this effort or the membrane. Most work in their limited half-cell environments, but these types of fuel cells are beset with best intentions gone unrealized. Two years on, we should be expecting some full cell results with the new materials shown in this effort.
- The project is relevant to the objectives of the DOE Hydrogen and Fuel Cells Program. This project is focused on the development of advanced materials, such as catalysts, membranes, electrodes, and membrane electrode assemblies for DMFC application, which is expected to be capable of fulfilling cost, performance, and durability requirements established by DOE and are very important for the commercialization of DMFC technology.

### Question 5: Proposed future work

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

- The future work described is aligned with the proposed work of the project.
- Proposed future work is a sensible extension of the prior work. This project is nothing unexpected, just a continuation of what has been started.
- The project is still about Pt-Ru (with some small addition of the third element). It would be good to see some design work on materials that are really novel.

- The researchers are proposing more of the same. This is a mature project and, in the final year, the team should be down-selecting, demonstrating scalability, and able to meet cost/performance targets and durability. None of these are discussed in the future work.
- There is a proper delineation of the future work. This reviewer recommends more tests in SFC hardware with proper, long-term, chronoamperometric data interspersed with start-stop conditions.

### Project strengths:

- Catalyst work is very good, especially work on developing catalysts for ethanol and DME.
- This is an outstanding group of scientists. The catalysts are designed with a basis on a deep understanding of electrocatalysis, syntheses, and the transformation of fundamental science to real applications.
- The new materials concepts, including catalysts and membranes, are juxtaposed with characterization and a portable fuel cell partner (SFC). Good collaborations are shown. The results in some cases are promising, albeit in controlled, half-cell conditions.
- The team is well organized and capable of developing a DMFC membrane and MEAs. The team is composed of respectable research organizations with adequate expertise. Overall, the team is equipped with a necessary knowledge base, resources, and industry/academia/national lab mix that is required for the success of this project.
- New catalysts, and focus on high activity/low PGM is a strength. The team is cognizant of performance, cost, and durability issues. Low-risk work with methanol is balanced by high risk with ethanol and DME. Overall, this is a strong effort.
- The approach is good and involves experimentation and modeling guidance. The team is very strong and well assembled. This project has good planning, collaboration, and coordination. The results and progress are impressive and could have a significant impact on the portable power market using direct liquid fuel cells.

### Project weaknesses:

- The combination of a large number of research organizations, which may be a management challenge for the prime organization, is a weakness.
- Coordination among the seven thrust areas could be better. It seems that each group works pretty much on its own.
- The catalyst concepts are not new or novel and suffer from the same issues described in literature dealing with their stability. The membrane effort is also not new and follows the same lines of previous efforts. Proof of a remarkable breakthrough awaits actual fuel cell results in SFC hardware.
- Oxidation of small organic molecules on PtRuX (X = Sn, Pd, etc.) electrodes has been an “attractive” research direction for decades. The PIs should go beyond PtRu systems; should avoid Cu; and, because of stability problems, should replace $Sn/SnO_2$ with more stable non-noble elements.
- The lack of cohesion among the different focus areas and lack of long-term testing (1,000 hours is only a 2-month test), is a weakness. This is the fourth year of the project and the testing should have been done for all of their promising catalysts. For ethanol and DME fuel cells, complete conversion to $CO_2$ is the most important/interesting thing, but that is not shown here. The goals at the beginning are given in terms of cost, efficiency, and energy density. These are the targets and none of the work is reflected back on these targets.
- On the membrane research, it would be very useful to check how the TM membrane/electrode interface changes in the durability test to validate the hypothesis that matching water uptake to Nafion® can improve interface durability. Also, perhaps it is more useful to have the membrane swelling ratio data. The milestone for the ethanol oxidation catalyst is delayed. A $SnO_2$-modified catalyst made by microemulsion synthesis has a much lower activity than the one made by the Cu UPD method. No explanation was given. It is important to understand the cause, which can help further improve the catalyst. On the MEA durability test, the new results showed that catalyst layer cracking has no major effect. The question still remains unanswered as to why MEA failed faster in high methanol concentration operation.

### Recommendations for additions/deletions to project scope:

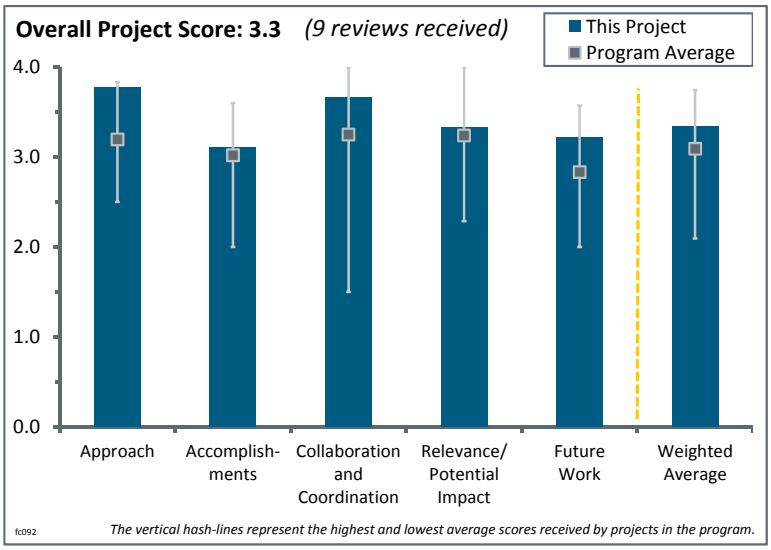
- One thing to add to this project is a cost analysis on the catalyst and membrane.
- Any use of Cu NWs should be avoided due to their well-known poisoning effect when dissolved. The proper choice of durability tests should be incorporated in this effort.
- Given the research team's strength in direct fuel systems, the team should drill down to the fundamentals of DME fuel systems and determine the viability of the success of DME technology as a competitive technology to DMFC.
- The membrane work should include a look at ethanol and DME crossover to bring it into the fold of the overall project. Efforts need to be done to show how this work moves toward DOE's goals for efficiency, power/energy density, and cost.

# Project # FC-092: Investigation of Micro- and Macro-Scale Transport Processes for Improved Fuel Cell Performance

Jon Owejan; General Motors

## Brief Summary of Project:

Expected outcomes of this project include (1) development of a validated transport model, including physical and chemical properties of all the fuel cell components; (2) public dissemination of the model and instructions for exercising the model through a project website that includes all data, statistics, observations, model code, and detailed instructions; (3) compilation of the data generated during model development and validation; and (4) identification of rate-limiting steps and recommendations for improvements to the plate-to-plate fuel cell package. Model validation with baseline and auto-competitive material sets will provide key performance limiting parameters.




## Question 1: Approach to performing the work

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

- This project has a wide-ranging analytical approach, which addresses major barriers.
- The project approach is logical in design and reasonable in scope. The project is well-balanced between practical in-situ characterization/diagnostic experiments and directly insightful modeling.
- This is an excellent project approach to improve the fundamental understanding of what limits state-of-the-art fuel cells and thereby enables commercially viable automotive fuel cells. Additionally, websites and publications ensure that understanding is available for others to access and use.
- This project has a very substantial and well-articulated, multi-scale approach that includes substantial diagnostics, methods development, and sensitivity studies. Unfortunately, there were no considerations for freezing conditions.
- The proposed model could be very useful; however, there are some doubts about the robustness of the model. With so many parameters, the physics is sometimes lost. It is not clear whether the team has done a sensitivity analysis to test how much the predictions of the model respond to changes in the parameters.
- The approach is great. It leverages General Motors' (GM's) work and keeps the project relevant to automotive applications. Making data available to the community in general via a website is a great strength of this project. One concern is that it is not clear if the baseline or the auto-competitive materials are available for other researchers to purchase. This might be needed if additional data on materials properties are required.
- This project has an outstanding modeling and experimental approach to understanding transport processes in polymer electric membrane fuel cell (PEMFC) membrane electrode assemblies (MEAs). It has contributed significantly to the knowledge of controlling factors in PEMFC performance and the design of improved electrodes.
- GM's approach combining ex-situ materials property measurements, multi-scale component level models, a 1+1D simplified transport model and extensive validation of the model with data from two material component sets would be difficult to improve on. The experimental effort is measuring the relevant parameters. The thin film measurements are now being attempted on relevant substrates.
- The principal investigator (PI) has done an excellent job of pulling together the approach to try and solve a complex modeling problem. As the project has evolved, the PI (with the research team) has been able to

link findings from the model development work to specific component attributes that have subsequently been employed to design new materials for fuel cell applications. The hallmark of any successful modeling effort is the application to and the improvement of real-world systems.

### 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 overall progress is very good. Although details of the model are not provided, critical aspects seem to be included. The goal of using the model to identify critical components is excellent.
- While the progress has been slowed this past year by the reorganization and relocation of GM, the overall progress throughout the duration of the project and the dissemination of the progress has been very good.
- This project has made good progress over the past year; however, it seems like there is still a lot to finish with only 15% of the budget remaining. Additionally, this project has made significant contributions towards overcoming barriers, but there are no real game-changers here.
- The project almost completed all its objectives and has good model dissemination to the community. Due to the high impact of non-active transition areas, it would be useful to include the results averaged over the load profiles rather than at the steady states only. Also, transient responses would be valuable to present.
- The project has achieved several relevant accomplishments and has made good progress. Of particular note is the work to understand the $O_2$ transport losses at the ultra-low platinum group metal (PGM) levels, which is a key limitation for achieving DOE's PGM targets for 2017 and beyond for many catalyst systems.
- The project's progress is broadly on track. This project has demonstrated good correlation between models developed and experimental results. The project is a little behind (approximately six months), although this is not a significant issue since the models are developed and the extra time is required to do sensitivity analysis and make the information public.
- There were numerous accomplishments in all areas. One area that stood out was the collaboration with the National Institute of Standards and Technology on the use of the neutron imaging techniques to monitor water transport. These studies are critical to improving the designs of the fuel cell "plumbing," the anode and cathode outlets. Pore and slug flow issues were clearly identified by neutron imaging, and the GM team used these to modify designs, which is excellent.
- Good progress was made both in modeling and in experiments. The project leverages a lot of GM efforts in this area. There are concerns about the errors for the auto-competitive material. It is not clear if this is similar to the baseline material, and if any other data (other than voltage on a polarization curve) is used for model validation. For example, it is not clear if the model can predict the water balance in the various material sets, or if it can predict the water profiles (from inlet to outlet) obtained from the neutron imaging. More needs to be done to convince the community that the model has predictive power.
- The project has provided a useful, very complete dataset for baseline materials; has developed a second auto-competitive materials set showing performance and water content; and has made the dataset publicly available. The gas diffusion layer (GDL) characterization work is important. Measurements of thermal conductivity as a function of saturation and compression are useful. The work characterizing the effects of droplets and films on $O_2$ resistance, the transition from slug to film to mist flow, and the channel effects have been beneficial. The model does not perform as well at high current density, with more than 20 mV error for more than half the cases shown for the baseline materials set (slide 19). In particular, it appears to systematically over-predict voltage at low temperatures. In addition, the model does not do as well at predicting current density for the auto-competitive dataset. There appear to be issues dealing with liquid water or high water saturation. The component model appears to use interfacial resistances at the Pt-ionomer and ionomer-gas interfaces as fitting parameters, and it is not clear whether or how these interfacial resistances would be adjusted for other materials sets. The data presented, which have been interpreted as indicating interfacial resistance at the Pt and ionomer interface, is not conclusive. The data on slide 16 appears to fit a simple logarithmic dependence of the resistance on the ratio of the ionomer area to the Pt area and does not appear to have a minimum, as the model with the additional interfacial resistances would predict. The $O_2$ transport measurements did not show any interfacial resistance. Most of the thin film studies showing changes compared to the bulk material appear to be on $SiO_2$ substrates, though it is unclear whether the substrates are Si or $SiO_2$. Thin film studies need to concentrate on films with representative

substrates. The project is moving in that direction, though it appears these studies may not be complete by the end of the project.

### Question 3: Collaboration and coordination with other institutions

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

- This project has good collaboration.
- Although many partners are involved, their specific participation is clear.
- This project has an impressive array of modeling and experimental experts.
- This is a large and diverse team that seems to have been well coordinated.
- The collaborators contributed significant material and data sets, including diagnostic analytical methods.
- The project appears to have managed collaborations between GM and its partners effectively.
- This project has good collaboration with other institutions, especially on more fundamental property measurements/characterization. It is not always clear from the presentation how relevant some of the items are (e.g., using TEM of thin films of an ionomer to elucidate transport characteristics and then inferring that this may be what happens in a catalyst layer).
- This project has great collaboration with various universities where the fundamental work is done at the universities and the modeling and validation is done at GM. This project has effectively used the transport modeling working group to foster collaborations.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- Posting a simple demonstration of the use of the model could be very interesting and useful to potential users.
- This project has made good contributions towards the DOE Hydrogen and Fuel Cells Program goals and objectives. The project has not made any breakthroughs though.
- The project addresses a key area for fuel cell understanding, namely, the impact that water production and its movement has on the operation of fuel cells (with a focus on the sub-component level).
- This project addresses transport processes that limit performance at full power. The performance at full power is instrumental in determining system size and cost. An effective transport model would help developers decrease transport losses and achieve higher power with smaller systems, decreasing the cost.
- Water transport is an important issue. The role of the ionomer in the catalyst layer, especially at the low loadings, is even more critical. The project is doing a great job in addressing these issues. Innovative ideas are lacking, but at least a good evaluation of the problem has been presented.
- The tools and knowledge developed in this project have the potential to impact the future direction of catalyst layer design and to guide the understanding of performance limitations in catalyst layers based on high specific activity catalysts with the ultimate goal of designing and fabricating high-performance electrodes to reduce PEMFC cost.
- The work is directly relevant to the Program's barriers of performance, water transport within the stack, system thermal and water management, and startup/shutdown. The results clearly indicate that the $O_2$ transport issue with low-PGM cathodes is a potentially significant barrier to achieving DOE's PGM targets.

### Question 5: Proposed future work

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

- The proposed tasks are appropriate and will complete the objectives.
- This project should focus on finding links from various ionomer studies.
- The proposed future work is appropriate based on the time left in the project.
- The project is in its final phase and looks as though it will conclude very successfully. Well done!
- The team primarily needs to complete documentation of the work that has already been done.

- The project is near its conclusion. The proposed work is what is needed to finish the model and complete the project.
- The project is ending and most of the GM people involved in the project have left, including the PI. GM should continue to engage with the various national laboratories and universities involved in the Program to address the problem of transport losses in low-loaded catalyst materials.
- The funded portion of this project has ended and it is currently in a no-cost extension for the few months left in the project. The future work is finalizing and disseminating the models, which is commensurate with the short time left in the project.

### Project strengths:

- This is a diverse team that provides open access to results and focuses on relevant issues.
- The model dissemination is very useful through the interactive website.
- This project's public distribution of the validation dataset has provided a valuable resource for the community.
- The experiments are integrated in a simple model, which will be useful to assess fuel cell performance and the role of the various components.
- Strengths of this project are the multiple-component approach to water management modeling (i.e., plate, GDL, and catalyst layer effects) and including the evaluation of an auto-competitive sample for longer term DOE targets.
- This project has excellent collaborations with universities and transport modeling working groups. The extensive data set was made public on the website and was shared with the community. This project sets a great standard for other DOE projects to follow.
- The project is well balanced between practical in-situ characterization/diagnostic experiments and directly insightful modeling. The project addresses the key barriers towards understanding and improving the transport issues for lower PGM MEAs. The publishing of all project data and models is of tremendous benefit to the PEMFC community at large.

### Project weaknesses:

- No real weaknesses are evident.
- The PI and several other project personnel have left GM.
- This project does not have a clear description of how the various components of the model may interfere with each other, thus making model results difficult to interpret.
- The model this project developed is somewhat specific to the architecture of the flow-field with the specific transitional areas studied, which limits its industrial applicability.

### Recommendations for additions/deletions to project scope:

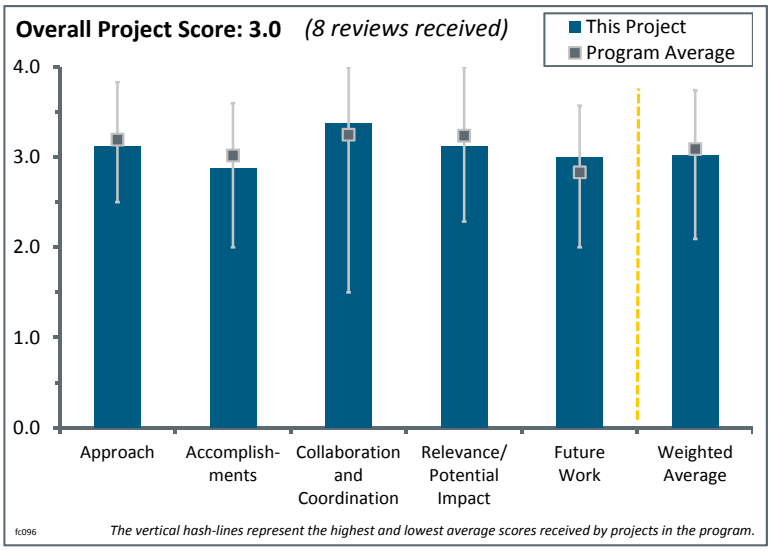
- Since the project is near completion, no deletions/additions are suggested.
- This project should work on better model validation in addition to the needed voltages from polarization curves.
- It would be interesting to see the model predictions for the material sets developed with the gradient features, including through and in-plane direction, or for the project team to produce narratives on the model's outcome, if such material is developed.

## Project # FC-096: Power Generation from an Integrated Biomass Reformer and Solid Oxide Fuel Cell (SBIR Phase III Xlerator Program)

Quentin Ming; InnovaTek

### Brief Summary of Project:

The goal of this project is to develop and demonstrate a fuel cell distributed energy system that operates with second-generation biofuels. The objectives are to establish a design to meet technical and operational needs for distributed energy production from renewable fuels; design, optimize, and integrate proprietary system components and balance-of-plant in a highly efficient design; and demonstrate the technical and commercial potential of the technology for energy production, emissions reduction, and process economics.




### Question 1: Approach to performing the work

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

- The approach, which is now focused on natural gas, makes sense.
- The project has been focusing on (1) improving system design and operating efficiency and (2) performing economic analysis.
- This project went well and reached all milestones. InnovaTek did a very good job on reforming technology development and system integration.
- It is difficult to independently validate how well the principal investigator (PI) is doing in meeting project goals and objectives. No data or performance metrics are really provided.
- The approach is to develop and demonstrate a fuel cell distributed energy system that operates with second-generation biofuels. The system is based on InnovaTek's steam reforming process and a Topsoe solid oxide fuel cell (SOFC). Non-food biofuels include pyrolysis oil and biokerosene, which is processed locally, and is to be demonstrated in Richland's renewable energy park and tied to the grid.
- This project is focused on developing the biomass reformer. The fuel cell stack is outsourced. Early market entry is assumed with natural gas fuel. There are other fuel cell developers who are also working on 5 kW systems. It is not clear what differentiates InnovaTek's product from others. The cost for 5-kW systems at the production rates mentioned in the study are very optimistic.
- The project develops technology for the production of fuel-cell power from renewable biofuels. Fuel cells yield more useful energy from a fuel and reduce $CO_2$ emissions and criteria pollutants. Bio-kerosene is a refined fuel and will likely be expensive. The analysis presented indicates that the fuel needs to be $3.50 after taxes to be economically feasible or attractive.
- This project is targeting cost, durability, and performance and it appears that two of the three are addressed. Durability has not been addressed in the approach. A full bill of materials estimates, an analysis of the cost and design through modeling and simulation, and the cost of power versus financial benefits are all excellent pieces of this project's approach. The reviewer would like to see the improvements that contributed to taking the system from 37.5% efficient to 41%. It is not clear how much is attributed to advances in SOFC stack technology versus reformation improvements versus revised flow paths to enhance thermal energy.

### 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 achieved the milestone of system efficiency greater than 40%. There are reduced part counts. Details on the system's operating characteristics are lacking.
- The accomplishments to date are very good. They have reduced the costs of various components that they are developing. The initial performance is promising.
- For the bio-kerosene reforming, the catalyst performance was very stable for more than 900 hours. The SOFC was operated at 750˚C. Since the reformate had approximately 10% methane at 750˚C, it is not clear if there is any carbon deposition on the fuel cell electrode.
- The reviewer would like to see the performance of the reformer for both bio-kerosene and natural gas, and would also like to understand the cost projections with variable volume. The production volume is not specified in the analysis; it is a key parameter. Efficiency values in the presentation did not specify what fuel was used.
- Thus far, the program has hit its milestones, met the go/no-go criteria, and addressed the performance barriers for this program. Achieving over 40% efficiency is important, especially when it is being accomplished on a bio-kerosene. The reduction of reformer complexity and optimized flow paths would take advantage of the heat created in the system and will bring the system closer to meeting the criteria and barriers set out to be reached through this project.
- InnovaTek's proprietary catalyst reforms bio-kerosene during long-term tests. It is an integrated system that produces power from bio-fuel. The measured efficiency needs to be higher to be attractive. A different SOFC may produce better results. It is not clear how many hours were actually measured, if the total system electrical efficiency was 27.49%, and if the test ran 200 hours. The test conditions, such as temperature, utilization, etc., were not specified. It was unclear if the stack showed degradation or coking.
- The stack was operated at 65% efficiency. The high efficiency was achieved by operating at a very high voltage and low current and power densities. The low power density will lead to higher stack costs. The reviewer suggested showing the results of a tradeoff analysis between capital cost and efficiency. The reformer has been operated for 900 hours and the reformate contains approximately 10% methane. While 4%–8% methane is desirable for stack cooling, the higher methane content may lead to excessive cooling at the entrance. The extent of stack cooling resulting from internal reforming in the anode is not clear. This data was not presented. Stack current density and durability have improved. Economic analysis is optimistic, assuming 45% net alternating current efficiency.

### Question 3: Collaboration and coordination with other institutions

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

- The PI seems to have a capable team assembled.
- This project is collaborating with partners very well.
- InnovaTek has good cooperation with the fuel cell industry, national laboratories, and government.
- This project should pursue continued collaboration with the fuel cell developer (Topsoe Fuel Cells).
- InnovaTek is working with manufacturers and a national lab and is supporting students at universities.
- Leveraging regional advantages and a cost share with Impact Washington will help this program reach a larger audience. Leveraging the work of other DOE-funded projects increases the efficiency of DOE investment and prevents duplicative work. Collaborating with local businesses in creating $H_2$ from renewables is the best way to bring the cost down in this particular region.
- InnovaTek seems to be limiting its focus to fuel processing and systems integration. It should narrow further as total system manufacturing, warranty, etc., is going to be extremely challenging for such a small company. It would be better to see a systems integration partner that can help with some of the systems integration expertise and customer relationships/specs input that will be hard for a small company to develop.
- The strategic partners are well selected. Pacific Northwest National Laboratory and Washington State University-Bioproducts, Sciences & Engineering Laboratory provided bio-oil made from wood sawdust.

Boeing provided bio-kerosene made from camelina. The City of Richland Electric Utility provided the site for field demo. Mid-Columbia Energy Initiative is a strategic partner.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The project fully supports the objectives of the Hydrogen and Fuel Cells Program (the Program).
- The work is certainly relevant to and the Program's R&D goals. Bio-fuel is a renewable energy with a C effect. Completing this project is the way to advance technologies and generate jobs in the states.
- The critical work here is the biomass reformer. If InnovaTek can meet cost and performance goals on the biomass reformer, the project will be successful.
- This technology will help shift the primary energy source for $H_2$ from fossil fuels to renewable non-food biomass. Use less fuel through high-system efficiency by effective thermal integration and off-gas recycling. Provide an alternative method for distributed power generation near the source of the feedstock, enhancing grid stability.
- The technology for fuel processing and the stack is being advanced by this project. The progress in stack technology is good and needs continuing support to become competitive. The fuel processor and stack technologies need to address sulfur in fuels since most fuels, including natural gas, contain sulfur and without the broader fuel application it will be difficult to succeed commercially.
- One solution to utilizing bio-fuels is through the reformation of liquid fuels. They are easy to transport and store. This project has a high potential impact on proving that the efficiency and performance of using this resource is feasible. Making use of renewable, distributed, non-food-based biomass, demonstrating the feasibility and efficiency of it, and providing power has a large impact.
- If successful, the project could be relevant to DOE's goals of deploying stationary fuel cell systems for residential and light-commercial applications. It is a long road and will require significant extension of operating lifetime and extreme reliability (both of which will cost a lot of money), as well as a company with a balance sheet to service the customer with a warranty. The outcome of this project could show that a small SOFC system with a reformer might be a good concept to develop.

### Question 5: Proposed future work

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

- The proposed future work is vague.
- The system testing in the next phase is very critical.
- The proposed future work is consistent with the project plan and the technical progress.
- For the for-profit company, the target is to make a profit. InnovaTek is on the right track to make products.
- The project appears to be partially funded with that assumption that the future work identified makes sense.
- A higher measured efficiency will be obtained in year two with better insulation and design improvements.
- The economic analysis is needed and will be valuable in guiding the path of this project towards commercial development. A continued effort toward improving efficiency and durability is needed, but the strategy is not clear. Methane conversion in the reformer needs to improve. It is not clear what is limiting the stack current density and how it will be improved.
- The proposed future work appears to be the next logical step; however, more clearly defining how durability is going to be addressed would strengthen the project. An accelerated stress test of some kind could provide insight into the prospect of reaching the 40,000-hour life predicted. Additionally, a reforming catalyst that lasts 40,000 hours has not been proven or predicted yet. It is not clear if there is an estimated number of hours of operation that would require a reformer overhaul. Partnering with a local business to provide a real-life, long-term demonstration of the system would also prove useful and provide an excellent learning opportunity.

### Project strengths:

- Development of the biomass reformer is a strength of this project.
- Development of the fuel processor and system design and integration are strengths.
- The project has demonstrated improvements in durability and performance. More than 40% efficiency has been demonstrated. The economic analysis shows a path to commercialization.
- It is difficult to independently ascertain the project's strengths beyond the claims made by the PI. Very little data was presented that is specific to the technology.
- Achieving high efficiency from reforming biomass and producing power from non-food biomass are strengths of this project.
- The durability of the SOFC and reforming system is still an issue. Some long-term tests are needed. On/off tests with cycles are also needed for end users.
- This project has a good focus on natural gas as a bridge fuel. InnovaTek seems to have limited its scope to systems outside the stack. Limiting the scope and leveraging partnerships makes sense.

### Project weaknesses:

- There are no weaknesses, just challenges for the commercialization.
- It is recommended that more emphasis be placed on evaluating the system's operating characteristics and the effects of key operating parameters on system performance.
- There should be more focus on durability and identifying the expected maintenance schedule and the cost and time associated with it, including the operational availability.
- Some weaknesses in the presentation were omissions of critical information. For example, efficiency was quoted without specifying what fuel was used. Another example was the capital estimates for the units were reported without referencing the production volume. Also, it would make sense to have even more partnerships in the system integration domain.
- The high efficiency has been demonstrated at the cost of low-current density and higher capital cost. The economic analysis is based on a very optimistic price of bio-kerosene and anticipated future efficiency values. The methane yield in the reformer is high. The presentation did not clearly distinguish between the actual experimental work from which data was obtained and the projected scenario that was used to estimate the performance and cost.
- Very little data was presented that was specific to the technology. No information was presented regarding the reformer technology, catalyst, and method of operation. It is not clear from the presentation materials how the system is to be used (continuous use, back-up, or stand-by). This information is important to know since the temperature cycling may significantly impact the SOFC, reformer durability, and long-term performance. A slide is given regarding catalyst durability, but these data appear to be for continuous operation. It was not clear if there was a concern with poisoning by the odorant levels in natural gas. The PI does not explain the source of performance decrease with time.

### Recommendations for additions/deletions to project scope:

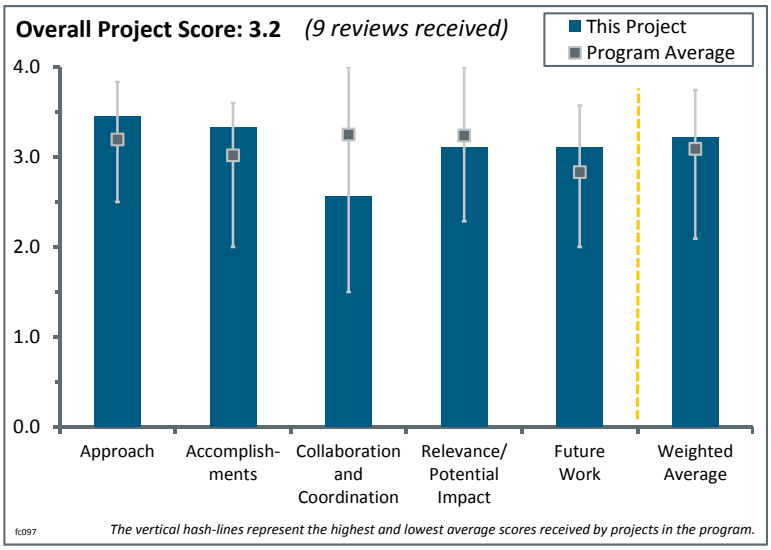
- The project should develop additional partnerships for system integration.
- The project should partner with FuelCell Energy in the SOFC area. Surely they can produce a 1 to 5kW SOFC stack.
- The PI should provide more technical details within the obvious constraints of protection of proprietary information.
- The project should focus on the performance and durability of the components and the integrated hardware in the laboratory. The researchers should investigate operations with sulfur in the feed to the reformer and the stack. They should also study the tradeoffs between efficiency, current density, and cost, and they should defer the field demonstration.

## Project # FC-097: Stationary and Emerging Market Fuel Cell System Cost Analysis – Material Handling Equipment

Kathya Mahadevan; Battelle (presented by Vince Contini)

### Brief Summary of Project:

The 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 models and cost estimates. The applications modeled include primary power (including combined heat and power), backup power, auxiliary power units (APUs), and material handling equipment (MHE). Fuel cell types modeled included polymer electrolyte membrane (PEM) and solid oxide fuel cell (SOFC) technologies. Multiple production volumes and fuel cell sizes are modeled. The project also addresses cost reduction needs for non-automotive applications by performing cost analyses of fuel cell system designs.




### Question 1: Approach to performing the work

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

- The approach is systematic, thorough, and has focused in the last year on the MHE market. Gathering stakeholder input and feedback throughout the project is important for the success of the project.
- The methodology is excellent; however, as with any model, the accuracy of the inputs are critically important. Some of the cost inputs appear to be one to two generations old.
- The project directly targets cost and manufacturability barriers, which in turn will affect customer acceptance. Starting with fuel cell systems for MHE is appropriate, as are the assumptions about production volume and the bill of materials for the 10 kW and 25 kW systems.
- This is a good method for system costing; however, it is surprising that the cost estimates are two orders of magnitude higher than light-duty automotive cost estimates (as per Strategic Analysis, Inc. [SA]). It would be beneficial to benchmark the models against each other.
- Studying the cost at multiple production levels is a valuable step of this project. It seems like the process of gathering vendor quotes could be influenced by many factors. Some of the specifications were included, but the ranges of inputs (or number of inputs) from vendor quotes were not clear.
- The approach to estimating production costs appears to be consistent with other DOE-sponsored cost analyses, so results can be compared when differences in system configurations are taken into account. The costing methodology used by Battelle is used by SA and other companies and is a robust method for estimating manufacturing costs.
- The project is effective in addressing most of the stated barriers, but the project is very dependent on the design for manufacture and assembly (DFMA) software and many of its embedded assumptions, and it is using generic designs. The researchers could make more effective use of their industrial partners to explore the extent to which DFMA assumptions are not necessarily applicable, and the extent to which generic designs are not representative of what is or will be deployed.
- As identified in slide 5, the approach is systematic and appears to cover the relevant areas. This project has good validation from the industry. The balance of plant (BOP) is a difficult area for obtaining good cost data due to the breadth and variety of system components, and it represents a major cost driver. System

integrators understand this and are working to get the parts count down, but this will remain difficult for the project to address.
- This project takes a very detailed approach to determining the costs associated with manufacturing MHE fuel cell systems. Most of the modeling inputs seem reasonable. One exception is the assumed battery change time in the cost of ownership results. While there are certainly battery change times of 30 minutes, these are the exception, not the rule, in the high-productivity sites where fuel cells have a play. Since the recharge time of fuel cells is one of their primary benefits, it may make fuel cells look more attractive than they really are.

### 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 researchers appear to be making very solid progress toward meeting their stated project objectives.
- The team has made good progress representing system complexity and modeling completeness since last year. The detailed manufacturing cost and total cost of ownership results should be useful to industry. Also, the focus of the work has sharpened to include only relevant system sizes.
- This question is really not applicable since this is cost modeling. It is providing a projection of what the future cost might be, but really it is not doing anything to drive down the cost. Once again, the accuracy of the model is determined by the quality of the inputs to parameters.
- This project has done great work with PEM systems; however, it would be at least informative to examine the effects of current density and Pt loading. The accomplishments state that SOFC/APU work has been partially completed. It would be beneficial to have shown this data (stack level for example) in relationship to PEM.
- The progress is excellent. A thorough cost analysis for two different classes of PEM fuel cell forklifts has been completed at various production levels. The BOP manufacturing costs contain two high-cost items identified as additional work and other components. A high-level breakdown of these items would be beneficial.
- The comparison between the MHE and light duty vehicle fuel cell system costs was good. The systems are similar enough in scale and application that this comparison is a useful check on the assumptions and methodology in both DOE-funded studies. Overall, the approach used for the MHE fuel cell system should be applicable to the other systems that are studied in the future.
- Overall, the project has done a good job in developing its cost models and inputting data. The models have shown where R&D effort needs to be applied to lower costs (slide 23). In regard to the capital cost assumptions, there is a large variation between $3 million and $20 million in total capital cost. This appears to be for the Columbus, Ohio area. It is not clear if all this equipment can fit on one acre of real estate—real estate is very expensive at $125,000 per acre. The project should take corporate overhead and profit margins out and stay with the hard costs. Each corporation is different and the competitive situation is a driver, particularly on the profit margin. For slide 21 on Pt cost, it was not clear if the values in brackets should be swapped and what the effect was on the bottom line number of $3,423.
- Progress has been somewhat greater than was the case last year. The conclusion that systems that do not operate 24/7 result in a value proposition for the MHE market that is not favorable is consistent with general expectations and experience. The system schematic appears consistent with MHE, but it is unclear if the system schematic represents all of the MHE systems being deployed or if the system and operating conditions have been vetted with the MHE developers. There appears to be a significant discrepancy between the BOP cost estimate of Battelle and that of SA. It would be instructive to determine where the differences lie. The MHE analysis appears to include silicone gasket material. Other alternatives should be investigated due to the tendency of the material to migrate into areas of the cell that are detrimental to performance and durability. Progress in the SOFC APU area has not been as good as in the MHE area.
- It was unclear which were new analysis results when the current presentation results were compared to the 2012 DOE Hydrogen and Fuel Cells Program Annual Merit Review system selection chart. The 10,000-hour lifetime is too low for fuel cell MHE to provide savings when compared to battery MHE. It was unclear what the reason was for selecting a 10,000-hour lifetime, thus changing out the fuel cell system every 3 years, basically on the same schedule as batteries. It was also not clear how 10 kW and a 25 kW

MHE systems were identified for the base cases. It was expected that the final estimate, with a markup, would be a price number and not a cost number. Steel tanks have been used for a while in order to reduce the cost. Steel should thus be used as the baseline, not an opportunity for cost reduction. The PI did not mention if integration into the forklift was investigated as an opportunity to reduce cost. It is unclear where the life cycle cost assumption comes from. For instance, the battery change-out time and the hours of operation per year are on the high side. The time for fuel cell refueling is higher than the average identified in the National Renewable Energy Laboratory's reporting of the American Recovery and Reinvestment Act systems. The lifecycle cost analysis assumes \$8/kg for $H_2$, but it does not look like a monthly service charge for $H_2$ infrastructure was included in the operating cost for fuel cell MHE. Assumptions for maintenance of batteries and fuel cell MHEs are not provided.

### Question 3: Collaboration and coordination with other institutions

This project was rated **2.6** for its collaboration and coordination.

- Input from industrial partners and their feedback ensures the success of this project.
- Collaboration appears extensive regarding fuel cell stack and system design. Numerous collaborators appear to be involved that have materials, stack components, and system design expertise.
- This project needs to partner with an OEM who can provide actual volume price quotes for BOP components. Of course, the OEM will have to remove the supplier information from the quotes.
- There is a broad group from the industry providing input; however, the only company still currently active in the MHE market is Plug Power. It is understandable that they may be less willing to contribute, since they are still trying to make a business of this market. Their absence, however, is notable.
- It can only be assumed that there has been collaboration. There were no slides describing the degree of collaboration, so this is difficult to judge. It would have been useful if the project presentation followed the format requested.
- The speaker referenced getting feedback from the industry, but the work to date appears to have been solely done by Battelle. The degree of industry participation is unclear, and it is therefore recommended that it be strengthened.
- It is positive that the number of collaborators significantly increased from the 2012 review, although a leading fuel cell MHE manufacturer is not included. It was expected that there would be some collaboration with a similar project at Lawrence Berkeley National Laboratory.
- The project has a well-selected list of collaborators in the industry; however, the presentation did not make clear the extent to which these collaborators were consulted to seek key inputs on model results, particularly regarding cost sensitivity assessments. Also, it is not clear whether or how the researchers made efforts to reach out to make comparisons and seek feedback from other DOE-funded national laboratory efforts that are conducting similar cost analyses.
- Plug Power is not on the list of collaborators. They are one of the leaders in forklift sales.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The project is directly relevant to a growing area of fuel cell deployment.
- This project has no real impact. It might drive people to work on only more expensive components, but that is only a good thing if the inputs in the model are correct and it does not seem like they are.
- This is an important market segment for the DOE. Understanding the costs and potential areas of improvement can be accomplished through this type of analysis.
- The cost is a key barrier that needs to be overcome in order to achieve market acceptance of $H_2$ and fuel cells. Developing accurate cost models using industry data is very important to identifying where DOE program dollars need to be invested for the good of the overall program.
- The cost analysis of materials handling fuel cell systems is well aligned with the DOE objectives. It establishes a cost baseline for the current state-of-the-art systems and identifies key components for potential cost reduction.

- In general, the project aligns well with the needs of the Hydrogen and Fuel Cells Program; however, the system boundaries set up in the cost model tend to restrict the systems cost analysis somewhat and the focus on generic design and the use of DFMA assumptions could limit the ability of this project to anticipate a broader range of cost impacts in fuel cell deployment.
- It is important that the cost projections for non-automotive systems be at the same fidelity as that for automotive systems for the same reasons, namely to assist DOE in setting appropriate targets, particularly since there is no equivalent to the U.S. DRIVE Partnership in the non-automotive space. The project is very relevant and needs to achieve the same level of detail and fidelity as the SA work on the automotive system.
- The cost analyses from this project will provide a solid baseline and future estimates for the total cost of ownership for fuel cell systems. The analysis appears to be mostly from first principles. It is recommend that a teardown analysis of an existing system may provide a stronger case for the cost estimates than the informal industry survey approach, which appears to have been used. The team identified BOP as a significant factor in the cost of 10 kW and 25 kW systems. Input materials are also a significant factor in costs (75%–83% of the total system cost). This brings to light the fact that these costs need to be reduced in some way. For example, the lithium-ion battery, the $H_2$ storage tank, and the power electronics were identified as the top three cost items in both the 10 and 25 kW fuel cell systems. However, other than recommending a steel tank for $H_2$ storage, a choice that also has safety considerations, no recommendations were presented on pathways to lower cost.

### Question 5: Proposed future work

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

- The future plans are adequate.
- This project needs to focus on getting more accurate cost inputs.
- The future work seems light, given that 90% of the APU assessment is already completed.
- The planned future work should be relevant to the fuel cell industry. The project methodology should be relevant to these other areas as well.
- The work done on other systems is important, but it is also important to continue to refine the MHE cost analysis for comparison with the automotive projections being developed by SA.
- The team needs to consider stronger recommendations for where future manufacturing R&D should be directed to address the manufacturing costs for fuel cell systems for the applications being studied. The team should also clearly identify what the barriers are to reducing component or sub-system costs.
- In spite of this reviewer's minor concerns about the relevance of this work to the Program, the researchers have been very effective in building their future plans on past progress and are focused on the barriers they have identified.

### Project strengths:

- The methodology is a strength of this project.
- The interaction with stakeholders at all levels of the analysis is a strength.
- This project includes a detailed analysis of the costs associated with this technology.
- This project has a good modeling effort and has identified the cost drivers and areas where R&D effort needs to be applied.
- The cost analysis appears to be using a straightforward approach to estimating fuel cell system costs. The results so far, and especially the level of detail provided, will be a useful resource for those in the industry.
- The estimates at various production levels and the details at the manufacturing and stack levels are both strengths of this project.
- The list of collaborators is extensive and can provide valuable insights into the assumptions and methodologies employed in this study.
- The researchers appear to be making very solid progress toward meeting their stated project objectives. The researchers have been very effective at building their future plans on past progress and are focused on the barriers they have identified.

### Project weaknesses:

- This project needs to refine some cost models with better data or provide options analysis, particularly for capital costs. It looks like the data may have been from one source.
- A leading fuel cell MHE manufacturer is not a collaborator. The life cycle cost analysis is missing pieces (like maintenance assumptions) or has numbers that should be updated.
- The ability to get accurate costs is a weakness of this project. Companies do not want to share this information, so it is hard to predict costs at high volumes. The researchers need to present the future cost as a range and not an absolute number. It is not clear what the uncertainty is.
- Researchers should consider activities that could allow a broader range of cost assessments that address geographic and use variations. The researchers could make more effective use of their industrial partners to explore the extent to which DFMA assumptions are not necessarily applicable and the extent to which generic designs are not representative of what is or will be deployed.
- This project appears to be one of three proceeding in parallel that have strongly overlapping objectives. It was not clear what benefits this approach provides to DOE's program mission. It is unclear how the results from this project will be reconciled with each other as it concludes. The life cycle cost analysis for fuel cell MHE versus battery-powered MHE seems incomplete. There appear to be applications for which the value proposition favors fuel cell MHE already. It is not clear if the cost analysis assumptions need further input from the industry partners or, even better, from companies that are operating fuel cell MHE already.

### Recommendations for additions/deletions to project scope:

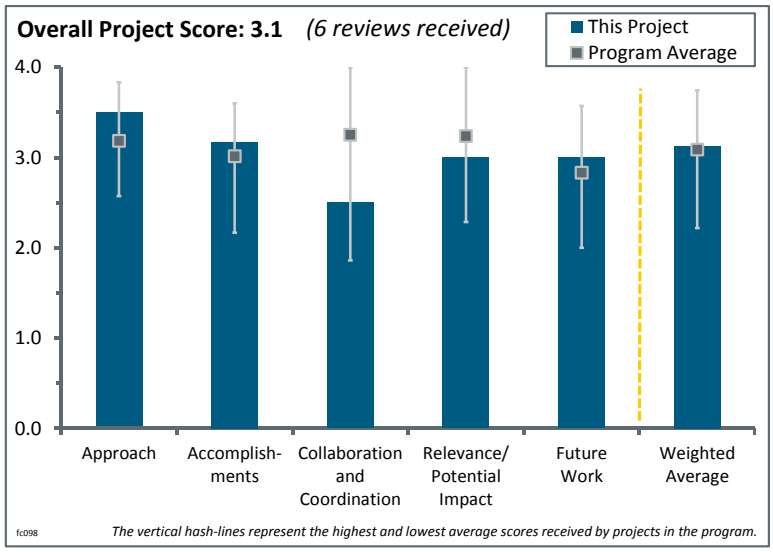
- This project should make more use of the model performance evaluation to set research priorities. It should also make more effective use of industrial partners in assessing results of the DFMA analyses.
- There are very different numbers from this analysis and its sister analysis by Strategic Analysis. It would be beneficial to benchmark the models to understand their agreements and differences.
- This project should solicit feedback from Plug Power, a key manufacturer of PEM fuel cell systems for forklifts. The researchers should also publish a report on PEM fuel cell MHE cost analysis.
- The project should take a more detailed look at battery swap times for the different classes and include these as a sensitivity in the life cycle cost analysis, as they are essential to making the business case for fuel cells in MHE work.
- It is recommended that the scope of this and the related projects be clearly defined so that each has a primary area of focus. Based on the presentations at the AMR, it seems like we are likely to get three answers to the same basic question, and it is not clear that the answers will be entirely compatible with each other.
- Since a dispersed catalyst system is being employed in the analysis, a comparison could be made between the system and the nanostructured thin film system used by SA. It would be necessary to ensure that both systems are modeled with the same fidelity to make the comparison meaningful. Alternatives to silicone gasket material should be investigated due to the tendency of the material to migrate into areas of the cell that are detrimental to performance and durability. The definition of overall plant efficiency should be explicit in the presentation. Every opportunity should be taken to compare the MHE system design and cost projections to that of SA for the automotive market. There is much to be learned from this exercise. Differences in the costing of BOP need to be examined.

## 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 objective of this project is to provide a comprehensive cost analysis for stationary and materials handling fuel cell systems in emerging markets, including ancillary financial benefits. The framework of the cost analysis includes life-cycle analysis and design for manufacturing and assembly (DFMA) analysis with additional data emerging on carbon credits, health and environmental externalities, end-of-life recycling, and reduced costs for building operation. System designs are explored to meet the lowest manufacturing cost and total cost of ownership goals as a function of application requirements, power capacity, and production volume.




### Question 1: Approach to performing the work

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

- The approach seems to be consistent with industry norms for cost analysis and the total cost of ownership models.
- The methodology is excellent; however, as with any model, the accuracy of the inputs are critically important. Some of the cost inputs are one to two generations old.
- It is not clear if the material handling equipment (MHE) was dropped off the task list or if it was completed prior to the 2013 Hydrogen and Fuel Cells Annual Merit Review (AMR).
- The proposed approach is correct and the tools seem to have been adapted to reach the objectives. The low-volume productions are well adapted for the start phase of the commercialization. The integration of regional impacts is well done.
- This project has a very good approach overall. The authors consider production volume as a primary variable and also go down to quite a low volume, which is important. The authors also look at what is procured and the different volume sensitivity of these procured items and stack parts. They also consider appropriately different system configurations for different applications. Considering externalities to the overall value proposition probably makes sense at this emerging state of the technology. The challenge is that the externalities can vary quite widely depending on where the product gets deployed and who the customer is.

### 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 project has shown good overall progress to date. This is sort of a slow-paced program, which presumably is a function of the budget allocation. Hopefully, this effort can be maintained by DOE.
- The first key deliverables are only just becoming available now, making the accomplishments difficult to gauge. It appears that the project team is on track at this point.

- The manufacturing cost model that excludes many high-cost items, such as research and development (R&D) costs (especially at the low-production volume levels), is incomplete and could be misleading. The addition of the environmental/health impact is valuable.
- This question is really not applicable since this is cost modeling; it is providing a projection of what the future cost might be, but it really is not doing anything to drive down the cost. Once again, the accuracy of the model is determined by the quality of the inputs to parameters.
- Significant results are shown this year. The whole system is considered, including the tank. The stack and balance of plant (BOP) costs for different production rates are described well; however, some results are surprising, such as the BOP and reformer costs remaining almost constant regardless of production rate (100–10,000 units). This is not in accordance with other AMR-presented studies and is difficult to consider for components like compressors and humidifiers.

### Question 3: Collaboration and coordination with other institutions

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

- The collaboration appears very fair. The impression is that Lawrence Berkeley National Laboratory (LBNL) is working alone, with Ballard Power Systems (Ballard) bringing just the stack and system specifications.
- This project needs to work with an original equipment manufacturer (OEM) who can provide actual redacted cost quotes. It seems that some of the project's input costs are off.
- It seems that the LBNL group is making good use of capabilities elsewhere, including at the University of California (UC), Berkeley, and some fuel cell manufacturers. Getting a little more help from some of the stack part producers would be appropriate.
- The collaboration on this project seems to meet minimum standards. It is not clear how much support is coming from key partners at UC Berkeley or Ballard. The report suggests LBNL is doing most of the work on the program with some consultation from the named partners.
- Industry collaborations decreased since 2012 and no longer include a MHE system industry representative. Most of the results are focused on combined heat and power (CHP) but the collaboration with fuel cell CHP manufacturers is weak. Some collaboration with a similar project at Battelle was expected.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- Using a systematic and consistent approach to look at both production volume and application type makes a lot of sense. Making sense of various externalities will probably represent a bit of a challenge.
- The project team is developing a total cost of ownership model to help identify and address cost barriers. The team is also looking at a lifecycle carbon analysis to address $CO_2$ (or greenhouse gas) emissions issues.
- This project has no real impact. It might drive people to work on only more expensive components, but that is only a good thing if the inputs in the model are correct—and it does not appear that they are. This project cannot be used to suggest what to work on until the accuracy of the inputs are improved.
- The cost analysis used will contribute to determining key cost drivers and provide insight for the direction of R&D priorities. Fuel cell stationary systems, in particular with reformers, represent a very significant part of fuel cell commercialization. As the cost studies focused mainly on transportation in previous years, this study completes the cost panel.

### Question 5: Proposed future work

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

- The remaining scope of work seems commensurate with the budget remaining.
- Basically, the future work simply includes analyzing the systems yet to be completed and considering externalities. This makes sense.

- The proposed work is correct and monetizing health damage is a very good point. This parameter may have more impact in the near future than reducing $CO_2$ emissions. Mapping the effects for different regions should be useful to prioritize the fuel cell deployments.
- The researchers need to work with an OEM to get real quotes.
- It is unclear if the total cost of ownership model will only include the manufacturing costs.

### Project strengths:

- This project is addressing critical needs.
- The methodology is a strength of this project.
- Strengths of this project include the life cycle cost analysis for different regions and the monetization of health damage.
- The consideration of different applications and volume production levels with one single modeling program is a strength of this project. The inclusion of a systematic DFMA model is meaningful and different.
- A great deal of background work has been completed, hopefully laying the ground work for many results in the coming year.
- It is appreciated that the cost analysis is being done at several production volumes. It looks like the large break in labor and amortized capital is between 100 and 1,000 units per year. The next break is going from 1,000 to 10,000 units per year, but with lower marginal benefit, as expected.

### Project weaknesses:

- There are no major weaknesses identified.
- The lack of volume effect on the cost of BOP components is surprising and differs from other studies. It should have been detailed in the presentation. The parameters used for the parameter study are different from those used in Strategic Analysis and Battelle studies. Therefore, the comparison is more difficult.
- The ability to get accurate costs is a weakness as companies don't want to share this information. It is hard to predict cost at high volumes. This project should present the future costs as a range and not an absolute number. It is not clear what the uncertainty is.
- This project appears to have a high degree of overlap with two other projects currently funded by DOE. It is recommended that the scope of each be clearly defined to eliminate redundant work where possible. Also, please make sure that the results from these projects are being reconciled against each other so that when the final results are available they are consistent. The project team should more clearly identify the pathways to reducing manufactured cost besides increasing production volume. It is not clear how manufacturers can reduce the materials costs for these systems. The project touts $CO_2$ reduction benefits, but this benefit is not relevant to the national energy policy, as there is neither a carbon tax nor a cap-and-trade system in place at the federal level. As the research team is primarily based in California, it is understood why this is of interest, but it is not relevant to the DOE program currently. The team should review why the BOP cost does not scale strongly with production volume. This may be a result of the assumption that the BOP components used in the largest system essentially scale down to the smaller systems.

### Recommendations for additions/deletions to project scope:

- An annual update of the existing models should be carried out: the update should not wait for the end of the project as presented in the planning.
- This project should consider a collaboration with project FC-083, Enlarging Potential National Penetration for Stationary Fuel Cells through System Design Optimization run by the National Renewable Energy Laboratory.
- It seems like there is the potential for overlap between this program and project FC-097 on the CHP cost estimates. It is not clear how DOE will make sure its funding is being used wisely for these analyses. DOE may be considering a teardown analysis to develop a fuel cell system bill of materials (BOM) and thereby generate manufactured costs. The current BOMs seem to be developed by surveying the literature and manufacturers, which seems less rigorous (or more open to manipulation). It is recommended to add an

analysis to identify the top 5 to 10 systems that contribute most heavily to the overall fuel cell system cost and then provide recommendations on how to mitigate their effects on fuel cell system cost.

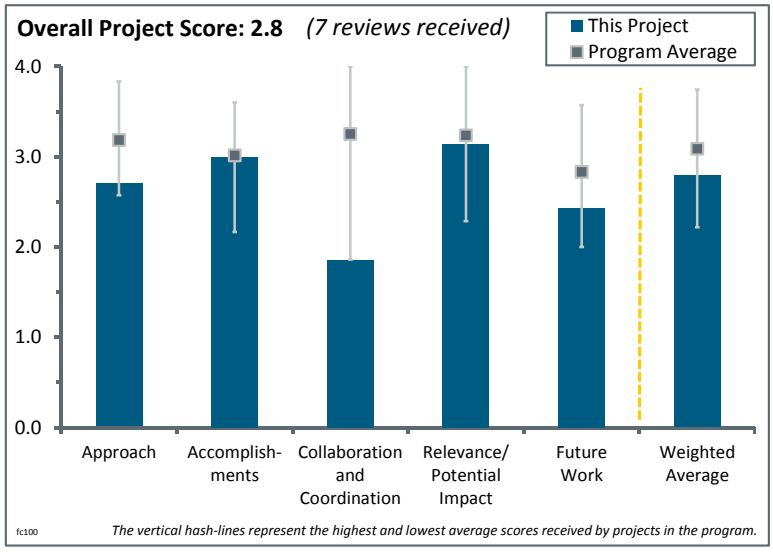
- The researchers should look at two things. First, some of the cost numbers, even at low volume, would portend that there should be some markets where fuel cells can break through. It is not clear what the barriers are that prevent adoption. The researchers should clarify if the cost numbers are right, or if there are other costs that swamp direct costs (i.e., assume that a company invested a couple hundred million dollars to make a product) and they make 33% gross margin on a 100 unit/year market, with a ramp of some rate. It is not clear when the investors will get paid. At least for their own purposes, this quick analysis should be done. Second, benchmarking to the Japanese New Energy and Industrial Technology Development Organization (NEDO) program for small stationary CHP should be done. Those systems were considerably more expensive than projected in this report (though they were quite small). What can be learned from those data to calibrate the "should-cost" here and the "does-cost" of the real world?

## Project # FC-100: High-Aspect-Ratio Fuel Cell Catalysts

Brian Larsen; National Renewable Energy Laboratory

### Brief Summary of Project:

The objective of this project is to produce novel, high-aspect-ratio, nanostructured, platinum (Pt)-based catalyst materials with increased activity and increased Pt utilization. The synthesis of Pt-alloy extended thin-film electrocatalyst structures (ETFECS) using three different alloying metals allows for maximization of Pt ETFECS specific activity. The development of methods to increase ETFECS surface area allows for increased mass activity of the Pt ETFECS. Finally, ETFECS are integrated in membrane electrode assemblies (MEAs) and evaluated for effectiveness.




### Question 1: Approach to performing the work

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

- This project was very straightforward and directly focused on the objective of reducing Pt loading.
- Novel, high-aspect-ratio Pt structures are being developed.
- The approach is sound, but it would have been good to include a pure Pt baseline for comparison. Maybe it was a 2012 objective, but no data was presented.
- Pt-alloy ETFECS is a reasonable approach to meet U.S. Department of Energy (DOE) cathode catalyst performance and durability targets; however, the National Renewable Energy Laboratory's (NREL's) approach for optimizing these materials was not explained.
- This work involves the fabrication of catalyst formulations and testing of these in the fuel cell environment. The approach is to address targets, numerical values set by DOE. The goals are to achieve performance by using the precious metals more fully, the result of additional dispersion.
- The approach described seemed to be disjointed; the continuity between the different preparative methods to enable a comparison between factors in catalyst and preparative procedure was discontinuous.
- This project created high-aspect fibers to reduce Pt loading in polymer electrolyte membrane fuel cell electrodes. However, it is not clear that this approach will actually reduce Pt loading versus creating a high dispersion of supported Pt nanoclusters, since reducing particle size for higher dispersion will only result in shorter fibers and, ultimately, high-aspect-ratio will no longer be a factor.

### 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 project was completed on time and within budget with excellent results.
- The project has met most of the goals and the performance tests have been performed.
- This project has successfully made nanofibers and demonstrated catalytic activity.
- Progress is satisfactory. The idea was to make nanowires of Pt alloys—one set of Pt-Ni alloys and a second set of Pt-Co alloys. Successful alloy structures were prepared and the data show good results.
- The accomplishments are good, but there is no science. It is not clear why this works.

- The project was shown as 100% complete, but no durability testing was done. This testing is important because leaching transition metals from alloy catalysts is one of the limiting features of these types of catalysts. Beginning-of-life mass activities in MEAs and rotating disk electrodes (RDE) are still below DOE targets.
- Because the principal investigator (PI) has left, the work is in disarray and the plan to complete the effort and provide a high degree of technical understanding of the effect of alloying on catalyst performance seems doubtful. It was fortunate that Shyam Kocha was available to describe much of the work for which he was not responsible, as he defended the project. It is not clear what advantage ternary alloys would offer with the limited understanding provided on the alloys presented. It may be relevant to investigate other methods of characterizing the Pt catalyst to establish a correlation with the observed difference in performance relative to the baseline. It is also unclear what ratio of Pt to alloy metal is optimum relative to the preparative method used and how this is important.

### Question 3: Collaboration and coordination with other institutions

This project was rated **1.9** for its collaboration and coordination.

- It does not appear that any collaboration was required for this project. There should be an "N/A" category for this question so that the overall score is not affected.
- There was no collaboration with outside organizations.
- The presentation did not show much collaboration.
- There is no evidence of any collaboration with other institutions.
- There were no collaborations. Even if the project is a success (and, arguably, it is) there is no path forward.
- It was not clear that this was coordinated with any other effort, although the General Motors work certainly has some important parallels.
- The presentation showed no collaboration with other institutions, although clearly this omission is an oversight given that success is based on participation by a variety of outside vendors and colleagues. In this presentation, they were not described.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The structures are unique and the performance significant.
- This project is aligned with critical areas of fuel cell cost and durability.
- Reducing the Pt content in fuel cells is critical to achieving cost reduction targets. By definition this project does that by developing and testing three different alloys.
- The project represents an interesting approach; however, galvanic displacement may be expensive and difficult to manufacture and handle for these materials.
- If the project had been carried out in a more systematic manner and details associated with the impact of the preparative procedure and catalyst involved were more cohesive, this effort could have provided a very valuable understanding of how preparative procedure variables can affect the activity and stability of the resultant catalyst.
- There has been considerable effort on achieving improved Pt electrode performance with a metric of the number of watts resulting from a certain mass of Pt. This work encompasses decades of effort. Because Pt has a high price, using less of the metal is one way to cut device costs. This work is clearly relevant. The result was that the Pt-Co alloy electrodes, utilizing the high dispersion afforded by this approach, resulted in superior performance.

### Question 5: Proposed future work

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

- Most of the proposed work has been accomplished.
- The suggested future work simply extends these tasks and includes looking at ternary (three element) alloys, along with a continued effort to incorporate these highly dispersed materials into usable electrode structures.
- The future work includes (1) "Synthesize ternary Pt-alloy ETFECS with sacrificial, 'leachable' metal (e.g., Fe, Mn)" and (2) "Increase performance at high current densities;" however, there are no specifics as to how NREL plans to do these two things. No durability tests are planned.
- There needs to be some focus on getting the in-situ performance to match the RDE data by structuring the catalyst layer. The project should not go into ternaries at this stage. The durability of these structures must be investigated in-situ. Characterization must go beyond microscopy. It is not clear what the crystal phase is or what the ratio of Pt to Co is. More work needs to be done here.
- Because the PI has left and the effort does not seem to have been carefully and systematically pursued, the balance of funding does not seem adequate to bring this effort to a meaningful conclusion.
- There are no comments on the proposed future work—the project is complete.

### Project strengths:

- The project team has accomplished the objectives on time and within budget.
- This project has a novel approach for meeting DOE goals.
- This idea is great idea and has a neat synthesis route.
- NREL is studying potentially relevant materials.
- The experimental details of this synthesis process require a skill set of considerable ability. This success involves an excellent experimental team. That team makes the difference. Good people are the primary strength.
- The plan to investigate preparative procedure variations on the performance and character of different catalyst species is a worthwhile and valuable endeavor and would assist in understanding the importance of advancing the techniques of catalyst preparation. Combining catalyst preparative variations with electrochemical performance evaluation is the correct methodology.

### Project weaknesses:

- This approach is a potentially expensive method to generate high activity.
- This project has no science, no partners or original equipment manufacturers, and no future work.
- This project lacks a systematic approach to design improved materials and durability testing.
- The project did not seem to be well organized in coordinating between preparative nuances and the various catalyst species being investigated. The amount of time spent on what was presented does not seem to allow for time to complete the work, unless another investigator can be assigned to finish the task.
- It is not clear that this work will really reduce Pt loading versus just resulting in a high dispersion of supported Pt nanoclusters. Reducing the particle size for higher dispersion will only result in shorter fibers and, ultimately, a high-aspect-ratio will no longer be a factor.
- There is a long history of Pt-Co alloys in fuel cell technology, but stability has been a previous problem. Depending on testing conditions, enhanced activity can show some endurance, maybe to 1,000 hours, but the Co slowly dissolves and activity loss occurs. If the test period is only a few hours, this result will not be apparent. However, when moving towards 30,000 hours of durability testing, this dissolution will usually result in the same performance that would have resulted with a pure Pt formulation. If that is the case, there is perhaps no advantage in alloying. It just does not pay. There has to be some longer term testing before this advance can be fully understood. The nanowires always look so fragile. Unfortunately, making fuel cell electrodes involves some pressure and fuel cell testing usually involves compression on the test fixture. While not a weakness really, the test results will be easily influenced by the test fixture design.

### Recommendations for additions/deletions to project scope:

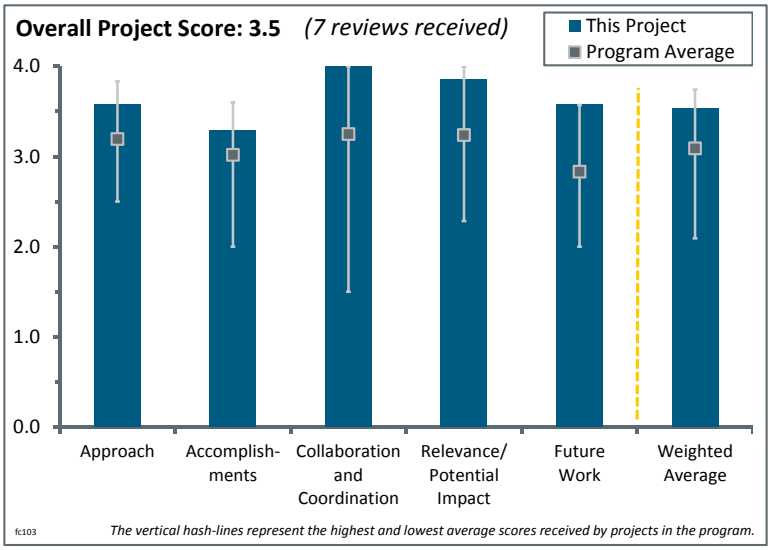
- Any continuation of this work should be rolled into Bryan Pivovar's ETFECs project.
- The project should do some durability testing. A great deal of work has to be done to get the catalyst layer to work with these materials. The next phase of funding should be used to sort this out.
- It was highly considerate of Shyam Kocha to present the task, considering that the PI had left and Shyam was only peripherally involved. Because of his background and understanding, it would seem appropriate that he help seek a qualified individual to bring the task to some logical and meaningful conclusion, perhaps with the assistance of others who have been working in this arena.
- It is smart to encourage skilled experimentalists to continue to explore ways to approach Pt fabrication that achieve more watts with less Pt. The next task here should be some replication, proving that the result can be obtained repeatedly. The other task of critical importance is building these complex micro wires into useful electrodes. There also needs to be some consideration of the amount of energy expended in spreading the Pt into a high dispersion contrasted to the amount of energy that investment will recover. Even at this early stage, it makes sense to ask that question, and perhaps think of less energetic approaches. In truth, today, the high-performance polymer electrolyte membrane electrodes have a cost well beyond the cost of Pt in those electrodes.
- There are no recommendations. The project has been completed.

## 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 air management hardware efficiency, including compressor/expander efficiency greater than 65%/70% at 25% flow by 2017, combined motor/controller efficiency greater than 90% at full flow by 2017, and compressor/expander input power less than 8/14 kW at full flow by 2017. Secondary objectives include conducting a cost reduction analysis and achieving a fully tested and validated air management system capable of meeting 2017 project targets.




### Question 1: Approach to performing the work

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

- This project features a very clear approach, a good and clear list of deliverables, real-world validation, and good risk mitigation analysis. It is a well-managed project.
- This project features a highly focused systems engineering approach to improving air compressor technology.
- The approach is sound and offers an interesting alternative to high-speed centrifugal turbomachinery approaches. The researchers plan to reduce parts count and lower rotational speed (to allow conventional bearings and motors); application of near-net-shape rotors supports reduced system cost.
- The project approach is very good. It builds on Eaton's extensive experience with Roots blower technology and appears to be logical and comprehensive. Every subcomponent in Eaton's current technology is being looked at to determine where cost savings can be achieved while still meeting performance targets. The project is focusing on a lower speed alternative to turbo machinery, with its costly air bearing system for high-speed operation.
- Eaton presented a detailed discussion of its approach. The approach builds on the experience of Eaton and incorporates modeling methodology to develop designs at a greater rate. The researchers build on the experience of Argonne National Laboratory (ANL) to provide fuel cell performance data and fuel cell systems understanding that will accelerate the development of the compressor/expander. Eaton has coupled the modeling efforts with hardware manufacturing and systematic testing of the components. Eaton moves from component hardware development to prototype development and testing with a major fuel cell stack manufacturer.
- Eaton is aware of cost issues and how cost is affected by balance of plant component and system simplicity and integration. The single-shaft compressor expander motor configuration will reduce cost and efficiency losses. Modeling is being used to assess the impact of configurations without the need for multiple hardware builds. A roots blower "mule" is being used to cheaply and quickly test concepts ex situ.
- The project could be of value to developers if compressor costs are decreased. Investigators have done well in partnering with Strategic Analysis, Inc. (SA). Weight and volume targets are accounted for by the decision to go to plastic rotors, along with use of fewer bearings and a more integrated rotor design. Finite element analysis on the rotor has shown that there is a feasible solution.

### 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 team has made excellent progress.
- The project has met all milestones to date and is on track to pass the June 2013 go/no-go decision point. The go/no-go criteria were not discussed quantitatively.
- The progress made in less than a year shows promising prospects. Major modifications/improvements of compressor-expander design have already been designed. There are very good complementary modeling and validation activities. The test stand is ready.
- Baseline compressor testing was conducted, showing fair/good results. Modeling of the expander and overhung rotor motor was conducted, but not completed. Substantial progress has been made, but there appears to be substantial additional work before the July milestone.
- The accomplishments during the first year are impressive. The team appears to have hit the ground running, so to speak, and both modeling and experimental results are very good. Trade-offs are being examined to achieve the most impact on system cost while meeting performance requirements. Despite the excellent progress, it is not clear if the project will meet the DOE 2017 targets even if completely successful. Increasing expander efficiency to 80% from the current 64% will be difficult, and the presentation does not provide a path forward that can ensure success.
- The project is new and test results are only just becoming available. Eaton appears to be systematically addressing each of the topic areas. Eaton is sharing data with ANL and is reporting compatibility of their own performance data with the ANL model. The data on expander experimental results were confusing; two of the three expanders were identified for additional testing, although only one of the two had tests described. It is unclear if the other one is not going to be tested. Compressor performance work is in the early stages without any conclusions available. The team is developing a model for compressor and expander computational fluid dynamics (CFD). The design of the overhung motor rotor analysis is underway. The test stand is under development and no data has been reported. The team has conducted expander plastic rotor analysis and has demonstrated 20,000 rpm, but materials properties may not be acceptable.
- It is unclear whether the plastic and aluminum materials would contribute to contamination. The project team needs to confirm noise, vibration, and harshness after new materials and design. More detail is needed on the test stand setup. This is especially true in understanding the accounting for the humidity inlet to the expander.

### Question 3: Collaboration and coordination with other institutions

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

- There is excellent collaboration with industry partners.
- The interaction between ANL and Eaton has been excellent and productive.
- The collaboration is excellent. Analytical support is being provided by ANL, SA, and Kettering University, and final prototype testing is planned with an 80-kW system at Ballard.
- A strong team was put together for making hardware, developing models that evaluate the performance of "real" hardware, and performing a detailed cost analysis. The team has a lot of capability.
- The project features a good mix of industry, institute, and university partners. The partners cover all of the necessary areas of know-how. The group is as big as necessary and as small as possible—it promises efficient and effective work.
- Simulation collaboration with ANL and cost modeling with SA are good. Collaboration with Ballard on inserting a real system between the compressor and the expander will be good. CFD with Kettering University should help with the rotor design.
- Partners include a national laboratory, a university, a stack system developer/supplier, and a cost analysis specialist.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

This project was rated **3.9** for its relevance/potential impact.

- Development of a low-cost, high-performance compressor/expander is critical for PEM fuel cell advancement.
- The project has high relevance to the DOE Hydrogen and Fuel Cells Program (the Program). The air handling system is a significant contributor to the system cost. Any means of reducing the cost should be investigated.
- Development of a new air intake system is critical to providing the weight, volume, and cost savings to commercialize fuel cells for many applications, particularly automotive. The technology has been overlooked until now, but it is now being studied. This is a long-overdue project.
- The single-shaft compressor/expander motor configuration will reduce costs and increase system efficiency.
- The parasitic losses in the air system of a fuel cell are a key issue. The project is addressing this in an excellent way.
- Eaton seeks to develop a robust, high-efficiency compressor/expander/motor unit optimized for fuel cell use. Because Eaton is an automotive industry supplier, it fully appreciates the need for reliability and has designed its system accordingly. Consequently, the potential impact of the project is substantial because there is a clear and strong need for a reliable (and low-cost) air system.

### Question 5: Proposed future work

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

- This project features well-organized future work with a schedule for the completion of activities.
- There is a very clear plan for the right activities. Validation in an 80-kW system is a real challenge; it is also proof of the confidence of the partners to achieve this goal, which is fully supported by the impression one gets from the presentation.
- The future work plan is very good. Prototypes will be fabricated and tested in year 2, and an updated cost analysis will be completed. If completed, these activities will enable DOE to determine if the concept will meet Program targets.
- The plan is concise, the go/no-go criteria are set right, and the project has identified the right tasks to do. The go/no-go decisions every July offer the capability to off-ramp if the technology does not work out, which is good.
- The plan is logical: Period 1 is designing; Period 2 is building and testing at Eaton; and Period 3 is testing with a fuel cell at Ballard.
- The proposed future work is in line with the project schedule and milestones, but the first budget period go/no-go criteria need to be quantified. More discussion of cost analysis would be helpful.

### Project strengths:

- The experience and expertise of the team members will greatly help this project.
- This is a well-designed and managed project. It utilizes industry partners and has made excellent progress.
- An excellent team made excellent progress in year 1. Most projects with multiple collaborators and subcontractors take considerably longer to form a team and get the necessary paperwork in to DOE.
- Eaton knows how to produce technology and develop a product at significant cost savings. Targets have been well planned. Collaborations are with the right parties.
- The concept builds off of existing machinery for which much experience and technical detail is known. Eaton is a very credible developer and future manufacturer of the systems. Eaton knows the requirements very well.

### Project weaknesses:

- More detail is needed on the laboratory setup (expander test stand). This will strongly influence "Period 2" in the work plan.
- Eaton is working on plastic expander housing. It does not seem that Eaton is aware of potential fuel contamination issues from the plastic component. The team should consult with the National Renewable Energy Laboratory on system-derived contamination.
- The project does not estimate the potential cost savings if the work is successful. This analysis probably exists and has been presented to DOE, but there is no indication in the presentation of the cost savings possible if the project is completely successful. Without this analysis, it cannot be determined if the proposed effort will result in a device that meets the DOE targets.
- Much work remains in Period 1I. Further results would be expected if the researchers are going to make the July design deadline. The expander efficiency is reported at only 64%—short of the 73% 2011 status and well short of the 75% 2015 target. It is unclear why efficiency went down and what the plan is to raise it. Compressor efficiency is reported at 74% at full flow, but the performance maps do not match this high level. An explanation of this discrepancy was not given. Much is made of reduced rotational speed to avoid "costly" bearings. However, foil bearings should not be considered costly. Thus, a driving element of the researchers' design appears to be faulty. The overall pace of the project is a bit slow. One would think that a derivative product such as this could be developed in less than 3 years.

### Recommendations for additions/deletions to project scope:

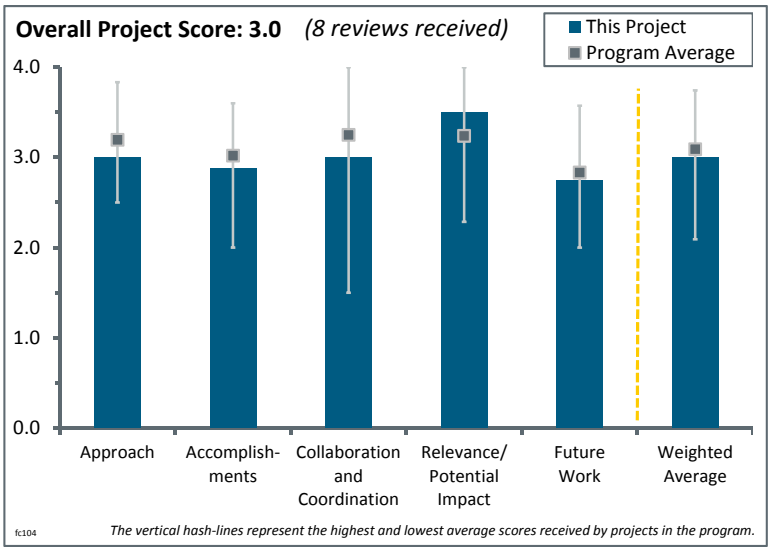
- The research team should keep up the good work.
- A cost analysis is planned. A preliminary cost analysis conducted during the design phase would have been useful input into the final design.
- The team should present an analysis that indicates the estimated cost savings for each individual change in component specifications. The sum should be a reasonably reliable indicator regarding the ability of the Roots technology to meet DOE performance and cost targets.

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

Andrew Steinbach; 3M

### Brief Summary of Project:

The objective of this project is to develop a durable, low-cost, robust, and high-performance membrane electrode assembly (MEA) for transportation applications. The project's approach will optimize integration of advanced anode and cathode catalysts, based on 3M's nanostructured thin-film (NSTF) catalyst technology platform, with next-generation perfluorosulfonic acid proton exchange membranes, gas diffusion media, and flow fields for best overall MEA performance, durability, robustness, and cost.




### Question 1: Approach to performing the work

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

- This novel approach both maximizes the performance and minimizes the amount (and consequently cost) of Pt in a fuel cell catalyst by maximizing Pt utilization.
- This project was exemplary in terms of the thoughtful plan and approach that was presented. It was clear that the strong research culture at 3M informed the systematic, but still creative, attack on the complex problems of electrode assembly properties and performance.
- This approach for developing an MEA that meets targets is well planned and comprehensive. The project uses an aggressive technological approach that is undoubtedly necessary to meet targets, particularly operating the cells at high temperatures (90°C) while using thin (<25 µm) and low equivalent weight membranes. Even if durability testing appears satisfactory, it is questionable that such a combination of temperature and materials provides a "safety factor" for avoiding membrane failure that is sufficient for commercial application.
- This project features a good balance of basic research and applied work that has led to a better understanding of the NSTF system and improvements in performance. Studies of the structure of NSTF catalysts before and after surface treatments, conditioning, and dealloying have been used to guide cathode catalyst improvements, leading to better peak power performance. Membrane studies have led to understanding how base metal leaching affects membrane and MEA performance. Water management work is integrated with the Lawrence Berkeley National Laboratory (LBNL) project. More of the effort and focus should be directed toward what appear to be the limiting factors for NSTF technology—water management and overcoming low-temperature operational issues.
- This project aims at further optimization of the NSTF format in the form of MEAs and with the use of bimetallic catalysts. The principal goal of this effort is to meet and exceed the U.S. Department of Energy (DOE) 2017 targets for MEA performance and loading, which include 0.44 A/mg of Pt at 0.9 V (iR free) and 0.720 µA/cm$^2$ MEA specific activity. In addition, the total metal loading target for the MEA is 0.2 mg/cm$^2$ or 0.2 gm/kW. Here, the primary focus is the NSTF cathode performance improvement via dealloying and annealing and electrode integration in an MEA format. The approach toward these goals follows the expected lines.
- The approach is very rigorous and the team is working very hard in a certain direction. The team should consider doing a full impedance analysis to better understand the results from the high-frequency resistance

tests to discern whether there is an impact from another effect in the MEA. The researchers might also look at a broader range of test conditions.

- Durability testing is shown to be completed by quarters 7 and 11, which is far too late in the project to be useful for informing major integration decisions. Given that the project is an integration project with no component development, it is curious to see a few slides on the continued $Pt_3Ni_7$ cathode catalyst development. It is very likely that the project will have to revert to doing some component development in order to achieve integration goals. The success of many of the integration tasks will be dependent on cell design and assembly parameters (e.g., thermal mass, compression force, and channel geometry). The project needs to state how the results can be interpreted to be universal for developers.
- This project appears to essentially be a continuation of what 3M has historically been doing to develop NSTF-based MEAs. The NSTF catalyst architecture is very promising because it appears to offer a route to both high activity and improved stability relative to conventional dispersed catalysts. However, after more than a decade of development, there are still no commercially viable NSTF MEAs. The principal reason is that NSTF MEAs are extremely sensitive to both temperature and impurities relative to conventional MEAs. Therefore, it would seem that DOE's focus on thin-film catalyst architecture should be to determine if these fundamental barriers to the commercialization of NSTF can be overcome. Doing the same thing over and over again should not be expected to yield a different result. Again, this project is essentially just a continuation of what Mark Debe did for a decade; namely, continue to tweak the catalyst compositions and treatments (e.g., improve intrinsic activity), and change the membranes and/or gas diffusion layers (GDLs) (e.g., to alter the water management). However, 3M does not appear willing to actually change the catalyst-layer composition or architecture, which severely limits the probability of success here. In fact, the principal investigator (PI) states that this project is limited to "optimization of existing components and processes" and that there shall be "NO COMPONENT DEVELOPMENT." This limitation is very unfortunate because making NSTF successful requires much more than optimization; it requires a new catalyst layer architecture.

### Question 2: Accomplishments and progress toward overall project and DOE goals

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

- Good progress has been achieved over the relatively short duration of the project.
- The performance of 0.48 A/mg Pt at 0.9V for $Pt_3Ni_7$, as shown on slide 7, is impressive. There was a careful tracking of catalyst composition as a result of catalyst preparation, which helps make the results clear and understandable.
- Good progress has been made in the short time the project has been active. The project has already made improvements in decreasing platinum group metal (PGM) loading and increasing performance at high power.
- The mass activity is promising with SET processing of $Pt_3Ni_7$. A controversial point has arisen with regard to the assignment of ohmic resistances. Physical explanation needs to be provided for the "variable" cell area that is used to get iR-free curves to overlay each other. This may be due to protonic resistances that vary, but it is not accounted for in a high frequency resistance (HFR) measurement. The achievement of 20 $\mu g/cm^2$ Pt on the anode appears to be outside the context of adding an oxygen evolution reaction (OER) catalyst. Another 3M project shows that at least 29 $\mu g/cm^2$ is needed in the presence of an OER catalyst. The question exists then as to whether the two projects agree with each other. Proton pump data for the low-loaded anode would be interesting to see. It may also be good to see testing with $H_2$ dilution on the anode (40%–100% $H_2/N_2$).
- It appears that the properties that have been achieved are quite good, but the surprising, relatively low benefit of the SET on the $Pt_3Ni_7$/NSTF cathode activity was perplexing. The project is still relatively young, so perhaps the cause of this low benefit can be identified. Overall, the systematic and scientific approach instills confidence that the basic premise of the work will ultimately pay off with good results.
- The major barrier to commercialization of NSTF is the high sensitivity to operating conditions (as shown on slide 4 for Tasks 2 and 6), yet the progress on these tasks is delayed or has not even started. In addition, no real progress will be made in these key areas unless a different approach than what has already been tried is utilized, which does not appear to be in the plan. There does not appear to be a good fundamental

understanding of what is limiting the NSTF cells, as evident in slide 15, which does not explain the dominant mechanisms controlling the performance.

- The researchers have a long way to go on all of their metrics. It is not clear from the presentation which metrics are system-dependent and even flow field-dependent. The reported 67% loss of mass activity looks like a big concern, so while there are many passing metrics, it seems like it would be helpful to rank the importance of the various criteria. They should be ranked "good" because they seem to be progressing on so many fronts, but some of the results, while reported to three significant figures, might be misleading because of the use of optimal test conditions.
- The initial performance at this early stage is modest compared to the previous generation of work, especially the work conducted using SET. The principal difference appears to be related to grain boundaries, size, and morphology of the crystallites. In this context, some attention should be paid to the density of the resultant crystallites because it appears to be significantly lower than the bulk values and previous-generation SET-treated samples. Some background into the logic used to select the elements for cathode electrocatalysts should be specified. For example, it is unclear why PtCoMn was chosen as a ternary alloy. In addition, because the structure of the interface is very different from the conventionally supported electrode MEAs, some additional metrics on the activity at higher current density should be adopted. It is strange to see $H_2$/air comparison at 0.9 V, where partial pressure of $O_2$ should not be a factor, and not have concomitant metrics at 0.65 V to see differences in the onset of mass transport. Because this work is aimed at MEAs and electrode structures, a He, $N_2$, and $O_2$ study should be part of this effort. In addition, it is strongly recommended that the team pursue some semblance of transport modeling to see what elements of the electrode structure, ionomer content, equivalent weight, etc., are crucial.

### Question 3: Collaboration and coordination with other institutions

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

- The team includes many smart people who are investigating issues with the GDL, etc.
- The collaborators appear to be making meaningful contributions and working closely with 3M.
- The presentation was particularly effective in showing the multiple contributions and their value to trying to figure out how to better tune the SET approach for the lower-PGM-content MEAs.
- The industry (General Motors) and university (Johns Hopkins University [JHU]) team is lean and effective. This team has brought a new valuable approach and insight into development and optimization of precious metal and possible non-precious-metal catalysts.
- The collaborations within the project appear to be working well. The project is collaborating with a transport project at LBNL. Collaborations with original equipment manufacturers may be beneficial in elucidating what effects proposed changes may have on the fuel cell system (e.g., moving water out of the anode in a system with anode recirculation would probably lead to higher vent rates and lower efficiencies).
- While collaboration with Oak Ridge National Laboratory is evident from the transmission electron microscopy data, the corresponding collaborations with other institutions such as LBNL, Michigan Technological University (MTU), JHU, and Argonne National Laboratory (ANL) are not clear. Maybe it is too early; for example, work at JHU only started in February 2013, almost at the point when this presentation was requested.
- The PI needs to have more interaction with partners that can help to determine the root cause of the MEA performance issues and be open to suggestions on how to resolve these issues. The primary focus of the project should be on the development of a viable MEA, not continuing to make minor changes to the catalyst compositions and treatments.
- For an MEA project, it is extremely surprising to see that the list of collaborators does not include a stack developer (either automotive or otherwise). The three national laboratories and two universities will not have access to the operating conditions and cell designs needed for context. ANL is being tasked with hydrogen oxidation and hydrogen evolution reaction kinetics studies, but the slides refer to personnel associated with modeling. The question remains as to whether this is a modeling study. It appears that some of the collaborations are focused on dealloying Pt-Ni catalysts. Perhaps there are other parties that have already gone down the same path with other morphologies that could help. The list may include Nenad Markovic or some automotive developers.

### Question 4: Relevance/potential impact on advancing progress toward DOE research, development, and demonstration (RD&D) goals

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

- The topic of the work is crucial to the long-term economic success of $H_2$ fuel cells, and it appears that the strategy being followed has a very good chance of significantly and positively impacting that topic.
- The project has strong relevance towards meeting DOE targets for low-PGM content and durability by improving electrode structures and mass manufacturability.
- As long as Pt is needed, this kind of approach is a valuable tool for lowering costs.
- The primary effort of meeting the 2017 targets is highly relevant and, if successful, the potential impact on the Program would be considerable.
- The project is apparently on the path to a high-performance catalyst.
- The goals of the project are well aligned with Program goals and objectives, which is why NSTF has deserved DOE's support for the past decade. However, the promise of NSTF will never be realized if the MEA performance issues are not resolved, and this will not be accomplished unless a different approach is undertaken.
- An optimized MEA with $Pt_3Ni_7$ NSTF properly conditioned with thin, durable membranes could be very valuable to developers. There is some question here as to whether an MEA integration project is appropriate for public funding. Integration activities are usually pursued with developer funding because integration is often fairly particular to stack design and operating strategy.
- This project is focused on integrating NSTF catalysts into an MEA. NSTF has been the most active and most durable catalyst, but it has issues with integration in an MEA due to water management. This project addresses optimizing the rest of the MEA to integrate with NSTF and overcome the water management issues. If successful, it would allow for low-loading, low-cost, durable NSTF catalysts to be utilized effectively in fuel cell systems.

### Question 5: Proposed future work

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

- The proposed future work is in line with the aims and objectives of this effort.
- The proposed future work is consistent with the approach and the experimental plan already underway.
- The proposed future work addresses issues that were delayed in the program, and they are important to give attention to. These topics are noted on slide 4 of the presentation, and the PI clearly has these in focus and on the "to do list," so the proposed future work plan looks good.
- The proposed future work is appropriate. It was stated that transient response and water management will be the main focus for next year. Area should be the main focus of the project because it appears to be the limiting factor for NSTF technology.
- Test plans should be expanded to intermediate temperatures.
- The study needs to better prioritize durability. The full suite of DOE-prescribed stress tests should be done at the beginning of the project to understand the response to relative humidity (RH) cycling, base metal dissolution, and other possible concerns. It would be good in the future work section to see what stack formats and operating conditions will be used for the integration activities. The systematic study of land-channel characteristics needs to be outlined.
- The team has to make clear the effect of electrode area normalization. If the flow field is the same, then the area of the flow field should be the correct area of the fuel cell electrode. One way to see if this is true is to leave the linear activation zone and run the cells in the mixed transport activation region (until the curves bend) to see if the curves bend uniformly without and with area normalization. Dealloying offers another way to make effect catalysts through thin-film nanoporous supports; this should be explored for precious and non-precious metal catalyst work.
- The team must focus on the key barrier, which is the poor performance of the MEAs under realistic operating conditions. This reviewer was optimistic that having a new PI on 3M's NSTF program would also result in a new approach, but this does not appear to be the case. Instead, the plan appears to be to continue to tweak (or "optimize") the catalyst compositions, when the "elephant in the room" is the

unacceptable sensitivity of the resulting MEAs. The PI must be open to making substantial changes to the catalyst layer composition and/or architecture to resolve this key barrier. The team does not appear to understand the root cause of the MEA performance sensitivity. For example, it should be apparent that 3M's hypothesis on why changing the anode GDL somewhat helps the performance is flawed, because it states in a 2010 *ECS Transactions* paper, "While a significant and consistent performance difference was observed between two anode GDLs at both 30 and 50 C, no significant difference in product water distribution out the anode and cathode was observed." This paper does not agree with their stated hypothesis in the same paper that the performance improvements are due to "the amount of water leaving the cathode was also reduced and thereby the $O_2$ transport restrictions were reduced." Simply changing GDLs without actually understanding the mechanisms responsible for the change in performance is not an efficient way to address the key barrier. The root cause must be determined, and then truly effective mitigations may be efficiently developed. The root cause may be differences in saturation levels (not flux rates); this hypothesis would lead one to explore different GDL options. Additionally, the limitation may not be $O_2$ transport under some conditions, which may lead to an entirely different approach.

### Project strengths:

- This project features good teaming and innovation.
- This project features a strong, experienced, and productive team.
- The project is leveraging many years of DOE and 3M investment in catalyst improvement.
- This project features good collaboration and integration of the work at subcontractors and 3M—transferring basic knowledge and understanding of the system to the application in an MEA.
- The highly systematic approach was particularly impressive and is a clear strong point.
- The main strengths are in the NSTF morphology and performance. The scalability and potential to surpass the DOE 2017 targets for performance and durability are strong.
- The project team is working on one of the most promising catalyst architectures to date for PEMFC MEAs with high activity and good inherent stability. The project lead is also an MEA supplier, so the development of successful technology should also result in commercial products. The lead is a polymer company, which is the competency required to develop and manufacture viable MEAs (i.e., much more likely than a company that specializes in making catalysts).
- The investigators have years of experience developing a high-specific-activity catalyst that avoids support corrosion. 3M has shown the ability to generate the data needed to complete the project. 3M fabrication processes have the ability to generate the materials needed to complete the project. MTU is the right party to have on the project for water management modeling, especially with regard to pore network modeling.

### Project weaknesses:

- Although the routine characterization work is being very well handled, the area normalization is cause for some concern. A little more thought needs to go into the data analysis.
- It seems that some of the tasks have not been achieved as planned and were delayed. It was not completely clear why this happened, but there was some mention of an equipment or facility availability problem. In any case, it seems that the team is committed to achieving the various tasks in a timely manner, so this should not be seen as a serious problem.
- The main weakness is that this effort may not surpass the previous generation's NSTF, multicomponent, Pt-based catalysts. There is no clear approach, either computationally or in improving electrode structure through a scalable transport model. In short, the novelty of this project is unclear.
- The project results will be sensitive to cell design and operating conditions. Being an integration project, it overlaps with activities usually pursued by a stack developer. However, no stack developer was listed among the collaborators. The future work plan focuses first on beginning-of-life characteristics and later on durability.
- The relevance and effects of RH are glossed over. Lower HFRs are needed to achieve targets; yet, where noted, the cells are operated relatively dry. Because it would be difficult to avoid saturation at the anode, these conditions would seem to have significant stack and system ramifications.
- The team is doing the same thing over and over again and expecting a different result. The project is not focused on the primary issue that is preventing the commercialization of NSTF technology, namely,

unacceptable MEA performance sensitivity. There is an apparent lack of expertise in diagnosing what limits the performance of an MEA under various operating conditions.

- This project is a cause for concern. While all of the data looks really good, it is well known that 3M has a history of showing great laboratory results that do not translate well to practical stacks. The question is really whether they are carrying out the "right" tests. The MEA performance is all evaluated under "standard" test conditions, but such tests were really only validated in stacks for carbon-supported Pt nanoparticles. There is no assurance that the MEA test conditions for the 3M catalysts are best assured by the methods developed for Pt/C due to the significant differences in the catalyst layer structures. It would just be a shame to do all of this development and then fool everyone (including themselves) by testing in the wrong conditions. The path looks excellent, but it might be too test dependent. This criticism goes beyond just 3M; it seems to be a problem for the whole fuel cell community that standard tests unravel once off the path for Pt/C and related alloys.

### Recommendations for additions/deletions to project scope:

- The project should better prioritize durability. The project team should add a stack developer as a collaborator. The team should also seek to incorporate realistic cell design and conditions and to understand how results can be made universal for various designs and conditions.
- Fairly high operating temperatures (>88°C) are needed to meet targets. However, most testing, where noted, is performed at 80°C. In anticipation that the higher temperature affects longevity, particularly of the membrane, at least some of the durability testing should be done at the higher temperature.
- A clear modeling approach to understanding transport in the electrode structure is required. A fundamental effort needs to be applied to better choose the alloying elements. At the moment, this looks like a serendipitous route to selection.
- The project scope is reasonable and should lead to the desired goals. The academic team member, Ehlebacher, has published a dealloying process for making nanoporous supports to support Pt or Pt alloys (also formed dealloying). An electrode made with the nanoporous support and dealloyed catalyst as an electrode in an MEA should be examined for performance and durability while this team is still intact.
- The research team should focus the project on developing an MEA that performs as well as conventional MEAs under various operating conditions (i.e., performance is not severely degraded at 25°C relative to 65°C). The team should also be open to making substantial changes to the catalyst layers, as required to resolve the MEA performance issue (e.g., add ionomer and/or carbon to the catalyst layers, if needed). It should also quit working on "tweaks" to the catalyst composition until the MEA performance issue has been resolved. It should also include more interaction with partners that can help diagnose and/or model the MEA performance and determine the root cause, as well as propose mitigations.
- The team should perhaps carry out MEA testing beyond cold start-up and 80°C conditions—perhaps some intermediate conditions, such as 30°C, would be helpful to catch problems early. Some thought needs to be given to whether the tests developed for Pt/C in standard catalyst layers are relevant to the 3M catalyst layers, and the team should probably confirm the tests in a high-performance stack. While validating the test in a stack would be very time consuming, and might even be flow field dependent, it might save everyone a lot of grief early and allow 3M to make required changes while it still has time. While 3M is marching along to the DOE-approved methods, it might be worthwhile to question this overall approach.