

2017 – Fuel Cells

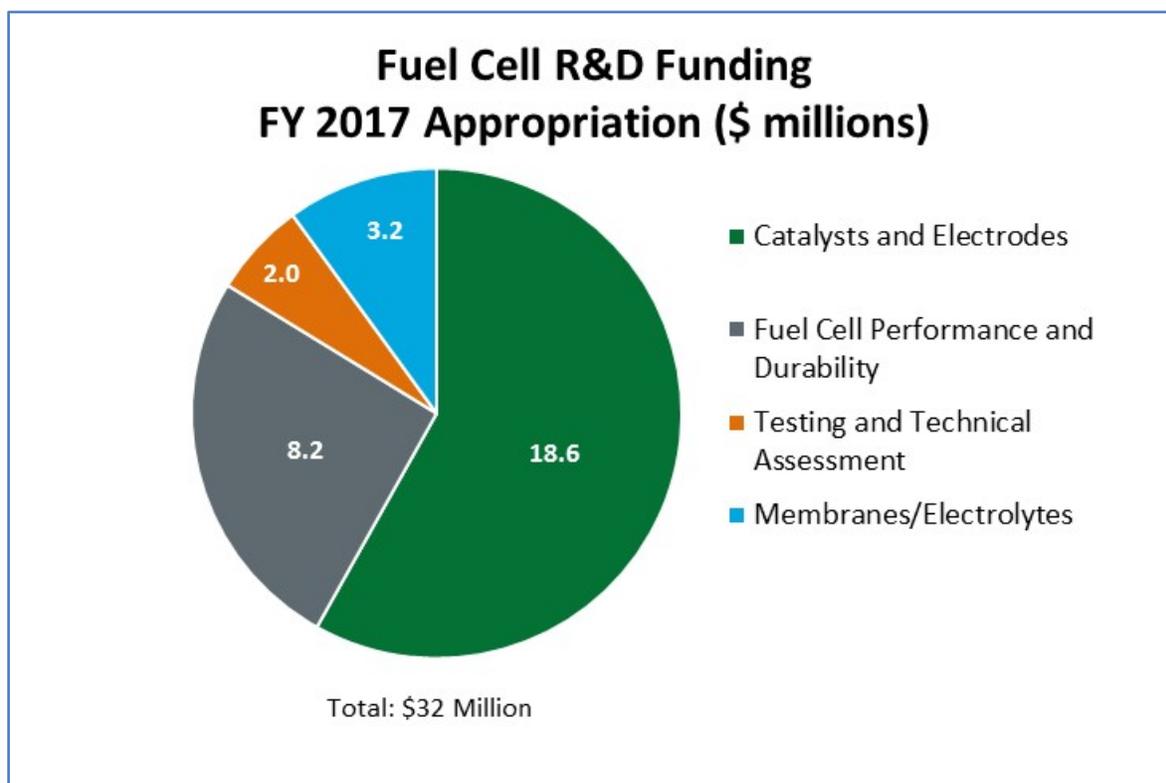
Summary of Annual Merit Review of the Fuel Cells Sub-Program

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

Reviewers commented that there was a good balance between near-, mid-, and long-term research and development (R&D) in the Fuel Cells sub-program, and they agreed that cost and durability are the major technical challenges. Reviewers praised the sub-program's approach to identifying and addressing these issues and noted its well-structured, collaborative, and well-managed projects as a strength. In particular, the consortia established by the sub-program, the Fuel Cell Consortium for Performance and Durability (FC-PAD) and the Electrocatalysis Consortium (ElectroCat), were lauded for their potential to transform fuel cell technology. Key recommendations included (1) increasing focus on technologies that will build on progress achieved thus far, particularly in fuel cell performance; (2) conducting more scaled-up analysis of performance and durability at the stack and fuel cell levels; (3) developing better transport properties for platinum-group-metal (PGM)-free catalysts and a better understanding and characterization of novel membranes; and (4) establishing clear and ambitious go/no-go criteria to enable ending projects that do not meet these criteria. Several reviewers encouraged continued and increased collaboration with relevant consortia and key industry partners such as 3M, General Motors, and others.

Fuel Cells Funding:

The sub-program received \$32 million in fiscal year (FY) 2017. The sub-program focuses on reducing fuel cell costs and improving durability. Efforts included approaches that will achieve increased activity and utilization of low-PGM catalysts, PGM-free catalysts for long-term applications, ion exchange membranes with enhanced performance and stability at reduced cost, improved integration of catalysts and membranes into membrane electrode assemblies (MEAs), and advanced fuel cell performance and durability. There was no funding in FY 2017 for balance-of-plant (BOP) component projects.



Majority of Reviewer Comments and Recommendations:

At this year's review, 39 projects funded by the Fuel Cells sub-program were presented, and 35 were reviewed. Projects were reviewed by between four and eight reviewers, with a median of six experts reviewing each project. Reviewer scores for these projects ranged from 2.5 to 3.5, with an average score of 3.1.

Catalysts and Electrodes: The scores for the 12 catalyst projects ranged from 2.5 to 3.5, with an average of 3.1. Reviewers praised the highest-rated project for its progress on improving the lifetime durability of PGM-free catalysts and the strength of its collaborations with university and industry partners. Reviewers recommended that the project increase its focus on longer-term durability testing of new catalysts. For the lowest-scoring project, reviewers noted fundamental flaws in the approach used to characterize the support stability of the platinum catalysts, citing a need for better understanding. Reviewers also expressed doubts over the project's overall relevance to meeting U.S. Department of Energy (DOE) fuel cell targets.

Fuel Cell Performance and Durability: The seven projects reviewed in this area are all part of FC-PAD, including the consortium overview. All projects scored above 3.0, with a range from 3.1 to 3.4 and an average of 3.3. Reviewers praised the highest-rated projects for their focus on component degradation characterization and electrode optimization, the strength of the teams and their access to a large number of characterization tools, and the design of their approaches. However, reviewers noted that the projects will face challenges if they do not foster stronger collaborations with suppliers, other DOE-funded projects, and original equipment manufacturers. Reviewers felt that the lower-scoring projects demonstrated strong project teams and that their approaches were reasonable, but identified the need to transfer rotating disc electrode findings to useful results at the MEA level.

MEAs, Cells, and Other Stack Components: Two projects were reviewed in this area, with one project receiving a score of 3.1 and the other receiving a score of 3.2. Reviewers felt the highest-rated project, which focused on novel bipolar plate development, utilized a sound cost-analysis-based approach and effective partnerships to achieve targets. However, reviewers expressed some concerns over the scalability of spray-coating for pre-stamped plates and suggested future work focus on analysis of a fully scaled-up system. For the second project, associated with dimethyl ether (DME) fuel cells, reviewers found the approach to be sound but noted that the project lacked a demonstration of the PtRuPd catalyst and remained short of stated targets. It was noted that the potential applications of high-temperature DME cells could help meet DOE's strategic goals but do not support specific polymer electrolyte membrane fuel cell development targets.

Membranes/Electrolytes: The four membrane projects reviewed received scores between 3.0 and 3.4, with an average score of 3.2. The highest-rated project, focused on developing improved anion-exchange membranes (AEMs) and MEAs, was one of the highest-rated in the sub-program, and reviewers commended the innovative approach to studying alkaline membranes, recognizing real progress in in situ testing. They suggested that future work focus on PGM-free rather than low-PGM catalysts. The other alkaline membrane fuel cell project also received praise from reviewers for producing stable membranes by eliminating sulfonamide linkages. It was suggested that the project place more focus on cost and performance. For the lower-rated project, focused on AEMs for high-voltage redox-flow batteries, reviewers found the project's approach reasonable and noted improvements in stability and progress toward other targets. Reviewers were uncertain about the project's relevance to Fuel Cell sub-program goals, given its focus on redox flow batteries, but noted that it could yield benefits in advancing hydroxide membrane technology. Recommendations included focusing future work on testing and improvements in high-temperature fuel cells.

Testing and Technical Assessment: Four projects were reviewed in this area and received scores between 3.0 and 3.4, with an average score of 3.3. Reviewers lauded both of the highest-rated projects for the variety of interesting and helpful findings that can help set realistic DOE cost targets and goals. Both projects were urged to continue work in analysis at the fuel cell system level. Reviewers commented that the lower-rated project was reporting impressive imaging advancements, but the improvements had not been directly linked to specific DOE goals and targets.

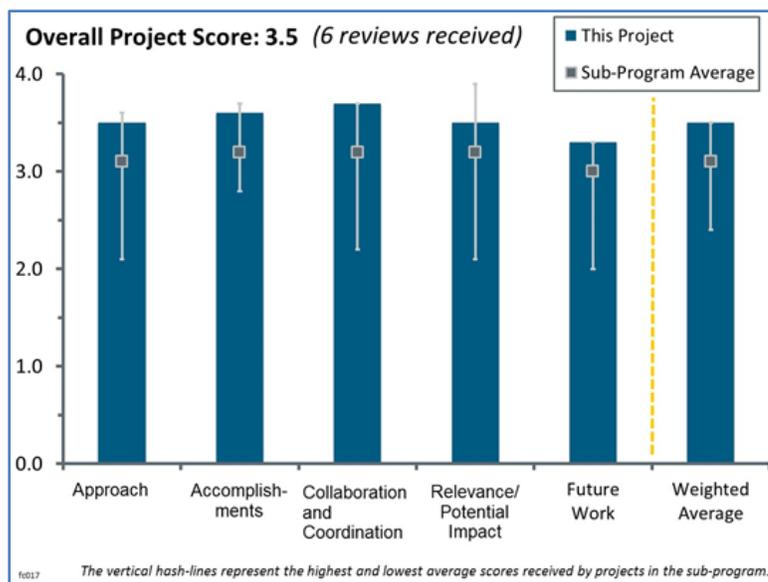
Project #FC-017: Fuel Cell System Modeling and Analysis

Rajesh Ahluwalia; Argonne National Laboratory

Brief Summary of Project:

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

Question 1: Approach to performing the work



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

- This project is certainly valuable to DOE and to Strategic Analysis, Inc. (SA) in helping to evaluate the impact of various cell materials and design options. It is unlikely that any fuel cell OEM is actually interested in these results, since OEMs have their own models that enable them to do this type of analysis. However, it is very important for the entire fuel cell community to be aware of system-level implications of various design/material options, so this project is certainly worth the modest budget.
- ANL employs a sound approach in developing and validating robust models for system design and analysis, which are predictive in nature and made available to the greater community.
- The team has developed a validated systems model that is essential to allowing DOE to guide component targets and project success and allows for high-volume manufacturing costs to be estimated.
 - The approach used in this project is relevant and ambitious. The developed model at the system level is able to achieve cost and durability projections, even if more degradation mechanisms and their impacts on durability should be studied and taken into account in the model.
 - A very good point of the developed tool is that different electrode structures and different catalysts have already been evaluated. Among them are recent developments and collaborations with other ongoing projects. This shows the ability of the developed tool to be updated and refined by integrating recent achievements, even if it does not prove the tool's versatility.
 - ANL does not address some aspects such as the validity of the passage from small-size cells to the expanded polarization and the applicability of results outside of the operating conditions and configurations used in the differential cell. This is important to address because the entire tool relies on this model. The investigators may also address the question of how to validate the durability projections at the system level when only individual components are optimized, and when system designs, architectures, and control are different from one original equipment manufacturer (OEM) to another. There is a lack of real validation with real systems.
- The goal of this task appears to be a detailed fuel cell system model to assess the effects of changing certain design parameters such as catalyst type, catalyst loading, membrane thickness, etc. If this assessment is accurate, then the effort is worthwhile.
- Received performance data are clearly analyzed, and the existing model parameters are fitted accordingly, thus enabling performance forecasts when altering operating conditions—it is not always clear whether these projections have been validated as well.

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.

- ANL reported on an impressive range of achievements that include predicting stack performance of de-alloyed PtNi/C, analyzing conditions under which humidification could be eliminated, exploring the application of pulse ejection, and evaluating the impact of increased stack inlet pressures. These results were much broader in nature than last year's scope and show great progress toward providing both specific predictions and suggestions for improvements in stack efficiency and cost.
- This is an excellent study of catalyst systems in the stack in collaboration with industry stakeholders and national laboratories. The partners are continuing to look at air, water, heat, and fuel management in the system to improve efficiency and cost.
- Several accomplishments were in line with the fiscal year 2017 planning. The comparative performance study of different state-of-the-art membrane electrode assemblies (MEAs) with Pt, Pt-alloy, and de-alloyed Pt-alloy catalysts provides valuable input to the industry. Good performance and cost targets were shown for the de-alloyed PtNi/C. However, investigation of the impact of the Ni loss degradation mechanism on not only performance but also long-term operation durability is very relevant and must be integrated in the model.
- The results of the performance degradation study help in understanding the breakdown of the contributions to the voltage loss. This is a good step; however, this part must be continued by including all the aspects and issues linked to the system (degradation mechanisms at stack level resulting from balance-of-plant [BOP] components, interaction between these mechanisms, and varying operating conditions/load profile).
- Many interesting results were generated over the past year, and the principal investigator is clearly responsive to suggestions. The emphasis on low-volume cost is very good.
- There has been clear progress toward comparative performance evaluation between four state-of-the-art MEAs with different catalysts. There is additional emphasis on the influence on operational conditions but no work mentioned (yet) on the updated projected durability relative to the life target of 5000 hours in the submitted version of this presentation (April 2017).
- The model is impressive, and some of the results given are interesting, such as the catalyst cost and degradation estimates. Other results are more confusing. For example, UTC Power was fielding hardware with anode recycle blowers in 1998 and pulse-width-modulated fuel injection circa 2010.
- It appears that the model results are being benchmarked, but this is unclear.

Question 3: Collaboration and coordination with other institutions

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

- The collaboration and coordination with other institutions are excellent and well balanced. The collaboration includes industry (component suppliers and OEMs), laboratories, and other consortia. The component suppliers and OEMs provide data to continuously refine the developed tool and to validate it. They also provide guidance/feedback to be continuously in line with the industry needs. Having different suppliers helps the versatility of the developed tool. The collaboration and involvement in other consortia allow use of the tool in the most recent component optimization.
- ANL continues with its impressive list of collaborations, engaging with various Fuel Cell Technologies Office (FCTO)-sponsored partners and other industrial collaborators. The nature of the engagement between this project and the Fuel Cell Consortium for Performance and Durability (FC-PAD) could be more clearly defined.
- There is a large number of collaborators including component suppliers, OEMs, universities, and national laboratories.
- There was excellent collaboration with multiple partners, especially SA. However, more collaborations with OEMs to obtain stack and/or system data could be very beneficial.
- The level of collaboration and coordination appears appropriate for this type of project.
- The information provided was not made public for all partners separately but appears to be fully integrated in the model. Comments or feedback on the fit between model and original data would be appreciated.

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

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

- A generic (versatile) multi-scale model for assessing the impact of component change at the system level that allows fuel cell system performance, cost, and durability assessment is a powerful tool for industry, laboratories, and the DOE Hydrogen and Fuel Cells Program (the Program). This model allows evaluation of performance, durability, and cost for new materials, components, architecture, and operating conditions for a fuel cell system at low cost, without running long-term testing. It is also a good tool to guide the industry's technical choices regarding the cost/performance and durability targets. The tool is not complete, and its validation is not achieved. Calibration is needed upon each new component assessment.
 - The project results are key inputs for the Program. Evaluating performance, durability, and cost for fuel cells will allow DOE to assess the advancement of the technologies and correlate their current status with the set of objectives and the means to achieve them (funding). Knowing this, DOE can update its objectives and targets more precisely and efficiently.
- The project is relevant in its role of predicting and directing stack development and DOE targets. Further validation or adoption of the various system-level predictions made in the presentation will solidify the relevance of the modeling work by ANL.
- The major impact here is on helping DOE and the entire fuel cell community understand the cost status (with SA), which is very important. The impact would be higher if ANL could show that their models have been validated at a stack and/or system level since there is always a large degree of skepticism with an unvalidated model.
- This project addresses multiple barriers including cost, performance, thermal and water systems management, air management, and start up/shut down. As such with a validated system model, its impact across the FCTO Multi-Year Research, Development, and Demonstration Plan is very high and is an essential component.
- Performance boosting and problem solving could be made more insightful when such a model is made accessible to the wider community.
- The project appears to be relevant, although this type of work is usually done by the fuel cell developer and not a national laboratory.

Question 5: Proposed future work

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

- ANL proposes to continue the excellent work that has been started. It is very nice to see that more durability will be included in the future.
- The proposed future work listed by the project team is in line with the results shown.
- The proposed future work is almost verbatim from the previous year's presentation; more thought should be invested here. Validation against a full stack system might be a good addition. Future plans should also include the ongoing development and dissemination of the ANL General Computational toolkit (GCTool) software package, which is a key component of the project approach.
- This future work is good, but ANL should add model validation at stack and/or system level.
- ANL provided a comprehensive list to achieve maximum performance, seemingly not considering stability issues.
- The future work appears to be appropriate.

Project strengths:

- The proposed tool is powerful for assessing the fuel cell system performance, durability, and cost for different components at the stack or system level. It should be applicable independently of the component, the application, the operation condition, or the system architecture and design (versatile). Another strength is that the good communication and different collaborations allow the tool to integrate large data sets and

feedback from different technology suppliers and users. Strong communication with other ongoing DOE projects allow refining of the model with the most recent developments (materials) and achievements (modeling).

- ANL continues to be engaged in very strong collaborations involving topics highly relevant to the Fuel Cells sub-program.
- The transparency of system-level performance and cost implications of various materials and design options is a project strength.
- A strength of the validated system model is that it can be applied from component, modeling analysis, stack, and system levels.
- The skills of the national laboratory have shown themselves to be a project strength.
- This model is solid, based on data.

Project weaknesses:

- The following are project weaknesses:
 - One weakness is the wide range of explored topics and the difficulty in assessing the final objective (fuel cell system performance), taking into account the interactions between individual components as they are optimized.
 - The project relies entirely on the developed model. Therefore, the validity of the passage from small cell operating condition to extended polarization should be addressed.
 - The model should include the degradation and aging mechanisms at the stack and system levels.
 - There is a lack of validation on real systems.
- It is not clear why a national laboratory is developing a fuel cell system and design tools. This is normally intellectual property (IP) for an OEM. It is unclear who has direct access to the results and whether they are considered IP.
- This work includes a system-level model that has not been validated at a system level, or even at stack level.
- Areas of uncertainty are not obvious.

Recommendations for additions/deletions to project scope:

- The project team should keep the driving objectives at the system level. That will help to lower the number of possibilities and save time and effort in exhaustive component evaluation. Durability studies must be continued and completed with degradation mechanisms, impacts on performance, and aging. Including new stack and BOP component suppliers is relevant.
- General Motors should be persuaded to share stack or system data (which could be done under a non-disclosure agreement) to validate ANL's model under at least a few key operating conditions with at least one system-level configuration.
- The project team should rethink how to protect the laboratory from perceived threats from the marketplace.
- Projections toward lifetime vs. performance would help highlight the best mode of operation.

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

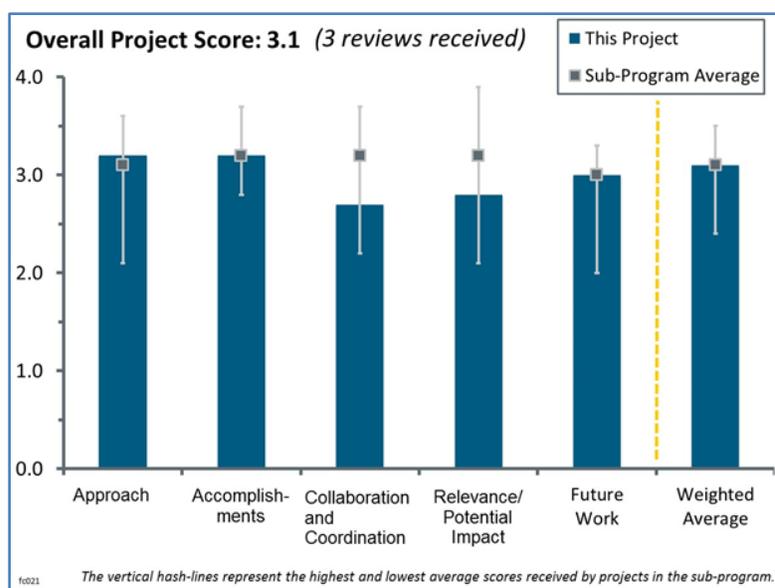
David Jacobson; National Institute of Standards and Technology

Brief Summary of Project:

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

Question 1: Approach to performing the work

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



- The National Institute of Standards and Technology (NIST) maintains a national user facility for neutron imaging of water transport in operating fuel cells. NIST pursues facility improvements through collaboration and feedback with testing partners and the fuel cell community. NIST provides free access for open research or fee-based access for proprietary research. NIST operates the neutron imaging facility and test stands in a user-friendly environment.
- The work that the project team is doing is fantastic, but as the team members admit, they are nuclear physicists looking at fuel cells. As such, it is ever more important that they have close ties and connections with those doing the work in fuel cells, particularly the original equipment manufacturers, but these interactions seem weak at best. It is hard to know the problem that they are solving. In that regard, they are doing brilliant work to improve resolution, but it is not at all clear what that increased resolution will buy the fuel cell community or the problem that it is trying to solve. Karren More's work would provide the obvious blueprint for their interaction, but there must be just as much of a call for this resource.
- Intention and implementation is clearly focused on providing the best tool for imaging in situ fuel cell conditions, moving beyond current limitations and incorporating backup plans.

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 work the project team is doing is excellent. The progress toward project targets is good but is very loosely linked to DOE goals. The potential certainly seems to be there, and the investigators cite the work with General Motors to shed light on cell corrosion as a good example of how it could work.
 - There was really nothing here except a demonstration of what the technology can do using baseline materials.
- Despite some delay in the work as explained by the team, the presenter clearly presented how progress was made on centroids and why this was important, and advances in further improving timescale of measurement.

Question 3: Collaboration and coordination with other institutions

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

- NIST listed a number of partners, users, and collaborators from academia, national laboratories, and industry. The last call for proposals received six new submissions for fuel-cell-related projects.
- There is a clear focus on the needs of the community.
- This is the weakest part of the project. Normally there is not as much weight placed on the collaboration rating, but this project can have an impact only if there is strong collaboration. It is hard to see where collaboration exists here, especially outside of the national laboratory community.

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

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

- This project is actively engaged in developing state-of-the-art neutron imaging capability for fuel cells and making it available to the community of fuel cell researchers and developers. The latest call for proposals received six new submissions. Experiments planned for summer 2017 will investigate electrolysis, solid oxide fuel cell, alkaline electrolyte fuel cell, and polymer electrolyte membrane contaminants.
- A highly specialized tool has been developed, enabling users to visualize the situation at hand in great detail. It could gain further value by providing insight into off-spec conditions that may occur and cross-correlate these with other characterization measurements such as electrochemical impedance spectroscopy so that researchers can identify, recognize, and visualize problems without being in the beamline themselves.
- The investigators need to work harder to make their work relevant. Having a stronger interaction with someone whose main focus is fuel cell performance would be a big improvement. It would be better if the project was led by someone in the fuel cell community.

Question 5: Proposed future work

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

- Enhanced resolution in experiments taking less time is welcome, as are more measurements to ensure reproducibility under fuel cell operating conditions.
- The main focus is on further increasing resolution, but the rationale for why this is important has not been presented. The investigators mention that there will be more demand if they improve the resolution or decrease the sample time, but it is not clear why. For example, with 2 μm resolution on a 5-s time scale, we could observe water formation and release from the catalyst layer and determine the time and length scale on which this occurs. NIST needs a compelling reason to further improve the technique; otherwise, efforts should be more focused on outreach and using current tools.
- Owing to unexpected delays at NASA, the target completion dates in the ongoing project on the neutron microscope to reduce the image acquisition time to 10 s for 20 mm spatial resolution and to improve the spatial resolution to 1 mm with 10-minute acquisition time have been postponed to 2020 and 2021, respectively.

Project strengths:

- NIST has fantastic imaging capability. There is great work on the physics and improving characterization capabilities.
- NIST has a unique facility, staff capabilities, and experience.
- NIST provides a unique facility that is open to the entire community, encouraging learning from the field.

Project weaknesses:

- Collaboration needs improvement. This project works only if the researchers are intimate with the challenges and problems in the fuel cell community and how the researchers can help.
- The localized analysis needs to be representative for a situation under evaluation, which is always a question.

Recommendations for additions/deletions to project scope:

- The team should not focus on continual improvement of the device, but should work with those in the fuel cell arena to ensure that the team's work is relevant.
- The vast number of achievements and results need to be easily accessible to educate and inspire users to submit more compelling proposals.

Project #FC-052: Technical Assistance to Developers

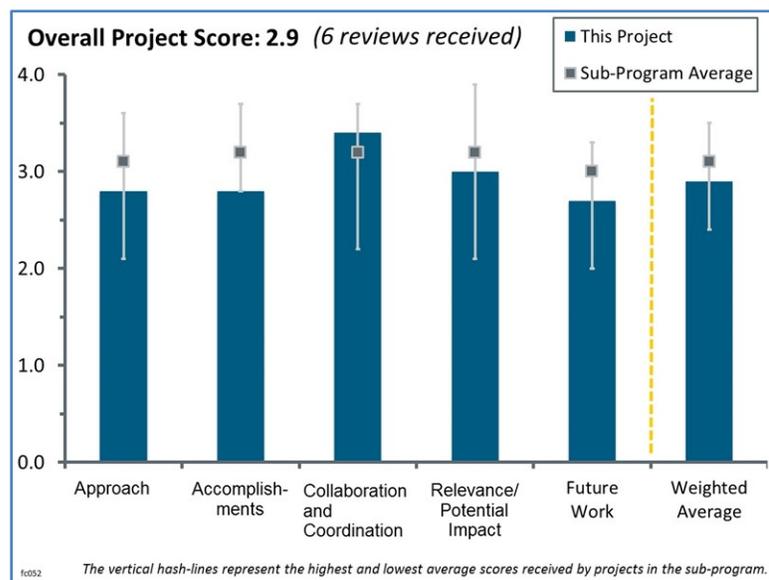
Tommy Rockward; Los Alamos National Laboratory

Brief Summary of Project:

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

Question 1: Approach to performing the work

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



- The aim of the project is for the LANL group to provide technical assistance to the testing and validation of materials, to validate single-cell test protocols, to assist durability working groups, and to make technical expertise available to the U. S. Department of Energy (DOE) and the Fuel Cell Technical Team. The proposed approach is rational but requires improvement.
- The strength of this project is the utilization of a combination of sputtering systems, which, in combination with physical vapor deposition (PVD), provides sufficient knowledge to create unconventional classes of coatings with desired thicknesses and other properties.
- The approach to characterizing low-platinum-group-metal (PGM) catalysts should be improved. Simple testing provides no information on the interesting activity and stability of these materials. Furthermore, the proposed approach for probing the “mechanism” that governs the activity of non-PGM materials is insufficient for such an ambitious task.
- It will be very important to expand this project to provide concrete information about the differences and similarities between rotating disk electrode (RDE) performance and performance in real fuel cells, which is a subject about which the fuel cell community has still not found any final answers. The principal investigators (PIs) should consider pulling together experts in both RDE and fuel cell testing to address this important issue.
- Given that the project deals with the stability of both the active and supporting materials, the PIs should consider utilizing inductively coupled plasma-mass spectrometry methods to quantify the dissolution rates of their materials.
- LANL does careful work, primarily in response to fuel cell community requests filtered through DOE and to address important controversial questions, such as whether oxygen reduction reaction (ORR) activity of non-Pt catalysts correlates with the reduction–oxidation reaction (redox) potential.
- The potential overall impact of this kind of service project is less than that of more tightly focused laboratory projects properly coordinated with industry. The PVD efforts and the fuel cell/catalyst work are so different that they should be undertaken under separate projects to allow for improved direction and review.
- The approach for this project is not leveraging the resources and technical know-how of the national laboratories but is instead acting as a service provider for technical developers. There is no transparency in which projects/topics get selected. This project should be ended in fiscal year 2017. A completely alternate approach to the developer can be established by:
 - Creating an online portal for accepting incoming requests from developers
 - Understanding how the developer is willing to pay for using the national laboratory resources
 - Identifying the technical benefit of the project to DOE goals

- Overall, the approach is to help those in the field to access capabilities that they do not have. However, the selection process was not clear, nor was whether the work provided is addressing critical barriers. It would be good to see how many new projects and carryover projects there are per year, including those that were not selected.
- It is not really clear how this work necessarily makes significant contributions to key barriers, nor how what is done is decided (besides the statement that assistance is “as directed by DOE”).

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

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

- The work showing a lack of correlation between redox potential and ORR activity of non-Pt catalysts is an important contribution to fuel cell development, addressing recent data from a reputable group that claimed such a correlation despite earlier work that found no such correlation. The new data should prevent developers from following a false pathway toward improvement of non-Pt catalysts.
- The University of California, Los Angeles, shape-controlled catalysts show unusually low initial activities for catalysts of this type but have shown unusually good stability, at least in RDE testing. It is not clear that one should expend effort on complex catalysts with little improvement in activity versus standard Pt/C.
- The PVD work on catalysts and bipolar plate (BPP) coatings was sufficiently encouraging to serve as seed data for more detailed projects on these topics.
- Overall, there has been progress, and the project has helped academia and industry. However, it is not clear how much of the activities address the DOE goals. The selected highlights were interesting and wide in scope, so it is hard to see cohesiveness. The selection of projects is a bit confusing, and it would be good to understand if there is a priority based on DOE goals.
- The PIs presented testing of Pt₃Ni catalysts doped with Mo. To be able to compare the activity and stability of these materials, the obtained results should be compared with the state-of-the-art PtNi and/or PtCo catalysts. It is also very important to acquire information about the particle size and shape before and after testing. An independent RDE measurement is also required to confirm that addition of Mo indeed affects the stability of the catalysts. Without such a comparison, it is very difficult to evaluate the Pt₃Ni/Mo system.
- The PIs have also shown results for the ORR on Fe-N-C catalysts in two electrolytes. It is puzzling why the diffusion currents are different in perchloric and sulfuric acids, while in the kinetic region the activities are the same. For the next review, the PIs should establish a much better protocol for exploring electrochemistry in the RDE configuration. It would be highly desirable to simultaneously assess the stability and activity of the catalysts. Furthermore, although it is very important to establish correlations between the redox potential of cations and the onset of the ORR, it is not clear that the proposed experiments will resolve this issue. In situ spectroscopy(s) is (are) required to gain insights into such complex relationships.
- The examples provided clearly yielded useful results for those who received assistance, but it is not clear that these results yielded significant progress toward key DOE goals.
- The support of the working groups, as well as the Fuel Cell Short Course, is arguably the most beneficial work here, since this work obviously has impacts on a much broader group than simply working on the needs of individual customers. Assistance to a Canadian company (Blue-O Technology) is also a questionable use of U.S. taxpayer support in this manner.
- Since this project does not generate any ideas or intellectual property on its own, the accomplishment needs to be measured on how efficient the service request system is working. No metrics were provided on the total number of requests or the percentage of projects that were completed on time. There is no definition of measurement of metrics.
- In addition to the evaluation and assistance to the developers, the project should include benchmark data for a standard or best-of-class material so that the developers could understand the level of achievement.

Question 3: Collaboration and coordination with other institutions

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

- This project provides facilities and expertise that allows collaborators with less complete fuel cell facilities to make meaningful contributions to the field of fuel cell development.
- The PIs have established excellent collaboration with many partners who are part of this project.
- There are obviously a number of collaborations here since this is a necessary part of this sort of work. A potential improvement would be if the results of these collaborations were disseminated more broadly. For example, it might be a requirement that any work done on this project should be published so that others can potentially benefit from the results (e.g., use the National Institute of Standards and Technology model for free use of government assets).
- It is a highly collaborative project. It would be good to coordinate perhaps with other laboratories or capabilities as needed. There is a question about how much of the work is just data-gathering versus truly collaborative and jointly performed research.
- The project would benefit from listing all of its competencies so other developers can relate.

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

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

- This project is highly relevant with a potentially high impact on the DOE Hydrogen and Fuel Cells Program. Note: If the investigators are able to fully integrate fuel cell testing with fundamental science, then the project will contribute significantly to DOE efforts for full implementation of electric cars.
 - The project provides an important service to the community. This provides impact, assuming that the correct and higher-valued projects are chosen.
 - It is not clear whether this is duplicative to other efforts such as Small Business Vouchers, ElectroCat, the Fuel Cell Consortium for Performance and Durability (FC-PAD), cooperative research and development agreements, etc.
- Since this is primarily a service project, the relevance and potential impact are dependent on the importance of questions brought into play by the collaborators. The current case is a mixed bag of good and mediocre subprojects.
- LANL worked on multilayer coatings for BPPs, which could be an extremely expensive approach, and it is unclear how this will reduce BPP costs. Other projects also did not show any significant progress toward meeting DOE goals since the PIs did not report any metrics toward the Pt utilization in grams of Pt per kilowatt.
- The project has a minor impact toward overall goals.

Question 5: Proposed future work

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

- In general, the proposed future work is reasonably well organized. One direction that needs to be improved is developing methods that are capable of resolving the issues of catalyst stability and a strategy to provide fundamental reasoning for why Mo improves the stability of PtNi alloy catalysts. Further development is also needed on a strategy for optimizing the physicochemical properties of nanoparticles to improve the activity and stability of these catalysts.
- Testing stability in real fuel cells is an important step in the evaluation of cathode materials. However, without having a reliable balance with the fundamental understanding of the driving forces that control catalyst activity and stability, it will be impossible to know what types of materials we should synthesize.
- It was hard to judge the future work, as it depends on the projects being added. The existing collaborations and continuing projects seem adequate in terms of making progress.

- The project's potential impact could be significantly improved by requiring that companies that request assistance must agree to publication of at least some of the key results.
- The planned future work seems to be just completing present activities and responding to questions that arise in the future. This service project does not seem to initiate activities on its own.
- It is not clear why the researchers are working on durability of materials that do not show promising beginning-of-life results.

Project strengths:

- The leading PIs have proven in the past that they are able to develop and execute similar projects. The methodology is rather well developed.
 - A big strength of the project is the utilization of many tools that can explore the feasibility of implementing various types of noble and non-noble catalysts in polymer electrolyte membrane fuel cells.
- This project leverages core expertise, provides critical help and service, and is making technical progress on the issues presented and requested.
- There are good collaborations with multiple parties.
 - National laboratory resources are used to help multiple customers.
 - This project supports DOE working groups.
 - This project contributes to the Fuel Cell Short Course.
- The resources, technical personnel, and national laboratory know-how are strengths.
- The project makes catalyst testing and PVD available to the fuel cell community.

Project weaknesses:

- One key weakness is the lack of a clear path toward understanding and minimizing the dissolution of catalyst components during fuel cell operation. For non-noble materials, there is no alternative direction if the proposed systems do not work as planned, and it does not seem likely that these materials will work.
- It appears that this work may duplicate other efforts and mechanisms. Justification for choosing projects is needed, and it is unclear if cost share is provided and if it should be. It is unclear if there is a priority for industry versus academia or U.S. versus non-U.S. projects.
- There is limited impact on major barriers, limited dissemination of results, and a generally small impact on the broader fuel cell community.
- The project responds to limited-scale requests from the community rather than pursuing a coherent set of ideas toward the improvement of fuel cells. There is a lack of approach and leverage.

Recommendations for additions/deletions to project scope:

- Overall, with some tweaks, this project can be enhanced. This is a great project for the community and should perhaps be expanded to other national laboratories.
- One suggested addition would be a clearer connection between single-cell testing and fundamental understanding of the processes.
- More dissemination of key results should be required when individual companies are provided with assistance (e.g., publication in technical journals).
- It would be better to use the funds and facilities currently dedicated to this project for projects with clearly defined goals rather than for loosely defined service to the field. It might be good to dedicate such a service project specifically to support projects funded by the Small Business Innovation Research program rather than making it available also to organizations with their own extensive facilities and capabilities.
- The whole project should be terminated. However, if the DOE Fuel Cell Technologies Office decides to keep the project, then the high-value addition would be an online portal wherein any developer can request testing with some additional details on what the cash contribution would be.

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

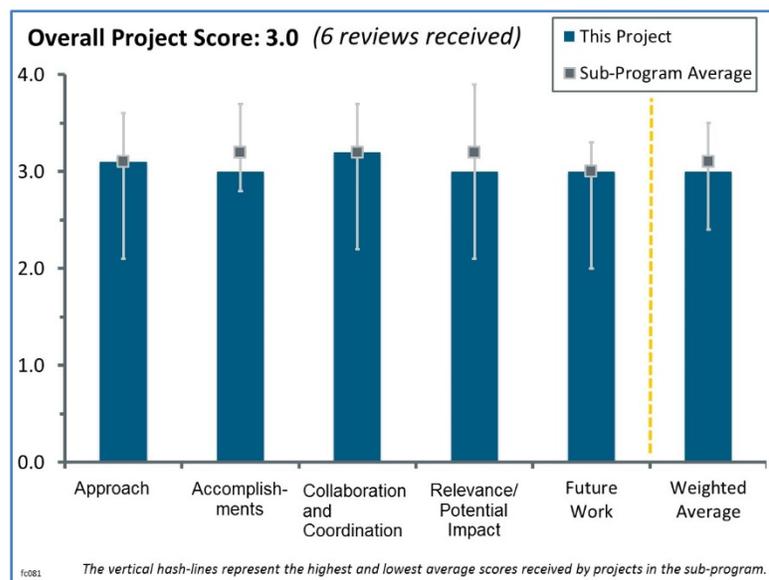
Jennifer Kurtz; National Renewable Energy Laboratory

Brief Summary of Project:

The fiscal year 2017 objectives of this project are to (1) receive and analyze new laboratory durability data, (2) update and publish the durability results, and (3) include electrolysis data. The National Renewable Energy Laboratory (NREL) will (1) develop a snapshot of the state-of-the-art fuel cell durability, (2) uniformly apply analysis methods to developers' voluntarily supplied data from laboratory testing, and (3) provide an independent assessment and status of state-of-the-art fuel cell technology.

Question 1: Approach to performing the work

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



- The approach being used by the project team is sensible given the nature of the data being collected. The National Renewable Energy Laboratory (NREL) team has a solid track record with this sort of data collection and aggregation, and the summaries are appropriate. Presumably, the full composite data product (CDP) report contains more views of the data that might help the broader community understand trends in fuel cell durability.
- The project aims to gather durability data (mostly from industry), analyze that data, and produce detailed data products (DDPs) and CDPs. The amount of data gathered, submitted voluntarily by suppliers, increases over the years despite the difficulties associated with data protection and the required 1,000-hour minimum operation condition, which demonstrates the efficiency of the approach (receiving DDPs is a good return on investment for data suppliers). The approach used is a statistical analysis that processes a large number of data with plenty of heterogeneities (different technologies, suppliers, operating conditions, testing protocols, operating ranges, etc.), which do not allow for fine analysis. The degradation fitting is based on a segment linear fitting, which is one simple option, but other fittings could be tested. Voluntary data collection can be limiting when data supplied is incomplete, as it was for fiscal year 2017 (numbers related to backup power and bus analysis were the same as in 2016 because of a lack of 2017 data). The use of a 10% threshold for power degradation for all applications is still questionable (metrics used in the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan [MYRDDP] are 20% for backup power and for stationary applications [1–10 kW]).
- It would be more informative if the request for information (of course on a voluntary basis) also included durability testing conditions and temperature.
- The approach is limited by the data suppliers are willing to share. Given this, the project team has systematically improved their data collection and analysis.
- The data collection method is limited because NREL does not have leverage with U.S. companies and most companies consider data intellectual property. A nicely phrased request would help to some extent. A demonstration in how this might be in the individual company's best interest (a "carrot") might work better. Small companies often have limited test budgets. If NREL can assist a company with either test support or test funding, the company may share the data under a non-disclosure agreement in which release of data requires the company's approval.
- Many of the voltage degradation trend data products represent an averaging of steady, duty-cycle, and accelerated data. For some applications, particularly automotive, this averaging will put the application in a

fairly poor light. System mitigations for stack failure modes are commonly implemented for automotive operation so that averaging data from accelerated testing of stack components with system or vehicle-level lifetimes will certainly lower lifetime estimations. Accelerated tests exist for the purpose of understanding failure modes, not for providing a representation of component lifetime in a vehicle setting. The approach does not address how information such as current density and Pt loading bins might be used to understand emerging technology trends. It would be interesting to see voltage degradation as a function of either current density or Pt loading. Current density and Pt loading should be compared to each other as well to see whether particular applications have been more adept at making the most of low Pt loading. Perhaps there are lessons learned that other applications could derive. As in years past, the voluntary data inputs severely restrict the meaningfulness of the data. Given the impossibility of any other route, the practicality of this project is questionable.

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 results are interesting. A voltage–current plot is useful to the researcher; however, a voltage–current plot at times requires perspective of the relevant task. A loss of 70 mV may or may not be germane, and a loss of 25% of the available power resulting in only 50 mV/cell margin on the power electronics might also be useful to the researcher. Depending on the context, a fuel cell with given performance might still generate power but may fail to run a vehicle. Current density might not be a good metric. For example, a PAFC unit might operate at 300 mA/cm², but last with negligible performance degradation for 80,000 hours. An alkaline fuel cell on the other hand might operate at 10,000 mA/cm², but last with negligible performance degradation for only 200 hours. It is not clear which is better. The project team should be careful when mixing data from different applications and technologies. A cross-plot of current (I)–voltage (V) curves (i.e., changes in voltage as a function of fixed current density and time) can be useful, and derivative plots can help detect internal (decay) and external (step change) issues.
- There is appreciable difficulty in harmonizing test data in multiple formats from multiple sources. It is a credit to the NREL team that they continue to get data from manufacturers. It is good that the team used the limited set of more recent results, although it would have been good to see the early results compared directly to later results. Given the patchy nature of the data, perhaps a rolling three- or four-year window should be used.
- Although it is good to see that some information is coming back with respect to the current density and Pt loading bins, the project has not been able to derive much that is useful so far from either, with the possible exception of the current density versus application plot. The Pt loading versus year trend is not likely to reflect the overall industry trend, which is unfortunate. It can be seen that automotive applications use the highest current density, followed by materials handling vehicles, but these trends were fairly well known within the community. The challenge for this project is to go into greater technical depth with the small amount of information gathered. The short stack and full stack plots for automotive operating hours are interesting for status reporting to the public, but being removed from operating and materials context (e.g., power density, operating conditions, Pt content, and membrane thickness), this data does not allow the development of any deep understanding as to how durability is being achieved. Because of this, the project team cannot say that it is assisting in overcoming the barriers that developers face.
- The project does not address any objectives set in the MYRDDP, but it is meant to provide an assessment of the status of fuel cell and electrolyzer durability and cost. It is, however, a valuable tool for DOE to follow the evolution of the technology regarding the targets, even if it is difficult to draw fine conclusions regarding durability since the analyzed groups do not distinguish between various fuel cell technologies or designs. Durability assessment depends also on the fuel cell utilization (control strategies, hybridization, etc.) that is not taken into account. Stationary application analysis is achieved independently of the range, while DOE's targets address residential applications (1–10 kW) and distributed generation (100 kW–3 MW) differently.
- The project is doing about as well as expected given the limitations of the data gathering (i.e., dependent on supplier willingness to disclose information). In addition, progress was shown in presenting the current density and cathode platinum group metal (PGM) loading segmentations, as requested in previous project reviews.

Question 3: Collaboration and coordination with other institutions

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

- The project team has been very successful in convincing several data providers to share data. However, the issue remains that this data is provided on a voluntary basis, and that makes the project very dependent on the goodwill of data suppliers to provide quality data, complete information about the testing conditions, testing history, and so on. That can lead to unreliable conclusions despite the team's sound analysis. The number of supplied data sets have increased over the years (50 datasets since May 2016), and the same partners have provided data over several successive years, which means they are satisfied with the resulting analysis quality of the CDPs and DDPs. The project team has excellent collaboration with other partners, given the facts cited above. The collaboration and the coordination seem to be good.
- The project team is canvassing as many suppliers/developers/stakeholders as it can. The response rate is outside of the team's control but appears to be decent. One can always hope for more thorough responses, but at least the project team is trying to get the information and has been responsive to adding extra categories (cathode PGM loading, max current density) with at least some supplier response.
- NREL is the only funded participant on this project. Although there is an impressive list of participants among fuel cell manufacturers, this group is not collaborating among themselves. Nevertheless, given that the data collected is not pre-competitive, this is an appropriate level of interaction, so no change from current levels is recommended.
- Given the reluctance of most organizations to provide data, it is commendable that 23 developers were willing to provide data. Furthermore, it is understandable that identification of developers cannot be provided. Collaboration could be enhanced by working more closely with developers than through just an email with an attached spreadsheet. The project team should think more about what developers could derive from the information while still maintaining the confidential boundaries crucial to enticing data sharing. If there are data products available only to those that share and those data products contain technical depth, the sharing of data might become even more widespread.
- Additional collaboration may be needed to advance this project. NREL should start thinking about "carrots."

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

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

- The data being collected is extremely useful in gauging the progress of the fuel cell and electrolyzer industry toward DOE's durability goals for various applications. The project should include graphs or tables that can get at trends over time, perhaps with a sliding three-year average of results. The project should also evaluate how the main project deliverable, the CDP, is being publicized so that people in the industry can find it. Also, perhaps there are venues other than the Annual Merit Review and the U.S. DRIVE Partnership's Fuel Cell Technical Team meetings at which this project's results can be presented.
- There is potential to both understand the operation of the fuel cell and help some of the struggling companies out there.
- The analysis has an impact because it allows DOE to track the trends of durability and cost evolutions, to assess technology advancements, and to correlate their status with set objectives and means to achieve them. The analysis would have a greater impact with more relevant information (separate analyses for different fuel cell technologies, proposition of durability values based on different thresholds, etc.).
- As it is, the project does not advance progress toward the goals and objectives of the Fuel Cells program. The best results from the project still confirm only the widespread opinions that exist within the fuel cell community. While the project has progressed in terms of obtaining current density and precious metal loading information, the information is still not being used in a way to clearly show technical trends. Even at its best, if the project completed all of its objectives, the results might be only a very good status report. However, the project would not be able either to provide direction with respect to closing gaps or to advise

why gaps have been closed. The best the project can do is indicate where there is no longer a technology gap.

- The project has relevance to the Hydrogen and Fuel Cells Program, as it gives a durability assessment that is at least data-driven. It is, however, of virtually no relevance to developers without much higher contextual data (all of the information pertaining to loading, electrode type, generation, membrane characteristics, detailed cycle data, acceleration factors, temperatures, pressures, relative humidity, voltage cycling, etc.). This is not a criticism of the project team but of the project goal.

Question 5: Proposed future work

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

- The plan for future work is more of the same, which is perfect. The project should keep up the good work.
- The proposed future work listed by the project team is expected and in line with the results shown.
- It is agreed that the project should try to better characterize the acceleration factors. However, unless given very detailed time histories (including cycle data, voltage, temperature, relative humidity, and pressure vs. time), the project may have to rely on the supplier estimates of the acceleration factor. The principal investigator is encouraged to add “estimated acceleration factor” to the data request to get that information. The supplier estimate of the technology readiness level (TRL) value should also be added. In addition, if possible, the project should ask for suppliers to test to an agreed-upon durability test profile (e.g., Fuel Cell Technical Team durability test) to provide a consistent baseline.
- The project team acknowledges that it could do more work in trying to understand relationships between cost (or PGM loading) and durability, which are important. The project team should seek to explore even more technical relationships from the data it has. The relationship of current density and cost to operating conditions is probably fertile ground for trying to understand technology trends. Categorization of accelerated tests is mentioned in the future work and is very much needed to have any hope of making these data meaningful. However, the project must decouple accelerated stress test data from stack duty cycle data. Greater emphasis needs to be placed on interactions with developers. The project team needs to brainstorm how the process could be made more valuable for developers so that more developers contribute data.
- The proposed future work appears rational. The project needs to find “carrots” to get the original equipment manufacturers to play.

Project strengths:

- This project offers an independent and uniform analysis of data from key stakeholders involved in fuel cell technology development. This is an important tool for DOE in estimating the progress of current technologies regarding the objectives set in the MYRDDP and the involved funding, and for data suppliers to assess objectively the evolution of their technologies (DDPs). The analysis provides a general overview and trends of the evolution of the durability and cost values from different applications. Based on the analysis, targets for electrolyzer technology could be defined.
- This project brings together and synthesizes data from multiple sources, covering several manufacturers, technologies, and applications. The project team does a good job of synthesizing the information received into summary reports that support the DOE Fuel Cell Technologies Office’s mission.
- There is access to data from many prior years as well as access to considerable fuel cell expertise at NREL. The project has demonstrated the ability to collect data from numerous developers.
- This project offers the ability to share the data on a publicly accessible website and to leverage DOE to be able to gather the data.
- The dedication of the researchers (performance engineers) is a project strength.

Project weaknesses:

- The data analysis is applied uniformly, and different fuel cell technologies/ranges are analyzed within the same classification. Sometimes, in the same class (automotive, for instance), different balance-of-plant designs and architectures, hybridization, and control strategies could be applied, and the operation objective

could require a unique trade-off between performance, efficiency, and durability, which makes the fuel cell durability completely different. In the current analysis, the projected value for backup and stationary (1–10 kW) applications seems to be underestimated: a value of 20% voltage degradation seems to be a more adapted metric to assess voltage degradation (MYRDDP). (In general, durability could be given as a function of different metrics values.) A key point in durability studies is linked to the operating conditions (cycling, load profiles, etc.) and the past fault and the exposure time, etc. This kind of information is difficult to obtain (even with the goodwill of suppliers) and to correlate with the durability projections.

- Confidentiality boundaries prevent the project from providing deep technical insight to show relationships between degradation and operating conditions or materials. Continued mixing of accelerated data with stack duty cycle and steady-state data convolute the results shown by application. Application durability should be reflected by the mode of operation most likely to be observed in the field. The encouragement of developers to submit data is still not strong enough. The project team must consider what rewards could be made available to developers that share data. The results of the project, even if fulfilling objectives, may provide only a good status report. The results do not contribute to overcoming technology barriers.
- The main weakness is that the data shared are those volunteered by the participants. Thus, they may not be the most representative results for the state of the art in industry. Also, while there are reasons for using cell data, it would be helpful to correlate these test results to full fuel cell system durability results.
- The project is limited by supplier willingness to disclose detailed information required to develop a better understanding of durability status and degradation mechanisms.
- There is a lack of leverage to access more data.

Recommendations for additions/deletions to project scope:

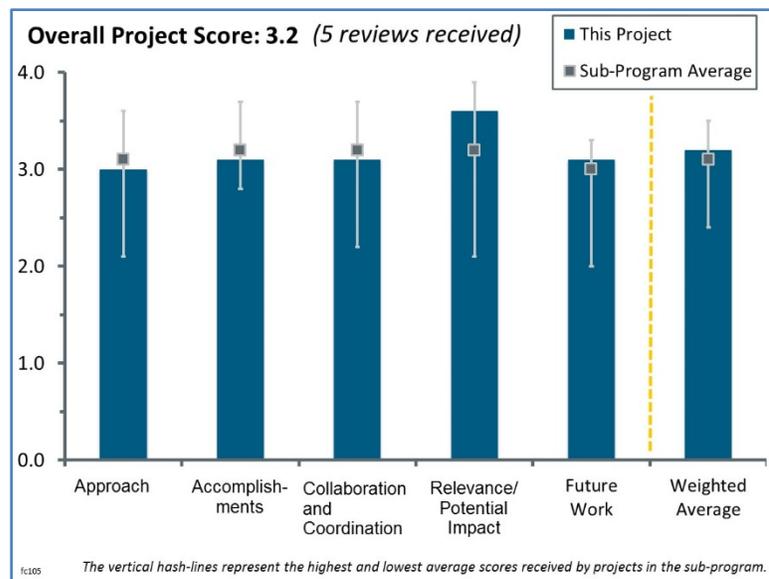
- No changes are recommended.
- The use of Pt loading bins and current density bins for the different technologies being surveyed represents an improvement over prior years. It would be interesting to compare the Pt loading bins versus the current density bins for like datasets to see whether there is a trend. There could be approximate “gram-per-kilowatt” bins derived from there that would begin to show the different levels of technology are being used. The information supplier should be asked to specify TRL. This would help to distinguish results that are more experimental from those that are already incorporated into a product. The project team needs to break out accelerated tests into various categories or by acceleration factor. Some operating conditions data (e.g., temperature) would benefit the analysis.
- The data should be analyzed by technology and by application for fuel cells and electrolyzers and to assess the durability with different metrics (from 10% to 20% is more compatible with backup/stationary applications [MYRDDP]). DOE-funded projects could be highly encouraged to share their generated data with the project team. Degradation, fault, and failure history should be included in the metadata template. The project should elaborate on fitting approaches other than linear.
- The project team is encouraged to include the acceleration factor in the data request. The team is also encouraged to request that suppliers supply data to a fixed durability cycle. The project might require a new project structure to enhance the data collection quality needed for true durability/degradation analysis. Such a project may require supplier compensation and involvement of the Fuel Cell Consortium for Performance and Durability for both project scope/management and analysis.
- Particularly for durability data, there should be a clear distinction between laboratory results and fleet results. Laboratory testing is commonly done in a more aggressive manner with both materials and test conditions.
- The project should work on “carrots,” perhaps offering to test or reduce data with various companies.

Project #FC-105: Novel Structured Metal Bipolar Plates for Low-Cost Manufacturing

C. H. Wang; TreadStone Technologies, Inc.

Brief Summary of Project:

Bipolar plate (BPP) cost is a major portion of total fuel cell stack cost. The project's goal is to develop low-cost metal bipolar plates to meet U.S. Department of Energy performance and cost targets, specifically a cost of <\$3/kW, corrosion of <1 x10⁻⁶A/cm², and resistivity of <10 mΩ·cm². The approach is to coat a stainless steel bipolar plate substrate with semiconductive doped titanium oxide (TiO_x). The project is addressing titanium oxide's challenges to increase the conductivity, increase the bonding strength to the substrate, and increase the coating composition uniformity. The project is also developing a large-scale manufacturing process for the coating technology. In addition, the project is investigating the relationship between the processing conditions and the doped titanium oxide properties for production quality control system development.



Question 1: Approach to performing the work

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

- The approach encompasses deposition and fabrication of BPPs, along with characterization at Oak Ridge National Laboratory. The work continues from previous Small Business Innovation Research (SBIR) efforts growing doped TiO_x interlayers for BPPs using a multi-step sputtering and oxidizing batch process. The difference here is that the project will evaluate alloy compositions using a new approach. The team starts with a cost analysis performed with a partner.
- The approach is technically sound and should provide a coating with the desired properties. High-volume manufacturing is still in question, but no showstoppers are evident.
- This project is a continuing effort of a previous project (FC-105), so the team has a good understanding of the barriers: high manufacture cost and missing of dopants (high resistance) on the surface of TiO_x-coated-metal BPPs. The work also proposes a feasible and integrated plan on fabrications, characterizations, and performance tests. A better fundamental understanding of the dopants (Ta, Nb) on the surface of TiO_x with surrounding electrolytes might better guide the experiments. Also, some modeling work should be included on the design of the “Mosaic” target for the desired atomic mixing, based on the dimension of each strip of the “Mosaic,” the distance between the target and the substrate, etc.
- The project is new, and future work identifies approaches to meeting the cost barrier. The project established cost analysis values and addressed resistivity issues.
- The approach of doing a physical vapor deposition (PVD) coating of the titanium barrier layer and TiO_x sprayed coating on a pre-stamped plate may not be a solution capable of reaching high production volumes.

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 been making nice progress in modifying the deposition process and bringing the dopant to the TiO_x surface to reduce the surface resistance. The proposed milestone has been reached as planned. The report also shows high-resolution images of the thickness and roughness of the surface layer, which is very important to understanding the microstructures and properties of the coating.
- For the startup of the project, good progress has been made in cost analysis, and previous SBIR work provides a pathway to superior corrosion resistance. The modified process suggests improved distribution of Ta and Ti can be achieved.
- The basic concept has been proven. Low contact resistance and superior corrosion have been achieved. Excellent adhesion of the coating on the substrate is anticipated. Projected high-volume manufacturing cost is lower than that of competing technologies.
- The project started six months ago but only got under contract recently. It is unclear what the full assumptions were that went into fabrication cost analysis. Number, type, and throughput of sputter system(s) would be required to get to volume to bring costs down to below targets.
- Compared to previous year results, the new accomplishment seems to be optimizing the layer composition and cost analysis. It is unclear if any of these deliverables are proportional to the amount of funding.

Question 3: Collaboration and coordination with other institutions

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

- It looks like the researchers have a good plan with Hawaii Natural Energy Institute (HNEI) for testing once they have a prototype.
- The team has set up effective collaborations on cost analysis, film depositions, microstructural characterizations, and performance tests. Some collaborative work is suggested on the modeling of the deposition and fundamental understandings of the materials, if the team does not have such expertise.
- Partners are adequate for this phase of the development and include Strategic Analysis (SA) for cost analysis. A stack integrator would certainly be beneficial in terms of BPP insight and in-cell testing.
- Researchers report results of work with SA. There is testing at HNEI to be initiated.
- The project is collaborating on a “what must get done” basis. However, the project could use some external help with the correct analysis techniques and leveraging existing national laboratory capability to do the analysis with higher accuracy (e.g., copper metal detected during the x-ray photoelectron spectroscopy depth profile on slide # 9).

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

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

- The cost of a BPP is a significant portion of fuel cell cost. The project aims to develop highly conductive and stable BPPs and manufacture the product at a low cost. Therefore, the project is closely relevant to the Hydrogen and Fuel Cells Program goals.
- Reducing the overall cost of the BPPs is highly relevant to meeting DOE project objectives. The potential impact of this project is high, provided that the goal is reached and the cost analysis done correctly. It looks like the cost analysis is either optimistic overall or missing the cost of the PVD titanium protective layer.
- The cost of the BPP is one of the main contributors to the overall stack cost. This project seeks to develop a coating that mitigates corrosion at an acceptable cost.
- If successful, this project could resolve corrosion issues and fuel cell poisoning by corrosion products from BPPs.

- If costs can be realized and technical targets met, there is potential in this BPP technology, although it is difficult to get low-cost coatings with a line-of-sight batch-type process.

Question 5: Proposed future work

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

- Proposed future work looks fine, given the scope and funding for this project.
- Future work logically follows progress to date: process development, coating characterization, and durability evaluation. Pre-forming coating will be evaluated.
- The proposed work is mostly well planned and achievable. However, the plan does not provide a target number for the performance. There is little risk analysis. Some modeling on the deposition and a better understanding of dopant segregation on the TiO_x surface should be included so that the proposed work can be carried out more efficiently.
- The approach uses the PVD process, which is slow compared to production requirements for BPPs for automotive fuel cells. The project should evaluate the throughput of PVD process and determine whether it will meet required production rates/costs for 500,000 fuel cell systems per year.
- The patterned sputtering approach suffers from getting away from the type of deposition that will eventually be used (alloy sputter target). The issue is that the alloy target has preferential sputtering of Nb versus Ta, which the researchers have seen in the past. This will also happen inside the chamber unevenly as many BPP assemblies are placed, leading to different compositions and morphologies on the surface. The approach does not address this issue and works only on optimizing an unachievable at-scale “optimal” composition.

Project strengths:

- The project aims at an important component for fuel cells, BPPs. The project is based on the product of a previous SBIR project, so the team has a good starting point and clear understanding of the challenges, such as high manufacturing cost and low conductivity on the surface of TiO_x coating. The team has proposed the approaches to overcome the challenges and provided some initial results to demonstrate the feasibility. The team also has established collaborations on depositions, characterizations, and durability tests.
- A primary project strength is the innovative coating on stainless steel plates and the concept developed by TreadStone.
- The approach has the proven capability to meet technical targets at low volume.
- A creative, technical person is the principal investigator.
- It is an innovative concept.

Project weaknesses:

- There is a lack of discussion on a fundamental understanding of the surface solubility and segregation of dopants (Nb, Ta) in TiO_x . The computational effort on achieving an optimized “Mosaic” target is not presented either. No risk analysis is provided.
- Processing using PVD may be too slow for automotive applications. Multiple PVD units may turn out to be too expensive. This needs to be evaluated by SA.
- It is unclear if there is any serious thought on durability and stampability of this coating to enable a pre-coated strip approach.
- The high-volume capability to achieve cost and compositional (performance) targets is questionable.
- The lack of participation of a stack integrator is a project weakness.

Recommendations for additions/deletions to project scope:

- A more fundamental understanding is recommended on the design principle of the coating, such as the segregation of dopants on the surface and bulk of TiO_x . Also, the development of targets for depositions can be expedited with the aid of computation.

- The project should clarify cost analysis and evaluate capability to achieve compositional targets at volume across the deposition space of a scaled-up deposition system.
- Adding a good analytical characterization technique of the fresh and aged samples of this coating is recommended.
- The project team should consider other coating compositions based on theoretical assessment.
- The project timeline is too short and should be extended by six months.

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

Andrew Herring; Colorado School of Mines

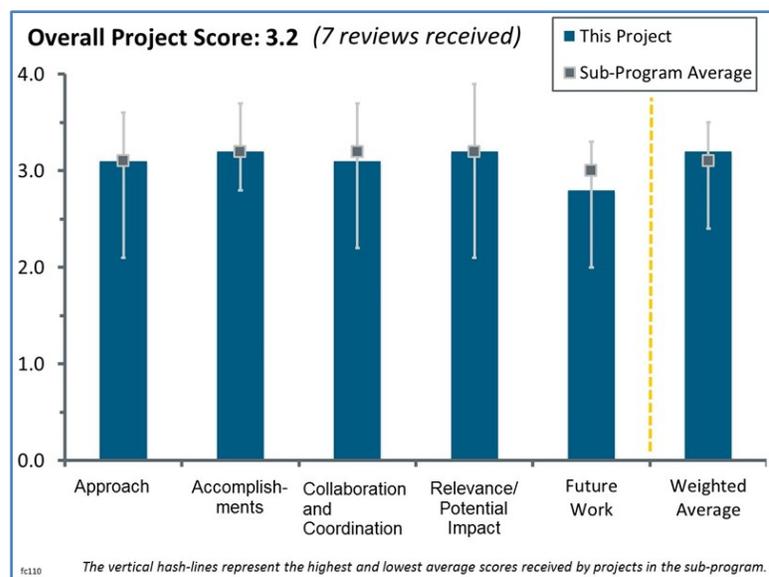
Brief Summary of Project:

The overall objective of this project is to demonstrate a low-cost hybrid inorganic/polymer from superacidic inorganic functionalized monomers with (1) area-specific resistance (ASR) $<0.02 \Omega \text{ cm}^2$ at operating temperature of an automotive fuel cell stack (95°C – 120°C) at low inlet relative humidity (RH) ($<50\%$) and (2) 50 cm^2 membrane electrode assembly (MEA) with desired mechanical properties and durability. The current-year objective is to incorporate the best hybrid polymer system into an MEA and deliver a 50 cm^2 MEA with all desired properties for third-party testing.

Question 1: Approach to performing the work

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

- This work continues a long line of investigation from the principal investigator (PI) on the use of heteropolyacids (HPAs) in polymers as proton conductors. The work for this review period settled on a specific fluoroelastomer system and HPAs, which the project team used to create membranes that were tested in the team's laboratory for conductivity and by a partner (Nissan) as MEAs. A key advance is the ability to make membranes that are thin, strong, and flexible. Conductivity values are very impressive at high humidity; work at higher temperatures (above 100°C) and lower humidities is needed in general and to address one of the milestones. The researchers say this work is planned with the National Renewable Energy Laboratory (NREL). MEA studies used conventional gas-diffusion-electrode-based electrodes sandwiching their membranes. Performance was very good and tracked with expectations for the membrane conductivity and thickness.
- The approach involves design of an improved polymer electrolyte membrane (PEM) through tethering of an HPA to a fluorinated backbone polymer. The fluorinated backbone should provide good chemical stability and gas transport, while the HPA should provide high conductivity even at low RH. This approach has been pursued by the PI and team for many years now, but it remains promising. The team is also working on integrating the new membrane into MEAs, which is appropriate at this stage of membrane development.
- The approach initially focused on addressing the performance, and indirectly the cost barriers, by attempting to prepare membranes that can operate at high temperatures under low RH. Earlier work indicated good conductivity for the Generation 1 polymers under hot and dry conditions, which could help reduce system costs. Recent work has not shown conductivity or ASR data for the new fluorocarbon-based HPA membranes under hot and dry conditions, but has focused on high RH conditions. It is not clear that these membranes will be less expensive than current perfluorosulfonic acid (PFSA)-type membranes, as the Colorado School of Mines (CSM) membranes rely on fluorocarbon polymers as well and are likely to have costs similar to other fluoropolymers. Advantages for this type of membrane are expected to come from the ability to operate under low RH. Performance under these conditions needs to be demonstrated.
- The scientific method includes appropriate controls. From the presentation, this seemed to be consistently lacking. For example, the PI cannot compare water transport of different thicknesses. Also, during the presentation, the Nafion™ sample was referred to as 2 million, but the label indicated 1 million. This made



it unclear (1) whether it was even known what control sample was tested and (2) whether anyone took the time to actually put a micrometer to it to verify. The PI could easily make the novel membranes the same thickness as the control. Not all data slides specified the type of control. The project team should be specific and reference accordingly. There are many different types of Nafion: standard, high, and low equivalent weight, as well as solvent-cast and melt-extruded films. All of these variables have impacts on fuel cell performance. Regarding the cost barrier, it is certainly understandable how \$3 million would be secretive; however, the PI certainly could add rationale. For example, it is unclear what the cost and source of the inorganic active group are, what the resin cost of a “standard” fluoroelastomer is, and what the range of costs for all fluoroelastomers is. The Synquest prices and logic through benefit of scaling should be shown. It is assumed that the cost of the polymer is the largest component; however, do not underestimate the cost of converting into a film. It is unclear what the incremental estimates via the different methods are. A cost of \$20/m² is an extremely low amount. Additionally, thickness is a critical factor. The work is feasible and integrated, and this is a good project—this criticism applies to the design.

- The approach follows mature chemistry modified for film-making capability, conductivity, and crossover properties, even at 120°C. The approach includes testing against U.S. Department of Energy protocols and 50 cm² cell testing. The project should be careful about drawing conclusions based on 80 μm films.
- The approach is challenging since this is such a new materials development project. There needs to be a good balance with actual fuel cell testing/demonstration versus material fundamentals understanding.
- The overall approach is feasible, but the plan for thinner membrane fabrication is not clear.

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.

- After a rocky start to the project, the team has been making good progress since last year. The work so far has indicated that HPA content needs to be kept below a threshold level to maintain good mechanical properties. Also, the mechanical properties are highly sensitive to the membrane processing conditions, with the only membrane showing good mechanical properties having been pressed at 180°C and 6000 psi for five minutes. It seems likely that the optimal pressing time would be different for different HPA contents. Therefore, it might be useful to further explore this space by constructing a plot of mechanical properties versus HPA content on one axis and pressing time on the other axis. The chemical durability and gas crossover results shown are misleading because they compare a thick HPA-based membrane (80 μm) with a much thinner Nafion 211 membrane. This sort of comparison should never be done unless the membrane thickness is the same. The PI indicated plans to repeat these tests with a thinner HPA membrane. The PI also indicated that 3M will examine degradation products to look for evidence of degradation modes that may harm performance without strongly affecting the open circuit voltage. These are crucial tests to run, and it is good that the PI seems committed to running them. The MEA results presented from Nissan are very encouraging. Considering the struggles the project experienced in earlier years, the level of achievement at this point is quite impressive. The H₂/O₂ performance looks especially good, whereas the H₂/air performance is good at low current but shows mass transfer limitations at high current. For some reason, the testing used extremely high flow rates of 2.0 and 4.0 liters per minute for anode and cathode, respectively. These flow rates are quite a bit higher than what is needed for normal differential testing and likely caused significant pressure drop. The team should check the pressure drop under these conditions and report the actual cell pressure, which is probably quite a bit higher than the claimed 0 kPa_g. Furthermore, fuel cell pressures should always be reported as absolute pressures, not gauge pressures, especially since two of the partners on this project are located at high altitude. Also, the testing should be repeated with lower cathode loadings. The use of an unusually high cathode Pt loading prompts questions about whether there may be issues at more normal loadings. Considering the high loading, the performance of the baseline N211 MEA looks a little low.
- Progress toward conductivity and crossover targets is excellent. Crossover prevention is particularly good with these materials, which may ultimately be one of their greatest strengths. Durability is also good, which the PI attributes to the fact that HPAs catalyze peroxide decomposition.

- The project has managed to make perfluoro-HPA membranes with very good conductivity under high RH conditions and has been developing processing techniques to repeatedly make good membranes. The project has not shown progress toward the DOE target of achieving an ASR of $<0.02 \Omega\text{cm}^2$ at 120°C and water partial pressure of 40 kPa, which is lower RH than the project target of 50% RH. The progress toward durability and permeability targets is not convincing when using an 80 μm thick film (that had an order of magnitude higher ASR than the target), as the thickness has a large impact on both permeability and durability.
- Accomplishments are very good, but some of the data seem to be from 80 μm films (usually $<25 \mu\text{m}$), so caution must be exercised when making conclusions. The process has been scaled from 10- to 100-gram batches. The process steps were reduced from seven to three. Stability at open circuit voltage is excellent. Cell performance is very encouraging for the unoptimized MEA (slide 18). Research and development on the electrode ionomer compatibility with the CSM membrane is needed. All project targets have been met but not simultaneously.
- Fundamental materials property advancements are hard to judge since they compare to a much thinner NR-211 membrane. A better comparison to a Nafion benchmark should have been done. Last year's reviewer comments were to compare to NR-212, which has similar thickness, rather than the much thinner NR-211. That comment did not appear to take since this year there are still plenty of NR-211 comparisons in which the difference in thickness confuses the comparison.
- There is good progress on concept demonstration. Cost needs more attention. Durability was addressed, but it is well known that the molecular junctions of dissimilar materials are the weak links. More attention to durability (in the holistic sense) could be beneficial.

Question 3: Collaboration and coordination with other institutions

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

- Coordination in this evaluation period with Nissan Technical Center North America (Nissan) has been very strong and was responsible for nearly all MEA results. Collaboration with 3M helped the PI work through selecting and using fluoropolymers. Collaboration with NREL was not much emphasized this period but was useful in earlier project phases and in other joint projects.
- Nissan and 3M bring substantial knowledge and expertise to the project, as do NREL and Hamrock. Cooperation is excellent.
- Collaboration with Nissan for in situ MEA testing is good.
- Collaboration between CSM and 3M appears to be working well, and 3M's participation has been beneficial in improving the film processing. Collaboration with Nissan seems to be working satisfactorily and should increase as MEA testing gets in full swing. The collaboration with NREL is not so clear, and it is not clear what NREL has contributed to date, and what NREL's future role is. Membrane testing results, especially under hot and dry conditions, were not shown. Testing shown appears to have all been done at Nissan and CSM, and future work is detailed at CSM and Nissan with no mention of NREL.
- The interaction with Nissan is valuable for MEA testing. The described role of NREL in Year 3 seems almost identical to that of Nissan. NREL could offer increased value to the project through the national laboratory's transport diagnostics capability, especially given the high apparent mass transport losses exhibited in the hydrogen/air MEA performance.
- Better collaboration with the Fuel Cell Consortium for Performance and Durability (FC-PAD) characterization techniques could have been done. Many questions remain about the MEA performance issues being attributed to the Nafion used in the catalyst layer; however, the newly developed polymer would be a worse candidate for the catalyst layer since the oxygen transport is much lower for the new polymer. This is a good property for the membrane but not so good a property for the catalyst layer.
- It is not clear why 3M was not participating/giving consultation more. 3M should have been able to provide valuable insight that could have made the project more efficient and robust. There are many Nafion fuel cell experts (ex-DuPont) that are happy to consult. The PI should consider adding them to the team as both chemical and application inputs. A film manufacturer would be a good addition.

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

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

- The project is highly relevant. Development of improved membranes is one of the most relevant things a project can do.
- Efforts to develop a membrane with the desired properties at high temperature and low RH are wholly consistent with DOE goals, objectives, and targets.
- If the approach can meet the cost target, \$20/m², the project well supports the Fuel Cells sub-program goal.
- The concept is good, and the practical progression is pleasantly anticipated.
- The project is relevant and has potential to provide an alternative type of proton-conducting membrane for polymer electrolyte membrane fuel cells (PEMFCs). The potential will depend on the cost and any potential performance advantages versus PFSA membranes. There appear to be conductivity advantages in the membrane, but durability and performance with membranes with relevant thicknesses at high and low RH conditions still needs to be demonstrated. Low RH conditions at high temperature are of particular interest, as performance there could enable lower-cost systems.
- The PI's approach is unlike any other being pursued for PEMFC electrolyte materials. There are still issues to be resolved (e.g., his materials are so far not suited for use in electrodes, and little is known about how they might interact with catalysts), but there is no reason to think that the issues will not be addressed with further work. Cost is likely to be reasonable, and performance and durability of membranes appear excellent. This work provides an imaginative and potentially very attractive alternative to PFSA and related materials.
- Although the material appears to be promising at the outset, many trade-offs in performance creep in on the way to making an MEA, and at the end of the day, the overall performance remains on par with Nafion, which greatly diminishes the impact on the DOE goals.

Question 5: Proposed future work

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

- The future work is focused on testing these membranes in MEAs and determining MEA performance. Ex situ testing of the membranes should also be done, and testing should be to the DOE targets, including 120°C and water partial pressure of 40 kPa, not just to 80 kPa or 50% RH.
- Future work is focused on defining chemistries, structures, and processing conditions to meet all technical targets simultaneously in a single membrane.
- The future work for CSM and Nissan looks reasonable, though there should be more of an emphasis on low-humidity testing. No future work was shown for NREL.
- The future plans sound good. How cleanly it will be carried out is questionable, considering the current state of reporting. More and different accelerated degradation testing should be conducted. A plausible plan on combining different (desirable) attributes that will pass all of the hurdles should be developed. Cost was not addressed at all; the presenter indicated that "3M says it's okay," but costs for post-polymerization are unclear.
- The project has essentially ended (July 2017).
- The project ends very soon, and proposed future work is appropriate.
- It is not clear how to fabricate thinner membranes with good mechanical stability.

Project strengths:

- The project has developed an innovative membrane system that relies on a different proton conductor from previous PFSA or hydrocarbon systems and should provide different transport and physical properties. Along with providing membranes with different properties, this could offer potential to change ionomer–catalyst interactions and reduce the negative impact of ionomer adsorption on the oxygen reduction reaction catalyst.

- Materials have good properties as membranes (high conductivity, apparent good mechanical properties, and good durability) and are able to be used in MEA fabrication and testing. MEA behavior is as expected for a membrane having the ASR of these materials.
- The team has amassed considerable experience in tethered HPA systems. The recent progress in incorporating HPA membranes into MEAs with reasonable performance is encouraging.
- The project did make a lot of progress toward the development of a new PEM polymer and even demonstrated it in an operating fuel cell.
- Mature/scalable chemistry for the membrane fabrication can be attractive to industries. There is strong collaboration with Nissan and NREL.
- A strength is the concept of highly functional components being combined to provide new capabilities.
- The project team is very good.

Project weaknesses:

- Progress has been made in preparing membranes that can be incorporated into a functional MEA, but the membranes seem finicky compared to conventional Nafion, requiring very specific processing within a narrow range of conditions to yield acceptable mechanical qualities.
- So many properties of a good PEM membrane need to be realized at the same time, so it is difficult to achieve them all. For example, we can have all the greatest properties of the membrane, but if the costs of manufacture are much too high, then no significant advancement is made toward the DOE goals.
- More work is needed to understand how to process materials to make strong and flexible membranes. More parameter space on processing, coupled with more careful mechanical studies, should be informative. Integration of the membranes with electrodes, either with the project's ionomers or other ionomers, was beyond the scope of this project but should be addressed going forward.
- The project has had delays and has not provided data on the current membranes at high-temperature, low-RH conditions. The project does not appear to be targeting the DOE target of 120°C with water partial pressure of 40 kPa.
- Study and optimization of the interface between Nafion in the electrode and the CSM membrane is needed but not in scope.
- There is a sloppy scientific method (at least as presented—perhaps the work has been done but was just not shown).
- Plans for thinner membrane fabrications and how to meet the cost target are not clear.

Recommendations for additions/deletions to project scope:

- The project should include testing over the whole range indicated in the DOE target table, including at 120°C with water partial pressure down to 40 kPa.
- There is a lack of real cost evaluation or at least comparisons/approximations. These should be added. The effect of film formation techniques (processing) on performance should be added. Forethought and probing on molecular degradation should be considered. There should be more accelerated degradation testing; for example, it is not clear what happens with freeze-thaw cycles or with mechanical stressing of the film.
- The project team should review the test conditions on slide 25.

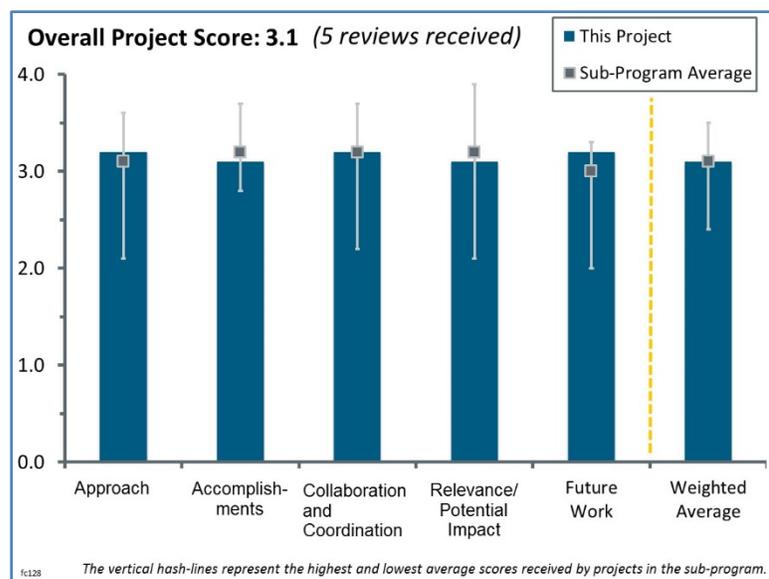
Project #FC-128: Facilitated Direct Liquid Fuel Cells with High-Temperature Membrane Electrode Assemblies

Emory DeCastro; Advent Technologies, Inc.

Brief Summary of Project:

Direct dimethyl ether (DME) is a carbon-neutral hydrogen carrier that can be used both for internal combustion and as cost-effective fuel for auxiliary fuel cell power systems in automotive transportation.

This project will demonstrate direct DME oxidation with high-temperature membrane electrode assemblies (MEAs) and a Los Alamos National Laboratory (LANL) catalyst. DME is expected to significantly outperform state-of-the-art direct methanol fuel cells (DMFCs). The project will incorporate the new ternary anode catalyst in gas diffusion electrodes designed for high-temperature MEAs, evaluate performance with two different high-temperature membranes (polybenzimidazole [PBI] and tetrapyrindine sulfones [TPS]), and optimize structures and reaction conditions.



Question 1: Approach to performing the work

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

- The project addresses durability, cost, and performance barriers for portable power, direct liquid-fueled systems. The project is well designed to address issues with DMFCs by looking at an alternative direct DME fuel cell utilizing high-temperature polymer electrolyte membrane technology. This technology can take advantage of the higher temperature, lower crossover of DME fuel cells in high-temperature polymer electrolyte membranes and higher volumetric energy density of DME. Part of the approach is based on potential advantages of utilizing Pd to break C-C bonds, but the majority of the work to date appears to be focused on PtRu anodes and not the PtRuPd catalyst. The approach to prove the concept for direct DME by optimizing the PtRu anode and then develop the PtRuPd catalyst and anode seems to require additional effort than if the project would prove advantages of PtRuPd first and optimize the anode only once.
- The project team has spent a considerable amount of time doing baseline testing and quantification with traditional catalyst and membrane materials. This effort will be invaluable in the assessment of new MEA components and is something that is often lacking when addressing unfamiliar systems including unique fuels. A very systematic approach is used to assess the impact of a range of operational conditions and material properties. The project team may want to consider a more combinatorial approach to determine if there are parameters that interact to yield a certain outcome. It may help to more quickly isolate the optimal operational conditions and material properties. More fundamental insight into the mechanism by which the ternary catalyst selectively drives the C-O bond breaking will either aid its implementation into the fuel cell or point to other catalytic materials that may have enhanced DME oxidation performance. Overall, the desired outcomes and barriers are well detailed, and the plan appears to be feasible. With baseline testing complete, the hope is that progress will be accelerated during the next few quarters.
- The approach taken seems to logically progress from DMFCs to DME-operated high-temperature fuel cells on PBI membranes, incorporating new catalysts. The project addresses a unique case competing with DMFC with greater potential power density at lower loading and lower crossover.
- The CO poisoning from the DME oxidation is shifted from high temperature. Perhaps it would be possible to include a calculation to show what the up limit of temperature to completely eliminate the CO poisoning

will be. Also, perhaps the operating temperature can be further increased (i.e., beyond 200°C) to improve the performance. Finally, using an additive to improve the DME solubility in DME is a good approach, but the team needs to be more specific on the working principle and the expected results.

- It is difficult to determine whether the work is addressing all of the key barriers to commercialization of the technology, as the end-use application is not well defined. Certainly though, crossover, power density, and catalyst cost would be important for most applications.

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.

- Accomplishments related to performance benchmarking and operational parameter analysis have been satisfactorily completed. Some performance metrics have been met, specifically with the PtRu/C catalyst. Sufficient progress has been made to this point, and with the down-selection of materials, attaining the other performance metrics is in sight. PtRuPd/C was identified as an ideal catalyst early on in the project. However, it appears that little success has been achieved with this material, potentially because of a lack of insight into the properties of the ideal active site or ensemble for DME electro-oxidation.
- The results to date are encouraging, and with better catalyst layer development incorporating the Pd-containing catalysts, the performance targets should be achievable.
- The project is clearly pushing the state-of-the-art performance and understanding for this novel technology. The only weakness is the uncertainty regarding whether the technology really applies to DOE goals.
- The project has made significant improvements in the anode activity over the past year, increasing the activity of the PtRu anode by approximately 15 A/mg_{PtRu} at 0.5 V, and matching anode activity of DMFC with 1 M MeOH. The project is still short of the target 75 A/mg_{PtRu} at 0.5 V by a substantial amount. The project has not demonstrated the advantages of adding Pd and that it increases C-C bond splitting in the direct DMEs over PtRu.
- The project is still about 60 mV from the Fuel Cells sub-program target. The project team should further improve the PtRuPd catalysts and optimize the corresponding electrode structure to achieve the target.

Question 3: Collaboration and coordination with other institutions

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

- The project makes great use of national laboratory resources and other collaborators.
- This is a good complementary team. The project team should emphasize the effort from the University of Connecticut.
- The collaborations are appropriate for this phase, but establishing strong collaboration with a system integrator/commercialization partner needs to be accomplished to ensure the technology progresses beyond this funding.
- It is not clear how much LANL is contributing to the project other than contributing PtRuPd catalyst material. The results from LANL indicating efficiency of the PtRuPd catalyst to split C-C bonds or convert DME to CO₂ are lacking. The project could potentially benefit from some imaging experiments (possibly in situ x-ray imaging or neutron imaging), which could provide information on the phosphoric acid penetration into the anode.
- There should be greater interaction between collaborators or further evidence showing interactions in the presentation, for example, a task breakdown, Gantt chart, or work breakdown structure.

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

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

- Expanding fuel cells to a range of other fuels is key to their adaptability to a range of applications including backup power, large-scale storage, and transportation/portable power. The proposed work fits well within the Fuel Cell Technologies Office (FCTO) Multi-Year Research, Development, and Demonstration Plan.
- The use of the liquid fuel is always a good approach for polymer electrolyte membrane fuel cell (PEMFC) applications on transportation. Keeping this exploratory effort is one of the options for liquid fuel for future PEMFCs in vehicle applications.
- The technology could be a potential enabler for phosphoric acid fuel cells (PAFCs) operating on DME, which may have niche market applications.
- The project partially aligns with the FCTO goals. It does not align with goals to aid development of PEMFCs. High-temperature, direct DME fuel cell development does not help with hydrogen PEMFC development, help develop the market, or help increase PEMFC production, supply stream, or hydrogen infrastructure.
- The project seems appropriate for incubator funding, but the potential commercial applications for the technology do not seem to fit the large-scale impact that DOE typically funds.

Question 5: Proposed future work

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

- The future work addresses the appropriate issues: activity of PtRuPd and integrating it into an MEA.
- The project has appropriate milestones and risk mitigation.
- The proposed future work is appropriate.
- Proposed future work focuses on MEA scale-up and use of additives to manipulate catalyst layer hydrophobicity and DME solubility. There needs to be a risk mitigation strategy for the PtRuPd/C catalyst, as its initial performance is low, and there are concerns related to the phase and segregation stability of this ternary alloy at elevated temperature, low pH operational conditions. Future effort should also consider other types of carbons, assessing them for their impact on transport and catalyst layer stability.
- More details on future work would be beneficial to reviewers.

Project strengths:

- The project is advancing the state of the art and the understanding of a novel DME fuel cell technology. The fuel is interesting because of its high energy density and means to reduce CO₂ emissions. This technology could ultimately find use in applications with weight sensitivity. A strong team is working on important barriers for this technology.
- The use of a high-temperature membrane is an effective way to deal with CO poisoning in hydrogen fuel, which is a very much less emphasized topic in the current Fuel Cells sub-program portfolio. For future fuel cell vehicle applications, the CO poisoning is still an issue that needed to be addressed. This project is a good exploration of the CO poisoning issue.
- There was a well-defined baseline performance for traditional materials. This project attained relatively high activity for DME in comparison to DMFC. The project used a systematic approach to understanding and optimizing the impact of operational parameters.
- The project provides a direct liquid-fueled fuel cell with potential advantages in power density over DMFC and reformed methanol fuel cells.
- This project has good partners and an approach to demonstrating PAFCs operating at intermediate temperatures on DME.

Project weaknesses:

- This project needs a risk mitigation strategy for PtRuPd/C catalysts. More fundamental insight into the performance and use of half-cell electrochemistry of catalysts could help to optimize their composition and morphology. This would also be helpful in assessing the properties of any new catalyst to determine what parameters, such as type of carbon, need to be changed to get efficient integration of the material into the catalyst layer.
- The project needs more fundamental aspects on the catalyst development. More characterization of the PtRuPd catalyst is encouraged to build the property–structure–performance relationship to better guide project development.
- The pathway for DME looks like a long shot compared with hydrogen and battery electric vehicles for transportation applications. Maybe this could work in limited markets. Catalyst loadings, power density, temperature, and cost are high compared to commercial low-temperature fuel cells.
- It is not clear whether this technology will ever have large-scale impact. The cells still require relatively high precious metal loading. Additionally, it is not clear how complex the system will be for this technology.
- The project addresses a smaller market, which does not help develop hydrogen-based PEMFCs and diverts resources from that goal.

Recommendations for additions/deletions to project scope:

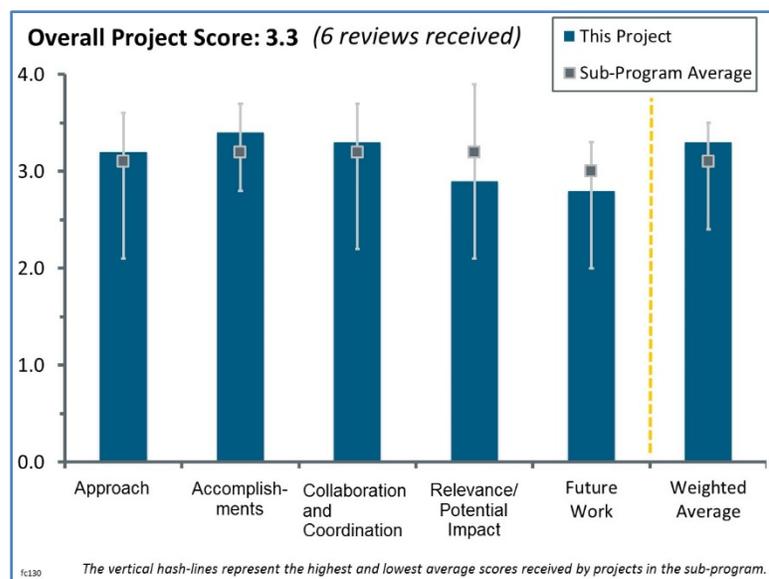
- The team should add some initial system design and report on the expected complexity and cost. This will help determine how competitive the technology can be with competing technologies and where future work should be focused. Determining the likely applications that would first adopt the technology would be beneficial to better define the barriers that need to be addressed.
- Characterization of the PtRuPd catalyst using transmission electron microscopy (TEM), x-ray diffraction (XRD), and rotating disk electrode (RDE) methods is highly recommended. Measurement of H⁺ conductivity within the gas diffusion electrode is highly recommended as well to guide the use of the additives.
- More fundamental assessment of DME electro-oxidation on PtRuPd/C and half-cell analysis are recommended.

Project #FC-130: Development of Platinum-Group-Metal-Free Catalysts for Hydrogen Oxidation Reaction in Alkaline Media

Alexey Serov; University of New Mexico

Brief Summary of Project:

This project will enable integration of platinum-group-metal (PGM)-free anode materials into an optimized membrane electrode assembly (MEA) structure. The resulting PGM-free-based anion-exchange membrane fuel cell (AEMFC) is expected to demonstrate significantly improved peak power density (up to 250 mW/cm²). Objectives include developing PGM-free electrocatalysts for hydrogen oxidation reactions (HORs) in alkaline media, scaling up the catalysts to 50 g batches, synthesizing a new type of ionomer for the AEMFC, and fully integrating the PGM-free catalyst with the ionomer into the MEA.



Question 1: Approach to performing the work

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

- The project is well focused on technology barriers for AEMFCs, particularly on replacing PGMs. The project was well planned using promising non-PGM HOR anode catalysts with a small amount of work on novel alkaline ionomers. HOR work showed promising results in both rotating disk electrodes in microelectrode experiments. A significant finding is that the ionomers appear to poison catalysts over time, which presumably will have a negative impact on fuel cell performance. MEA fabrication went very well, and MEA performance showed rapid and significant improvements to meet milestones. MEA tests showed an unusually large effect of temperature (e.g., in one case, peak power increased by nearly five times between 60°C and 80°C). The cause of this large change is not well understood, but it seems likely that it is not due simply to activation-energy-related effects. Overall, the final project results are quite strong for a cell with a PGM-free anode.
- There is a good materials design approach to meet unique project objectives (e.g., a PGM-free anode in an AEMFC). This project has met project objectives for peak power density. As pointed out by a number of audience members, this is a significant achievement.
- The team adopted a bimetallic nickel alloy supported over high-surface-area carbon. The new approach made significant progress over last year.
- This project is within three months of completion. The principal investigator's (PI's) approach is sound and well formulated. Project targets have been generally met or exceeded, most importantly including the second go/no-go design point of 250 mW/cm². Questions remain, for example, as to why there is such a large increase in power density with temperature for an MEA with NiCu/KB. It was surprising that there was no obvious mass transfer resistance/limitation for an anode 80–100 μm thick.
- The approach is fine, but the PI did not seem to really follow it. Instead, it appears that a myriad of materials and combinations of materials were tested, such as (1) commercial AEMs and ionomers, as well as ionomers provided by Los Alamos National Laboratory (LANL), and 2) both non-PGM catalysts and PGM catalysts. Additionally, there did not really appear to be any rationale to the combinations integrated into MEAs. For example, slide 8 claims certain combinations of ionomer and catalysts work well together, and then only one of these combinations is presented as an MEA.

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.

- Progress toward milestones and DOE goals appears to be excellent. All milestones were met, some by large margins.
- The project made significant strides this year and met a second go/no-go decision point.
- The approach demonstrated all project objectives with the possibility of a no-cost extension.
- The project team has met the project goals. The project had an end date of 5/31/2017, but the PI has requested a no-cost extension to 8/31/2017. The project goals have already been met, according to the AMR presentation. New project goals for the no-cost extension have not been identified. A “Future Activity” slide was included in the AMR presentation, with general research topics listed, but new MEA performance/durability targets were not identified.
- Although there has been some good technical progress, overall performance is still very poor. The milestones are really not compelling. There is no testing of MEAs on air—not simulated air or actual air (with CO₂).
- The project meets its own target, but the target is very low from a practical standpoint.

Question 3: Collaboration and coordination with other institutions

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

- It appears that there has been good collaboration with the team members, and new participants have been added as well.
- Good collaboration was demonstrated with all team members.
- Coordination with Pajarito Powder and EWII Group appears to have been very well coordinated and essential to achieving milestones. LANL work on HOR diagnostics was valuable for a semi-quantitative examination of HOR kinetics. LANL ionomer participation appears to have been relatively minor, with nearly all results reported for Tokuyama ionomers. The project reported some promising results for one LANL ionomer but did not follow up on them. Tufts work on water imaging is impressive, though not so well integrated. Overall, the effort did well in meeting milestones, though the fundamentals behind why improved performance was obtained were a bit lacking.
- Collaborations with LANL, EWII Group, and Pajarito Powder were described. It appears that ionomers developed at LANL were not incorporated into MEAs, so this collaboration is weakly tied to the project goals/accomplishments. There was little or no information/discussion on Pajarito Powder’s scale-up efforts.
- The project team consists of multiple partners from universities, national laboratories, and industry.

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

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

- This achievement has significant impact. It opens the door on future studies in other alloy systems. In the NiMo and ternary systems examined, the project goals were met. Durability studies to determine long-term chemical stability are warranted.
- The project does a nice job toward advancing the goals and objectives of the Program.
- Alkaline fuel cell research represents one of the Fuel Cell Technologies Office’s (FCTO’s) long-term research and development focuses.
- Demonstration of MEA-level performance using a non-PGM anode is important and potentially transformative. More knowledge is needed about the mechanism and rate of HOR reactions and the role of ionomer poisoning, which apparently can be significant. Closer attention to water management will also be needed.

- Unless the more serious issues with AEMFCs are resolved, such as sensitivity to CO₂ and membrane stability, work on HOR catalysts will have no use.
- It is not clear that this work will have any real-world impact unless some future breakthroughs arise.

Question 5: Proposed future work

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

- Optimizing the carbon-supported NiCu alloy should be continued. It represents a good choice in finishing the project with the remaining time.
- There appears to be more future work listed than is likely to actually be achieved in the time/budget remaining. The plans are good but should probably be prioritized.

Project strengths:

- The project strengths are as follows: (1) the PI and project team have developed/tested a Pt-free high-activity anode for an AEMFC, which is a significant breakthrough, and (2) the project team has met all of the milestones and go/no-go design points.
- This is a high-risk project. It is good to have some in the FCTO portfolio, although it may be less important with the Advanced Research Projects Agency–Energy. This project has made good progress and has a good approach/plan.
- A non-PGM anode was demonstrated in an AEM fuel cell.
- The team achieved the second go/no-go target with promising performance.
- The project used a novel approach and delivered on project milestones.

Project weaknesses:

- Project weaknesses include the following: (1) the MEA power densities are not yet competitive with those obtained with Pt-based or PGM-free polymer electrolyte membrane fuel cells, so it is not exactly clear whether there is a pathway for commercializing this technology, and (2) fuel cell performance data with a NiCu/KB MEA and air were not presented.
- Mechanistic details regarding how HOR occurs and how ionomer can poison the catalyst are lacking. A very strong link between cell temperature and MEA performance is important but unexpected and is not yet understood.
- The project does not appear to follow the plan. Performance is too poor to make an impact.

Recommendations for additions/deletions to project scope:

- Presumably, testing on “non-simulated air” means testing on actual air (with CO₂). If so, this should be the highest priority with the best MEA, to date, to see whether there are unique issues with these catalysts and ionomers in the presence of carbonates. This should be done before more optimization.
- This project will end in three months. There is no need for additions/deletions to the project scope.

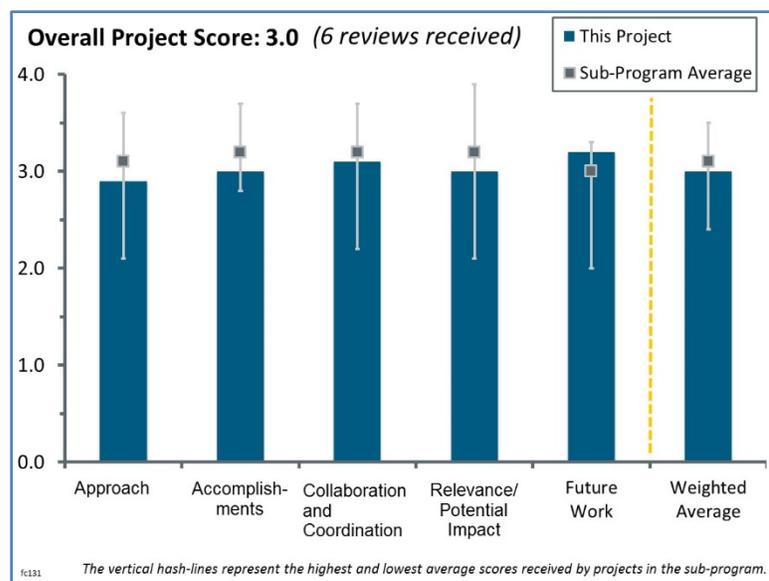
Project #FC-131: Highly Stable Anion-Exchange Membranes for High-Voltage Redox-Flow Batteries

Yushan Yan; University of Delaware

Brief Summary of Project:

This project aims to develop a class of anion-exchange membranes (AEMs) with very high oxidation resistance for high-voltage cerium redox flow batteries (RFBs) and other alkaline-membrane-based electrochemical devices, such as fuel cells and electrolyzers. Cerium RFBs show potential to offer high-performance and low-cost electricity storage solutions for renewable energy, and stable AEMs are the key missing element in making cerium RFBs a viable technology. Stable AEMs can also be used for hydroxide-exchange membrane fuel cells, for improving cell durability and performance, and for highly durable AEM electrolyzers, lowering hydrogen production costs. This project will

contribute to knowledge of polymer chemistry and membrane technology that will help advance the design and development of polymer electrolytes for electrochemical devices.



Question 1: Approach to performing the work

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

- The approach is to work on an RFB with high potential and low capital costs. This is an excellent choice, as making an AEM work in this environment will rapidly get the technology leveraged. This is a logical polymer development project that starts with gaining chemical stability, followed by mechanical strength, and finishes by demonstrating some modest scale-up capability.
- The project seeks to develop a stable alkaline membrane for use in RFBs, enabling the development of a double-membrane flow battery as demonstrated in a separate Advanced Research Projects Agency-Energy (ARPA-E) project. The approach of combining a stable cation with a stable backbone to achieve high membrane durability for RFBs is reasonable.
- The approach is effective and comprehensive, and the methods are sound in terms of membrane characterization and synthesis.
- The approach for membrane fabrication was well designed and reasonable. The project was integrated with the National Renewable Energy Laboratory (NREL) testing system.
- This project is focused on the development of a membrane for flow batteries. Although some of the fundamental properties of new materials can be used in both applications, targets for this work are not clearly defined. For example, the project's reported membrane conductivity of about 5 millisiemens/centimeter (mS/cm) is much too low for any meaningful automotive fuel cell application, for which typical polymer electrolyte membrane conductivity is more than 100 mS/cm. Furthermore, 5 mS/cm is not an adequate conductivity for flow batteries. Nonetheless, no target for membrane conductivity was given.
- The approach to making an alkaline membrane, using 9MeOTTP-PBI and oxygenated (methoxyphenyl) tetra substituted phosphorous cations in polybenzimidazole (PBI), is reasonable. However, the rest of the approach to the work is rather poorly designed. Using a liquid acid and base solution separated by a neutral salt solution for the RFB is impractical. These liquids will never be separable in a practical device, let alone by a Donnan electrostatic barrier. The conductivity is mostly liquid and not membrane conductivity, and it

is not so high. The authors should try to test the 9MeOTTP-PBI in pure water to see what the anion conduction is.

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 overcame initial stability issues with the first polymer system by switching to a PBI backbone. This improved polymer system demonstrated adequate stability. Building membrane electrode assemblies (MEAs) was delayed, as first materials were not suitable for testing. It is to be hoped that MEA testing will now be accomplished with the more stable materials.
- The team transitioned away from polysulfone backbones over the past year, finding that PBI-based backbones should have better oxidative stability. The strategy for ionic tethering also changed, with an iodobutyl linker replacing the bromomethyl group used previously. The new linking strategy appears to successfully avoid the crosslinking issue that arose with the earlier linking strategy. Changes to the cation itself, which involved replacing methyl groups with methoxy groups, were also made in an effort to improve the conductivity. Still, the conductivity of this ionomer seems rather low compared to other alkaline ionomers, and far lower than acidic ionomers. It is difficult to determine whether this should be considered a successful material since no targets exist for alkaline ionomers and the project milestones do not indicate a target conductivity. The new ionomer material passed the specified chemical durability test with less than 20% loss of conductivity, but it appears that the chemical stability of this ionomer is still insufficient. Significant conductivity loss occurred, and chemical changes are apparent in the nuclear magnetic resonance spectra. Further work is needed to understand and prevent the degradation. Since DOE does not have any published goals or targets for this application, it is not possible to assess progress toward DOE goals.
- This project is a low-technology-readiness-level (TRL) project. The principal investigator (PI) has finished most of the milestones successfully.
- The PI emphasizes that this is an AEM project and not a fuel cell/flow battery project. Even so, showing the performance in relevant applications (either flow battery or fuel cell) would provide the additional information. The stability of the “traditional” AEM was limited to 40 hours, and this work demonstrates stability in solution tests for 100 hours. It is unclear whether this test would hold in any operating cell.
- The conductivity is for mixed liquid and membrane electrolytes and is low, on the order of millisiemens. This work may have started with a good idea, but execution has been poor. The team should concentrate on optimizing the performance of the 9MeOTTP-PBI polymer and membrane in water.
- Conductivity and stability goals should be defined so that one can gauge the progress toward the goals. It is very difficult to see whether any actual progress has been made.

Question 3: Collaboration and coordination with other institutions

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

- The addition of Giner Inc. and Xergy as team members that can help with reinforced membrane fabrication strengthens the project. Overall, it looks like a good and appropriate mix of capabilities within the team.
- The collaboration is very good. The PI from the same institute was involved. The formal PI kept contributing and even changed job duties. The collaboration with the NREL is excellent.
- The Delaware team needed help and seems to be getting it from membrane optimization. It is admirable to seek help when needed, but this may be too little, too late, to get useful results (a high-conductivity hydroxide-conducting membrane).
- There is collaboration with a national laboratory to build MEAs and with industrial partners to fabricate the reinforced membranes. The project needs collaboration with a national laboratory or university to study the in-depth physical chemistry and mechanics of materials so that they can be improved from a scientific basis. Also needed is collaboration with a redox flow company or potential original equipment manufacturers.

- Not much cost-sharing collaboration was seen. It makes more sense to collaborate on fundamental material measurements rather than fuel cell testing at this point in this project.
- The work includes partners at NREL, Giner Inc., and Xergy. However, it is unclear where they were involved and to what extent.

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

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

- DOE needs to invest in AEMs, as they have potential to be game changers in polymer-electrolyte-based technologies. This project is an excellent example of building a membrane for a device that needs an AEM to be functional.
- This is a relevant project and will contribute greatly to the Hydrogen and Fuel Cells Program (the Program).
- Improvements in AEM are of importance to many electrochemical flow systems, and the project demonstrates that its polymer membrane is resistant to oxidative resistance and shows significant improvements in comparison to the state of the art. However, the scalability of this material is in question.
- The project seeks to advance RFB technology, which is mentioned in the Program's Multi-Year Research, Development, and Demonstration Plan but does not represent a major part of the Program. Since the Program has not established specific goals or milestones for RFBs, it is difficult to assess the likelihood that the project can contribute to meeting the Program's goals. Since flow batteries represent a secondary area for the Program, the relevance of this project to Program goals is limited. However, the membranes being developed may also be relevant for hydroxide exchange membrane fuel cells, which adds to the relevance of this project.
- The making of a hydroxide-conducting membrane gives relevance to the Program. The authors did well to try phosphorous cations and have evolved this class of AEM, but the results are still not clear, such as determining the conductivity of the best 9MeOTTP-PBI membrane in pure water. The redox flow aspect was a diverging distraction.
- Membrane conductivity needs to improve by a significant amount before any meaningful relevance/impact toward the Program goals can be realized.

Question 5: Proposed future work

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

- The four proposed future work areas as given by authors are:
 - 9MeOTTP-PBI polymer synthesis
 - Investigate conditions for large batch production of 9MeOTTP-PBI polymers
 - 9MeOTTP-PBI [polytetrafluoroethylene (PTFE)]-reinforced membrane preparation
 - Improve thickness uniformity of 9MeOTTP-PBI PTFE-reinforced membrane
 - Work with Xergy to prepare PTFE-reinforced membranes
 - Work with Giner Inc. to prepare electrolyzers
 - 9MeOTTP-PBI PTFE-reinforced membrane characterization
 - More comprehensive tests of reinforced membranes, including oxidation stability, conductivity, [ion-exchange capacities], water uptake, and mechanical properties
 - RFB/fuel cell test
 - PBI PTFE-reinforced membrane in cerium RFBs and fuel cells

The first three work areas are fine, but the fourth should be dropped. Under proposed research work area 2, the work on electrolyzers should be dropped. The project should just characterize the best 9MeOTTP-PBI membrane in water to get the maximal useful result from this project. Phosphate cations are interesting; it would be good to see what the project has in this respect.

- The future work looks reasonable. Improved understanding of the degradation mechanism and the development of pathways toward improved conductivity should be emphasized.

- The project is close to completion and has been able to meet its proposed targets and milestones.
- The proposed future work is very feasible and meaningful.
- Future work focuses on modest scale-up, quality assurance and quality control issues, and reinforcement of material in membranes. It does not appear that the material is ready yet for device testing.
- The project has essentially ended (end date of August 2017).

Project strengths:

- The project has met its proposed targets on time (except for the one-month delay on the fourth-quarter milestone) and has successfully synthesized an ionomer that appears to have improved properties over the incumbent technology.
- This project developed a new type of highly stable AEM. Stability and conductivity were improved compared to the other membranes. This provides a new electrolyte film for both fuel cells and RFBs.
- The project builds off of earlier work performed by the PI in an ARPA-E project and successfully leverages that earlier work. The project benefits from the fact that the membrane under development could be useful in multiple applications, including the flow batteries as well as hydroxide-exchange membrane fuel cells.
- The project has the wide scope of work to innovate new materials.
- The approach to use immobile cations for a hydroxide-conducting membrane is novel.
- New AEMs are a strength.

Project weaknesses:

- A lack of targets or guidance from DOE makes it difficult to determine what this project should be shooting for. While progress and achievement of milestones is being demonstrated, the membrane conductivity is rather low and may be insufficient for commercial applications.
- The weaknesses of the project are the low transport properties, the synthesis yield, the limited direct collaborations (no data was presented in the presentation), and the limited applied results under electrochemical operating conditions.
- Because this membrane will also be used for fuel cells, the stability at higher temperature should be improved, and the property and performance at higher temperature should be given.
- The work was poorly executed. The best part was the evolving membrane, but to date, even this has not been brought to any tangible clear conclusion for conductivity in pure water and stability.
- Very little progress was made on materials with good conductivity.
- It is not clear that this technology can be scaled.

Recommendations for additions/deletions to project scope:

- The project should compare results versus commercial membranes as a reality check that the phosphonium-based membrane is on track to be competitive. This was shown for oxidative stability but not for other relevant metrics such as conductivity.
- The project should delete everything except optimizing the proposed research work areas 1, 2, and 3, which are fine. Work area 4 should be dropped, and the electrolyzer work under proposed research work area 2 should be dropped. The project should just characterize the best 9MeOTTP-PBI membrane in pure water for conductivity and stability to make clear conclusions.
- Electrochemical experiments would provide complementary information for assessing the true performance and value of this membrane.
- The project should improve high-temperature stability and test high-temperature properties and performance for this new type of AEM.
- More polymer science and device integration, maybe beyond the flow battery, are recommended.
- As the project has essentially ended, this question is not applicable.

Project #FC-132: Innovative Non-Platinum-Group-Metal Catalysts for High-Temperature Polymer Electrolyte Membrane Fuel Cells

Sanjeev Mukerjee; Northeastern University

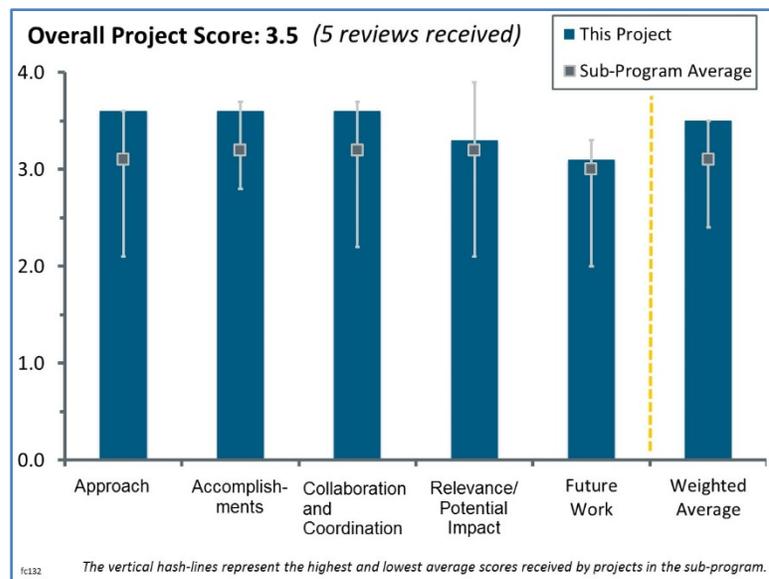
Brief Summary of Project:

This project is investigating the use and development of platinum-group-metal (PGM)-free electrocatalysts that would allow for high performance in high-temperature (HT) polymer electrolyte membrane fuel cells (PEMFCs) based on polybenzimidazole (PBI)-type membranes. A successful outcome will enable HT polymer electrolyte membrane (PEM) technology to be less dependent on Pt resource availability and lower membrane electrode assembly (MEA) costs by at least 50%. Benefits include increased energy efficiency and improved U.S. energy security.

Question 1: Approach to performing the work

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

- This work builds upon prior work from the principal investigator (PI) on non-PGM oxygen reduction reaction (ORR) catalysts in other cells (e.g., at lower temperature). Multiple Fe-N-C catalysts were made and tested by rotating disk electrode voltammetry, showing reasonable ORR kinetics, though significantly less than Pt/C. Fuel cell measurements with PBI MEAs at NEU and the University of South Carolina show good behavior at high gas pressures. Transport limitations in the electrodes appear to be important and are under study but were not finished in time for this presentation. Overall, very good performance is achieved with non-PGM cathodes. Durability studies showed very good results, which is perhaps the reason that alternate supports (e.g., tantalum and tungsten carbides that were proposed initially for consideration) were not studied.
- This project is pursuing a promising approach to develop PGM-free catalysts for HT-PEMFCs. The use of PGM-free catalysts enables elimination of a high-cost component while also potentially avoiding the phosphate anion poisoning issue that has limited performance of previous phosphoric acid fuel cells (PAFCs). By further developing several related PGM-free catalysts based on carbon and nitrogen-coordinated iron centers developed for low-temperature PEMFCs in a previous effort, this project is leveraging previous work toward a new application.
- The project approach in developing metal-organic framework catalysts (Northeastern University [NEU]) and Fe-based catalysts prepared using silica template (University of New Mexico [UNM]) is promising and shows good activity and durability. The catalysts studied in this project have shown encouraging results for the mitigation of phosphate anion adsorption.
- This is a well-thought-out and well-planned approach. The non-PGM catalyst for a HT fuel cell is an innovative approach. The use of a PBI membrane is an effective and valid approach.
- The project applied several previously U.S. Department of Energy-funded PGM-free catalyst syntheses and achieved reasonable performance. It would be more desirable if the PI could elaborate the design principle in applying these PGM-free catalysts under HT operation.



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.

- Performance levels are not yet matching performance of conventional PAFCs, but significant cost reduction, through the elimination of Pt from the cathode, has been achieved. The project has met targets for MEA performance using an Advent North America (ANA) anode catalyst and a Pajarito Powder cathode catalyst. NEU/UNM cathode catalysts seem to be slightly behind the Pajarito catalyst. The project is close to meeting final performance goals. The durability results are encouraging, but the improvement in performance during the 48-hour hold makes it difficult to interpret. The MEA should have been fully conditioned to reach its maximum performance prior to the start of the durability testing. Since it was not fully conditioned, the simultaneous conditioning and degradation make it impossible to say how much degradation occurred. Last year, the PI showed results from a voltage loss breakdown analysis. This year, the PI did not show such results, though he indicated that such analyses are still being performed. It is okay to show results from previous years where needed, but these should be clearly labeled as such. Several slides from this year contain the same data as presented last year, with no labeling.
- Multiple synthesis techniques were applied to address the catalyst performance. Advanced characterization techniques were used to understand the catalytic active site and influence (or non-influence) by phosphate adsorption. The project made good progress in catalyst/MEA performance.
- All milestones were met with a non-PGM cathode, which is impressive. Durability tests in particular showed very good results.
- This project has produced a significant number of results, and most or all of the project milestones have been achieved.
- All major targets have been met, and the project successfully passed all go/no-go points.

Question 3: Collaboration and coordination with other institutions

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

- NEU and UNM have had a good collaboration on PGM-free catalysts for several years now. The inclusion of Pajarito and ANA in this project is clearly critical since they are providing the anode and cathode, respectively, that enabled meeting performance milestones.
- This project has an impressive collaboration with a non-PGM catalyst manufacturer, membrane developers (both university and industry), and academic institutions. The contributions from different collaborators are very significant.
- Collaboration was important to the project, with catalysts coming from two partners (NEU and UNM), scale-up being accomplished by another partner, and assistance with fuel cell testing being provided by another partner (University of South Carolina). This report showed ANA as providing materials only. The degree of collaboration is reasonable relative to the project scope and goals.
- This is a very complementary team. From the presentation, all parties work effectively, and the project is well coordinated.
- The team has multiple partners, including universities and small companies.

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

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

- The project is highly relevant to the Hydrogen and Fuel Cells Program (the Program), as it is following a promising pathway to improved performance and decreased cost of HT-PEMFCs. Furthermore, advancements in PGM-free catalysts developed through this project may prove helpful in developing PGM-free catalysts for low-temperature PEMFCs as well. If successful, the project could have a large impact in accelerating commercialization of stationary fuel cell technology. The idea that this technology will help

enable flare gas utilization seems improbable. Flare gas is currently vented or flared because it is uneconomical to capture and use. There is no chance that flare gas would be converted to electrical power for transmission since it would be easier and cheaper to transport the flare gas than to build electrical transmission lines. Similarly, there is little chance that it would be used to power local operations using a fuel cell since this can already be done more cheaply using combustion.

- This project can effectively reduce the cost and durability of the HT PAFC and improve the overall efficiency of the PAFC system. Non-PGM catalysts also help with CO tolerance.
- This project addresses HT-PEM cathode catalyst development and is well-aligned with the Program goals for combined heat and power (CHP) systems.
- Achievement of targets with a non-PGM cathode is impressive and potentially important.
- This project is aligned with the mission of the DOE Fuel Cell Technologies Office.

Question 5: Proposed future work

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

- Future work was well planned.
- The PI indicated plans to continue efforts to achieve higher performance with both UNM and Pajarito catalysts. Further durability testing will also be performed including some temperature cycling testing. The possibility of scaling up to the 100 cm² level was also mentioned, which seems to be a necessity to meet the Quarter 8 milestone.
- Focus on transport limits will be important going forward. The low volumetric active site density in these types of catalysts is a limiting factor, and transport studies will help to reveal how much these limits affect overall performance.
- The team has proposed performing a list of future work, including catalyst down-selection to achieve better hydrogen–air fuel cell performance, durability studies, and MEA scale-up, until the end of the project period.
- More aggressive and longer durability/corrosion tests should be included, given that high stability was found in the short-term test.

Project strengths:

- This project is well planned, well organized, and well executed. The project team is a supplementary and comprehensive team with well-balanced expertise and capabilities. The project is well coordinated and well managed to achieve all project targets and pass all milestones.
- Demonstration of strong performance of a non-PGM cathode in this fuel cell format is impressive and a project strength.
- The project has a well-defined approach and strong collaboration with industry and universities.
- The project met several milestones. Promising durability at MEA level was demonstrated.

Project weaknesses:

- There is no obvious weakness in this project.
- The durability test should be carried out for significantly longer than 48 hours, particularly when initial promising stability was found. The presentation should be laser-focused on the major achievement, since this project is approaching its end. With this high funding level and limited presentation time, topics such as the need for Pt replacement, the market on micro-CHP, and review of others' work should be significantly minimized or removed.
- This catalyst class suffers from transport limitations due to the low volumetric density of active sites. This is not a weakness of this PI or his ideas but of these catalysts in general. It will be valuable to the field as a whole if solutions to this problem can be found.
- While significant progress is being made, the challenge of making a business case for this technology remains substantial. Significant additional progress would be required to enable a cost-competitive product.

Recommendations for additions/deletions to project scope:

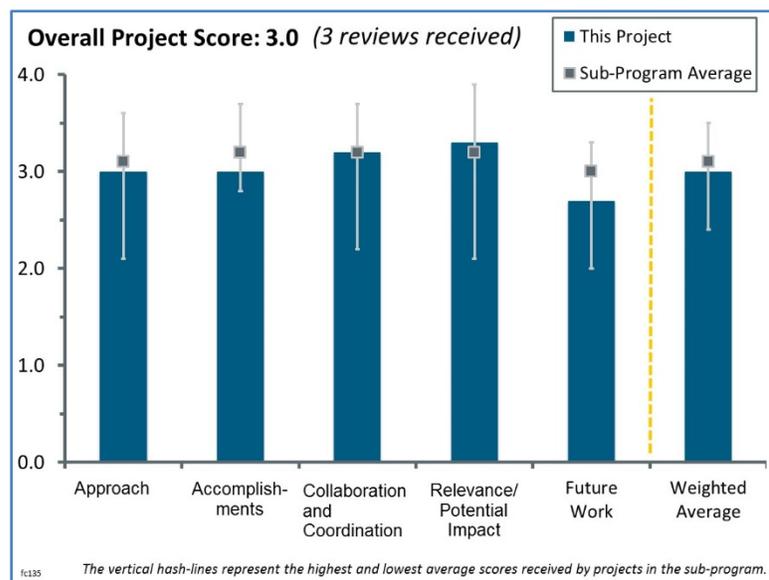
- Longer durability testing should be added to the project. The PI indicated plans to do so, but it is not clear how long they plan to test the durability. Addition of accelerating stressors, such as more thermal cycling (greater than the planned 50 cycles), and inclusion of inlet water vapor should also be used to look for durability issues.
- The only question is for the durability of the chronopotentiometric testing results, as there was no performance loss after 48 hours. After carefully looking over the data, the performance after 48 hours was slightly increased. The team may need to explain this phenomenon.
- Demonstration of durability for durations longer than 48 hours and a corrosion test for more than 3 hours would be a path forward for successful PGM-free HT-PEM technology.

Project #FC-135: FC-PAD: Fuel Cell Consortium for Performance and Durability

Rod Borup; Los Alamos National Laboratory

Brief Summary of Project:

The Fuel Cell Consortium for Performance and Durability (FC-PAD) coordinates activities related to the denoted development areas and supports industrial and academic developers. This effort aims to advance performance and durability of polymer electrolyte membrane fuel cells (PEMFCs). Researchers will develop the knowledge base and optimize structures for more durable and high-performance PEMFC components; improve high-current-density performance at low Pt loadings; improve component durability; and develop new diagnostics, characterization tools, and models.



Question 1: Approach to performing the work

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

- The approach of coordinating the investigation of the performance and durability of fuel cells through a consortium composed of the best available experts of national laboratories in a five-year project is excellent. It allows for a deep understanding of the different mechanisms involved and the impact of novel materials or structures and ensures a long-term continuity of the knowledge. The addition of complementary projects with new industry and academic partners completes this approach in a very efficient way. The addressed barriers are well-defined and -structured.
- The aligned approach of the FC-PAD consortium will have pros and cons, but on the whole, should provide more effective leveraging of the individual capabilities of the national laboratories involved. As a result, the overall advancement of fuel cell performance and durability should be accelerated.
 - The plan to include industry through four funded projects and with 30% support of the national laboratories' activities will provide an effective means of ensuring that the industry is engaged, that a reasonable portion of the national laboratories' work is relevant to industry, and that the work is well integrated with a broad range of activities.
 - On the cons side, care will need to be taken that administration of the consortium does not outweigh the benefits of coordination. Not all test plans and approaches should be decided by consensus, and the national laboratories should use their expertise to provide guidance and help set priorities.
 - The six component and cross-cutting thrusts have been well chosen to contribute to required fuel cell membrane electrode assembly (MEA) advancements.
 - The focus on pre-competitive-level activities is appropriate and will provide significant support to industry, whereas the competitive-level work is appropriately carried out by industry-led projects.
 - The technical work shown on the Ce migration appears well developed, with behavior of the Ce characterized through a number of methods that are providing a consistent picture and quantified parameters, which will be useful to others.
 - While the objective of evaluating/benchmarking different materials is useful and appropriate, it is not clear how the results shown (i.e., for the perfluoroimide acid [PFIA] versus perfluorosulfonic acid [PFSA] membrane) are different from what the supplier would also be doing and has shared

via U.S. Department of Energy- (DOE)-funded work. This type of work may be more useful if conducted on a broader range of materials.

- The main objectives of this work are to improve component stability, durability, and cell performance with optimized transport. Surely degradation analysis to couple experimental observation with modeling is conducted, but there is no indication of countermeasures on how to make these improvements. On top of that, it would be better to show a quantitative impact on the performance and durability.

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's accomplishments and progress are effective. The project clearly addresses the DOE targets in terms of performance and durability of MEAs and the associated components. The main focus this year was on membrane additive migration, PFIA water uptake and conductivity, and microelectrode studies.
 - The high number of results is reflected in the numerous publications and presentations, which makes the results accessible to the whole scientific community.
 - Different effects of contamination have been presented. It might be interesting to try to better link the evolution with a quantitative impact on the performance/lifetime in a real PEMFC.
 - Moreover, it is not clear whether the contamination of the membrane can be differentiated from that of the ionomer in the active layer.
- It is relatively early in the project, about 1.5 years into the consortium, and it is difficult to assess effectiveness against overcoming barriers. In addition, the consortium is focused on increased understanding, and the objectives of the project are high-level in nature, without clear milestones. It is also not clear how much of the progress has been made under this project, as opposed to follow-on from previous work. In terms of accomplishments, the Cerium migration and modeling has shown clear and valuable progress. The work on cation effects on conductivity is showing useful information, the membrane conductivity ionomer model is a valuable approach, and the effect of sulfate contamination is relevant.
- Degradation analysis became better than the previous project and has been conducted well because not only experimental observation, but also modeling, are well coupled.

Question 3: Collaboration and coordination with other institutions

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

- Collaboration between the different partners appears to be well structured, well managed, and very efficient, even if the material transfer agreements (MTAs) or intellectual property management plans (IPMPs) are still in progress. New collaborations were also started with DOE-awarded FC-PAD projects (under Funding Opportunity Announcement [FOA] DE-FOA-001412), including industrial partners 3M, General Motors, and the United Technologies Research Center, as well as with numerous non-FOA activities supplying materials or performing specific analysis.
- The very nature of the consortium and the interactions established through the FOA ensure that collaboration is an essential part of the work. It appears that the national laboratories are well coordinated. The interactions with the industry partners are just beginning, and how effective this will be will require proper assessment next year.
- Surely some collaboration exists; however, how to correlate one another organically is not well visualized to finally achieve the goal.

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

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

- The FC-PAD consortium and project is, as Electrocat, crucial for the Fuel Cell Technologies Office and represents a very high potential to significantly advance DOE in achieving Multi-Year Research, Development, and Demonstration Plan targets. The main targets are improving fuel cell durability and performance while decreasing the cost through a better understanding of the mechanisms involved, leading to a better mitigation solution. The high number of publications and presentations allows a very good diffusion of the knowledge learned to the fuel cell community.
- Performance and durability of the MEA are critical to meeting the long-term targets of fuel cell commercialization. These must be met at low cost. While the industry will focus on designing to appropriate trade-offs with cost, FC-PAD is focused on those activities that will enhance the understanding and provide the input to the industry to do so.
- Current activities should focus on indicating countermeasure/direction of the improvement, as well as the degradation analysis to finally achieve the goal.

Question 5: Proposed future work

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

- The future work proposed is in accordance with the results already obtained and aligned to achieve DOE goals.
- The activities outlined under future work are relevant and appropriate. However, in terms of milestones, only three near-term joint milestones were selected, and these are insufficient to understand the timelines and scope of the work to be accomplished. The stratified electrode layers are of questionable manufacturability, and it is unclear if this work is of any value.
- More effort on how to improve the performance and durability should be made with quantitative impact to meet the final target.

Project strengths:

- Extensive national laboratory capabilities will be coordinated and applied in a synergistic manner toward polymer electrolyte membrane issues affecting durability and performance. The multiple principal investigators are all of excellent caliber with many years of experience. The work is generally conducted in a systematic manner, with extensive characterization to support hypothesis and models. The use of models is routinely used to help support understanding, and development of appropriate parameters will help others for use in industry models. The interactions with industry are expected to be positive, will help to guide the work, and will allow industry to access the very extensive national laboratory capabilities.
- FC-PAD gathers the national laboratories' core competencies and the associated considerable amount of equipment to achieve the DOE goals. This project seems to be further improving collaboration between the national laboratories. Organization of the project into six thrust areas appears to be very relevant to cover all the required fields to advance fuel cell technology. The high number of publications and presentations allows a strong and quick communication to the fuel cell community.
- There are excellent human resources from various national laboratories.

Project weaknesses:

- No specific weakness was identified. A difficulty may be to finalize the IPMPs and MTAs, in particular for the state-of-the-art materials from industry.
- The large amount of administration and coordination is likely to take some focus away from the technical work and management. Is it unlikely that FC-PAD will truly get state-of-the-art materials, though the

researchers will likely get materials which are adequately close. The electrode structures to be studied are of uncertain relevance. Model predictions of the structures should be conducted as a first step.

- It was unclear how to link each analysis to finally indicate quantitative performance gain. Currently, it seems that only material-based analysis has been conducted. To communicate further with partners/manufacturers, how to improve performance as a device, and not only the material itself, is preferred.

Recommendations for additions/deletions to project scope:

- FC-PAD is focused on MEA components. Quantification, the link of these results to single-cell and even to stack performance evolution, should be taken into account. When considering a stack, bipolar plates also have an impact on its performance and durability through the aging of the bipolar plate itself or through contaminating the MEA, even with the presence of a coating. This may be added in the FC-PAD investigations. Differential cells use a serpentine design, which is not used in the current stacks. It has to be ensured that extrapolation of FC-PAD results remains valid in these new stack designs.
- It is better to apply a typical performance model consisting of measurable parameters to predict performance gain based on the individual excellent degradation analysis.
- Timelines and the description of milestones should be improved.

Project #FC-136: FC-PAD: Fuel Cell Consortium for Performance and Durability – Components and Characterization

Karren More; Oak Ridge National Laboratory

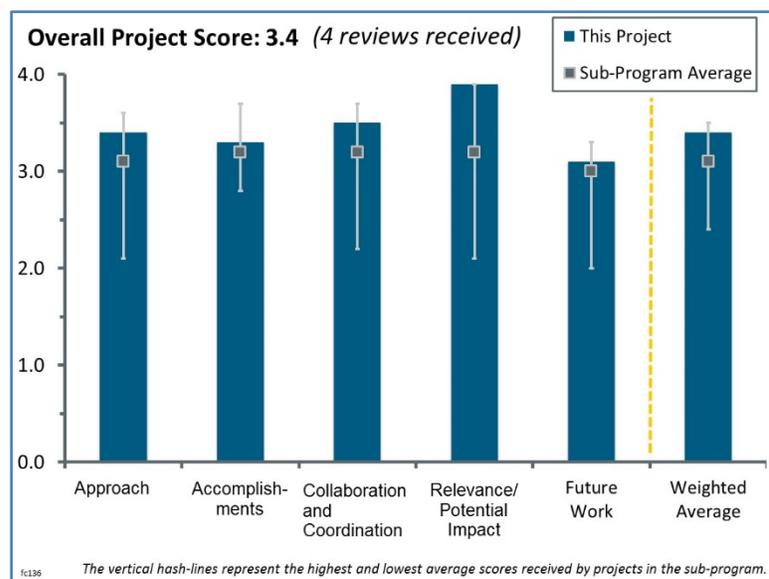
Brief Summary of Project:

The Fuel Cell Consortium for Performance and Durability (FC-PAD) coordinates activities that advance performance and durability of polymer electrolyte membrane fuel cells (PEMFCs). FC-PAD efforts include six complementary thrust areas, including one on electrocatalysts and supports. This thrust area aims to realize the oxygen reduction reaction (ORR) mass activity benefits of advanced platinum-based cathode electrocatalysts in high current density, with air performance for over 5,000 operating hours, and with low-platinum-group-metal (PGM) loading.

Question 1: Approach to performing the work

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

- The approach of coordinating the investigation of the performance and the durability of fuel cells through a consortium composed of the best available experts of national laboratories in a five-year project is excellent. It allows a deep understanding of the different mechanisms involved and the impact of novel materials or structures, and it ensures a long-term continuity of the knowledge. The addition of complementary projects with new industry and academic partners completes this approach in a very efficient way. The addressed barriers are well defined and well structured. This is a thrust area of FC-PAD, and the objectives and approach are very clear.
- Development of a new, faster accelerated stress test (AST) is a very positive development and was strongly needed, as the old protocol took too long to complete. It was not mentioned, but one assumes this is still under nitrogen, which may not be representative of duty-cycle degradation. A comparison of the failure fingerprint to Pt dissolution under actual operation is required for validation of the protocol. Results will differ as a result of cathode catalyst layer (CCL) water generation when cycled under air.
 - The AST does appear to be independent of carbon type, but it is unclear whether the Pt particles were all the same size as well. If not, then it would not be expected to see overlapping electrochemical surface area versus cycle number results for all three catalysts. It is unclear whether any catalysts have been screened that do not overlay on top of the three shown.
 - The study of PtCo catalysts is well designed and highly relevant. The Pt and Co dissolution rate measurements as a function of potentials and operating conditions, under inductively coupled plasma mass spectrometry (ICP-MS) and microscopy characterization, provide valuable information to characterize degradation rates and to compare catalysts. Going forward, it may be important to understand in which acid these tests were performed. The use of H₂SO₄ will delay oxide formation (versus in HClO₄), but binding with bisulfate will enhance chemical dissolution, so it is not clear which acid will result in faster dissolution.
 - The different membranes used make it harder to directly compare catalyst results. This is likely one failing of the approach of testing “state-of-the-art (SOA)” materials supplied by partners in that it will be more difficult to draw understanding and make correlations, as opposed to systematic studies. Additionally, it seems that there is incomplete information on some of the materials, which will also make it harder to draw conclusions. There is characterization and



- specification to be done within the project (e.g., x-ray diffraction [XRD], Co composition, and high-surface-area carbon) that will mitigate this, but only partially.
- Regarding the question raised on how much Co remains within the ionomer in CCL: when load is applied, Co from the membrane will transport to the CCL (migration). Thus, evaluation/calculation of Co in the CCL will be greatly underestimated if values of Co are used based purely on the ex situ (scanning tunneling electrochemical microscopy [STEM]/energy dispersive x-ray spectroscopy [EDS]) analysis.
 - Standardized rotating disk electrode (RDE) protocols are important to enabling cross-laboratory comparisons, and the understanding of the ionomer effect on ORR kinetics is important to interpreting data.
 - Presently, many national laboratories do not use Nafion™ in their RDE inks. It appears that the recommendation on slide 21 is to start using Nafion. It is unclear whether this indicates that the national laboratories will all be switching back to using Nafion. The optimal RDE ink seems to depend on the goal of the researcher. It is not clear whether the U.S. Department of Energy is interested in having RDE mass activities that match membrane electrode assemblies (MEAs) (the ink should contain ionomer), or activities that represent “intrinsic” values (no ionomer). The presence (or absence) of ionomer in the RDE ink is a major cause of discrepancy between the literature-reported values for mass activity.
 - It is not clear what the statement regarding the $i_k > 2$ is referring to, since the data shown are for lower values. Perhaps this is referring to Pt/C powders.
 - Since the recent introduction of the FC-PAD consortium, the concept of “Approach” has changed somewhat from a well-designed project with milestones and goals to a more community-wide collaboration, and this project becomes one with more of a supporting role rather than a leading role. Furthermore, this project combines and reports on the efforts of several laboratories.
 - By applying SOA characterization techniques, it is expected that the bottleneck can be revealed gradually.

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.

- Significant progress has been made, and the accomplishment is solid and well coordinated.
 - AST development and refinement is interesting, with an acceleration of a factor of 5 in the testing duration. The effect of the water in the dissolution of nanoparticles has been demonstrated. It was not clear whether the test could be further accelerated by increasing the relative humidity.
 - Regarding electron spectroscopy for chemical analysis (ESCA) measurements and the Pt and Co dissolution measurements, it would be interesting to establish a quantified correlation between them.
 - The results of quantifying Co loss are interesting, but the remaining question now is how much Co remains within the ionomer in CCL and how it will be measured.
- Key parameters, such as Co dissolution amount and ratio of nanoparticles of inside/outside carbon support, have been quantified by SOA techniques, which can help in analyzing and improving performance by communicating with performance and modeling teams.
- Progress looks good, and relevant activities are underway. However, it is early in the project, and it is difficult to assess how much was done in this project and how much is building on previous work. This project also suffers from a lack of clear milestones. The objectives, while appropriate, are fairly high-level in nature, and it is difficult to assess the overall test plan. The identified activities are important and will provide valuable input to the overall DOE objectives.
- The project is more of a support role, so the responsibility to meet the goals has been somewhat diminished since it is a shared responsibility across FC-PAD. Sometimes innovation is required to advance the SOA toward the goals. However, sometimes good, shared information on the properties and characterization of the SOA allows others to provide innovation. Thus, more characterization of the SOA components should be included in this project.

Question 3: Collaboration and coordination with other institutions

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

- There is excellent collaboration with other institutions to evaluate their SOA MEAs. Industry partners that receive DOE support should be required to have the FC-PAD team evaluate their components.
- Collaboration between the different partners appears well structured, well managed, and very efficient, even if the material transfer agreements or intellectual property management plans are still in progress.
- According to the representative partners' opinion, characterization by the latest technique at Oak Ridge National Laboratory is quite powerful and can act as a hub of the analysis with exceptional techniques and equipment.
- The very nature of the consortium and the interactions established through the funding opportunity announcement (FOA) ensures that collaboration is an essential part of the work. It appears that the national laboratories are well coordinated. The interactions with the industry partners are just beginning, and how effective this will be will require proper assessment next year.

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

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

- The FC-PAD consortium and project is, as Electrocat, crucial for the Fuel Cell Technologies Office and represents a very high potential to significantly advance DOE in achieving the Multi-Year Research, Development, and Demonstration Plan targets. The main targets are improving fuel cell durability and performance while decreasing the cost by developing a better understanding of the mechanisms involved, leading to a better mitigation solution. The high number of publications and presentations allows for a very good diffusion of the knowledge learned to the fuel cell community.
- Advanced characterization techniques and AST development are critical to supporting the fundamental understanding of performance- and durability-related processes occurring in PEMFCs. This information will feed into the supply chain and fuel cell designers to advance the materials and designs used to meet the long-term objectives of the industry.
- Characterization results here can find the root cause of the degradation and quantify key parameters that can predict the performance gain. Therefore, whether the target is achieved can finally be transparent.
- This type of project, one with a common set of characterization and evaluation techniques of industry-wide fuel cell components, is critical to advancing technology toward the goals.

Question 5: Proposed future work

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

- This project includes world-class characterization techniques and technique development. The extensive national laboratories' capabilities will be coordinated and applied in a synergistic manner toward polymer electrolyte membrane issues affecting durability and performance. The extensive characterization provides significant understanding to experimental studies and will support model development. The interactions with industry are expected to be positive and will help to guide the work. They will allow industry to access the very extensive characterization capabilities.
- FC-PAD gathers the national laboratories' core competencies and the associated considerable amount of equipment to achieve the DOE goals. This project seems to be further improving collaboration between the national laboratories. Investigation of industrial SOA catalysts is a strength. The high number of publications and presentations allows for strong and quick communication to the fuel cell community.
- Some future work would be for the FC-PAD team to develop new characterization tools. This would be an excellent utilization of the innovative resources at the FC-PAD laboratories.
- The future work proposed is in accordance with the results already obtained and aligned to achieve DOE goals.

- The proposed future work is relevant and necessary. However, there is some concern with how the FOA partners' samples will align with a preferred systematic study approach and how much information will be shared to allow mechanistic understanding to be developed. There are neither timelines nor milestones provided for the future work. The use of a very small cell for a durability test may result in significant variability because of the small sample size under study.
- It is quite important to reveal the bottlenecks. It is necessary to feed the characterization results back to the performance analysis and to find the essential countermeasure and finally predict the gain.

Project strengths:

- The common characterization techniques applied to different SOA MEAs are an excellent strength of this project. The development of these outstanding techniques does not need to be duplicated industry-wide, and having them in a common shared national laboratory resource is a great asset to the industry.
- The project strengths include SOA characterization techniques and facilities with excellent researchers.

Project weaknesses:

- No specific weakness was identified.
- The characterization work will be dependent on samples received, and it is not clear how coordinated the work will be with all of the FOA support work. It is likely that some of the samples will have proprietary restrictions. The many variations in the FOA samples will make it difficult to draw direct conclusions (e.g., the MEA samples had both different membranes and different catalysts).
- The project has weaknesses in having fuel cell industry-wide participation in the characterization efforts.
- More quantification and linkage to the performance prediction/modeling are needed.

Recommendations for additions/deletions to project scope:

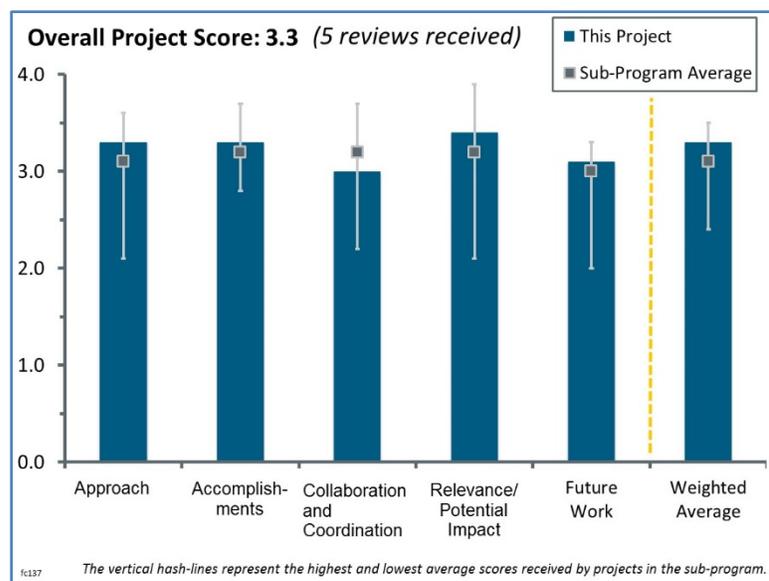
- The project scope could be expanded to include the development of new innovative characterization tools.
- The project is expected to characterize and quantify other key parameters that are controllable and critical for performance.

Project #FC-137: FC-PAD: Fuel Cell Consortium for Performance and Durability – Electrode Layers and Optimization

Adam Weber; Lawrence Berkeley National Laboratory

Brief Summary of Project:

The Fuel Cell Consortium for Performance and Durability (FC-PAD) coordinates activities that advance performance and durability of polymer electrolyte membrane fuel cells. FC-PAD efforts include six complementary thrust areas, all of which contribute to the electrode layer integration studies. Optimizing electrode layers and mitigating transport issues are vital to meeting U.S. Department of Energy targets. This project is identifying state-of-the-art catalysts; optimizing the catalyst layers; developing diagnostics to help resolve problems with high current density and low loading; and mitigating the problems through the use of novel electrode design, components, and diagnostic techniques.



Question 1: Approach to performing the work

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

- The overall approach is comprehensive. It is important that all the information feed into microstructural and molecular dynamics modeling. There is a broad range of activities, which are potentially too many. The highest-priority work should be the thin-film understanding, microstructural modeling, local resistance analysis, and ink model studies. Conditioning effects are also important. The characterization of ink structures helps to understand the ionomer structure. The electrode microstructure analysis via detailed characterization provides significant model input. The influence of the environment on the ionomer thin film is providing some interesting results. In terms of the electrode structures, there are some very interesting structures being developed. The use of model structures and understanding transport effects should provide good data for model validation, and the ability to control structures and confirm intended transport effects will provide good insight. However, it should not be expected that these structures can be manufactured as commercial structures, at least in the short term. The hydrophilic microporous layers (MPLs) using carbon nanotubes are also providing some interesting results. Overall, the combination of unique structures (catalyst layer [CL] and MPL) and enhanced characterization techniques will significantly advance understanding and eventually lead to the ability to tailor designs.
- The approach is excellent: coordinating the investigation of the performance and the durability of fuel cells through a consortium composed of the best available experts of the national laboratories in a five-year project. It is allowing a deep understanding of the different mechanisms involved and the impact of novel materials or structures, and ensuring a long-term continuity of the knowledge. The addition of complementary projects with new industry and academic partners completes this approach in a very efficient way. The addressed barriers are well defined and well structured. This is a thrust area of FC-PAD, and the objectives and approach are very clear. In particular, using a synergistic combination of modeling and experiments is very relevant.
- This FC-PAD project should use and report on commercially available materials and components. Detailed studies on new structures that are not commercially available or easily made are less valuable. For example, the stability study on the catalyst ink is a result that can be well related to industry efforts, whereas the

wettability study of a special carbon nanotube MPL, from a grade of SGL Group carbon material that is not available, is of less help. However, in general, nearly all of the work helps the industry as a whole with understanding the electrode performance; thus, the score was an “Excellent.”

- Because the CL is a key component governing the performance, where both electrochemical reaction and mass transport occur simultaneously, modeling is quite helpful to understand phenomena there and finally find key parameters in terms of performance/degradation.
- This is a very mixed approach, as there are too many disconnected projects that have a small likelihood of success compared to a concerted effort. Additionally, much of the effort is spent on ionomer layer resistance for low loading. The work seems very good, but it does not correlate well with what General Motors (GM) is seeing (ability to have very low loadings with no additional mass transfer losses). Much of this work will be very catalyst/ionomer dependent, and the researchers need to be very selective about what they look at. It is unclear why they have added the patterned catalyst layers, as this certainly has a long history, and it is not obvious that anyone is interested in it. A better understanding and some guidelines for ink/slurry stability and formation would certainly be helpful.

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.

- Significant progress has been made, and the accomplishments are solid and well coordinated. Main accomplishments concern the development of new catalyst-layer architectures, unraveling the origin of local resistance, and the exploration of the ink stability, dispersions, and fabrication methods.
- Accomplishments and progress toward a specific goal are difficult to measure in this project since it is more of a fundamental understanding/supporting role. This should be considered a good feature of the project, so perhaps DOE should consider modifying this question a bit to better reflect the mission of the FC-PAD laboratory team members.
- It is relatively early in the project, about 1.5 years into the consortium, and it is difficult to assess effectiveness against overcoming barriers. In terms of accomplishments, there has been excellent development of techniques and the ability to link a suite of characterization and diagnostics methods to investigate phenomena. In addition, a number of interesting structures have been made.
- As much of the project goals are “a better understanding,” this is difficult to judge. Along those lines, the work on better understanding of the ionomer resistance at low loadings is very important, but here coordination with other groups becomes very important. The investigator needs to ensure that what the project is measuring is relevant (i.e., whether these are the same catalyst/ionomer structures that are being used). When the data/conclusions differ, the team needs to actively work together or repeat the work of others to see what the bottom of the differences is.
- A catalyst ink study is needed from the industry aspect to finally guarantee the performance by process. A transport study is also helpful to find the bottleneck and finally improve hydrogen contaminant detector performance.

Question 3: Collaboration and coordination with other institutions

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

- Collaboration between the different partners appears well structured and well managed. New collaborations also started with DOE-awarded FC-PAD projects (Funding Opportunity Announcement [FOA] 1412) that includes industrials such as 3M, GM, and the United Technologies Research Center (UTRC), as well as with numerous non-FOA activities supplying state-of-the-art (SOA) materials.
- The very nature of the consortium and the interactions established through the FOA ensure that collaboration is an essential part of the work. It appears that the national laboratories are well coordinated. The interactions with the industry partners are just beginning, and how effective these will be will require proper assessment next year.

- Collaborations with other institutions should be expanded. Reporting on industry components provides the best utilization of efforts by the laboratories. Industry partners that do not allow sharing of this type of analytical work with the community as a whole should not be supported by DOE.
- Most of the collaboration is with other national laboratories. There is really good work being done outside of the laboratories. This seems to be partially corrected, moving forward.
- More collaboration with the electrocatalyst/kinetic team, performance team, and/or original equipment manufacturers is desired in terms of dominant factors and design of the performance.

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

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

- The FC-PAD consortium and project are, as is the ElectroCat (Electrocatalysis Consortium), crucial for the Fuel Cell Technologies Office and offer a very high potential to significantly advance DOE in achieving Multi-Year Research, Development, and Demonstration Plan targets. The main targets are improving fuel cell durability and performance while decreasing the cost through a better understanding of the mechanisms involved, leading to a better mitigation solution. The high number of publications and presentations allows a very good diffusion of the knowledge learned to the fuel cell community.
- This project has very direct and significant relevance to electrode performance. Focus should continue to be on the SOA industry-supplied materials and components. Divergence from these sets of materials for the purpose of understanding is also very relevant. Therefore, this project has a high level of relevance.
- The investigation into the thin-film losses, the catalyst ionomer interactions, the ionomer morphology, and the impact of the conditions on the ionomer structure is critical to achieving commercial fuel cell targets. The focus is currently on beginning-of-life structures. It is anticipated that at some point these techniques and understanding will also be applied to understanding degraded structures.
- The work seems to be a step behind the researchers working to develop the catalysts. That is, they look at a problem facing catalyst developers and try to find the root cause, but by that time, developers have already moved on. For the work to have real impact, it needs to develop a fundamental understanding that could *lead* to catalyst development. If there were a catalyst/ionomer structure like this, then that could be achieved. If they can do this, it will truly help the developers.
- This activity is focusing on part of dominant factors governing the performance, which can finally clarify how to achieve the goals.

Question 5: Proposed future work

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

- The activities outlined under future work are relevant and appropriate. However, no timelines or milestones are presented.
- The work on the catalyst ink could be expanded to give more understanding as to how ink properties relate to resulting electrode structures and ultimately fuel cell performance levels. There is a large degree of interaction in these materials and the processing steps that is not well understood by the industry. Industry is mostly working on this as an “art form” rather than a fundamental understanding. This FC-PAD project could help all industry with more work in this area.
- The future work proposed is in accordance with the results already obtained and aligned to achieve DOE goals. However, regarding the exploration of different CL structures, there are so many routes that choices will have to be made and explained at the next review.
- Adding 3M and GM as collaborative partners is a big improvement and will hopefully guide the work. In so doing, the team will likely find little interest in patterned CLs. This is not really a section to address this issue, but having ~ten laboratories work separately on different issues and then trying to tie them together is an incredibly ineffective/inefficient way to fund research. A concerted effort at one location, or possibly two, would be much more effective. It will be interesting to hear more about their work on the inks, as this is a little-studied area. Much of the other areas seem as if little has changed over the past years.

- Catalyst ink should be addressed more to control CL microstructure. In addition, ionomer network and coverage on the electrocatalyst can be studied further, which can govern the performance as well. Therefore, to understand the phenomena around Pt/ionomer, it is important to quantify the microstructure and properties affecting performance.

Project strengths:

- The focus on the understanding of the electrode layers for low catalyst loadings is likely the most important area to achieve low-cost electrode structures. The principal investigator is a leader in this field, and the continued support to focus in this area is expected to drive significant progress. The collaboration and coordination between the national laboratories and the FOA partners will result in the most comprehensive approach possible to study the critical phenomena. The combination of the characterization, experimental studies, and incorporation into models is likely the greatest strength of the project.
- The strength of this project is for the FC-PAD team to have access to a wide variety of SOA materials and components. This access should be able to give the team a deep understanding of the fundamentals of the mechanisms that dominate the electrode performance.
- FC-PAD gathers the national laboratories' core competencies and the associated considerable amount of equipment to achieve DOE goals. Investigation of industrial SOA materials is a strength. The high number of publications and presentations allows for a strong and quick communication to the fuel cell community.
- This project has exceptional modeling capability with coupling SOA characterization.
- This project has a truly talented group of researchers.

Project weaknesses:

- It is not clear how coordinated the FOA support work will be. If this results in non-systematic studies or incomplete studies because of proprietary information, this may delay progress.
- There is a lack of focus, both in goals the team is looking to achieve and in the team itself. It is extremely difficult, if not impossible, to get so many disparate pieces to work together.
- There is a challenge with open exchange of information that one hopes can be overcome with DOE help.
- This project needs further interaction with the kinetic team and performance/degradation team (FC-135).

Recommendations for additions/deletions to project scope:

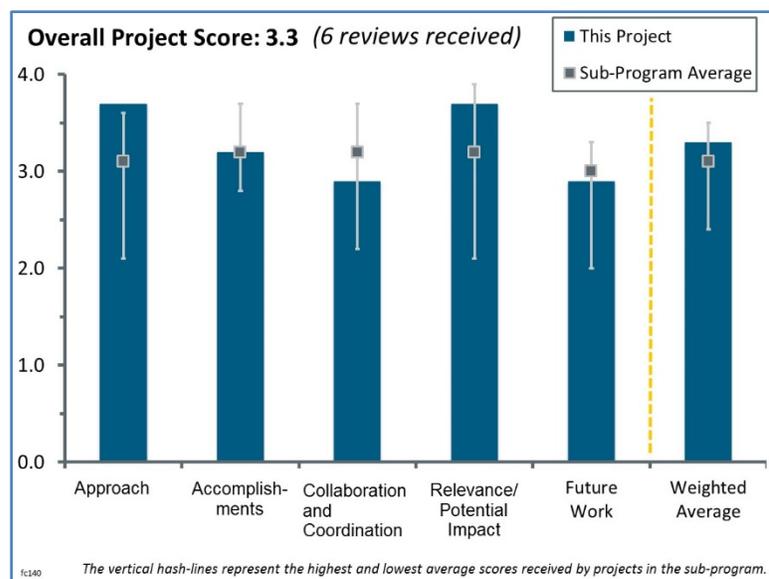
- It will be effective to consolidate some of the functions and roles between FC-135 and FC-137 in FC-PAD regarding performance analysis/prediction. Also, more communication with GM and UTRC is preferable for improving the performance in terms of design and prediction.
- Segmented catalysts should be deleted. Understanding why GM's new structures and, one could say, 3M's old structures do not see the mass transfer limitations that the team is trying to understand should be a big part of the work.
- There is no mention of how durability will be studied as part of this project.

Project #FC-140: Tailored High-Performance Low-Platinum-Group-Metal Alloy Cathode Catalysts

Vojislav Stamenkovic; Argonne National Laboratory

Brief Summary of Project:

A primary focus of the U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program (the Program) is development of highly efficient and durable Pt alloy catalysts for oxygen reduction reactions (ORRs) with low Pt content. This project will go from fundamentals to real-world materials to achieve rational design and synthesis of advanced materials with a low content of precious metals. Researchers are taking a materials-by-design approach to design, characterize, understand, synthesize/fabricate, test, and develop tailored high-performance low-Pt-alloy nanoscale catalysts.



Question 1: Approach to performing the work

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

- The overall approach is excellent. The principal investigator (PI) and team are applying strong fundamental understanding and state-of-the-art methods to develop and characterize novel catalysts. Increased focus is recommended on membrane electrode assembly (MEA)-level testing for activity and durability to help understand the large gap in mass activity between the rotating disk electrode (RDE) (e.g., 4 A/mg) and MEA (e.g., 0.5 A/mg).
- The approach from fundamentals to real-world materials is the only way to tailor the structure/composition in order to optimize durability/performance in Pt alloys for the cathode. This materials-by-design approach, by a better understanding of limitation, will secure the ability to industrialize new compositions aiming at the DOE target. Very good work has been performed since 2016.
- These are clearly unique catalyst structures, and the authors have well represented why they believe they may have higher specific activity while achieving high current density. Additionally, they have demonstrated scalability with no exotic steps.
- The approach of this project to prepare nanoscale Pt-alloy catalysts is unique and fascinating. Changing the size, shape, and composition of alloy particle morphology should have substantial impact on ORR catalytic activity and electrode performance.
- The approach is both aggressive (multiple tasks in parallel) and well designed, since it strives to address many potential risks (in a highly complex system) at early stages.
- The project team uses world-leading resources and capabilities to design catalysts from a fundamental point of view.

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 had impressive results in the past year in all key areas. (1) Fundamentals: The previous development of the RDE-inductively coupled plasma mass spectrometry (ICP-MS) was a great

contribution, and it is great to see the group using this tool effectively on these new catalysts, with interesting results. (2) Synthesis: The core team has continued to make excellent progress in developing new nanostructures. (3) Scale-up: The progress here is especially impressive. It is unclear whether this new one-pot process can be used to make nanoframes as well as nanoparticles. (4) MEA performance: It is also great to see MEA results, which are impressive when one considers how challenging it is to make a good MEA with a new catalyst.

- The work is progressing well toward scale-up synthesis reproducibility and 5 cm² MEA testing for demonstrating a real capability to reach the DOE target and reduce the cost.
- The team has made excellent progress in several areas. Use of electrochemical (EC)-ICP-MS is a very novel approach toward understanding the intrinsic stability of various catalysts, including different structures, compositions, and use of Au for stabilization. A new nanocage structure yields very high activity in RDEs, several times higher than the DOE target. It is unclear if the new structure is stable. Work toward scale-up of nanoframes, including development of a one-pot synthesis, is noteworthy. Additional work should focus on activity improvement in MEAs and durability assessment.
- Excellent mass and specific activities of the nano Pt-alloy catalyst have been demonstrated for fiscal year (FY) 2016. No further improvement has been reported in FY 2017. Some additional progress on characterization, particularly on transmission electron microscopy at Oak Ridge National Laboratory (ORNL), has been made. Scaling up and property validation of the catalysts are not trivial tasks; the team has good efforts on those tasks. However, fuel cell performance evaluation with the catalyst has seen very little progress, which is somewhat disappointing. The DOE target for the project is to use 0.125 mg platinum group metal (PGM)/cm² for an MEA. It is not clear how 0.04 mg PGM/cm² cathode loading can show an equivalent performance with the current state-of-the-art higher-loading (0.1 mg PGM/cm²) Pt-based electrode. The team should have made more effort to improve fuel cell performance in addition to the RDE measurement. Also, durability of the catalyst in MEAs is missing.
- It is good to see increased focus on catalyst stability in the catalyst design, as EC-ICP could become a powerful tool. However, most work is still on the RDE level, a very slow transition from RDE to any demonstration in an MEA. The only scale-up and MEA evaluation was with spherical PtNi catalysts that were similar but inferior (activity and stability) to existing alloy catalysts. It is unclear what the project milestones and go/no-go criteria are.
- Progress in the past year has been slow. Compared to where the researchers were last year, not much more has been accomplished. A year ago, they had many of these amazing catalysts, had demonstrated their potential in RDE, but had miserable fuel cell performance. A year later, they have even more new catalysts, more evidence of their potential, and more poor fuel cell performance. More effort should have been put into demonstrating that RDE results can translate into MEA results, and if not, why not.

Question 3: Collaboration and coordination with other institutions

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

- This project has excellent collaborations with team members (with others at Argonne National Laboratory [ANL] and with partners at national laboratories). The collaboration with the Fuel Cell Consortium for Performance and Durability to obtain the MEA results is especially commendable.
 - The catalyst community position should simply be that RDE is a good screening tool and that they would welcome improved methods to translate this into MEA performance projections by those who can contribute to this challenging task.
- The level of collaboration is generally good. Work with electrode development/MEA integration partners should have higher focus.
- The collaborative work with ORNL is excellent. The interactions with Lawrence Berkeley National Laboratory (LBNL) and Los Alamos National Laboratory (LANL) are moderate. No interaction with original equipment manufacturer industries is a concern. Difficulty (or slow progress) with scale-up does not give much room for further evaluation at the industrial sector.
- The proposed approach needs a good coordination between the different institutions involved (LBNL, LANL, ORNL) for tailoring the composition based on design, characterization, understanding, and synthesizing end tests. Some international collaboration may also progress the work toward disruptive technology.

- On national laboratory projects, it would be preferable to see a centralized effort rather than work distributed to six different laboratories. This is not efficient. An industry partner would be a great help, especially in the fuel cell development area and to make sure that the work is relevant.
- ANL needs to get catalyst samples to other laboratories for evaluation. Inclusion of industrial partners would increase credibility. The project should avoid excessive delays in technology transfer.

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

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

- The project aligns well with the Program and research, development, and demonstration objectives. The project has the potential to advance progress if the MEA using the developed catalysts shows good mass transfer properties.
- This project is focused on a key barrier (catalyst cost), and the focus on maximizing the full potential of PGMs makes it a highly promising route to having real-world impact.
- If activity in MEAs can be improved and sufficient durability is demonstrated, the project has the potential to significantly reduce costs of polymer electrolyte membrane fuel cells (PEMFCs).
- This project supports and advances progress toward the Program's critical goals regarding the cost.
- ORR catalyst activity and stability remains the highest priority of PEMFC research and development.
- The authors have well documented the potential of these materials to make the relevant targets. However, unless some evidence can be given that these catalysts can be incorporated into catalyst structures, they cannot have an impact and will not gain anything but an academic interest.

Question 5: Proposed future work

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

- The future work appears to be outstanding.
- Future work shown in slide 27 is a repeat for current activities. No specific or focused work has been addressed, even though the PI indicated some remaining challenges on Slide 27.
- International collaboration could benefit future work with alternative approaches.
- There is too much "stay the course." The researchers have an obvious, huge problem in that their fuel cell performance looks worse than a non-PGM. Most of their future work is concentrated on new and modified catalysts, optimization, scaling, and characterization. After two years of very poor MEA results, it is very clear to an outsider where the effort should be.
- It is unclear if this was effectively planned (difficult to assess with limited slides).

Project strengths:

- The strength of the project is the PI, who is knowledgeable on nanoparticle preparation. The team also has a good background on electrochemistry and a good track record on advanced catalyst development. The approach of this project toward lowering PGM catalyst loading is unique and intriguing. Having good catalytic activities for the alloy catalysts in RDE experiments is outstanding.
- The project has wonderful new catalysts with a logical rationale as to why they will be stable, active, and able to achieve low loadings. This project has great characterization methods.
- The team has shown excellent progress in the scale-up of a high-activity catalyst. The use of tools (e.g., EC-ICP-MS) to understand durability brings excellent insight.
- The approach, overall quality, quantity of results, and scale-up were all project strengths.
- The materials-by-design approach was a project strength.

Project weaknesses:

- Scale-up of the nanoframe catalysts may be more difficult than for other advanced catalysts, which hampers other evaluation/validation processes of the catalysts. There is no clear pathway to improve high-current-density MEA performance using this type of catalyst.
- The researchers need to keep their eyes on the prize; the entire point is fuel cell performance, so they need to establish why RDE results are not translating. If there is a fundamental reason why these catalysts do not work in MEAs, then the researchers are wasting a lot of time, money, and resources.
- Activity of the catalyst in MEAs is approximately 10 times below RDE activity. Apparently, there is limited work on MEA-level testing and characterization.
- MEA testing was a project weakness.

Recommendations for additions/deletions to project scope:

- No additions/deletions are needed. However, more MEA work should be planned. The notable inconsistency in MEA performance between ANL and the National Renewable Energy Laboratory evaluation suggests that the work for synthesis of low-PGM materials with alternative supports and their effects on MEA performance may be more weighted.
- The project should decrease focus on new nanocages until the activity of a nanoframe catalyst in an MEA is improved and MEA-level durability is understood.
- The project should reduce effort on new catalysts and catalyst scaling and increase effort on MEA development.
- The project should look for new collaboration at the international level.

Project #FC-141: Platinum Monolayer Electrocatalysts

Radoslav Adzic; Brookhaven National Laboratory

Brief Summary of Project:

This project aims to synthesize high-performance platinum monolayer (ML) electrocatalysts for the oxygen reduction reaction consisting of a platinum ML shell on stable, inexpensive metal, alloy, metal oxide, nitride, or carbide nanoparticle cores. Three low-platinum catalysts will be developed that will meet the U.S. Department of Energy (DOE) technical targets for 2020.

Question 1: Approach to performing the work

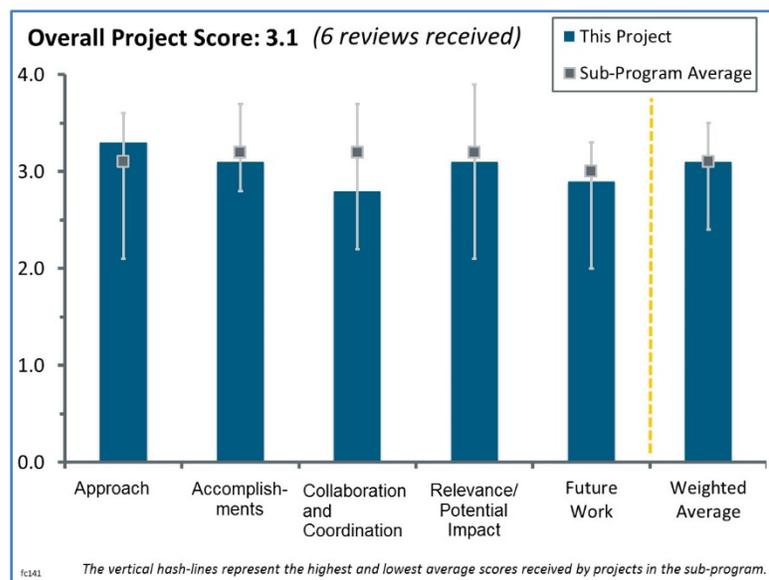
This project was rated **3.3** for its approach.

- The overall approach is good. The use of membrane electrode assembly (MEA)-level testing appears to be significantly increased over previous years, a key positive step. Development of non-platinum-group-metal (PGM) cores is key to the viability of this approach, and this activity level appears moderate.
- The selected approach addresses the main barriers related to both activity and durability of the electrocatalyst. The modification of the C support with oxides seems to be a very promising strategy.
- The approach is a good one that has a good balance of fundamental studies of catalysts manufactured and actual fuel cell performance.
- Many different technical concepts have been implemented and executed to a satisfactory standard to enable preliminary conclusions, falling just short of a ranking to indicate the most promising direction to achieve goals.
- The approach by Brookhaven National Laboratory (BNL) is to synthesize and screen Pt ML catalysts on inexpensive cores that enhance stability and activity while lowering cost. Nitriding to improve stability of non-noble metal cores, doping of Pt ML shells for improved stability and activity, and new catalyst supports are also being explored in this project. However, reported efforts spent on catalysts with expensive Pd-rich cores conflict with the defined approach.
- The team has studied a wide range of PGM and low-cost cores as support for platinum, and the researchers continue to develop highly active catalysts. However, focusing on one catalyst's material to pass all DOE-suggested accelerated stress tests on an MEA might help with understanding the limitations of this material group.

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.

- MEA testing of nitride-stabilized Pt/PdNiN/C was an important achievement of the project. It appears that additional effort is needed in electrode layer optimization to achieve the full potential of the catalysts. Characterization of the changes to the catalysts after MEA assembly and testing was very limited and would provide some critical insight into the structural and compositional changes that are limiting performance in the MEA. Progress made on the NbO₂-doped carbon black supports was promising and should be explored in more detail. Work on Au- and Pd-coated metallic W cores did not seem to directly



relate to this project. The Pd-rich Ni aerogels, while an interesting concept, do not seem to offer much cost savings because of the high cost of Pd.

- Progress is as follows:
 - MEA tests of nitride-stabilized Pt/PdNiN/C systems show lower MEA performance than expected. The principal investigators (PIs) state that the performance would be improved with a “single membrane to be used next.” However, no information about that membrane is provided in this report.
 - Stability tests of this membrane show low stability, and the PIs argue that this is due to C corrosion and a decrease of the electrochemical surface area, possibly due to Ostwald ripening. Thus, MEA implementation is still a challenge.
 - Introduction of new nitride cores containing refractory metals is shown to be a promising avenue.
 - Gold-doped Pt MLs are demonstrated to increase activity and stability. However, cost might be an issue.
 - Ir-doped cores showed very good stability during cycles. The results at 10,000 cycles are weird and not explained.
 - Doping of the C support with Nb oxides appears as the best result.
 - Synthesis of W-based cores is also shown as a very interesting approach, although some challenges point to the synthesis and electronic effects. How to overcome these challenges is not discussed in this report.
- Technical progress shows promise to meet the milestone requirements when considering the catalyst (stability) improvement in ex situ experiments; however, demonstration at the MEA level may provide practical difficulty, as experience with the aerogel support structures indicates. If, indeed, some innovative solutions cannot be validated during the project, a key should be used that correlates ex situ and in situ performance results to predict expected progress against milestones.
- The degree of MEA-level testing of catalysts has improved and is key for truly characterizing system-relevant activity and durability. Development at MEA level is needed. A large amount of work incorporates PGM-containing cores. It is unclear how much PGM is contained. More work on cores with substantially reduced PGM or PGM-free cores is needed.
- Catalysts showing high activity have been developed (e.g., the Pt monolayer on a Pd₂₀Au aerogel). However, there might have been some trouble with the high activity on an MEA.
- It appears from the fuel cell performance curves that progress toward DOE goals is not as good as one would expect from the approach in this project. For example, the ML of Pt on a non-PGM core should yield a large improvement of Pt utilization: kilowatts per gram Pt. However, the comparisons at low current densities appear to have roughly similar Pt utilizations. At higher current densities, the utilizations are better than the benchmark commercial Pt/C-based gas diffusion electrodes. The validity of the comparisons to the benchmark being attributed entirely to the catalyst, and perhaps not somewhat to the electrode structure differences, is questionable.

Question 3: Collaboration and coordination with other institutions

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

- The PI collaborates with Los Alamos National Laboratory (LANL) researchers and with a number of industries and academic institutions. However, it is not clear whether Dr. Zelenay, the co-PI, reports separately. There is no discussion regarding his participation and no publications with his name.
- There is a clearly engaged collaboration on material synthesis and characterization, but cell testing and interpretation could receive more emphasis to validate results against milestones by solving stability issues.
- The project is able to leverage a good cross-functional team. It is unclear whether the licensing agreements for four patents are under this project or under some previously funded DOE projects.
- The degree of contributions from collaborators is unclear. The collaboration team appears well suited for work.
- Collaboration with LANL was mentioned, but it was not clear what parts were from LANL.
- BNL could do much better in highlighting the role and contribution of their collaborators. MEA testing was performed in this year’s project but, little was said about the efforts of collaborators on this front.

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

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

- It is good to see innovative catalyst materials and support structures being realized and prepared in “scale-up” 0.5 g batches. The overall impact could be further increased if such materials could be evaluated by relevant stakeholders apart from direct project partners to confirm findings and gain involvement.
- The project is perfectly aligned with the Hydrogen and Fuel Cells Program goals and objectives.
- This project has a high potential to improve power density and durability.
- The project is directly addressing key activity and durability barriers for polymer electrolyte membrane fuel cells. The potential impact is unclear. The cost of catalysts with multiple interlayers deposited on PGM-containing cores is a concern; multiple wet-chemistry steps with yield losses are likely.
- This project has potential to significantly increase Pt utilization in fuel cells, but the fuel cell test results do not appear to show this.
- The preliminary results of the MEA testing were rather disappointing. It seems much more effort is needed here to understand the factors limiting the realization of the high catalyst activity in an MEA.

Question 5: Proposed future work

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

- The proposed plan follows the approaches discussed in this report. Most ideas look promising and feasible. Possibly the largest challenges are related to the synthesis. It would be good to factor the cost into the potential solutions being tested.
- Many different pathways that could work are suggested, but a fast method for screening performance is recommended (as mentioned in the 2016 presentation) to speed up iterations and recognize (lack of) progress faster.
- Future work should include a scale-up of some promising materials to the 10 to 100 g scale. Then, it is recommended that the project share these materials with commercial MEA manufacturers or Fuel Cell Consortium for Performance and Durability (FC-PAD) laboratories that can make MEAs with identical techniques to the baseline Pt/C catalyst materials. FC-PAD should be more integrated into this effort.
- Future work should largely focus on bringing PGM-free cores to technological maturity and fully evaluating MEA-level tests for activity and durability. Understanding of Cu contamination (slide 7) is needed.
- Additional advanced physical characterization of the MEA-tested catalysts is sorely needed and should be an emphasis of the future work.
- Proposed future work seems quite ambitious in developing various catalysts.

Project strengths:

- The great experience and knowledge of the PI and his team are project strengths. Since the PI is retiring, it is expected that the team will continue with his line of work. The great instrumentation existent at the national laboratories is also a strength. The collaborations with industrial partners are a project strength.
- This team has a high level of skill and execution regarding materials synthesis and characterization. BNL has great experience and knowledge on the subject.
- The project is developing catalysts that, conceptually, could greatly reduce the PGM content of fuel cell electrodes.
- The project has a good approach, and good techniques for understanding analytical and fundamental catalytic activity have been developed.
- The team is highly competent in imagining and synthesizing new catalyst concepts with high performance in rotating disk electrodes.
- This project has an excellent team and very innovative synthesis.

Project weaknesses:

- The project seems to be moving in too many different directions. The continued work on Pd-rich cores seems to contradict the project objective to utilize inexpensive core materials.
- While improved over the past, additional work on MEA-level performance and durability is needed, especially with PGM-free cores.
- Poor collaboration leaves the results and progress limited and in question.
- There are too many catalyst options, and none have shown better hydrogen–air performance at MEA level.
- Individual approaches appear disjointed in this presentation.

Recommendations for additions/deletions to project scope:

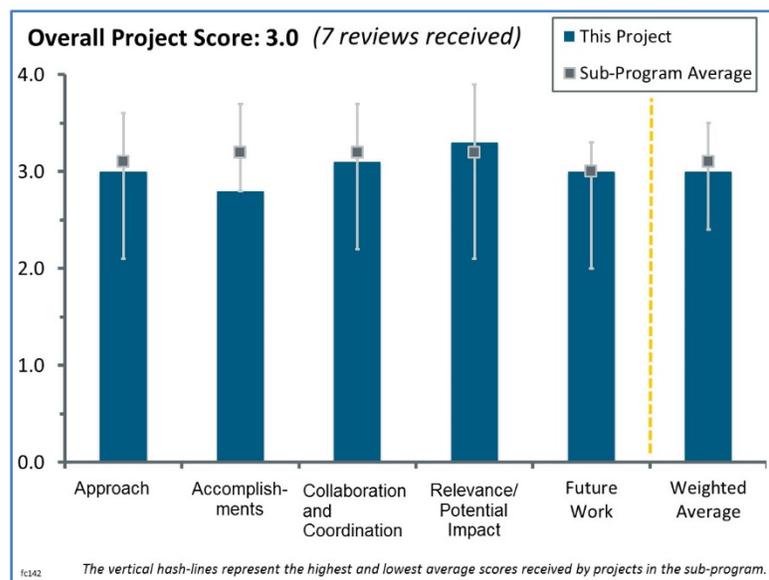
- Further integration of theoretical studies (possibly with existing or new collaborations) would be very helpful to understand the successes and improve the designs.
- Performance milestones appear to turn into stability requirements in attempts to maintain high activity of the MLs, and as a suggestion, the milestones should be defined as such.
- The cost of complex/multilayered structures needs to be understood. MEA-level durability testing is needed.
- A scale-up of the material synthesis and teaming with FC-PAD is recommended.
- MEA testing with 5–10 g scale batch catalysts is recommended.

Project #FC-142: Extended-Surface Electrocatalyst Development

Bryan Pivovar; National Renewable Energy Laboratory

Brief Summary of Project:

Platinum catalysis remains a primary limitation for fuel cell commercialization. This project is developing durable, high-mass-activity, extended-surface platinum-based catalysts for decreased fuel cell cost, improved performance, and increased durability. Researchers are focusing on novel extended thin-film electrocatalyst structures (ETFECs), a particularly promising approach. Parallel efforts include novel extended nanotemplates; atomic layer deposition (ALD) synthesis of platinum–nickel nanowires; and membrane electrode assembly (MEA) optimization and testing including multiple architectures, compositions, and operating conditions.



Question 1: Approach to performing the work

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

- The project team has accomplished the proposed work with a well-defined, systematic approach assessing the impact of synthesis conditions on material properties and their impact on device performance. The feasibility of the project is not in question, as many of the early targets related to material development and synthesis have been met. However, the performance of the extended catalysts is still significantly lower than the target, and significant effort will be needed to bring the performance up.
- The approach of synthesis of ETFECs for meeting the U.S. Department of Energy's 2020 objectives of improved cost, performance, and durability is very relevant. Low-platinum-group-metal (PGM) loading in ETFECs has the potential to overcome the cost barrier of fuel cell implementation. The team has demonstrated that the use of nano-structured Ni template as a platinum deposition substrate has the potential to meet the low-PGM loading objectives.
- Using Pt-Ni nanowires with ALD Pt coatings is a novel approach for synthesizing fuel cell oxygen reduction reaction (ORR) catalysts. The project researchers correctly recognized that spontaneous galvanic displacement would not work, so they moved in a different direction, with ALD coating of nanowires. The ALD results look promising.
- The overall project approach is good. The utilization of PGM-free nanowires as substrates has great potential for the development of active and stable thin-film electrocatalysts.
- The nanowire approach is a good one for developing highly active platinum sites. However, the basic flaw with this approach is that there is either too much transition metal or the electrochemical surface area (ECSA) is low. It looks like the project is trying to optimize between these two tough trade-offs.
- It is expected the project will investigate the mechanism of each method (catalyst fabrication) and how it works, or does not work, rather than try to fill out the development gaps (gap to target value). For example, if hydrogen annealing shows better specific activity, it is expected to investigate why.
- This project continues to work on ALD synthesis of Pt-Ni nanowires to achieve the project milestones.

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.

- Significant progress has been made in the area of materials synthesis. Cost-effective scale-up of both the sacrificial nanowire growth and ALD deposition of Pt has been demonstrated. Sufficient operation has been demonstrated at low ionomer contents. The close match in performance metrics between rotating disk electrodes (RDEs) and MEAs is very promising, as this is not often the case with other types of electrocatalyst form factors. The dramatic improvement in activity with acid leaching of the core Ni metal is a significant accomplishment. However, developing a protocol for this that is amenable to industrial manufacturing scale-up is complicated. The mechanical stability of the catalyst layer against compression resulting in transport resistances must be addressed. Long-term durability of the catalyst must also be addressed. Strategies for improving the ECSA must be developed.
- Progress is as follows:
 - The team's decision to use a commercial source of Ni nanowire for making Pt-Ni nanowires helped the team accelerate ALD activities. Study and down-selection of Type 4 Ni nanowire among four different types of nanowires supplied by the vendor is good progress. The team has also optimized the oxygen treatment and hydrogen annealing steps for ALD-deposited Pt-Ni nanowires.
 - From the presentation, it seems that the team has done a significant amount of work with Pt-Ni made using spontaneous galvanic displacement (SGD) and compared the product with Pt-Ni made using ALD. As fabrication of Pt-Ni, SGD is not economically viable, and it is not clear why the team is devoting so much effort to it. Understandably, Pt-Ni (SGD) shows better oxygen transport resistance and better low relative humidity performance, most likely a result of the absence of Ni, as compared to the Pt-Ni (ALD) MEA. The focus of the work is Pt-Ni (ALD), and the overall cell performance of MEAs containing Pt-Ni (SGD) and Pt-Ni (ALD) are almost identical. Therefore, the team needs to focus more on the method of Ni removal from Pt-Ni (ALD) MEAs.
 - From slide 19, it seems that low Nafion® content (2%) is providing a higher ECSA for ALD-synthesized Pt-Ni MEA. This shows that the ALD-synthesized Pt-Ni is doable if the team finds out an efficient method of Ni removal.
- There is good progress toward identifying and apparently resolving Ni nanowire supply issues. There is good progress on ALD development. Reasonably high activities have been demonstrated (but perhaps not as high as previous galvanic displacement). While some progress on de-alloying has been made, it is unclear whether this has translated into substantial improvement in MEA performance under hydrogen-air; all data presented were under oxygen. This is the key first technical challenge this project needs to address, with MEA-level durability next. It is unclear whether the Ni dissolution issue can be sufficiently resolved. MEA activity is relatively poorer than RDE. Little evident work has been conducted toward understanding the gap.
- Accomplishments and progress for the past year has been adequate. Hydrogen annealing of ALD-coated nanowires appears to be an effective approach to achieving high ECSA and specific activity. The performance of catalyst nanowires prepared by SGD was not particularly impressive. Some contradictory results were present.
- Hydrogen annealing and acid leaching seem to have helped these catalyst types achieve satisfactory polarization curves. Bigger batch sizes is also a good accomplishment. Low ECSA (<40 at beginning of life) is causing some concerns.
- A significant amount of research such as batch scale-up, hydrogen annealing, and MEA studies has been carried out in the second year.
- Hydrogen-annealed ALD shows higher activity, and an explanation of how it works was expected. Durability and stability of acid leaching to remove Ni core nanowire to make hollow tubes is still in question.

Question 3: Collaboration and coordination with other institutions

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

- The collaborations and coordination with other institutions such as the University of Colorado Boulder, the Colorado School of Mines, and ALD Nanosolutions seem to be very effective. The synthesized material from ALD Nanosolutions is post-processing and characterized by other institutions, which is going on well. There is no necessity to further coordinate with the University of Delaware, as that collaboration has ended. Overall, the task coordination and data flow between the collaborators seem to be moving well.
- Collaborative research tasks are well integrated into the overall project work plan.
- The project makes good use of national laboratory and university partners and resources.
- The project has established good collaboration with three universities and an industry.
- Collaboration exists; however, it is hard to see how much work and catalyst fabrication approaches are discussed in the team.
- The principal investigator (PI) should be conducting discussions with catalyst suppliers (e.g., Johnson Matthey or N.E. ChemCat Corporation) to figure out whether this solution is truly scalable.

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

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

- The project is very relevant to DOE's Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan. The project goals are aligned with DOE's 2020 target of achieving 0.125 g/kW combined PGM content in both electrodes. The success of this project will have great impact in bringing down the overall MEA and hence fuel cell stack cost, which will increase its commercial feasibility.
- Moving away from traditional carbon-supported materials can lead to dramatic improvements in the operational lifetime of polymer electrolyte membrane fuel cells (PEMFCs). Extended surfaces can also be tuned to yield improved activity of nanoparticle-based catalysts. Almost as important as the material development is the ability of the team to nearly match MEA performance metrics with RDE values. This is notoriously difficult to do, but it is an important step in the development of this and other next-generation catalytic materials.
- There is a need for high-performance (high-activity) and durable ORR catalysts for fuel cells.
- The project objectives are relevant to DOE's catalyst activity and cost targets.
- There is good relevance to the DOE target, mass activity, and catalyst durability.
- The project addresses key commercialization barriers for PEMFCs. The ultimate potential impact is unclear unless Ni dissolution issues can be demonstrably resolved.
- RDE-level activities are now being translated and matched at MEA level. While this is promising, there is not a single hydrogen-air pol curve. Therefore, it is not clear how this can be translated to the Fuel Cell Technologies Office Program goals of < 0.125 g Pt/kW.

Question 5: Proposed future work

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

- The proposed future work, namely ALD scale-up to prepare 10 g batches and fuel cell performance improvement, is very relevant to achieving the project goals.
- The proposed future work is well-thought-out and is aligned with the project goal. The team has appropriately focused on optimization of hydrogen annealing and acid leaching, which is needed for scale-up work. Co-deposition of Pt with Ni/Co to further enhance ECSA and specific activity may be a good avenue to explore, given the data from last year that showed some promise as a Pt-Co catalyst. The proposed "nanotemplate" work cited on slide 22 indicates that the critical-to-quality (CTQ) characteristics of Type 4 nanotemplates from the supplier do not meet the commercial specifications and need to be validated by the team. If so, the team needs to be upfront with the supplier and should try to "fix" the

nanotemplate specification as soon as possible. Any uncertainty in nanotemplate CTQ may delay the upstream activities for the team and delay the progress of the project.

- Reasonable future works are defined to the project goal. It will be important to identify critical characteristics for catalyst layer structure optimization for ALD catalysts for the fuel cell test rather than just engineering optimized catalyst layer fabrication.
- The mass activity of the extended materials is still considerably below the target. There does not appear to be a clear plan to address this other than further optimization of synthesis and leaching conditions. The project team should consider looking into the possibility of increasing porosity to enhance the ECSA and mass activity. The mechanical integrity of the catalyst layer, especially for the hollow structures, needs to be addressed.
- Project work should focus on the development and demonstration of catalysts in the MEA toward some reasonable measure of DOE activity and hydrogen–air performance targets. It is possible/likely that a large effort will be needed toward the development of sufficient de-alloying methods or a new, stable nanowire core. ALD scale-up is premature in light of technical issues.
- The team should be accelerating this work and conducting a factorial design of experiments to include catalyst preparation (e.g., annealing H₂ vs CO, acid leaching, and a few other factors) and testing these catalysts at an MEA level to understand the impact of these factors on activity, hydrogen–air performance, and accelerated stress test durability. The amount of future work proposed does not seem proportional to the funding level.
- Future work will focus on improving the mass activity from the current level of 240 mA/mg to 440 mA/mg at 0.9 V. The required increase in activity, which is to be achieved in just a few months (by 9/30/2017), appears to be a sizeable task. Future work should also focus on catalyst durability; the PI did not address the durability milestone (<40% loss in mass activity after 30,000 voltage cycles) in his listing of future work tasks.

Project strengths:

- The following are project strengths:
 - The scaled-up process for making ALD-coated nanowires is a project strength.
 - This project demonstrated the ability to run with low ionomer content in the catalyst layer.
 - MEA performance matches RDE performance.
 - There was high activity without carbon in the catalyst layer.
 - Leached material is active and minimizes Ni contamination of the MEA, or at least significantly decreases it from the previous case.
 - Technoeconomic analysis supports the viability of the ALD process at a large scale.
- Project strengths including the following:
 - There is a need for new, high-performance ORR fuel cell catalysts, and the PI's approach is novel and potentially significant.
 - The experiments have been carefully carried out, and progress has been made by switching from an SGD to an ALD approach.
 - The project team members are well qualified, and the work shows a nice level of collaboration.
- The team is well composed of qualified scientists and a PI who are capable of conducting the research and development work. ALD Nanosolutions has technology required to use different deposition substrates to make Pt-M-type nanowire and hence create a non-morphology of Pt catalyst that possesses high ECSA and specific activity.
- The project conceptually has the capability of generating substantially active catalysts. Very high mass activities in RDE have been demonstrated previously.
- The project has so far showed an ALD Pt-Ni nanowire catalyst with significantly high specific activity.
- The project has various methods to fabricate the low-loaded catalysts to be tested.
- This is a good fundamental concept and a very innovative team.

Project weaknesses:

- The team needs to come up with an effective post-processing step, especially the acid-leaching step to make the MEA fabrication process viable. Any leftover of Ni in the electrode will diminish the cell

performance and affect its durability. The team also needs to ensure a supply of nanotemplates with consistent quality for this project. Any variability in nanotemplate quality may slow down the progress of the project.

- The following are weaknesses: durability is still a question, the pathway to higher mass activity is still unclear, mechanical stability of the catalyst layer needs to be addressed, and process development for the scale-up of the leaching process is needed. The project could consult 3M for its expertise.
- The following are project weaknesses: (1) it is questionable whether the project team will achieve its go/no-go decision metric of 440 mA/mg at 0.9 V by 9/30/2017, and (2) in addition to the mass activity metric, it is not clear whether the catalyst will exhibit the required durability after a metal dissolution accelerated stress test.
- The project does not have capable analytical methods to investigate how a fabricated catalyst works or does not work. For example, hydrogen annealing on an ALD catalyst shows higher specific activity, but not enough investigation was pursued to find how it works.
- Much too little progress toward resolving Ni dissolution is evident.
- The mass activity target is yet to be achieved.
- There is a lack of focus and traditional catalyst supplier collaboration.

Recommendations for additions/deletions to project scope:

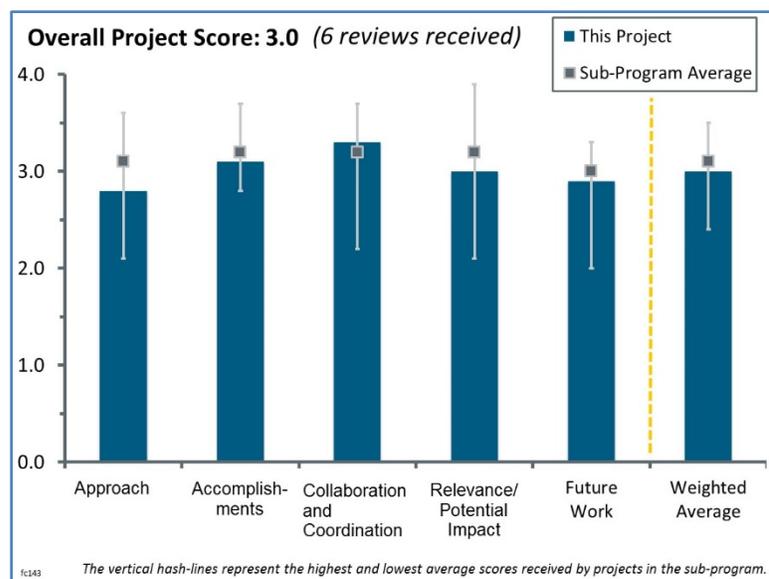
- The project should focus on approaches to fill the knowledge gap rather than the developmental gap or performance gap to the target. The project is expected to investigate how the hydrogen annealing can work for an ALD catalyst as a knowledge base that may be applicable for other materials.
- The project team should focus on ALD-coated nanowires. The mass activity go/no-go decision point (440 mA/mg) may need to be pushed back to a later point in time. Durability is an important issue; residual Ni in the nanowires may be an issue.
- For durability testing, the project should (1) assess mechanical properties of formed catalyst layers for solid and hollow nanofiber electrodes and (2) consider creating nanoporosity to improve ECSA and, consequently, mass activity.
- The project should predominantly focus on working toward Ni dissolution minimization to levels that will allow for acceptable MEA-level performance.
- A factorial design of experiments on how to optimize the catalyst is recommended.

Project #FC-143: Highly Active, Durable, and Ultra-Low-Platinum-Group-Metal Nanostructured Thin-Film Oxygen Reduction Reaction Catalysts and Supports

Andrew Steinbach; 3M

Brief Summary of Project:

This project is developing thin film oxygen reduction reaction electrocatalysts on nanostructured thin-film (NSTF) supports developed by 3M. The aim is to exceed all U.S. Department of Energy (DOE) 2020 cost, performance, and durability targets through developing two different NSTF-based structures, nanoporous thin film (NPTF) and ultrathin film (UTF) catalysts. The electrocatalysts will be compatible with scalable, low-cost fabrication processes. The project will integrate the catalysts into advanced electrodes and membrane electrode assemblies (MEAs) that address traditional NSTF challenges, which include operational robustness, contaminant sensitivity, and break-in conditioning.



Question 1: Approach to performing the work

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

- The approach taken addresses performance barriers and mitigates risk by tackling two different NSTF-based structures, an NPTF and a UTF approach, both showing potential to meet DOE and project targets. One of the structures will be down-selected for further work toward completing the project. Both structures can be combined into the dispersed NSTF-structured MEAs developed under a different project and thus address the flooding issue with the NSTF structure.
- The approach to performing the work involves adapting the long-standing 3M NSTF technology to produce two relatively new variations: UTF and NPTF catalysts. The materials are being tailored through investigation of different compositions and treatment methods, leading to different structures, and a variety of characterization techniques and computational methods are being used to interpret results. Overall, this seems like a reasonable approach to developing a better catalyst. However, the main issue with NSTF continues to be its integration into effective electrodes. 3M has made progress in this area recently, but until the problem is fully solved, the approach of investing in further development of NSTF catalysis is highly risky.
- The project has good focus on key targets including specific platinum-group-metal (PGM) activity and durability. The dual approaches (UTF and NPTF) with multiple alloys provide good options, but NPTF is clearly superior at this point. However, if loss of efficiency at rated power is important, as is stated repeatedly, then this should be added to the targets on slide 7, probably to replace “Loss in performance at 1.5 A/cm².” Also, better integration of modeling and high-temperature (HT) work with main catalyst development is expected in the future.
- The project keeps two approaches in terms of fabrication processes, NPTF and UTF. There is no clear explanation about the relationship between the two approaches. They seem to be independent. If so, it is unclear why two approaches are necessary. If they are related, it is unclear how outcomes from each process can be used.
- The work involves an extension and continuation of previous NSTF work funded by DOE. Although the surface area is increased by one of two methods, it is unlikely to be sufficient to result in improvements or

resolution of the fundamental issues with the NSTF that have led to its failure to be applied to automotive stacks. The fundamental problem is that the catalyst layer is thin and the surface area is low. The combination of the two issues leads to high mass transport electrodes and, consequently, poor high-current-density performance under real automotive operating conditions as well as an increase of Rlocal (impact/loss of local transport retention after electrocatalyst accelerated stress testing) at low electrochemical areas. The catalyst has high specific activity, but the catalyst layer on the cathode will not meet real targets under real operating conditions, as attested by most auto companies in the past.

- The project approach is sound, with the development/examination of two types of thin-film cathode electrocatalysts, NPTFs and UTFs. The project includes both experimental and theoretical tasks. The connection and usefulness of the theory in directing experimental catalyst development was not well established.

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.

- Over the course of a year, the duration of the project, progress has been made toward reaching DOE targets, and feasibility has been shown for meeting more aggressive 3M project targets using both the UTF and NPTF approaches. Plenty of data are presented, which speaks to the breadth of work being undertaken. Multiple alloys have been tested (some not disclosed), and in a unique application, Ir is used to suppress Pt dissolution of alloy structures.
- 3M has done an admirable job of testing a large number of candidate materials in this project. Progress has been made on improving the activity and the MEA performance of NSTF. It looks like 3M is closing in on some compositions and structures that may be able to simultaneously meet mass activity, power density, and durability targets. Progress has been made on high-current performance, but much of the MEA data fails to show the low-current region. When the low-current region is shown, performance is sometimes quite low, with extremely low open circuit voltages of less than 0.9 V. On slide 13, a value of “NA” is shown for performance at 0.8 V. Based on the shape of a typical polarization curve, it appears that this catalyst may have had near-zero performance at this voltage. While the project does a decent job of addressing high-current H₂/air performance and low-current H₂/O₂ mass activity, more attention to low-current H₂/air performance seems warranted. Data shown for UTF catalysts looked promising in terms of mass activity, but the high-current performance suffered from the extremely low cathode loadings used. Based on the presentation, some technical challenges remain related to limited surface area supports limiting the cathode loading, but the nature of this challenge remains unclear.
- The project has made good progress in its first year. The effect of Ir on durability represents an important contribution, multiple alloys being assessed, HT characterization being established, and computational approaches producing results. It would have been good to see more discussion on HT fabrication and its integration into the main catalyst development process. It is unclear what the process is: deposition, de-alloying, or annealing. It is unclear how repeatable the process is and how it compares to batch fabrication. For this project to cover the planned broad design space, harnessing of the HT approach needs to happen very quickly.
- There was significant progress on the project over the past year. Both NPTF and UTF cathode catalysts appear to work well at ultra-low-Pt loadings. Pt alloys, PtNi and PtNiIr, appear to work best. Many project targets have already been met. Although the mass activity of NPTF and UTF cathodes was very high, Pt alloy loading was insufficient to reach a power density of 1 W/cm². Also, it is unclear how the modeling results were used to eliminate unnecessary experiments and streamline the project work plan.
- NPTF shows good progress and is close to the 2020 targets for mass activity (low current) and rated power (high current). There is still a question about robustness, particularly sensitivity with relative humidity and hydration of the catalyst layer. That is a generic issue of an ionomer-free catalyst layer.
- Meeting the target of 440 mA/mg Pt is not sufficient and has already been met by most previous NSTF catalysts. The specific activity of commercial Pt/C is about 300 mA/mg Pt. The initial activity of PtCo/C catalysts varies from 400–800 mA/mg Pt at beginning of life, as seen in the literature and presentations from previous years. This should be clearly stated in the presentation for an unbiased look at the current state-of-the-art materials. Density functional theory and other modeling have never succeeded in the past in

predicting an ideal composition for a high-activity catalyst. It is not clear that work has value except to show that modeling has been combined with experimental efforts.

Question 3: Collaboration and coordination with other institutions

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

- Most of the key work is being done by 3M, but the principal investigator (PI) has assembled a good support team, with the characterization capabilities provided by Argonne National Laboratory (ANL) and Oak Ridge National Laboratory (ORNL) being particularly valuable.
- There is more than sufficient collaboration on modeling and other areas.
- Plenty of relevant work is being performed at multiple subcontracted institutions and was well represented in the slides. The interaction and leadership between the lead and subs appears to be well executed.
- 3M appears to be doing all of the main catalyst development in-house, with only ex situ characterization by ANL and ORNL and modeling by Johns Hopkins University (JHU) and Purdue University. It would help to clearly demonstrate the impact of external collaboration on progress in budget period 2. For example, on slide 14, it appears from the model that current densities are predicted to be significantly higher for UTF skin thicknesses of one to two monolayers and drop off substantially at three monolayers. It is strongly recommended that this prediction be tested experimentally.
- The collaborative aspects of the project were not explained well. There was some discussion of modeling work. The importance/need for some collaborations was not fully justified, especially for tasks during the upcoming year. For example, it is unclear how the HT development at JHU was coupled with a similar task at ANL.

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

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

- The project directly addresses the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan targets and tries to exceed them. The project has shown progress in both approaches taken to achieve the DOE goals, while only one will be selected to meet the final project goals.
- This project directly addresses and supports the Hydrogen and Fuel Cells Program's (the Program's) goals and objectives.
- Developing improved catalysts is one of the most relevant things that a project can do for the Program. However, the relevance of this project is limited by the robustness issues that continue to face NSTF. While the use of electrode interlayers has helped with this issue, the durability of these interlayers and their ability to provide robust operation throughout the life of the stack still need to be demonstrated.
- As a catalyst development project, this approach seems very effective and can be envisioned to translate well to other architectures. However, in the context of NSTF MEA development, it is not clear whether this project addresses the key issues facing this technology (e.g., membrane degradation).
- Development of a lower-PGM catalyst is effective to the cost reduction if it can improve mass activity (low current) and area-specific power density (high current).
- The goals are relevant but are unlikely to be achieved by these modified electrodes.

Question 5: Proposed future work

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

- The project has been planned for and is being executed effectively. Go/no-go milestones and key decision points are clearly laid out and presented and appear to lead to logical progression toward the final project targets.
- The proposed future work involves further development of both UTF and NPTF catalysts, which is reasonable and appropriate.

- Overall, future work is relevant to the project goals. The approach for the operational robustness (sensitivity with relative humidity and hydration of catalyst layer) is expected.
- The future plans flow logically from recent progress. However, it is not clear how HT development will be integrated with NPTF and UTF projects. The mention of other high-area supports, as an alternative for UTF, was surprising and would benefit from more detail.
- A complete change in direction that has been requested by reviewers for many years is necessary. The catalyst layer needs to be modified so that its mass transport is improved—it has got to be thicker, be porous, and have characteristics that are comparable to traditional Pt/C electrodes. Continuing in the current direction will not lead to achievement of the targets in a similar manner to traditional NSTF catalysts that have been studied for almost 15 years and are still not implemented in real automotive stacks successfully. At times, the investigators use English units such as “psig,” terms such as “AtmA” (atmosphere units of air pressure), and other such units that are jarring to the eye and outdated. Pressure should be presented with units of kPa or MPa. SI units are always absolute units by definition. The authors should not fill the slides with excessive data that muddles things up.
- Future work must focus on down-selecting either an NPTF or UTF cathode morphology. There is no mention of down-selecting on the “Future Work” slide. The criteria for making the selection do not appear to be well defined. It is not clear why further modeling studies are needed. The modeling work should help direct future experiments. A major effort must be directed to increasing the catalyst loading to achieve a power density of 1 W/cm². The issue of cathode flooding and the need to move away from ultra-thin cathode structures was not properly addressed. Many of the project’s cathode performance targets have been met, but future work should be better focused.

Project strengths:

- The team has demonstrated good early progress in the development of catalysts, with large design space yet to be explored. Modeling and characterization infrastructure appear to be in place. 3M has chosen good collaborators that can provide scientific insight into structure–function relationships.
- The project team has made two cathode layers with excellent mass activity and durability. Many project goals have already been met. Experimental data collection has been extensive. The project is on target to meet all project milestones and go/no-go decisions.
- The team is doing an excellent job of leaving no stone unturned in the search for compositions and structures that could further improve the activity and MEA performance of NSTF. Several candidate materials and structures are meeting or approaching multiple, relevant DOE targets.
- This project utilizes two unique approaches to decreasing PGM loading while increasing durability through alloying. Both approaches seem to work well, while Ir incorporation into the catalyst structure leads to stabilization.
- The project is yielding a high-performance catalyst with NSTF approaches.
- The highly active catalyst is a project strength.

Project weaknesses:

- It is not yet clear how high throughput will be harnessed to address the large design space. Regarding slide 9, hiding the iridium content is not appropriate for precompetitive, federally funded research. The same could be said for the identities of the various alloys. This is especially true if no intellectual property is claimed (slide 31). This project, while valuable, does not appear to directly address major issues with NSTF technology. So far, there have been relatively few scientific contributions via papers, presentations, and patents.
- (1) There must be a down-selection process during the next year, after which the project team will focus on only NPTF or UTF. (2) The need for modeling work next year has not been established. (3) The issue of cathode flooding and the use of very thin cathode layers were not adequately addressed.
- No matter how successful the team is at developing improved NSTF catalysts, operational robustness issues will continue to undermine the relevance of this approach to oxygen reduction reaction catalysis.
- It is unclear how segmented cells, strain measurements, and various modeling efforts help in resolving the ultimate barriers within this system to achieve the Program targets.

- The project has a weakness in its investigation for characteristics of an ionomer-free catalyst layer, particularly operational robustness.
- This project has a poor catalyst layer.

Recommendations for additions/deletions to project scope:

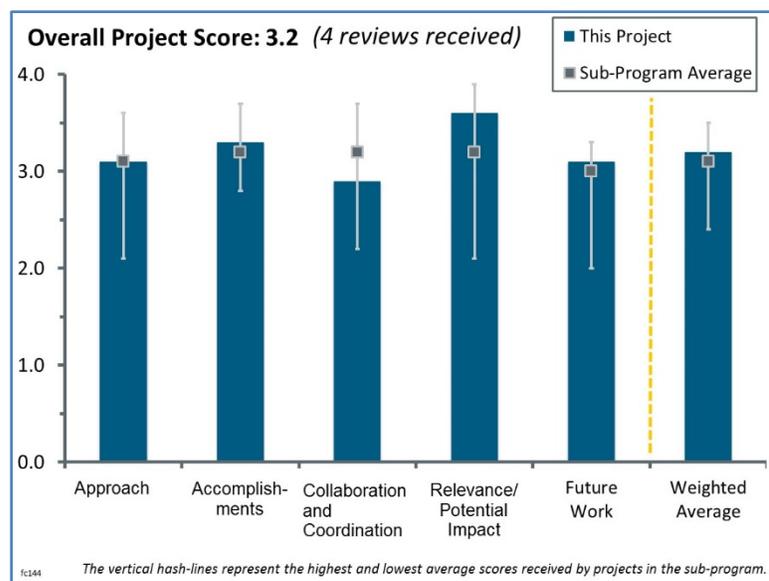
- The PI should down-select one of the two cathode morphologies. Modeling and experimental work should then be focused on this one structure. There should be better justification and integration of the experiments and theoretical work. The modeling work should help drive/direct experiments. Also, flooding issues need to be addressed.
- Very often the discussion revolves around small differences. Measurement and repeatability studies are encouraged to ensure that small differences are not significant. (A good example is in slide 8, on the bottom right.) A direct comparison with powdered NSTF and carbon-supported alloy catalysts would be valuable.
- While both UTF and NPTF approaches have merits, the project scope should be limited to NPTF unless methods of effectively incorporating UTF with good performance at reasonable loadings can be demonstrated.
- The project is yielding a high-performance catalyst with NSTF approaches. The project is expected to investigate characteristics of an ionomer-free catalyst layer, particularly operational robustness.
- Both approaches should be tested in dispersed electrodes prior to down-selecting to ensure that both can be incorporated into more conventional thick catalyst films.
- The project should focus on the catalyst layer and limit current studies that address the high-current-density performance.

Project #FC-144: Highly Accessible Catalysts for Durable High-Power Performance

Anu Kongkanand; General Motors

Brief Summary of Project:

This project aims to reduce overall stack cost by improving high-current-density performance in hydrogen/air fuel cells that meet U.S. Department of Energy (DOE) heat rejection and Pt-loading targets. Investigators will maintain high kinetic mass activities and mitigate catalyst degradation by using supports with more corrosion resistance than the current high-surface-area carbon (HSAC). The project takes a four-pronged approach: (1) improve oxygen transport with new carbon support, (2) reduce electrolyte–Pt interaction, (3) enhance dispersion and stability of Pt–Co particles, and (4) improve understanding and control of leached Co^{2+} .



Question 1: Approach to performing the work

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

- The project's approach is focused on overcoming barriers for polymer electrolyte membrane fuel cell (PEMFC) performance at high current densities. The project is well designed and addresses multiple causes of performance degradation at high current densities.
- The approach is well structured and stems from recent literature that clearly indicates that there are non-Fickian mass transport losses that present themselves as unexpected voltage losses at low-Pt loading. Three of the four different approaches correspond to the hypotheses that could be given for why these losses exist: (1) Pt buried in the primary carbon particle, (2) poor Pt–ionomer interactions, and (3) influence of leached Co or base metals from the alloy catalyst. Further details regarding the ionic liquid strategy would be useful. It would be good to understand how ionic liquids are selected and how they are incorporated into the electrode. The approach to situate Pt on the exterior of carbon primary particles has already reaped benefits that speak for themselves. However, it may have been good to report on the stability of PtCo/HSAC-e or PtCo/HSAC-f before declaring victory.
- The project has four distinct approaches that seem mostly unassociated with the others. This project was awarded under a catalyst section of a funding opportunity announcement. It is unclear why the project has such a large portion of electrode development. Apparently, multiple reviewers commented on this last year. The response seems overly simplistic and does not address the topic in which the project was awarded. Only one portion of the project seems to be relevant to a catalyst project.
- The project's approach is designed to address the relevant barriers. It is a little difficult to follow the approach because so much is going on in parallel on this project. There is concern as to whether Co dissolution can really be addressed with the proposed approach.

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.

- Significant progress has been made toward understanding the role of support porosity on Pt utilization in the oxygen reduction reaction.
 - The method development for evaluation of internal and external Pt nanoparticles on the support represents a significant accomplishment.
 - The DOE 2020 high-power target (8 kW/g_{PGM}) is exceeded using PtCo on a new HSAC.
 - On the other hand, the DOE 2020 target was exceeded on the HSAC that failed the accelerated durability test.
- A dramatic increase in specific power (kW/g) was shown due to reduction of interior Pt on an HSAC and through the use of PtCo. This is not just an empirical observation but is also justified by theories that suggest local mass transport resistances decrease when oxygen does not have access to Pt within a carbon particle. However, the PtCo is now in greater proximity to the ionomer, which could have implications for durability.
 - There was a good result on the finding that the carbon type has little influence on Pt being deposited in the membrane. However, it may be difficult to get from these results to a statement about the lesser role of coalescence, since coalescence would not be expected to contribute to a Pt-in-the-membrane band.
 - The project should quantify the ionic liquids in electrodes, as well as to see if ionic liquids are being lost from fuel cell operation.
 - New PtCo/C catalysts should also be tested for low-temperature performance.
 - The development of the CO stripping technique to quantify surface Pt may turn out to be one of the best outcomes from this project. This could be useful for many developers.
- The power looks great for Pt/Co/HSAC; however, simultaneously achieving high power and durability with the low-Pt catalyst needs to be demonstrated. There are many other interesting results that improve understanding of support effects, Pt–electrolyte interactions, and degradation mechanisms. Since so much is going on in parallel, it is difficult to gauge the extent of the progress toward the final objectives. Showing that the knowledge/results from parallel approaches can be utilized, or combined, to achieve the objectives should be presented in future work.
- The project seems to be using oxygen-limiting currents and mass activity as a successful predictor of carbon support development.
 - There do not seem to be any new catalyst synthesis results.
 - It is curious that the principal investigator (PI) does not seem to support the catalyst support accelerated stress test (AST), as this was developed by the U.S. DRIVE Fuel Cell Technology Team, to which the project team's organization is a primary contributing member. As this AST was developed with their companies' input, it seems that discounting the results from this test is inconsistent with their own guidance to other developers.
 - The researchers are showing that ionic liquids can promote rotating disc electrode (RDE) oxygen reduction reaction mass activity. (This has been seen before, specifically in Argonne National Laboratory's [ANL's] nanoframe catalyst project). However, this does not transfer to membrane electrode activity (MEA). As the PI apparently does not believe RDE is a valuable tool to predict performance, it is unclear why it is used in this case to suggest increased activity.
 - There is a question about the stability of the ionic liquids and whether they will actually be chemically stable in the long term, whether they will remain within the catalyst layer or simply migrate out and/or react with the cations and anions that are present in the MEA.
 - It is odd to claim a new capability of preparing an MEA from milligrams of catalysts; others have been doing that for more than 20 years.

Question 3: Collaboration and coordination with other institutions

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

- General Motors (GM) is a very strong developer, so their need for collaboration is less than for other primary investigators. This is somewhat reflected in how the project seems to have gone very well despite some subcontracts not having been signed yet. Other collaborators contribute a very specific input (e.g., a characterization technique), but little else.
 - Although the major findings so far appear to be from GM, there are plans to significantly involve most partners. 3M Company and National Renewable Energy Laboratory (NREL) will be

- involved in ionomer screening. NREL, ANL, and Carnegie Mellon University will be involved in understanding cobalt dissolution.
- It would be good to see more of the decision-making from Drexel University with regard to ionic liquid selection and how the ionic liquid is incorporated into the composite catalyst.
- Cornell University collaboration is spread between both characterization and materials development, which creates an even stronger collaboration.
- Excellent collaboration is established between multiple partners, as is made evident by results provided by Drexel University, Cornell University, Pajarito Powder, and ANL.
- Project partners include 3M Company, Carnegie Mellon University, Cornell University, Drexel University, and NREL. Several are not yet under agreement. To date, the interactions seem to be limited to pre- and post-characterization of materials.
- This project includes the efforts of a large team of experts. Integration of those results needs to be explained.

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

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

- Improving fuel cell performance at high current density is the most relevant aspect toward lowering the overall cost of fuel cell systems. Outcomes from this project have been introduced into the Strategic Analysis cost analysis, and this is exactly what has already happened.
 - The project seeks to improve the performance and durability of the cathode catalyst layer, which is the most relevant component toward improving cost and durability of the entire fuel cell system.
 - Of all projects in the Fuel Cells sub-program, this project is best situated to address the barriers to meeting the DOE targets.
- The potential for a large impact is built into the project plan. Fresh ideas/approaches for preventing the degradation of Pt/Co in an MEA are lacking; however, it is possible that improving the fundamental understanding of the degradation could produce fresh ideas.
- Higher-performing catalysts with improved durability at low Pt loadings are critical to the Hydrogen and Fuel Cells Program.
- The project directly addresses the DOE target to reduce PEMFC stack costs by reducing Pt loading, while maintaining high current density performance.

Question 5: Proposed future work

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

- Many aspects of the future work line up well with what could be expected: optimization of PtCo supported on HSACs, testing of ionic liquid stability, and exploration of Co stability during operation. The one difficulty with the future work is the prescription toward higher-activity oxygen reduction catalysts in spite of the work already done to achieve high current density performance with a very specific type of PtCo supported on HSAC. Perhaps the only design lever would be greater ordering inside the particle, such as has been proposed in this project for the collaboration with Cornell University. However, beyond atomic ordering, the options appear to be slim.
- The future work is well planned and focused on achieving understanding of PtCo degradation at high current densities, transport losses, and implementation of new ordered PtCo alloys. Development of catalyst support is not indicated as one of the priorities of the project.
- The only weakness with future plans is if the ionic liquid studies, as described, would really lead to a useful result in an MEA. It is not clear how the team will get ionic liquids to work in an MEA.
- The project looks to optimize PtCo on three different carbon supports. In the approach, ordered metallic alloys are proposed, but that does not seem to be present in the future work.

Project strengths:

- GM is significantly invested in fuel cell research and has a proven track record of fundamental studies in which performance or durability gaps are elegantly defined, followed by hypotheses and experiments to address the gaps. The primary investigator is well equipped to address barriers. The project has already delivered a catalyst concept shown to reduce the overall cost of a fuel cell stack. The project is delivering not only material concepts but also analytical techniques that can help other developers, such as the CO stripping method, for observing the percentage of Pt on the outside of primary carbon particles. In many cases, the collaborations are premised on both material and analytical inputs.
- The project addresses multiple aspects of high-current-density mass-transport losses (such as catalyst and support instability, and oxygen and proton transport through ionomer and carbon micropores) using both experimental and modeling approaches. This is a very well-coordinated effort between different team members, including industry, academia, and national laboratories.
- The project has a strong team, and the project goals are well aligned with DOE objectives. A wide array of approaches are being pursued to achieve those goals. Improvements in understanding catalyst supports and degradation mechanisms were provided.
- The project uses a good mix of characterization of in situ characterization to evaluate materials.

Project weaknesses:

- There do not seem to be any new catalyst synthesis results; these were proposed, but it is unclear when and how those are going to happen.
 - For a catalysis project, there seems to be little catalysis development; this seems to be an electrode development project.
 - There is no real indication that the ionic liquid RDE results will translate and/or be stable in an MEA. The PI does not believe in RDE results, yet he presents those to indicate the promise of ionic liquids.
 - There are inconsistencies with what this project presents and the guidance that the lead organization is giving other organizations.
- The exact method for producing the catalyst that has resulted in estimations of lower-cost stacks is still unknown.
 - The project has shown considerable success for high current density performance without evaluating durability.
 - The project has come to the conclusion that further mass transport losses (beyond those already addressed through placing Pt on the outside of carbon particles) are not worth addressing. Between this conclusion and the limited options for improving PtCo supported on HSAC, the project almost unnecessarily constrains the future prospects for improving fuel cell performance further.
 - Further details are needed in the ionic liquid study with respect to integration of ionic liquids into the catalyst.
- It seems like a path to a durable support that would maintain a sufficient amount of mesopores is not very clear, as the HSAC that performs best in catalyst tests fails in accelerated support tests.
- It is a little difficult to follow how all of the approaches being pursued on this project will be tied together. The approach for exactly how Pt/Co alloys will be made stable in an MEA was not clear. The approach for exactly how ionic liquids will be made stable in an MEA was not clear.

Recommendations for additions/deletions to project scope:

- The team should either show improvement and stability of ionic liquids in an MEA or eliminate that from the project scope. The team should compare this project with the GM Fuel Cell Performance and Durability (FC-PAD) consortium project and eliminate overlap between the two projects. There seems to be significant overlap. Perhaps this, as a catalysis project, should concentrate on developing new catalysts.
- The project is already well scoped. However, further details regarding the methods for making the PtCo on HSAC would be useful to developers. It might be good to have specific go/no-go decision points for the ordered intermetallic alloys from Cornell University and the ionic liquid catalyst composites from Drexel

University. It would be preferred for the project to provide some understanding of how the PtCo on the HSAC catalyst is suitable for scale-up.

- The project may benefit from down-selecting approaches that are yielding promising results and focusing on those areas. A plan to address Pt/Co stability in an MEA should be presented. A plan to address ionic liquid use in an MEA should be presented.
- Support development should be more emphasized in the future work.

Project #FC-145: Corrosion-Resistant Non-Carbon Electrocatalyst Supports for Proton Exchange Fuel Cells

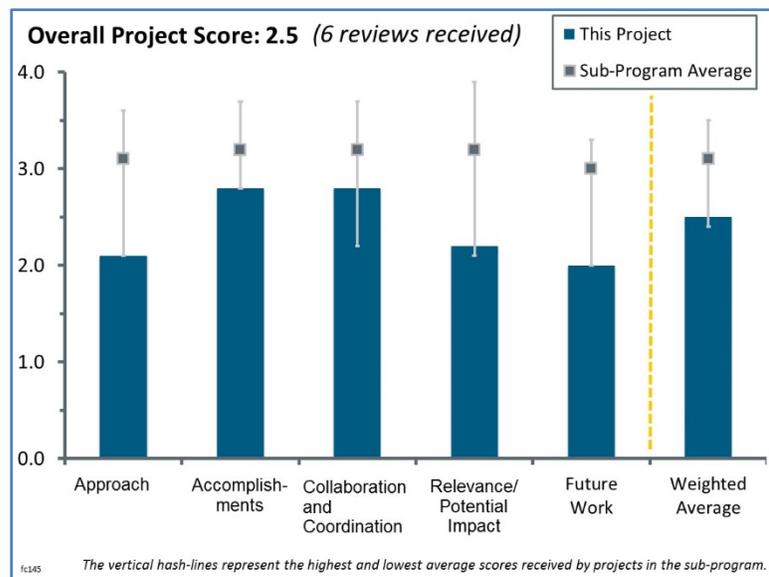
Vijay Ramani; Washington University in St. Louis

Brief Summary of Project:

Carbon's high electrical conductivity and low cost make it an excellent electrocatalyst support, but corrosion leads to kinetic, ohmic, and mass transport losses. This project is synthesizing doped non-platinum-group-metal (PGM) metal oxides as non-carbon alternatives. Along with being corrosion-resistant, the project supports would have high surface area, exhibit strong metal-support interaction with Pt, and demonstrate high electrocatalyst performance.

Question 1: Approach to performing the work

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



- Improving on carbon for fuel cell support materials remains a target that offers potential advantages for performance and durability. The material set chosen for investigation is very reasonable, and the application of multiple synthesis routes offers some opportunities for variability in structure and performance. The value of density functional theory (DFT) calculations is unclear.
- This work is very important in that both metal and support stability are key metrics to achieve commercialization. At this point, the reviewer has witnessed the talk, reviewed the 2016 and 2017 presentations, and reviewed some previous work on titanium dioxide–ruthenium dioxide (RTO) back to 2013. Perhaps there is more detail in the proposal, but it would be good to see more on the following:
 - It would be good to see data showing how much support electrical conductivity is enough to achieve performance targets. The project has materials from low to high conductivity and previous efforts on RTO. Perhaps there is a chart showing infrared or similar resistance losses in-cell versus support type. This would clarify an appropriate support conductivity target and give confidence to achieving 1.5 A/cm² targets.
 - The team's current activity is 0.05 A/mg_{PGM}. The project target is 0.30. There was not a clear path to achieve this in the 2017 presentation.
 - As electrode high current (1.5 A/cm²) is part of the team's targets, future presentations must address water management issues of a hydrophilic support such as doped metal oxides.
- This project looks at doped non-carbon metal oxides using a combination of DFT and experiment synthesis to develop new corrosion-resistant supports. There are few projects in this area at present.
- The Brunauer–Emmett–Teller (BET) target surface areas are the first indication that the project will probably not be able to make a practical fuel cell catalyst. Fuel cell catalysts made with supports that have >500 m²/g BET surface area often still have difficulty achieving at least 50 m²/g electrochemical surface area (ECSA) because of limitations of the triple-phase boundary, catalyst processing, and ink processing. The possibility of a 30 m²/g BET surface area support being able to achieve exactly 30 m²/g ECSA in order to obtain a roughness factor of at least 50 cm²_{Pt}/cm² at a cathode loading of 0.167 mg/cm² (which is higher than the DOE target for total loading) is extremely remote. The DFT calculations were performed to detect electrical conductivity for the supports. However, the porosity of the support materials may compromise the conductivity, especially for an oxide made as an aerogel. Given past experiences with platinumized metal

oxides being shown to be stable in a glass electrochemical cell, but then being shown to be unstable in a fuel cell, a better approach would seek to analyze stability directly in the context of a fuel cell.

- One of the critical problems of the previous project was the catalyst performance and dispersion of catalyst particles on the surface of the metal oxide support. This project tries to make higher physical surface area a target similar to a typical carbon support to be used for the polymer electrolyte membrane fuel cell (PEMFC) catalyst. However, there was no consideration as to whether the surface area of the metal oxide support has the critical characteristics for the catalyst dispersion. Toyota presented their metal oxide support materials at the 2017 Society of Automotive Engineers conference, and it achieved similar Pt dispersion to its high-surface-area carbon (acetylene black). Toyota showed good catalyst performance. This team needs to analyze why the previous project failed, and the approach should be developed based on that.
- This project is focusing on improving support stability while still using Pt as the catalyst. This is a fundamentally flawed approach because there is no indication that the learning from here could be translated to alloy catalysts.

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

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

- The project has demonstrated the synthesis of a number of target supports and the ability to deposit Pt onto a number of these structures. Some of the properties reported, like ECSA at $75 \text{ m}^2/\text{g}_{\text{Pt}}$ with durability toward potential cycling, seem to offer promise. The lack of any mass or specific activity data reported for these samples leads to concerns about the performance of these materials, as well as any potential advantages from strong metal support interactions as promoted as a motivation for investigating these specific families of materials. In past Annual Merit Review presentations by the project principal investigator (PI), novel supports often struggled to deliver reasonable mass/specific activity. Mass activity was called out as a target for these materials and needs to be reported with other relevant electrochemical measurements. Ta-doping for TiOx is shown to be best for stability, as it shows the lowest d-band center with Pt. However, Ta-TiOx was shown to be extremely low for BET surface area. Nb-doped TiOx appeared to have the best combination of surface area and conductivity. Surface area for Nb-TiOx was increased to $130 \text{ m}^2/\text{g}$, but the material has not yet been platinized, put in a fuel cell, or studied for durability. Sb-SnO₂ showed stability for rotating disk electrode (RDE) voltage cycling testing at 1.0–1.5 V. No fuel cell data were shown, although the project is still in its first year. BET surface area is extremely low for higher-conductivity Sb-SnO₂. Unless there is some way to increase BET surface area by at least one order of magnitude while still maintaining conductivity, the stability of the support will not matter since the required performance will not be obtained.
- The team has made significant progress in understanding the valence states and the critical factors in improving the conductivity of the metal oxide supports. It looks like there is only one metal oxide family than can meet both support surface area and conductivity targets. Beginning-of-life ECSA of $75 \text{ m}^2/\text{g}_{\text{Pt}}$ in RDE looks quite promising.
- This project appears to be using DFT successfully to guide the synthesis approach and determine the level of dopants needed to get the appropriate conductivity. The project is in its early stages, but a number of relatively high-surface-area materials have been synthesized with good conductivities. Some metal oxide support materials have had Pt deposited on them; some are in process.
- The DFT calculation showed expected results for an electrically conductive doped metal oxide. For the material screenings, the usage environment should be considered. The PI mentioned that these metal oxide supports would be used for both the cathode and anode. Therefore, materials stability should be shown in the reduction environment (anode) and oxidation environment (cathode).
- It was understood from the talk that this work was only five months in at the time of slide submission. From the talk and slides, it is a bit unclear what milestones to evaluate against, as they start in Quarter 7 on the 2017 talk. It is unclear if the team was in Quarter 2 or Quarter 4 at the time of submission. There is some added clarity in the 2016 talk. It is exciting that there seems to be a possibility for $>100 \text{ m}^2/\text{g}$ supports, but again, information about the status and progress is a bit confusing.

Question 3: Collaboration and coordination with other institutions

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

- Collaborations appear to be limited to Nissan and the University of New Mexico (UNM). Nissan is essentially being used as a fuel cell testing partner, but it is unclear whether Nissan is providing the automotive perspective that clarifies whether the materials being developed are viable for meeting requirements. The low-BET surface areas would seem to be an obvious indication that some automotive requirements may not be met. UNM is providing the silica templating technique for making higher-surface-area supports, which is a material input, as well as DFT calculations that do not factor much into whether a material can be made at high surface area. What the project genuinely needs is a catalyst supplier to step in that can suggest scalable ways of producing high-surface-area supports, as well as an automaker (such as Nissan) to draw the line on which materials can possibly meet the requirements.
- The team is well-rounded, with the specific background relevant to the project. The PI was unable to attend, and the co-PI who presented was not fully aware of all the subtleties of the project (although this is not necessarily a surprise and is not fully required for successful project execution). The dynamic of assessing this specific aspect was slightly more challenging because of the PI's inability to participate directly.
- As UNM gave the presentation for the Washington University in St. Louis (WUSTL), the communication seems to be good. Both UNM and WUSTL have made progress. Incorporating Nissan appears to be a later date intention and mostly for evaluation/testing.
- There seems to be a good collection of partners for this work. The Nissan Technical Center North America should perhaps outline more of its path to electrode optimization, especially in achieving operational robustness.
- Collaboration is planned, but currently each group's work is being individually pursued.
- The team needs to be collaborating with a traditional catalyst supplier (e.g., Johnson Matthey and/or Tanaka Kikinzo Kogyo K.K.) to understand the feasibility of these proposed synthesis routes.

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

This project was rated **2.2** for its relevance/potential impact.

- Supports that promote metal stability are likely more valuable than carbon stability. The possibility that this project might achieve both is quite exciting. Further, novel materials generation enhances overall industry understanding, leading to further progress.
- This project addresses the catalyst support durability target and thus the catalyst durability targets. It is unclear what the cost of these support materials will be and whether they will be able to address the cost targets. It will be critical to be able to support PtX alloys with high mass activity and achieve performance similar to carbon supports with improved durability.
- The development of improved materials relative to carbon as support materials is of clear relevance for performance and durability limitations of fuel cell systems. At this point in time, the state of this project does not suggest that a better material will be realized from these efforts.
- It is an industry consensus that the carbon corrosion problem is solved by a system solution rather than a materials solution. The system mitigation strategy is well developed and well implemented, although the DOE still keeps targets in this area. In other words, these system solutions enable the use of materials from a wider selection range and improve the performance and cost (less expensive materials might be used). Secondly, there is one industry project (no government funding) that already demonstrated good performance of a doped metal oxide support catalyst in a PEMFC. Pt dispersion on these metal oxide supports is similar to Pt dispersion on high-surface-area carbons. Optimized catalyst layers, such as ionomer dispersion, were achieved. This is a typical example of what the private sector can do. It is not clear why DOE should fund it.
- Thanks to system and seal mitigations for shutdown/startup operation, the prospect of a corrosion-resistant catalyst support providing benefits toward the Fuel Cells sub-program is practically zero. It may be good

for Strategic Analysis to find a way to estimate increased cost associated with (1) added system components for mitigation of high half-cell potentials on shutdown/startup, and (2) enhanced sealing materials associated with maintaining desired gas contents in the stack under vehicle soak conditions. If the increased cost is low or practically negligible, then it can be confidently said that the Fuel Cells sub-program should not be invested in projects such as this project.

- Very few automotive manufacturers need a stable support on the cathode side. Further, the project is reporting 0.4 mg/cm² loading as the state of the art; this is far from the DOE target of <0.125 g_{Pt}/kW. Therefore, it is unclear how this project, even if wildly successful, can reduce the cost and Pt loading of a stack.

Question 5: Proposed future work

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

- The proposed future work seems to refer primarily to FY 2017; the overall strategy of using modeling to guide the dopants to achieve appropriate conductivity during the synthesis with final testing by a vehicle manufacturer (Nissan) appears to be a good strategy, and less of an Edisonian approach.
- There seem to be good work and potential paths toward support conductivity and surface area. The following need more clarity:
 - Paths toward achieving activity targets
 - Overall pairing of the catalyst to the support; for example, a higher percentage of metal on the support might enhance the conductivity
 - Nissan Technical Center North America optimization plans
- The plan for making both surface area and highly conductive Sb-SnO₂ needs to be clarified. Greater emphasis needs to be placed on fuel cell testing. The glass cell electrochemical evaluation could almost be skipped in favor of what will probably need to be many iterations of ink processing in order to understand how best to optimize a cathode-electrode for a platinized metal oxide. Even without an optimized cathode-electrode, though, an un-optimized electrode could be used to provide an early indication of catalyst durability in a fuel cell environment. The project's plan for future work needs to move faster toward fuel cell testing.
- To continue this low-relevance project, it should focus on tasks to fill the knowledge gap rather than developmental gap. DFT calculation and modeling should be focused for (1) materials stability for anodic and cathodic environments and (2) critical characteristics to synthesize the materials. It is still questionable if the BET surface area is a critical characteristic for catalyst Pt dispersion.
- The proposed work is a continuation of the work already underway, involving increased synthesis of materials, characterization, and DFT modeling. It is not clear what is systematically going to be probed or why improved performance over initial efforts should be expected.
- Shown at the membrane-electrode-assembly level, this catalyst can get close to the DOE targets for Pt loading of 0.125 g_{Pt}/kW. The project should be stopped if it cannot show <0.25 g_{Pt}/kW (double the Pt loading on the DOE 2020 targets).

Project strengths:

- The project demonstrated the ability to make Nb-doped TiOx with decent conductivity at 130 m²/g. That provides some hope of an interesting material coming from the project. The project has an automaker (Nissan) as a partner, which should ideally provide a practical automotive perspective. Materials being used do not represent possible Fenton agents or other species that would be likely to cause quick membrane degradation or harm to other components.
- This project seems to be successfully coupling DFT with experimental synthesis to help guide the synthesis. Between WUSTL and UNM, the synthesis and electrochemical capabilities exist. Nissan appears to provide testing capabilities.
- Project strengths include DFT calculation, first principle modeling (UNM), and materials synthesis (doped metal oxide).
- Project strengths include the properties and potential of the support, as well as the team and expertise. The team achieved high metal surface areas.

- This is a relevant material set being investigated by a few select synthesis routes.
- The project has a good team and innovative scientists.

Project weaknesses:

- There is low activity at present and no clear plan to overcome it. This can be considered a support issue, as clearly different carbon supports enhance activity. There was no plan described for overcoming support hydrophilicity. There is a lot of data at present, and historical data from previous RTO efforts, but these are not organized into clear trends pointing toward long-term targets. It is understood that this is early in the project; however, such organization would be helpful.
- The relevance of the project is a weakness. First, industry consensus is that the system solution can work well for this carbon corrosion issue and enables wider selections of catalysts (with performance and cost improvement). Second, private sector studies already demonstrate good progress of doped metal oxide catalyst support for PEMFCs. (It is not clear why government funding is needed for a task that the private sector can do.)
- The project is working in an area that may have already been made obsolete because of system mitigations implemented in commercial fuel cell systems. The project is having difficulty obtaining high surface areas for all materials other than one particular form of Nb-TiOx. The project future work plan does not move fast enough toward fuel cell testing, especially for evaluating material durability.
- The value of DFT efforts has not been shown. The mass (and/or specific) activity of the synthesized materials has not been reported, even though other electrochemical data have been reported.
- There is not much thought to date, nor does the future work appear to address the cost of these materials.
- There is a lack of clear vision on how this project can reduce cost.

Recommendations for additions/deletions to project scope:

- It is highly recommended that the project funding be reconsidered based on the relevance. It is an industry consensus that the carbon corrosion problem is solved by system solutions rather than materials solutions. The system mitigation strategy is well developed and well implemented. Additionally, these system solutions enable the use of materials from a wider range of selections and improve the performance and cost (less expensive materials might be used). Secondly, there is one industry project (i.e., not government funded) that already demonstrated good performance of a doped metal oxide support catalyst in a PEMFC. Pt dispersion on these metal oxide supports is similar to Pt dispersion on high-surface-area carbons. This is a typical example of what the private sector can do, so it is unclear why DOE should fund this work. To continue this low-relevance project, it should focus on tasks to fill the knowledge gap rather than the developmental gap. DFT calculations and modeling should be focused on (1) materials stability for anodic and cathodic environments and (2) critical characteristics to synthesize the materials. These outcomes could be common knowledge and may be valuable for other areas such as metallic bipolar plates. It is still questionable whether the BET surface area is a critical characteristic for catalyst Pt dispersion. Optimization of the catalyst layer is necessary, but this is an engineering area.
- Fuel cell testing needs to be prioritized over glass cell electrochemical evaluations. In fact, it may be expedient to drop glass cell testing in the case of this project. Fuel cell testing should keep in mind low-temperature robustness. The hydrophilic electrodes may not perform well at temperatures experienced by a fuel cell stack warming up (e.g., 40°C). DFT calculations are not nearly as important as finding methods to deliver higher-surface-area supports. More effort needs to be shifted toward increasing surface area, or else the resulting fuel cell electrodes will not have high enough roughness factors at low Pt loading. The project targets should reflect DOE targets. There is no need to set the bar lower and expect a later project to resolve the gap.
- A small portion of the effort should be used to address the cost of materials and the cost of synthesis in comparison to more traditional carbon materials.
- The project should have intermediate activity targets (perhaps they are already there and were missed).
- The project should downscope the DFT studies to focus more on synthesis efforts.
- It is recommended that this project be deleted or refocused on developing an anode catalyst.

Project #FC-146: Advanced Materials for Fully Integrated Membrane Electrode Assemblies in Anion-Exchange Membrane Fuel Cells

Yu Seung Kim; Los Alamos National Laboratory

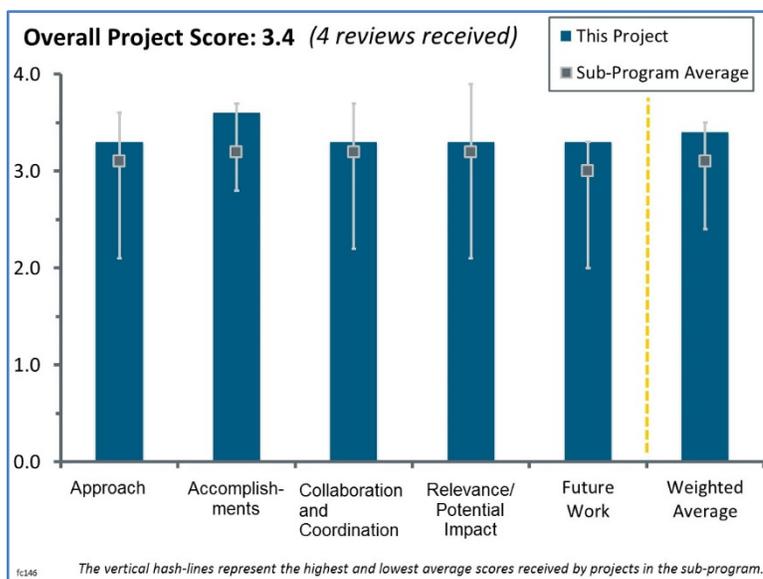
Brief Summary of Project:

This project is developing advanced materials for fully integrated membrane electrode assemblies (MEAs) in anion-exchange membrane fuel cells (AEMFCs), enabling fuel cell cost reduction without sacrificing performance. The improved anion-exchange membrane (AEM) materials are based on highly conductive and stable hydrocarbon polymers. The project also aims to address challenges with integrating catalysts and AEMs into high-performance MEAs. The approach involves (1) preparing AEMs without aryl-ether linkages in the polymer backbone and (2) developing different ionomeric binders for anode and cathode.

Question 1: Approach to performing the work

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

- This is an innovative way to get to the conductivity and at least the intermediate stability of hydroxide-conducting membranes by “eliminating” weak points. This should provide membranes so that other properties, such as the hydroxide and carbonate ratio in a membrane, can be established. Their transference numbers can be established, and net membrane conductivity can be established, which are critical points to know in order to find out whether this “alkaline membrane” concept is practical.
- This is an excellent approach to study non-polar hydrocarbon polymers with resonance-stabilized actions and to develop hydrocarbon ionomers. Although it is understood why precious metal catalysts are being used in this primarily membrane and ionomer development project, the true potential of these materials will not be realized unless they work with non-precious metal catalysts. It may be that developing them with precious metals leads to developing materials not compatible with non-precious catalysts and so they are ultimately worthless.
- The work addresses the barriers of cost, durability, and performance. This work does a particularly good job of addressing durability issues with alkaline membranes and alkaline membrane fuel cells. The work is well designed and has demonstrated the feasibility of AEMFCs. It is well integrated with other efforts in the AEMFC field. If aromatic groups are detrimental, as recent results seem to suggest, this could have impacts on the applicability of all three classes of polymers being studied in this project, as the Rensselaer Polytechnic Institute styrene–ethylene–butane–styrene block copolymers, the Sandia National Laboratories Diels–Alder polyphenylenes, and the Los Alamos National Laboratory poly, di, and terphenylenes all have substantial aromatic content and would be expected to have some poisoning effect on the hydrogen oxidation reaction (HOR). The goal of the alkaline work is to enable lower costs by enabling platinum-group-metal (PGM)-free catalysts. In that vein, the effort spent developing PGM electrodes and looking at interactions and poisoning of PGM catalysts with the ionomers diverts resources from the ultimate goals of a PGM-free alkaline system.
- It is difficult to address all the barriers at the same time with such a new material set. When demonstrating in a fuel cell, it should be with all aspects of the approach. For example, when demonstrating AEM in a fuel cell, the project should use non-PGM electrodes, as it makes no sense to work on developing and



improving PGM-containing electrodes, since we will get no closer to cost goals as compared to the use of PGM electrodes, as in PEMs.

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 team has demonstrated some very nice ex situ stability testing indicating these materials have much promise. The team is beginning to get an idea about carbonate/bicarbonate performance losses. There are interesting and insightful studies on HOR inhibition. Very impressive improvements in fuel cell performance have been achieved.
- There were good accomplishments on the fundamental material properties, such as conductivity and membrane film formation, and integration into fuel cells.
- This work is addressing the cost, electrode performance, and durability goals in a logical fashion with good progress.
- The project has made substantial progress in developing membranes stable under the basic conditions of AEMFCs. The project has made significant improvements in power density in H₂/O₂ conditions, achieving 1 W/cm² and surpassing the go/no-go metric for 2017 of 0.6 W/cm². The project has identified a new detrimental interaction between the catalyst and aromatic rings. However, in 2016, the researchers presented Fourier transform infrared spectroscopy (FTIR) data indicating alkyl ammonium groups adsorbing and interfering with HOR and guanidinium groups not adsorbing. The data they use to indicate poisoning by aromatic groups in 2017 uses benzyl trimethyl ammonium functionalized poly(phenylene) anion exchange membranes (ATM-PP) with a trimethyl ammonium cation, which they compare to a perfluorinated backbone with a guanidinium cation. Since the cation has been shown to adsorb previously, a better comparison to show the aromatic ring is affecting the HOR. It would compare the ATM-PP to the perfluoroalkyl backbone with the same cation functional group, and preferably a non-adsorbing functional group (i.e., with the guanidinium group on both the ATP and the perfluoroalkyl backbone). The project has slipped on the milestone to down-select an ionomer.

Question 3: Collaboration and coordination with other institutions

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

- The collaboration within the project appears to be working well. The move away from studying perfluoro ionomers (being studied by the National Renewable Energy Laboratory [NREL]) suggests collaboration rather than competition there as well.
- All team members have well-defined goals. The work on non-Pt catalysts seems on hold. High-performance cells (conductive membrane with fairly stable membranes with catalyzed electrodes) have still been made.
- There is good collaboration with other national laboratories and universities. A number of collaborators are listed that could do device testing for fuel cells and the balance of plant, but it is not clear that materials have been exchanged with these partners, and no results were presented.
- Better collaboration should be done with other laboratories working on these AEM membrane materials. Materials should be exchanged with NREL since they are also working on AEM and common test methods could be used.

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

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

- The project supports the goals and objectives of the DOE Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan. The work looking at performance and stability of the

alkaline membranes is well aligned with DOE goals. The work looking at interactions of the ionomer with PGM catalysts is less relevant, as PGM-free AEMFCs are the goal.

- This project is making progress toward DOE cost, performance, and durability goals. Some work should be on non-Pt catalysts and used in air at some point.
- DOE should absolutely study AEMs and the fuel cells derived from them. Major breakthroughs in this area will enable low cost and more versatile fuel cells, electrolyzers, and redox flow batteries. DOE needs to recognize that AEMFCs will take some time and investment to realize their potential, and the Department should not be too quick to cancel projects in this area. This project addresses many of the challenges in AEM development and cationic polymer development for ionomers in AEMFCs. That is critical if this technology is to get to the point at which it attracts much commercial interest.
- Since the progress on the materials is so pronounced, more effort should be placed on the non-PGM electrodes. If this is not undertaken, then the project is back to the same cost equation as PEMs, and not much relevance or potential impact can be realized.

Question 5: Proposed future work

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

- There is a very good plan to scale up polymers, down-select ionomers, and continue to study HOR degradation. Future work on MEA integration is much-needed, as is the fuel cell performance and degradation study.
- The proposed work does a good job of addressing membrane stability issues. The project should down-select to one membrane class to integrate into the MEA, but plans seem to continue with all three classes of membranes.
- The 2017 and 2018 work plans are reasonable. There is a good balance of needed fundamentals and useful, practical studies.
- The material is developed to the point that non-PGM electrodes should be developed for the full advantage of this new material to be realized for automotive fuel cell applications.

Project strengths:

- The project strength is the synthetic capabilities of the team and the focus on determining degradation points and chemical stability of the membranes.
- This is a good team generating good results and addressing DOE interests.
- New materials studies have potential for game-changing results.
- New AEMs are a strength.

Project weaknesses:

- The project needs to begin considering non-Pt catalysts and the effect of air at some point. It is logical that this has not been done yet, but this is needed if the “alkaline membrane” concept is going to be practical for automotive applications.
- Focus on the right issues needs to be shifted as progress is made in different areas.
- There is no non-precious metal catalyst work.

Recommendations for additions/deletions to project scope:

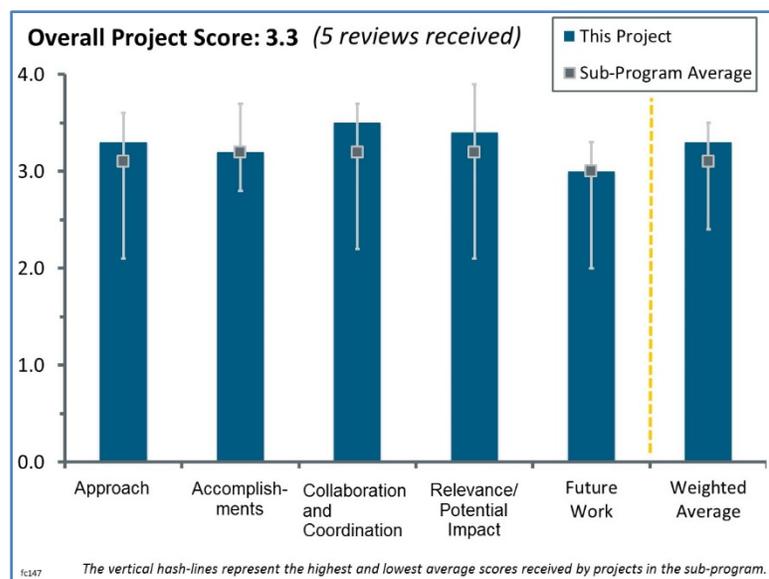
- Work on non-PGM catalyst electrodes should be added, even if the power density is not as good as in PGM catalysts.
- The project should start incorporating non-Pt catalyst studies in 2018, at least for comparison to Pt and Pt/Ru electrodes.
- This project should have less focus on PGM catalysts and interactions with/poisoning of PGM catalysts.
- Perfluorinated materials are already eliminated.

Project #FC-147: Advanced Ionomers and Membrane Electrode Assemblies for Alkaline Membrane Fuel Cells

Bryan Pivovar; National Renewable Energy Laboratory

Brief Summary of Project:

Alkaline membrane fuel cells (AMFCs) offer promise for improved performance and decreased cost. This project aims to develop novel perfluoro (PF) anion-exchange membranes (AEMs) with improved properties and stability, employ high-performance PF AEM materials in electrodes and as membranes in AMFCs, and apply models and diagnostics to AMFCs to determine and minimize losses (water management, electrocatalysis, and carbonate-related). Researchers will synthesize, characterize, and optimize alkaline-exchange membranes and fuel cells for performance and durability.



Question 1: Approach to performing the work

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

- The project addresses the barriers of cost, performance, and durability of alkaline fuel cells. The project focuses on alkaline membranes and alkaline membrane durability and performance. The approach of utilizing fluorinated backbones provides chemical stability of the backbone and advantages for forming a phase-separated structure, which has been found to be linked to good conductivity and mechanical properties. The approach of eliminating the sulfonamide linkage to increase stability is feasible and likely to be successful.
- The approach to developing an AEM is good, and the project is well designed and utilizes an effective teaming approach with modeling, industry, and academic team members.
- The approach of the project is well designed, integrating AMFC model development and diagnostics.
- This project is aimed at developing stable and conductive hydroxide-conducting membranes based on highly stable PF backbones. Oddly, short-term stability was found wanting, but the reason—namely, instability of the sulfimide linkage and short diamine linkages—appears to have been found and addressed. This is a work in progress.
- Had the proper chemists been involved, the design/progression of the tether could have been identified immediately to eliminate unnecessary work and be more efficient. The design was in series versus parallel and created plenty of work in which most people are not interested because they want to see a Generation 3 product. Both in the document and in the presentation, there seemed to be a lot of jumping back and forth on test subjects. Perhaps a Gantt chart in advance would help to streamline the work. It would be helpful if the project would verify that the chemical structures are correct for the PF tether versus the hydrocarbon tether; it became confusing to the outsider. Also, it is not clear where the composition details of the Tokuyama A201 and Tokuyama AS-4 were. It is also unclear which Nafion™ membrane was used. There are many different types. After a while, it almost seemed like the principal investigator (PI) was trying to impress by showing gobs of tests versus presenting a methodical analysis and evaluation. This can become baffling. As a side note, the reviewer has seen the flooding behavior/response in polymer electrolyte membranes (PEMs) before but, unfortunately, cannot remember the details.

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 progress is excellent. Good film-forming capabilities have been demonstrated with the industry team member, 3M. The properties of the membrane look promising. Good power densities have been shown in operating fuel cells, although on oxygen only. Given the good progress on membranes and materials demonstrated by this project, future work should be shifted to using non-platinum-group-metal (PGM)-containing electrodes and operating the fuel cells on air.
- It was quite difficult and an excellent accomplishment to sort out stability of the ionic groups. Work is on track to use this knowledge to meet goals. The non-Pt catalyst and cell using air should be addressed at some point.
- Last year, membrane stability was significantly increased by extending the chain between the sulfonamide and the quaternary ammonium group. This year, the Generation 2 membranes met the midterm criteria of maintaining an area-specific resistance $<0.1 \Omega \text{ cm}^2$ (after correction for cell electronic losses) at 60°C for 500 hours during testing at 600 mA/cm^2 . Degradation of the Generation 2 membrane at the sulfonamide site was determined to also be an issue. New chemistry looking at replacing the sulfonamide link to the backbone with aromatic linkages is being investigated, but results are not yet available. Membrane electrode assemblies (MEAs) prepared with Generation 2 membranes achieved power densities approaching 1 W/cm^2 in H_2/O_2 .
- Cost has been ignored. Analogies, or at least attempts to compare and rationalize cost improvements, should be presented. Cost mock-up through film formation should be taken into account. Performance and durability should be more thoroughly addressed with the Generation 3 membrane. As with any new membrane structure, severe accelerated degradation and physical/mechanical/compositional analysis should be conducted to understand the most likely forms of degradation. This should include possible gasket/perimeter failures. It seemed that the PI was achieving the different goals using different membranes.

Question 3: Collaboration and coordination with other institutions

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

- Collaborations outside of the project with other AEM fuel cell and AEM electrolyzer projects are apparent. Interactions with others outside the project have led to increased MEA performance by utilizing gas diffusion electrodes (GDEs) developed elsewhere. Collaborations within the project appear to be working.
- The National Renewable Energy Laboratory, Oak Ridge National Laboratory, Colorado School of Mines, and Lawrence Berkeley National Laboratory teams have well-defined roles for synthesis characterization and modeling of hydroxide-conducting membranes in different situations. Most work is in progress to address all issues.
- This project has demonstrated an excellent teaming approach with several small businesses, a large business, and academia, all bringing their own strengths to the project led by a national laboratory.
- It is clear that the PI is a networker. The only deficit: had a fuel cell fluoropolymer chemist(s) been involved, the compositional design/progression of the tether could have been identified immediately, and unnecessary work would have been eliminated. (Perhaps this was essential to aid in the modeling work, but it is doubtful.)

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

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

- Although excellent progress has been made and the goals and targets in the Fuel Cell Technologies Office (FCTO) Multi-Year Research, Development, and Demonstration Plan have been met, new targets need to be developed in order for the materials to begin to realize a more competitive approach as compared to the

conventional PEM fuel cell system. These targets should include the use of non-PGM catalysts and operating the fuel cells on air instead of pure oxygen.

- The project is relevant and aligns with the FCTO goals and objectives. AEM fuel cells could significantly reduce fuel cell costs by enabling PGM-free fuel cells, as alkaline membrane durability and availability have been issues. This project seeks to address these issues.
- This project has the potential to meet all requirements of an “alkaline membrane” to learn whether a practical hydroxide-conducting fuel cell for automotive applications can be made.
- The novel structure (and analysis thereof) is critical in understanding the potential for new membranes. It would be good to highlight the importance and benefit of this particular fuel cell technology and the conversion to this PF chemistry. The chemical durability aspect is understood, but it also seems cost is added. A cost-versus-benefit analysis could showcase rationale.

Question 5: Proposed future work

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

- The reason behind the surprising increase in performance for the GDEs needs to be determined. The future work on the membranes is appropriate and focused on what appears to be the weak point now determining durability. The work on modeling and AMFC implementation is important for getting a working fuel cell.
- The focus on Generation 3 polymers is critical. It is good that there is intention to do hydroxide and carbonate transport studies, for a practical system in air depends on this. Modeling validated by experiment is also admirable. There is no non-Pt catalyst work planned, so this should be considered when stability problems are behind the team and a reasonable fuel cell activity is achieved with Pt group catalysts for comparison to non-Pt catalysts.
- The project should incorporate visuals such as Gantt charts, flow diagrams, etc. on decision points. The project should also work through risk mitigation and contingency planning. The whole electrode issue is a mess. The team should figure out a way to achieve the goals without electrode development.
- Future work should include more emphasis on the remaining limitations of AEM fuel cells from a system and cost perspective, such as operating on air, and the use of non-PGM catalysts.
- The plan for Generation 3 membrane development is not clear.

Project strengths:

- New materials allow for big opportunities to develop major advancements in fuel cells. A good teaming approach has been demonstrated in this project.
- The project had a well-designed approach to develop novel AEMs using model development methods, nuclear magnetic resonance studies, and performance tests.
- Collaborations and interactions with the AEM fuel cell community are a key strength of this project.
- This is a good team working at a reasonable pace to overcome formidable challenges.
- Modeling and testing are project strengths.

Project weaknesses:

- The project still utilizes PGM catalysts, while the *raison d'être* for alkaline fuel cell work is the potential for PGM-free fuel cells. Alkaline fuel cell projects should move toward using PGM-free catalysts, or at least to loadings below PEM fuel cell anode loadings.
- The lack of a laser focus on only key materials and testing is a weakness. Cost rationale and estimates are a weakness. This project is susceptible to “not seeing the forest for the trees.” Perhaps the PI does, but in conveying it, it becomes a bit of a random cluster.
- There is no plan for non-Pt group catalysts.

Recommendations for additions/deletions to project scope:

- Electrode development should be deleted. The project should use the best commercial performer it can find and go with that. Modeling work should be de-emphasized. The project should build in more analytical

evaluations of the membrane before and after additional accelerated degradation (physical, mechanical, electrical, and application-developed).

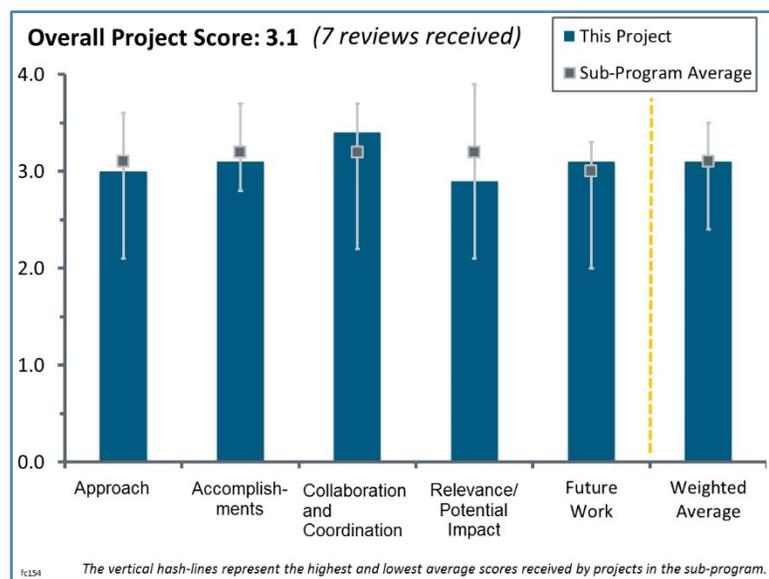
- The project should focus on the big challenges for AEM fuel cell implementation, such as operating on air, and using non-PGM catalysts.
- The team should consider two things: studies in air and studies with non-Pt catalysts.

Project #FC-154: Regenerative Fuel Cell System (Small Business Innovation Research Phase II)

Paul Matter; pH Matter LLC

Brief Summary of Project:

Fuel cells operated in a reversible manner are a promising potential energy storage solution, but high system cost is a major barrier. Development of a low-cost reversible fuel cell would be a key breakthrough for energy storage. This project, which builds on an earlier Phase I Small Business Innovative Research effort in platinum-group-metal (PGM)-free catalysis, aims to develop and demonstrate a reversible anion-exchange membrane fuel cell (AEMFC) that incorporates membrane electrode assemblies as regenerative stacks. System durability over 1,000 cycles will be demonstrated, and economic analysis of the developed system for use as a grid energy storage technology will be performed.



Question 1: Approach to performing the work

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

- This project has a good overall approach. The principal investigator (PI) mentioned using the Steward et al. technical paper “Lifecycle Cost Analysis of Hydrogen Versus Other Technologies for Electrical Energy Storage” (November 2009, publication number NREL/TP-560-46719) guidelines in planning the work. The project demonstrated 360 cycles with a non-PGM cathode. The project also demonstrated a range of catalysts that perform well. The project appears on its way to a successful 1,000-cycle deliverable.
- The team is trying to make a reversible fuel cell based on anion exchange membranes (AEMs). The approach is innovative and timely. The project has the right team members, with the National Renewable Energy Laboratory (NREL) providing hydrogen oxidation reaction (HOR)/hydrogen evolution reaction (HOR) catalysts and Giner, Inc., providing cell and stack design.
- The project’s approach is to utilize a carbon-based oxygen reduction reaction (ORR)/oxygen evolution reaction (OER) catalyst for a regenerative AEM-based fuel cell, and to show feasibility for a low- or non-PGM catalyst on the HER/HOR side of the cell. pH Matter LLC is collaborating with NREL and Giner, Inc., to integrate and demonstrate the performance of their catalyst. The project addresses a niche market where potentially low-cost regenerative systems may compete with batteries for energy storage. To date, the testing has largely been based on half-cell testing in concentrated electrolyte solutions. In pseudo-membrane-based cells operated under very short (one-minute) duty cycles, it makes it very hard to determine whether the project is feasible and whether the ultimate targets can be met.
- The project’s round-trip goals and strategy make sense.
- No rational approach for the project is addressed in slide 6, although the benefits of using a regenerative fuel cell system are obvious.
- The project approach involves developing a proprietary CNxPy oxygen catalyst and integrating it into a reversible AEMFC. This is a very challenging system to develop, with the unitized nature, the use of alkaline media, and the use of a PGM-free oxygen electrode all adding barriers. The project’s overall approach seems a bit too optimistic. At this point, it makes more sense for U.S. Department of Energy-funded projects to focus on just one grand challenge (such as reversible operation, or the use of alkaline

media), rather than trying to “shoot the moon” by incorporating so many high-risk aspects into a single project.

- The approach of this project did not give confidence that this work is very relevant. At a minimum, the project should do the following:
 - The data in slide 9 clearly shows Giner, Inc.’s baseline performance with previous metal cathodes. This project needs to compare its performance to this and clearly articulate the benefits, if any. For example, in slide 10, the voltage losses are significantly more when the PtIr catalyst is replaced with the COR-2 catalyst. It is unclear whether the cost of replacing the PtIr is enough to make up for this performance loss. If a system must be built that works at a particular efficiency at rated power, then the COR-2 catalyst system needs to be sized much larger, and it is not clear whether the cost saving from replacing the catalyst more than pays for this size increase.
 - It seems like the 42% efficiency target is not very high for an energy storage application. The PI needs to compare this to the performance of Vanadium flow batteries to at least make sure that this technology can compete with that on some basis.

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 taken a catalyst developed in previous years and integrated it into a reversible membrane electrode assembly (MEA). The MEA is based on a commercial AEM (the manufacturer was not disclosed because of a non-disclosure agreement), a proprietary CNxPy oxygen catalyst developed by pH Matter LLC, and a commercial PtRu/C hydrogen catalyst. PtNi nanowire catalysts developed by NREL were also examined for use on the hydrogen electrode. The fuel cell and electrolyzer mode performance are both lower than dedicated fuel cells or electrolyzers, but considering the avant-garde nature of this system (unitized, alkaline, and PGM-free oxygen electrode), the performance looks reasonable. Still, considering how effective some other published PGM-free catalysts are for alkaline ORR, the fact that the fuel cell performance lags so far behind the PtIr baseline is not encouraging.
- Much progress was made in 2017. Slides 7 and 9 are from last year’s accomplishments (the PI should have made this clear). The integration of the NREL work into the PI’s work was not obvious. It is unclear what the point was for replacing the Pt/Ru with Pt/Ni. The HyRoc-1 catalyst seems to be PGM-free and has similar performance at the operating point (200 mA/cm²) to the PtNi.
- The project has met its milestones; however, it is unclear why the targets have been set the way they are and what test hardware would be used to achieve them. For instance, the cells used to meet targets to date are operated under short duty cycles at 1:1 time duration, but the system will need to operate over longer timeframes to compete with batteries. If a 1:4 duty cycle is required, a discrete system makes more sense since the fuel cell and electrolyzer-active areas can be scaled separately to reduce the cost. Cost analysis should be expanded to compare discrete versions of the pH Matter LLC stack versus a fully polymer-electrolyte-membrane- (PEM)-based system operating at optimal current densities. It is also not clear based on a lack of catalyst composition or preparation description that the catalysts are indeed PGM-free, or how a carbon-based catalyst is stable at 1.6 V in potassium hydroxide.
- Some progress on measuring baseline catalyst performance was reported (slide 9). However, the slide does not contain much technical analysis. For example, it is hard to understand why the performance with gas diffusion electrodes is better than that with catalyst-coated membranes (CCMs). It is difficult to review without some information about the catalysts (e.g., COR-2 and HY-ROC). The justification to use Pt/Ni nanowire instead of Pt/Ru on the anode was not well rationalized. As the project is for developing non-PGM (or low-PGM) components, more efforts on COR catalysts, including comparison with other benchmark catalysts, should have been done.
- The overall performance goal is quite clear: 42% round-trip efficiency and >250 mA/cm² for fuel cells and >50 mA/cm² for electrolyzers. However, in a later section, it was not clear whether this target has been met or how much the gap is. The investigators claimed that the fuel cell met the goal, but it is hard to talk about fuel cells alone when the overall target is a round-trip efficiency. There is also confusion in the later slides, which refer to 200 mA/cm² for fuel cells, while the beginning of the presentation calls for 250 mA/cm².

- The current density for electrolysis mode is 40 mA/cm⁺, which makes the system cost very high for round-trip use. Cross-optimization is a significant challenge.
- The project is on cost and schedule.

Question 3: Collaboration and coordination with other institutions

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

- It seems like most data in the presentation are outcomes of collaboration. In particular, the cycling durability test by Giner, Inc., is helpful for the project. More interactions with universities and other national laboratories may be beneficial to the project to get more benchmark materials for the test.
- Giner, Inc., is an excellent team member that can offer good feedback and guidance.
- The project collaborated well with Giner, Inc., in the CCM development and testing.
- There is good collaboration among the team members. NREL clearly has provided its PtNiNW catalysts, although the MEA results are not as good as the ones for RDE.
- The project's collaboration with Giner, Inc., looks very helpful. However, the value of the collaboration with NREL is questionable. The project should be focused on using reliable commercial hydrogen catalysts, rather than trying to use the experimental and unproven PtNi nanowire technology that is still being developed by NREL in a separate project. A better use of NREL would be to use their MEA diagnostic capabilities to help understand the voltage loss breakdown.
- The team has all of the necessary expertise.
- There appears to be limited interaction with subcontractors, which may be a problem for integration into a commercial system that operates differently from the current duty cycle, electrolyte feed, etc. It is not clear what the work breakdown structure is between partners or responsibilities.

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

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

- This project contains both alkaline fuel cells and alkaline water electrolysis components. Therefore, it supports general goals and objectives of the Hydrogen and Fuel Cells Program. The product may still be far from the commercial product at the end of the project. Therefore, it may not help much on market penetration, but the potential impact still exists.
- This approach (i.e., of using a regenerative fuel cell for remote energy storage and power generation) is potentially much more cost-effective when compared to battery systems. The approach is very useful if deployed in island regions, wind turbine farms, etc.
- This is a project of strategic value, as AEM is the system in which completely PGM-free catalysts are possible.
- Relevance is not very high since there are no energy storage targets in the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan. However, the reviewer thinks this work is very relevant and should be continued *only* if it shows promise compared to other energy storage technologies.
- Unitized reversible fuel cells could have a big energy impact by helping enable intermittent renewables. However, the high cost, low performance, and low durability of the current technology means that a huge gap exists between current status and commercial requirements. The odds of this technology actually being deployed on a large scale seem small; therefore, the relevance is questionable.
- There is limited market and applicability of a regenerative fuel cell approach with the performance characteristics of an AEM. This is not only because of the low technology readiness level (TRL) of the individual components, such as the membrane and ionomer, but also because the economics do not work out. A PEM-based system, while still not ideal, would have a drastically lower cost, owing to the smaller cell size to achieve the same storage capacity. Cheaper still would be a discrete system.
- A 42% round-trip goal is too low to compete with batteries at 80%.

Question 5: Proposed future work

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

- The approach for the next budget period will aim to quickly transition the components from a dissimilar test cell and duty cycle to a commercial system that will struggle to operate under the required conditions for commercial applications, for example, having a potassium hydroxide feed to the membrane through wicking, and operating the anode on vapor during electrolysis operation. Before scale-up and five-cell testing, the targets should be met while operating the cell under the same conditions as the full-scale stack. As an aside, it does not make sense to utilize a carbon-based catalyst for the OER. Carbon oxidizes above 1.2 V, and it is unclear how it is stabilized by P and N. If this were true, the automotive fuel cell market would have switched to these as supports.
- The PI addressed the future work, which makes sense in light of the current project progress. However, no clear pathway is suggested to overcome the current system's low performance. It is questionable that the team can actually resolve those issues.
- Planning forward as described should enable the team to successfully demonstrate Phase II.
- In addition to what the investigators proposed, they should consider AEMs and ionomers with high-temperature stability so that they can raise the temperature and improve performance.
- The cell needs to be operated under more realistic conditions in terms of gases/humidity, potentials, and time period of charge (electrolyzer)/discharge (fuel cell) during the 1,000-hour durability test. A clear business case needs to be developed (e.g., a niche application) before the end of the project.
- It is unclear whether practical operating windows should have same current density for both modes. It is also unclear how to run this system in a real case.
- The future work is appropriate.

Project strengths:

- The PI has excellent expertise in PGM-free catalysts. The initial durability results look promising. Giner, Inc., offers great expertise in unitized reversible fuel cells.
- The team has good collaborations. The PGM-free catalysts developed from Phase I look promising. The project did a nice demonstration of excellent durability with the Pt-free electrode.
- The overall concept, when complete, will bring great benefit in remote regions.
- The demonstrated performance under the conditions used has allowed the project to meet targets.
- The team of Giner, Inc., and NREL is very good.
- This is a good project team that has made extensive progress.
- This is a good team on an excellent topic.

Project weaknesses:

- Regenerative AEM fuel cells do not make economic sense, and many of the components are at too low of a TRL to enable this technology, even if the concept was feasible. Carbon-based catalysts are fundamentally unstable and ill-advised for this application. Limiting the current of the electrolysis duty cycle coupled to wind that is variable also does not make sense; load following is one of the advantages of electrolyzers. The targets should be addressed at subscale, using realistic cell and operating conditions before scale-up to a five-cell stack that will inherently operate differently.
- The team should perform comparative cost-benefit analysis of non-PGM cathodes versus the potential performance enhancement that the Pt/Ni alloys, etc., can deliver. Then the team should perform measurements to substantiate the analysis results.
- The project is overly ambitious. It would be better to focus on one or two technological barriers, rather than try to develop a technology that faces so many challenges and is so far away from commercialization.
- The project provided very little information about the developed catalysts. If the structure of the catalysts is a trade secret, it should be a less favorable project, and DOE should reconsider supporting such a project.
- The project needs to have a clearer understanding of where the market is for this application, and what targets need to be met in order to make this technology successful.
- The understanding of real-life dual-mode requirement is not clear.

- The project is likely in need of a better membrane and ionomer.

Recommendations for additions/deletions to project scope:

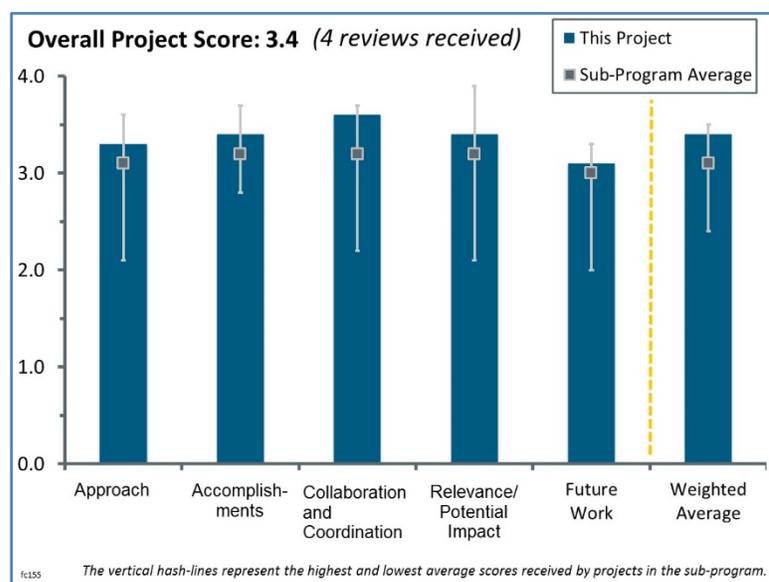
- The team should do a cost analysis comparing PEM-based discrete systems and a variety of operating duty cycles. The team should also do a long-term durability test in fuel cell and electrolysis mode at equivalent current densities. This would include catalyst characterization (pre- and post-mortem) to determine catalyst and support stability, and membrane short checks to determine whether the cell is shorting. The project should continue testing under relevant conditions, meaning relevant cell, materials, and flow rates, as will be used in the final device.
- The work with Pt-Ni nanowire for anode catalysts may be deleted, as the team has a Pt-Ru control catalyst, and the objective of the project is developing a non-PGM catalyst.
- New membranes and ionomers with higher temperature stability should be considered.
- It is necessary to match the current density 40 and 200 mA/cm².

Project #FC-155: Novel Ionomers and Electrode Structures for Improved Polymer Electrolyte Membrane Fuel Cell Electrode Performance at Low-Platinum-Group-Metal Loadings

Andrew Haug; 3M

Brief Summary of Project:

The objective of this project is to develop novel ionomers and electrode structures to improve polymer electrolyte membrane (PEM) fuel cell performance and durability. The focus of the ionomer development will be on combining high proton conductivity with improved oxygen transport. The project also seeks to understand and optimize novel cathodes that utilize nanostructured thin-film (NSTF) catalysts in powder form. These powder catalysts will be integrated with the ionomers to develop an advanced cathode of high activity and durability. State-of-the-art novel characterization and modeling techniques will be used to guide these development efforts.



Question 1: Approach to performing the work

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

- This is a well-thought-out approach with a good plan, milestones, and targets. The project mitigates risk by having two different strategies to minimize localized mass transport resistance in the cathode catalyst layer. Making powder out of NSTF is a new approach that is timely. The main issue (and potential strength) is that this results in completely different catalyst layers and interactions. It is unclear how much work will go into characterizing these structures, as opposed to figuring out if they help to improve localized mass transport.
- These NSTFs are a good approach to maximizing Pt utilization when the catalyst is decal-transferred, a path to minimizing the amount of Pt used in a fuel cell. It mostly addresses initial power density. It was not clear whether using powders derived from NSTFs would have some Pt “buried” by other Pt-coated whiskers overlapping another Pt whisker and another. It is also not clear whether the Pt would redistribute on a whisker. Maybe this was shown in the past, but this was not shown during this talk, so agglomeration of nanostructures on a whisker is one possible loss of Pt area per mass of Pt.
- Individual approaches are good, particularly ionomer characterization for the electrode. Powdered NSTF seems to be a developmental project and not suitable for the Fuel Cell Consortium for Performance and Durability (FC-PAD).
- Work on ionomers and NSTF represents strong technical contributions from 3M. However, there is not really a synergy between the two, and it seems almost like two separate projects: one on ionomers and one on NSTF dispersion. Exploring the ability of NSTF to be dispersed and incorporated into more traditional catalyst layers is of value, as is the work on novel ionomers (perfluoroimide acid [PFIA] and multi-acid side chain [MASC]). The ability of NSTF to break the trend for transport resistance losses at low surface enhancement factors may be due to the lack of ionomer in the electrode structure. More emphasis on local oxygen resistance and limiting current measurements would be useful to help elucidate the role of ionomers in these electrodes.

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 team has a clear and effective plan and is following its design of experimenting and measuring the surface area and performance as a function of Pt mass in a cell. This will lead to a clear understanding, whether or not the approach completely succeeds.
- There are good outcomes from catalyst layer modeling utilizing a pore-network model to identify characteristics of oxygen permeability and proton conductivity, as well as their limitations.
- Good progress has been shown in a short time, including encouraging ionomer and catalyst layer activities.
- The project is in the early stages and has good results for such early efforts. The work in ionomers has shown the potential advantages of MASC over PFIA and perfluorosulfonic acid (PFSA). The work on NSTF does not seem to have particularly high cell performance and seems to be significantly lower than the standard NSTF membrane electrode assemblies (MEAs). More direct comparisons between the performance of dispersed electrodes and standard NSTF would be beneficial. Additionally, the highest-performing NSTF samples would be of greater interest, as those studied to date do not appear to be the highest performers.

Question 3: Collaboration and coordination with other institutions

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

- The team led by 3M (M. Lindell, T. Matthews, J. Abulu, M. Yandrasits, A. Steinbach, M. Kurkowski, G. Weatherman, G. Thoma, I. Khan) has well-defined roles to address Pt dispersion, catalytic activity, and mass transport as a function of metal and polymer structures in the catalyst layer. Tufts University (Iryna V. Zenyuk, D. Sabarirajan, S. Normile) characterizes nano- and microstructure by direct imaging. Michigan Technological University (J. Allen, K. Tajiri, E. Medici, and team) is doing transport modeling. FC-PAD (Lawrence Berkeley National Laboratory [A. Weber, A. Kusoglu], National Renewable Energy Laboratory [KC Neyerlin], Oak Ridge National Laboratory [K. More], Los Alamos National Laboratory [R. Borup], and Argonne National Laboratory [D. Myers]) are helping with performance and durability validation. There is a challenge to correlating the nanostructure to modeling of transport when powders are used. The powders can bury some Pt by stacking the whiskers, and tomography and modeling will be needed to sort out whether this is a problem. FC-PAD will be needed to see whether Pt is redistributing in a whisker (as evidenced by loss of activity as Pt nanoparticles grow).
- Everything seems well organized, and there is good communication between the principal investigator, partners, and FC-PAD. The approach focusing on developing capabilities in budget period (BP) 1, and integration beginning in BP 2 is good.
- Good collaboration is pursued between industry and academia and also materials synthesis/empirical approaches and modeling.
- It is still early in the project, and it is not clear that Michigan Technological University had any specific contributions. This may be due to contracting delays. The project seems well coordinated with FC-PAD and can leverage efforts into this project.

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

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

- The project looks to take on the key challenges of fuel cell cost, performance, and durability by creating high-performance, low-loaded electrodes. The team's work in both novel ionomers and catalysts is interesting and relevant.
- The project has relevance to the Multi-Year Research, Development, and Demonstration Plan fuel cell performance targets for 2020 and is very relevant to current performance-limiting issues.

- This is one straightforward way to test Pt dispersion and stability in well-defined structures initially, and as a function of usage and time for catalysts in hydrated PFSA ionomer catalyst layers.
- Overall tasks of the project are relevant to the FC-PAD objectives, characterization, and analysis, not new development. This means that the project should focus on tasks to fill out the knowledge gap rather than the developmental gap. It is not suitable to work only on materials development to meet the target. Investigating why it works (or does not work) is expected. Electrode ionomer characterization is good. Powdered NSTF should be reconsidered to make it more relevant to FC-PAD.

Question 5: Proposed future work

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

- The full work plan is clear and concise, with appropriate goals and go/no-go decisions divided by task and subtask. Proposed work progresses logically and brings in collaborators to work on key issues.
- The team is following its design of experiment. It should lead to good evidence to make sound conclusions. It would be good to see some consideration of Pt ripening per whisker and the effects of stacking of whiskers in the imaging/modeling. This should be correlated with the performance to see whether these issues are relevant.
- The general plan to further study 3M's novel polymers and catalysts is fine. However, the specifics of how the project will investigate these areas lacks clear direction or next steps. This statement reflects general trends from the presentation, in which acronyms are used often without definition, and the presentation has the feel of needing further revision and thought for both layout and content. The specifics for next steps have not been presented in a clear, concise manner, nor are the key variables or learnings from the next steps presented in a way to lend confidence in obtaining positive results.

Project strengths:

- This is a good approach to minimizing Pt usage by dispersing in thin films. The new approach of using different amounts of whiskers of different Pt loadings with different polymer loadings is a good way to expand the design of experiment for Pt activity and transport of protons, water, and oxygen at Pt in the catalyst layer. Imaging and modeling need to perform validating and predictive roles, respectively.
- 3M's capabilities in novel ionomers and catalysts (NSTF) are a strength. Exploring dispersed electrodes as a pathway to overcome current NSTF limitations is another strength.
- Materials synthesis and empirical approaches for ionomer and electrodes are a strength. Pore-network-model application to the catalyst layer is a strength.
- This is a great team, work plan, and approach.

Project weaknesses:

- The approach is very good. It would help strengthen the argument if there was some consideration of other possibilities besides those in the original design of the experiment—possibilities such as the effects of Pt active area lost by whisker stacking and Pt agglomerating within a whisker.
- A clearer experimental plan and next steps are needed. Why specific approaches are going to be undertaken and what is expected to be learned at each step need to be clarified.
- MEAs based on NSTF powder are new. There are plenty of variables, but the initial data look good.

Recommendations for additions/deletions to project scope:

- It is suggested that the project reconsider its approaches to make them more relevant to the FC-PAD objectives of characterization and analysis, not new development. It is not suitable to evaluate the progress and go/no-go decisions based on the performance target. It should be evaluated based on the knowledge gained, such as characterization and why the technology works or does not work.
- This is actually two projects, and it would be preferable for the focus to be on dispersing NSTF rather than investigating multiple ionomers. Both approaches are important, but the more critical one is focus on dispersed NSTF, while other projects (perhaps within FC-PAD) focus on alternative ionomers.

- No real additions or deletions are needed, just some attention to detail—that is, the effect of whisker stacking and Pt redistributing on a whisker. Polymer addition and mixing are just fine.

Project #FC-156: Durable High-Power Membrane Electrode Assemblies with Low Platinum Loading

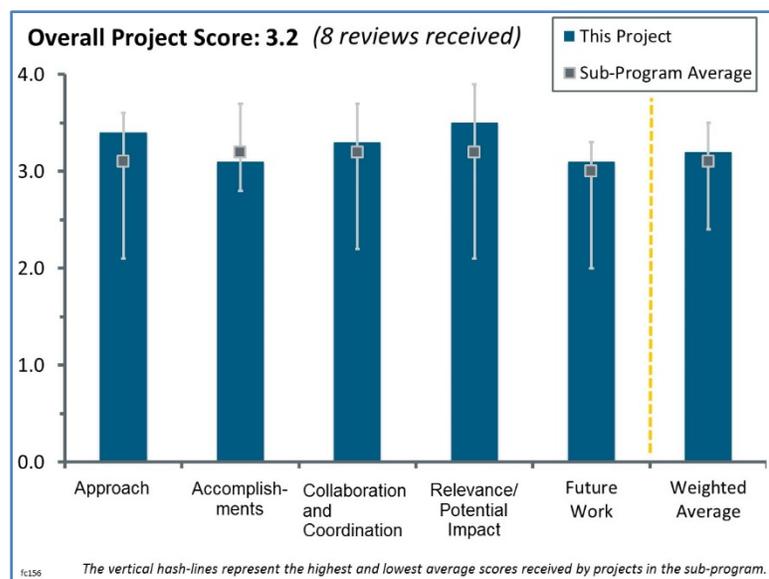
Swami Kumaraguru; General Motors

Brief Summary of Project:

This project seeks to improve the durability of a state-of-the-art (SOA) membrane electrode assembly (MEA) by identifying and reducing the stress factors affecting electrode and membrane life. Project tasks include (1) MEA optimization of a low-loaded electrode through down-selection and integration of MEA components, (2) durability studies of the developed MEA, and (3) development of a predictive model for degradation in different operating conditions.

Question 1: Approach to performing the work

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



- The approach for assessing electrode and membrane durability follows the standard DOE protocols. Nothing novel was demonstrated by the model approach, but it was nevertheless well defined. The Fuel Cell Consortium for Performance and Durability (FC-PAD) is well equipped and experienced to measure cerium ion migration in membranes, as the consortium published seminal work in that area. Segmented cells by their very design may introduce mechanical stresses within the catalyst-coated membranes that are not present in ordinary stacks.
- The project's work plan seems appropriate. The schedule looks to be based on three men for three years, plus operational cost and development hardware. The schedule looks aggressive, similar to an industry schedule, but obtainable.
- The project builds on good results from previous projects, projecting to have a SOA MEA within the first year after implementing a few iterations with select components. The only improvement could be to reserve time for optimization of this MEA so that it shows a stable performance at the targeted level as a whole.
- The project approach was well explained and logical and is integrated well with FC-PAD activities. The plan addresses relevant barriers.
- The focus on durability of SOA MEAs is clearly relevant. The approach is fairly comprehensive: first, screening potential SOA MEAs; then, focusing on a specific MEA for extensive durability studies; and finally, developing comprehensive models of the durability concerns. This is a reasonable approach, but one would expect General Motors (GM) to already have a SOA MEA, and much of the first-year optimization effort could likely have been avoided, and comprehensive durability studies could have started more quickly. More detail on model status and model development would have been beneficial, as it is not clear what the current status of the models are and how they will be further advanced.
- The project is to develop a SOA MEA, develop models for electrode and membrane durability, and determine benign operating conditions. Undoubtedly, there will be significant value in the work; however, there are a number of areas of potential concern.
 - It appears that the development of the SOA will be a benchmarking exercise. It does not appear that the design will be model-driven.
 - The electrode durability model does not appear to advance the SOA of modeling and will be based on empirical design of experimental data. The fuel cell industry should be moving beyond this type of approach and further build on the fundamental models based on mechanistic studies.

- Based on the use of an empirical approach and the U.S. Department of Energy, the determination of benign operating conditions will be MEA-design- and test-set-up-dependent. Furthermore, the selection of operating conditions is very dependent on specific application and system design. It is not clear whether the peroxide vapor test is representative of in situ mechanisms; otherwise, the membrane degradation studies/modeling will provide valuable insight. It is important to include voltage effects in the membrane degradation model. The studies shown on slide 12 for budget period 1 will provide valuable insight; however, these do not seem to be represented in the milestones. If these are included, valuable insight will be gained. Further, this type of work is also described in FC-137, but it is not clear whether the approaches and models will be in common with Lawrence Berkeley National Laboratory (LBNL).
- The objective of this project is to improve durability of the SOA MEAs by identifying and reducing the stress factors affecting electrode and membrane life. The project's approach relies on developing an electrode durability model for the power degradation rate caused by voltage cycling and a membrane durability model combining mechanical and chemical degradation with Ce migration.
- The project is new but planned. Its approach appears well designed and feasible.

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.

- This is a new initiative awaiting the official start of technical activities. The principal investigator (PI) discussed the planned activities for SOA MEA selection, test matrices for electrode and membrane degradation, model development for performance and aging, model validation, and model application.
- Even though the project has just started, it shows the potential to accomplish the goals.
- This is not a relevant question, as the project was just funded. The collaborations between the parties are well mapped; therefore, the project should make progress within the first year. Not Applicable is really the correct answer for this question; however, the site does not accept that input.
- As the project is just starting, this review criteria is not particularly relevant, as no accomplishments have been achieved or presented.
- The project just started in January, so it is difficult to really measure progress. Consequently, this question received a neutral score.
- The project has not started past contract planning. The accomplishments are appropriate for this point in the project.
- The project is new; there are no results.
- The project has just started.

Question 3: Collaboration and coordination with other institutions

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

- GM, as the prime contractor, has significant experience in this area. Both Giner and the University of Texas at Austin (UT) bring very specific, complementary competencies. The FC-PAD laboratories have complementary roles as well.
- It is early in the project to properly assess how effective the collaboration will be. Collaboration is likely to be good based on the FC-PAD/funding opportunity announcement structure, which has been designed to encourage collaboration.
- The project has Giner and UT as subcontractors. The project has significant contributions from the FC-PAD team.
- Considering the project objectives, there is a balanced consortium among the partners selected.
- The project's planned collaborations appear to be well suited for work to be done.
- The planned collaborations seem appropriate for the proposed scope of work.
- Project collaborations include the appropriate parties, with the exception of a tier-one membrane manufacturer.

- The project collaboration highly favors national laboratory researchers over academic or industry researchers. The universities have most of the same toys and can do the work for less expense and usually less time. Their undergrads are the next generation of electrochemists.

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

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

- This project is designed to have a large potential impact on DOE's goals related to fuel cell cost, durability, and performance. Understanding degradation dependence on operating parameters and creating a model for this degradation set this project apart from other projects in this area.
- Improving durability of SOA, low-loaded electrodes is a key need for industry. Development of an understanding of operational stressors and means to minimize will be useful.
- The focus on SOA MEAs with improved durability is critical for the advancement of fuel cell applications, particularly in vehicles. The breadth of what is being attempted is commendable but also makes the project perhaps broader and less focused on the most critical aspects. The focus on alloy catalyst durability and electrodes has been highlighted as a major driver for this work, and seems to be the most critical part of the effort. Adding membrane degradation, including complex and/or difficult approaches such as specific shorting defects, will be a challenge to achieving meaningful advances and runs the risk of reducing the advances in the catalyst/electrode area.
- The outset is good to provide the pathway to meet the cost, performance, and durability targets, but eventually it may provide an overview of benign conditions for individual components that could be mutually conflicting, thus not clearly giving direction or a single failsafe solution. It is recommended that in addition to the map of "operational danger zones," the final outcome could advise how non-SOA components could also meet DOE targets.
- The access of GM to SOA materials will likely result in the study of very relevant MEAs. The association with FC-PAD will ensure detailed characterization and modeling studies. The membrane degradation model and associated studies have not been done in detail previously and will be highly relevant. However, the MEA performance degradation model does not appear novel, and the design of experiment approach may result in dependencies that limit the applicability of the model for other systems.
- This task is to develop a new generation of polymer electrolyte membrane fuel cell with a lower cost and higher performance, in a short length of time. It will be interesting if the national laboratories can step up to the pace.
- The project is aimed at demonstrating a pathway to cathode and membrane life of 5000 hours with existing non-proprietary materials by defining implementable benign operating conditions.

Question 5: Proposed future work

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

- The project's future work is well planned and is expected to produce reasonable progress toward DOE and project goals.
- The project has yet to start. The team's plan looks sound. It is a standard electrode development program.
- The PI defined the future activities related to developing fundamental models, formulating correlations, validating and fine-tuning models, and applying models.
- The project is well laid out, with appropriate milestones.
- The focus on electrode durability from both an experimental and modeling perspective is very strong and needed. The efforts in membrane durability are less focused, attempting to include both mechanical and chemical aspects.
- The project's future workplan is good; however, the weighting of the selection criteria for SOA is a bit vague.
- The project plan seems reasonable. It was somewhat unclear whether any catalyst development is planned beyond year one. It may be required based on durability results in year two.

- The validation of the project's models will show true value of the work and make it transferable.

Project strengths:

- The project is unique from the many other cathode-related projects because significant effort will be placed on studying correlations between operating conditions and degradation. This will help build a nice predictive model for how long electrodes will last in the actual application. The project also considers real-world issues related to membrane degradation that are overlooked in other projects.
- GM has demonstrated its ability to make high-performance, low-loaded MEAs. The project has access to SOA MEA materials. The coordination/collaboration with FC-PAD will ensure use of detailed diagnostics and will feed into mechanistic models. The project has a fairly comprehensive and novel approach to membrane durability studies.
- The team has a well-designed project plan. Access to SOA materials makes the project highly relevant. Project organization and collaborators are good.
- There is strong teaming, particularly in the durability testing and characterization aspects of the project.
- The project team is a strength. The team showed strength by focusing on the durability aspects of SOA MEAs, including alloy catalysts.
- The project has a good starting point by taking the best-known components and supplier to work with, giving the project a proper chance of success.
- GM's project experience is a strength.
- The project has a credible team and partners.

Project weaknesses:

- All of the MEA development is in year one, but it seems that the majority of degradation characterization will be performed in year two. Feedback from the work in year two to further optimize the MEA should be added.
- The objective of establishing benign operating conditions does not seem particularly valuable, as operating conditions are very application-, component-, and system-dependent. It is not clear how coordinated the electrode modeling and mechanistic studies will be. MEA design appears to be by benchmarking, rather than model-driven.
- It would have been good to have seen participation from Gore, 3M, Ballard, IRD Fuel Cells, or other MEA manufacturers.
- The breadth of the project will be a challenge. Membrane durability efforts are not as likely to result in meaningful advances.
- The individual component approach may not capture (reversible) effects observed in reality when exposing the MEA as a whole to continuous operation.
- It is unclear how well, if at all, project results will be translatable to the field as a whole if the project is using GM-proprietary materials exclusively.
- The project has a high reliance on national laboratories being on budget and schedule.

Recommendations for additions/deletions to project scope:

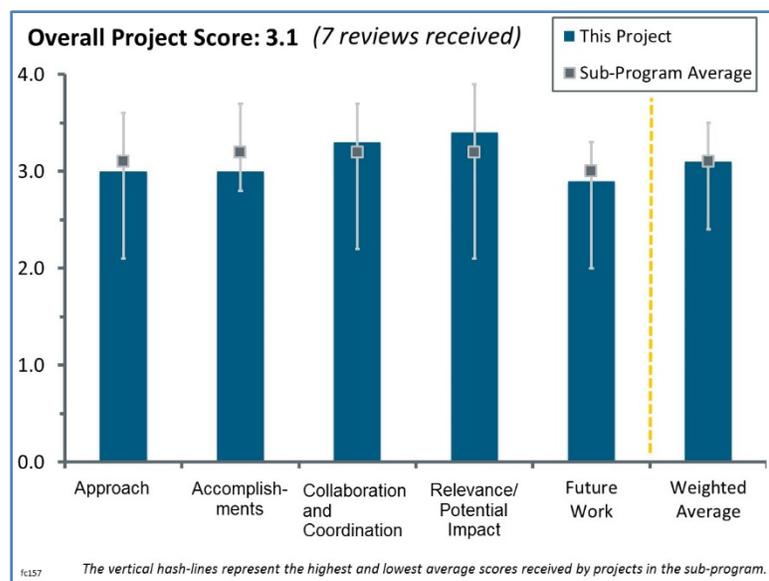
- The team should consider using lower-cost academics to meet budget and schedule. Clemson, Hawaii Natural Energy Institute, University of Connecticut, University of South Carolina, etc. have all demonstrated this ability in the fuel impurity testing. Additionally, universities are used to working under non-disclosure agreements.
- Definition of scope and experimental parameters may be needed when addressing the influence of mechanical stress together with chemical degradation.
- The project should consider focusing only on electrodes at the expense of membrane studies. For the level of funding, it is likely to have an increased impact and a clearer path to meaningful results.
- The team should ensure coordination with LBNL-led work on understanding thin-film losses and ionomer studies.
- Feedback from the work in year two to further optimize the MEA should be added.

Project #FC-157: High-Performance Polymer Electrolyte Fuel Cell Electrode Structures

Mike Perry; United Technologies Research Center

Brief Summary of Project:

The objective of this project is to improve the fundamental understanding of transport limitations in state-of-the-art (SOA) membrane electrode assemblies (MEAs) for polymer electrolyte membrane fuel cells and use this knowledge to develop and demonstrate high-performance MEAs with ultra-low catalyst loadings (ULCLs). Transport losses are a major barrier with ULCLs, but fundamental understanding of those losses is currently lacking. To gain better understanding of the nature of these losses in cathode catalyst layers, a detailed microstructure model of the cathode catalyst layer will be developed. This improved knowledge will then be utilized to develop improved MEAs that meet the U.S. Department of Energy's (DOE's) performance targets.



Question 1: Approach to performing the work

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

- The project is well aligned with DOE targets, as it addresses high-current-density performance losses for catalysts with ultra-low Pt loading. The project is well designed. It relies on a combination of SOA experimental work and modeling in order to clarify the origin of high-current performance losses. Although the focus is on catalyst structures, some of the proposed structures do not seem to be very relevant.
- Transport limitations at high current density and low loading remain as a barrier for the efficient deployment of fuel cell MEAs. The focus on Pt-only MEAs makes sense for focusing on local transport resistance but makes the tests less relevant for true SOA materials. Efforts along these lines have been common in the community lately, but the core team for this project has not been leading this area and does not have the strongest background to undertake these studies. The four simplified cases shown in slide 10 are key to connecting models to the experimental results obtained, and the complexity of the real system will result in significant challenges in connecting these efforts, with significant risk that little will be connected between the model and experimental results. No data have been presented yet to suggest that the models can be effectively applied to trends seen experimentally. There will be challenges in applying the model thin-film catalysts being developed to MEAs.
- The generic modeling approach (slide 10) seems to be good. The question is how to validate the model for each mass transport loss, total oxygen transport resistance, and local oxygen transport resistance. The project set go/no-go for this model validation. It is necessary to clarify the metric of the validation. It is also necessary to see effectiveness of operating conditions. Measurement of mass transport loss with the oxygen partial pressure method (various oxygen partial pressures) is not sufficient to distinguish total oxygen transport resistance from local oxygen transport resistance. Investigation of the novel thin-film catalyst later does not fit the Fuel Cell Performance and Durability (FC-PAD) consortium objective (characterization, not novel concept development).
- The project constitutes an effective combination of theoretical and experimental work to elucidate the causes of local transport losses in low Pt-loaded electrodes, a critical barrier to fuel cell commercialization, and to start on the road of mitigating these losses. The project seems to be overly

wedded to the idea that the local transport losses arise from details of aggregate structure, ignoring the possibility of losses right at the Pt-ionomer interface.

- The team has taken a good approach of understanding the fundamentals of transport limitations in SOA MEAs and then using the learnings to develop high-performing MEAs. Development of a microstructure model of cathode catalyst layer is expected to shed light on different modes of transport limitations. The project is new, and very little technical work has been done to assess the significance of the approach made by the team.
- The team has a good approach, but it might be helpful to add additional details on how the limiting case study MEAs are going to be fabricated.
- As the principal investigator (PI) noted, the project relies heavily on inventing methods to prepare electrodes with sufficient differentiation to identify model parameters. However, the PI did not share a clear plan to achieve these goals. The project team does not currently have that capability and likely will need to rely on FC-PAD to come up with a method. The characterization methods being used appear to be similar to previous projects. It is unclear what new insight the project will bring.

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 results presented were reasonable for a project at the beginning stages. The most interesting/useful results to date were in the area of model development. Isolating four limiting cases based on agglomerate morphology can help to provide insight into the catalyst layer structure/performance relationships. MEAs were fabricated and tested but did not provide any new insight or results that were not expected, rather just verification of the ability to synthesize relevant materials. The work in the area of thin-film catalyst development was useful at a rotating disc electrode level but will have significant challenges in transferring to useful MEA results.
- The project is new; it started in January 2017. The accomplishments achieved by the team in three months, which are cited in the presentation, are good. The prime (United Technologies Research Center [UTRC]) has organized the team and gotten the contracts signed. The relationship and communication tools to run the project are in place. The project is in a very early stage, and not much time has been spent on the project. The team's progress is satisfactory within the given time they have been involved in the project.
- Although none of the DOE targets have been exceeded yet, progress has been made toward development of a microstructured model of the catalyst layer, fabrication of SOA MEAs, and evaluation of activities of thin-film catalysts with porosity gradient.
- The project is still in an early stage but is already showing some good results.
- Even considering that the project has just started, it is disturbing that the Ion Power, Inc. MEAs are still just trying to match the performance of SOA low-loaded MEAs, and that no start has yet been made on producing MEAs with the controlled structures shown on slide 10. It is not clear that estimates of aggregate diameters and ionomer film thicknesses necessary to generate the experimentally observed local transport resistances have yet been made for the cases shown on slide 10. Such estimates should have been compared to Oak Ridge National Laboratory's measurements of actual electrode structures prior to generation of the proposal, and they need to be done early in this project to minimize wasted effort.
- The project just started.

Question 3: Collaboration and coordination with other institutions

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

- This project will likely be a showcase demonstrating the benefits of close coordination between industry projects and the characterization and theory capabilities of the national laboratories in the FC-PAD consortium. If Ion Power can actually control electrode structures to approach the limiting cases shown on slide 10, the collaborations in this project will be unusually fruitful. The presentation did not make it clear how the thin-film electrodes being made at the University of Arkansas at Little Rock will contribute to the understanding and mitigation of local transport resistances.

- The team comprises capable scientists and organizations to run the project. The team is in the early stage of coordination, and all collaborators in this project have past experience in working on a DOE project. As a part of FC-PAD, the team has access to the great resources from other FC-PAD member organizations and scientists involved in FC-PAD. If needed, the team can consult with other FC-PAD members.
- Although the project just started, there seems to be an excellent collaboration between Ion Power, UTRC, and the University of Arkansas.
- The project has reasonable collaborations with Ion Power to synthesize MEAs and a university collaborator to aid in catalyst synthesis. The project will have high dependence on FC-PAD core members. The connection with Lawrence Berkeley National Laboratory for modeling is one that will be particularly critical.
- The project has a good team and can leverage the national laboratories through FC-PAD further.

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

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

- This project is tightly focused on the local transport resistances, which now constitute the major barrier to the commercial application of low-Pt-loaded cathode electrodes, which is the key to economic viability of fuel cells in most applications. This project should generate the fundamental understanding of the still-mysterious local transport resistances needed to enable design of MEAs fully meeting DOE's performance targets.
- The project is relevant to DOE's 2020 target. Proposed performance comparison of SOA and high-activity oxygen reduction reaction (ORR) catalyst MEAs to understand the mass transport limitations is needed before developing a ULCL or thin-film catalyst layer (TFCL) to reduce the catalyst loading. It will be good to know how the increase in fluctuation rate affects the catalyst sites and affects the transport rates in an MEA. The understanding of such mass transport loss may help in reducing the mass transport loss and hence improve the performance of the ULCL and TFCL.
- The project has the potential for significant impact on the DOE Hydrogen and Fuel Cells Program, as stack cost is one of the most important metrics for commercialization of polymer electrolyte membrane fuel cells.
- Increasing the high-current-density performance of fuel cells is clearly a primary challenge to decreasing cost and increasing performance of fuel cell systems. The improved understanding of the mechanisms of overpotential losses can aid in mitigation of these losses.
- Detailed characterization of the cathode catalyst layer for the mass transport losses is critical for MEA development (performance and cost).
- The project is highly relevant and could have a big impact if the team can gain understanding of the root cause of high-current-density performance loss.
- The project does not intend to address DOE targets directly but attempts to identify key limiting parameters of high-current-density performance.

Question 5: Proposed future work

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

- As the project is very new, the proposed future work presents the details of the work proposed by the team in this project. The team has described the quarterly deliverables and the go/no-go stages for the duration of the first half of the project. The proposed future work for the duration of the first half of the project is aligned with the overall goal of the project.
- The project's proposed future work is well planned. It relies on a combination of modeling and experimental work to provide insights into the origin of transport losses at high current densities. There is no clear path to make outlined catalyst structures that are intended to be used for model validation.
- The question is how to validate the model for each mass transport loss, total oxygen transport resistance, and local oxygen transport resistance. The project set go/no-go for this model validation. It is necessary to clarify the metric of the validation. It is also necessary to see the effectiveness of operating conditions.

Investigation of the novel thin-film catalyst later does not fit the FC-PAD objective (characterization, not novel concept development). Measurement of mass transport loss with the oxygen partial pressure method (various oxygen partial pressures) is not sufficient to distinguish total oxygen transport resistance from local oxygen transport resistance.

- The proposed future work has a number of overlaps with existing results in the literature investigating high-current, low-loading operation, particularly work reported by General Motors. A major addition would be the ability to selectively create structures in the electrodes that approximate the structures in slide 10. At this point, it is not clear that the agglomerate structure can be controlled or influenced effectively through MEA fabrication modifications. For thin-film catalyst development, it is not clear that the thin films being developed can be transferred effectively to MEA incorporation.
- The project's proposed future work is sound; however, the team is heavily dependent on the ability to create and characterize the limit-case electrodes. Therefore, there needs to be some alternate factorial designs to improve the probability of success.
- The proposed work, if successful, should provide a clear test of the aggregate theory of local transport resistance. Insufficient information was provided to give any confidence that model electrodes distinguishing between the different transport modes as diagrammed on slide 10 can actually be grown. In the absence of such controlled electrodes, it is not clear that the project could generate clear conclusions about the true mechanisms of local transport.
- It is not clear what, how, or when tasks will be done, or by whom. The milestone criteria are also not specified.

Project strengths:

- The objective of attempting to understand the mass transport loss at the fundamental level is one of the important steps in the development of high-performing MEAs with ULCL- or TFCL-type low-catalyst-loaded electrodes/MEAs. The team consists of knowledgeable scientists and a knowledgeable PI, who are capable of conducting the proposed research. The team contains the FC-PAD consortium, allowing the FC-PAD team members to coordinate relevant information exchange with the project. This will help in the project's progress to maintain its direction as well as to input any relevant information that may come from other FC-PAD projects. The approach taken by the team is also a strength: working at the microstructure level to gather understanding of mass transport loss and to develop a model to direct future high-performing MEAs with ULCLs to meet DOE's 2020 target.
- Project members, in collaboration with the characterization capabilities of FC-PAD, are well situated to determine whether the local transport resistances observed for low-loaded cathodes arise from details of carbon and/or ionomer aggregates. The presentation evinced a good understanding of the complexities of local transport issues on the part of the PI.
- The major project strength is in applying unique UTRC expertise in modeling and diagnostics to elucidate the origin of high-current losses in the rationally designed catalyst layers.
- Quality experiment capability for MEA performance is a project strength, as is the modeling capability for mass transport losses.
- Modeling at the agglomerate scale is a good addition to the established fuel cell models.
- The project has a good team and approach.

Project weaknesses:

- Regarding Ion Power MEA, the initial result shown in slide 12 with the Ion Power-developed MEA (B1286) contains three times the Pt loading as the baseline SOA MEA (B1240), while it performs close to the SOA. As the team has identified, more work is needed to improve Ion Power's MEA performance to match the SOA MEA performance. It was not clear why the team chose the high-loading MEA from Ion Power, as it may be having conventional agglomeration issues leading to mass transport loss. In ULCL, the mass transport loss mechanisms could be different. There is another weakness regarding the density gradient electrode (DGE): in slide 13, the team has proposed the use of a gas diffusion layer (GDL) as the substrate. Using the membrane as the substrate will allow the porous top of the DGE to be against the GDL, and then the porous part of the electrode will allow better oxygen transport into the electrode to support the high kinetics of ORR. Using the GDL as the substrate will invert the configuration, and the porous part of

the DGE will be close to the GDL (i.e., away from the membrane, and the porous part of the electrode will be against the membrane), which will not allow better oxygen flow into the membrane–catalyst interface. The team needs to look into the configuration of the proposed DGE.

- It is not clear how the thin-film electrode work at the University of Arkansas contributes to the overall goals of the project. It is not clear how model electrodes with controlled aggregate forms and ionomer film thicknesses can be grown. The project apparently has not yet plugged the numbers into its math models to determine the sizes of aggregates and thicknesses of ionomer that would be required to give the experimentally observed local transport values. The estimated first-order dimensions may be large enough that the aggregates should be more obvious in microscopy than have been seen to date.
- The project weakness is in uncertainty with fabrication of catalyst layers with a given structure, both for carbon-supported and thin-film catalysts. Carbon support has not been given a proper consideration in the modeling and experimental work
- The project team is behind the SOA in the area of high current, low loading. The efforts in novel thin-film catalyst development are going to have significant challenges in extending to fuel cell tests.
- It is too early to say and unclear how they can help move the bar on the DOE 2020 goals.

Recommendations for additions/deletions to project scope:

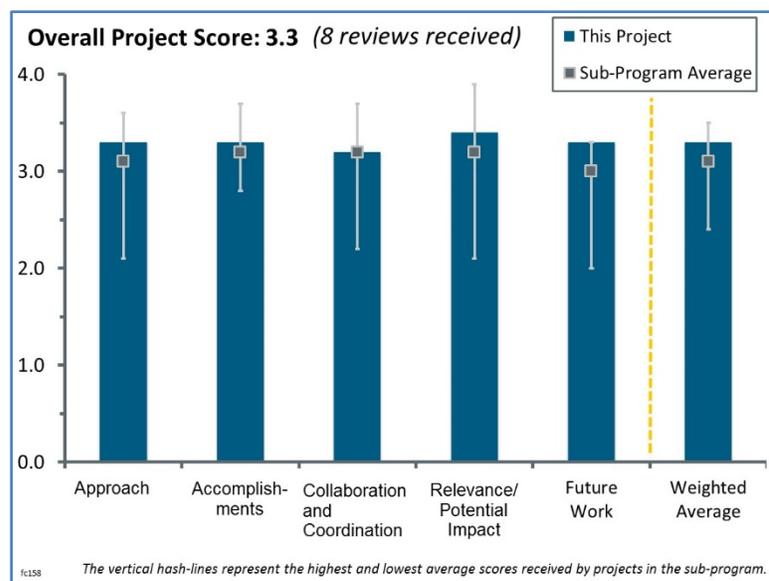
- It is suggested that the project clarify the metric of the validation of the mass transport losses model. The measurement of mass transport loss with the oxygen partial pressure method (various oxygen partial pressures) is not sufficient to distinguish the total oxygen transport resistance from local oxygen transport resistance. It is necessary to propose alternative methods (or additional measures). Investigation of the novel thin-film catalyst later should be reconsidered to make it fit with the FC-PAD objectives, which are focused on characterization rather than new concept development. The catalyst should be evaluated based on the knowledge gained, such as characterization and why it works or does not work.
- Novel catalyst synthesis seems to be of limited value. It would be better to reach out to others who have established thin-film catalysts to include such materials rather than develop them specifically within this project.
- It would be good to consider dropping the thin-film electrode work unless it can be logically connected to the rest of the project. The project should consider the possibility that the observed local transport losses arise not just from effects of aggregates but also from as-yet-unexplainable losses right at the Pt–ionomer interface.
- The team may consider including some low-Pt-loaded commercial cathode electrodes from other MEA suppliers for the study.
- Some pre-templated electrode structures should be added to enable limiting case electrodes.
- Carbon supports with different microstructures should be incorporated in both parts of the project.

Project #FC-158: Fuel Cell Membrane Electrode Assemblies with Ultra-Low-Platinum Nanofiber Electrodes

Peter Pintauro; Vanderbilt University

Brief Summary of Project:

Particle/polymer nanofiber mat electrodes are a promising alternative to conventional fuel cell electrode structures. This project seeks to better understand and further improve the performance and durability of low-platinum-loaded nanofiber mat fuel cell electrodes and membrane electrode assemblies (MEAs). Mat electrode MEAs with highly active oxygen reduction reaction catalysts for hydrogen/air fuel cells will be fabricated, characterized, and evaluated. The project will focus on nanofiber cathodes with commercial platinum–alloy catalysts and platinum–nickel octahedral catalysts containing various ionomer and blended polymer binders.



Question 1: Approach to performing the work

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

- This project looks to demonstrate a novel electrode architecture based on electrospun fibers. Based on initial results, this approach improves performance and stability because the electrospinning process results in excellent catalyst/binder mixing and forms a more porous morphology that is better for high-current-density operation.
- This is a novel approach to reducing Pt cost by dispersing Pt nanoparticles and polymer in nanodimensions on the active layer, catalyzing electrode reactions (mainly oxygen reduction in this presentation). There is a catalyst team (Georgia Institute of Technology [Georgia Tech]), an ionomer team (3M Company [3M]), a catalyst and polymer dispersing team (Vanderbilt University), and a performance validator (Nissan Technical Center North America [Nissan], with the help of the Fuel Cell Consortium for Performance and Durability [FC-PAD]). Therefore, the approach should be doable and checkable.
- Electrospun fiber electrodes are a new type of electrode structure that shows promise of good activity and improved and more durable high-current-density performance. This project's approach is appropriate for further developing this innovation and for understanding why the benefits occur.
- The electrospun nanofiber approach addresses the barriers of performance and durability. The project is well designed and feasible. It is well integrated with other efforts in FC-PAD.
- The approach of making nanofiber cathodes and MEAs is novel. This has demonstrated a significant performance and durability increase over conventional MEAs. The approach is also based on the principal investigator's (PI's) multiple years of experience in nanofiber development.
- The project approach of development of novel electrode structures directly addresses key DOE cost barriers.
- Nanofibers offer good opportunity. However, MEA is the key. The project has good collaborations.
- The PI and team are attempting to make high-performance MEAs based on electrospun carbon fibers. The premise of the concept is to provide a porous network for the electrocatalysts and a basis for evaluating more advanced electrocatalysts that other members of the team will provide. The approach is to make MEAs with the different catalysts and the ionomers and test them at standard conditions. There are several concerns:

- The researchers claim that the “electrode macroporosity, microporosity, and particle and binder interconnectivity become more critical when high-performance nanomaterial catalysts are used in fuel cell electrodes.” However, there is little or minimal attempt to correlate these parameters to the electrochemical results.
- Another major concern is that the porous materials will flood if used in a stack at low temperatures and will never be able to reach full power as tested on single cells. The test plan needs to be expanded beyond testing at around 80°C, in an attempt to replicate cold start.
- It is unclear how water will be removed from the porous materials if the fuel cell stack is frozen.
- The gas diffusion media will likely play a large role in the performance of these materials, but there is little or no attention to this vital part of the MEA.
- The test cell for the MEAs should also be described, along with how the cells are compressed; cell compression is key to the performance of porous materials and might skew the results if it is not carefully tracked.

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 results are excellent for initial performance and mass activity (even though electrochemical surface area is low) to date, far exceeding conventional catalyst spray coating, which is a state-of-the-art way to disperse catalysts in a gas-fed porous fuel cell electrode active layer.
- The project has made good progress toward its goals. Initial electrode optimization work has yielded modest activity improvements, and results appear superior to conventional spray-coated electrodes. The nanofiber electrodes appear to yield substantially improved hydrogen/air performance retention (slide 13). Development of Georgia Tech’s catalysts with high activity appears well underway and to be making good progress.
- The project has made good progress in the short time it has been active and since the non-disclosure agreements were signed. The project has prepared electrospun catalyst layers from commercial catalysts with improved performance over conventional sprayed or painted catalysts. The project has prepared shape-controlled PtNi and Pt-coated PtNi catalysts.
- The initial results with alloy catalysts are very encouraging. Rotating disc electrode results with the octahedral catalysts show unusually good durability for this class of materials, particularly with the added Pt overlayers. Activity is reasonable for this catalyst class.
- This project has progressed well in less than six months. The PtCo/C nanofiber cathode has demonstrated tremendous durability improvement. Shape-controlled Pt-Ni octahedral catalysts also show improved activity.
- The project had a delayed start, so the useful test data are limited. However, the results are very promising. The team has demonstrated good progress on electrode and MEA processes.
- Within the statement of work, the team has made progress. If possible, the researchers should carry out more in-depth physical characterization and have broader ranges for electrochemical evaluation.
- The project is mostly on track—just slightly behind on the full delivery of the catalyst.

Question 3: Collaboration and coordination with other institutions

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

- The team—Vanderbilt University (prime), led by Professor Peter Pintauro for polymer and catalyst dispersion; Georgia Tech (sub), led by Professor Younan Xia (project co-PI) for catalyst-making; 3M (sub), led by Dr. Mike Yandrasits for ionomers; and Nissan (sub), led by Dr. Nilesh Dale (project co-PI) as performance evaluator with FC-PAD for validation—covers all bases for executing and validating this nano-spinning approach to high electrode catalyst active layer activity in a perfluorosulfonic acid electrolyte membrane fuel cell.

- A good degree of collaboration is planned with Nissan, Georgia Tech, and the FC-PAD consortium. Advanced electrode structural characterization and modeling to help understand the apparent improved transport would be useful but does not appear to be part of the project.
- Collaboration and coordination with partners is in place. Coordination with the FC-PAD team is in place. Connections with the modeling efforts in FC-PAD are not yet apparent but could be beneficial to both FC-PAD and this project
- This project has a team with diversified experience. The collaboration of Vanderbilt University with Nissan, 3M, and Georgia Tech make this project team very strong.
- The project has a very good team; there are great collaborative efforts with 3M, Nissan, and Georgia Tech.
- The teaming with Nissan and other universities is good.
- The project is in too early of a stage to judge how well it will integrate with FC-PAD. The collaboration between synthesis and the testing centers seems to be working well.
- One expertise missing from the team is a partner who can perform electrode modeling. This would help to better understand the morphology effects at a fundamental level. Coordination with catalyst development projects may also prove useful as this project progresses.

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

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

- Electrospun cathode layers are one of the few novel electrode structures that show good promise both in kinetic activity and in high-current-density performance. The high specific activity of octahedral catalysts would lead to crippling local transport losses at high current density in standard electrode structures. The improved high-current-density performance seen to date with electrospun electrodes using standard catalysts gives hope of mitigating local transport losses on octahedral catalysts, possibly allowing practical utilization of their high specific activity that could not be achieved with standard electrodes.
- The Vanderbilt University, Georgia Tech, 3M, and Nissan team are well positioned to find a better understanding and further improve the performance of low-platinum-loaded fuel cell electrodes and MEAs. It is not clear what the long-term durability of these electrodes is because of the presence of 20–30 wt.% of hydrocarbon carrier polymer. This is an important issue to keep aware of when developing this technology. This may be a top priority of FC-PAD and Nissan.
- This is a novel approach to making electrodes that is showing very interesting results in the initial tests. Other reviewers may question the scalability of electrospinning since it is relatively new to the fuel cell community; however, this process has been scaled up for other applications and should not be a concern.
- Goals toward lower Pt loadings and high performance are in alignment with DOE goals, and the project is also testing durability. The larger question is whether meeting the DOE goals at a narrow test condition (i.e., 80°C at pressure) will translate to success at the stack level.
- The project supports the goals and objectives of the Fuel Cells sub-program to decrease cost and improve durability. The project provides a unique electrode structure with the capabilities to vary parameters in different ways from what can be done with conventional electrode structures.
- The project is directly relevant to addressing the key barriers of performance, durability, and cost. With access to high-activity catalysts, the project has good prospects for achieving several DOE 2020 targets.
- This project is very relevant to the fuel cell goals delineated in the Multi-Year Research, Development, and Demonstration Plan. The project aims to enhance fuel cell MEA performance and durability.
- The cost of nanofiber-based electrodes and MEAs for the proposed process needs to be justified for ensuring a competitive cost; yields are very important.

Question 5: Proposed future work

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

- The proposed electrochemical characterization work is substantial and should be helpful to the project toward meeting its first-year milestones. The milestones (slide 20) are largely operational, rather than

metric-driven. Support accelerated stress testing (AST) should be done early to assess whether this is a key issue with electrospun nanofiber electrodes.

- The team's design of experiment is very good. The team plans to inspect other carrier polymers that are either very stable or completely removed. The activity is already high, and the plan to do a design of experiments to vary the catalyst and polymer ratios to improve performance is logical.
- The project plans are an effective combination of novel electrode preparation and testing. The roles of the FC-PAD laboratories should become clearer as the project moves forward.
- The project builds very well on the initial data and input from team member Nissan.
- The proposed future work is appropriate and should help elucidate the differences between electrospun catalyst layers and traditional layers, and determine the factors limiting performance.
- The plan is for Vanderbilt University to make materials and Nissan (and FC-PAD partners) to do testing. There does not seem to be a good plan for developing a matrix to answer the questions about what type of catalyst microstructure is important, and how the microstructure affects the performance of Pt and other catalysts. Overall, the process seems boilerplate and not poised to answer general scientific questions—rather, just whether the team's MEA works or not.
- The proposed future research is reasonable. However, the PI should focus to some degree on the nanofiber MEA characterizations to elucidate the microstructures using in situ and ex situ approaches. It is unclear why the nanofiber MEAs show performance superior to the conventional MEAs.
- A high-level Gantt chart beyond 2017 should have been presented.

Project strengths:

- The project further develops one of the few novel electrode structures that show promise at both low and high current densities. Better understanding of how electrospun electrodes work could also clarify how standard electrode structures could be improved. The octahedral catalysts in this work have shown better durability than most such shape-controlled catalysts, and facile synthesis methods have been developed. Electrospun electrodes are a plausible, though still poorly understood, way to prevent excessive high-current-density losses on octahedral electrodes giving very high local current densities.
- Electrospun structure provides unique electrode structure and flexibility to provide different structures (for example, using a second needle to add additional ionomer in an independent intersecting ionomer pathway). The project has access to high-activity shape-selective catalysts and Pt-coated shape-selected catalysts that show good durability.
- This project has demonstrated novel approaches using nanofiber cathodes and MEAs. The project has shown good progress in less than six months and has a strong team with diversified experience.
- Nanofiber electrodes appear to have some strong advantages over conventional electrodes in terms of beginning-of-life performance and hydrogen/air performance retention after the electrocatalyst AST. The project has a strong team.
- The strength of this project is that it is developing a novel approach for electrode preparation that improves catalyst/ionomer mixing and porosity. The approach, in initial testing, demonstrated interesting results for performance and durability.
- The catalyst layer microstructure is important to the function of MEAs. The team has a good means of making highly porous microstructures, and a plan to test out their electrochemical performance.
- This project offers a novel and effective method to intersperse catalyst *and* polymer for producing a catalyst active layer. This project is very creative.
- The team is very well-qualified and -coordinated. The laboratory, university, and industry combination is very good.

Project weaknesses:

- Electrode structural characterization (e.g., nanocomputed tomography and transport modeling) is absent from the project. Such work would be useful toward development of further-improved electrode structures. It is unclear whether stability of support will be sufficient against the support AST.
- The carrier polymer's being made of hydrocarbon opens up questions about chemical and physical stability—especially in the intermediate (1,000 hours) and long term (5,000–10,000 hours). Tracking this

carrier polymer should be a top priority, and changing it may be needed to optimize this method's effectiveness.

- The conventional electrodes being used as controls are fabricated by spraying rather than by more manufacturable slot-coating processes, and thus may not properly represent the true state of the art. The expected roles of the FC-PAD laboratories should be better defined.
- This is a work-intensive project that may not lead to any mechanistic insights and might produce MEAs that do not work in actual fuel cell stacks. More work should be done on understanding why porous fiber mats might lead to higher-performance systems, whether they flood, and what gas diffusion media are ideal for operation with porous fiber mats.
- The correlation between the improved electrode performance and the microstructure is not well understood. Therefore, the in situ and ex situ approaches should be used to characterize the electrode microstructures. The PI should also discuss the scalability of the nanofiber approach.
- The project plan did not convey plans to model and understand the reasons that the electrospinning approach would improve durability.
- Mechanistic understanding of performance improvement and stability should be characterized to greater depth.

Recommendations for additions/deletions to project scope:

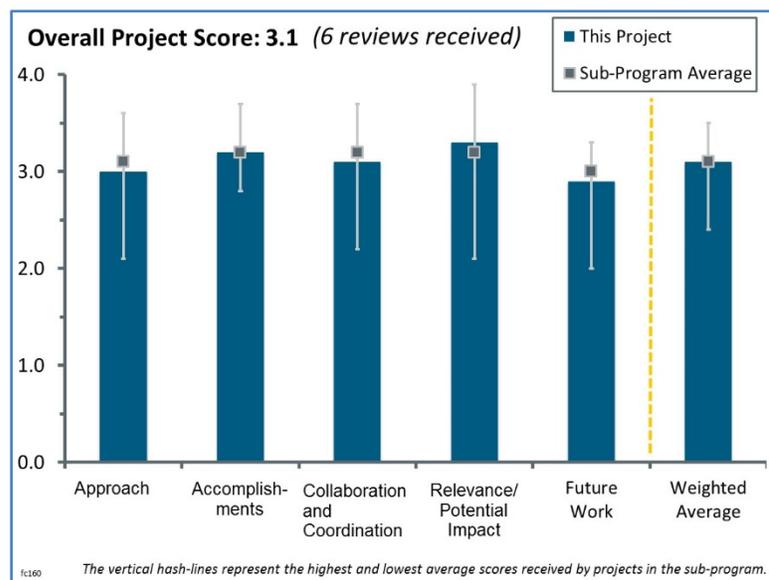
- Performance should be compared to state-of-the-art MEAs that were not made in-house. It is possible that the electrospun electrodes were better mixed because the team was not mixing the conventional electrodes well enough. Plans for modeling the electrode performance and degradation should be added to better explain what the new approach does at a fundamental level. The team should also consider coordinating with other projects/suppliers that are putting effort into optimizing catalysts, as collaboration with those entities could be mutually beneficial.
- FC-PAD has shown some advantages for electrode designs with varied Pt loading that the researchers believe leaves room for areas for water transport. Using dual electrospinning, this project could look at a more uniform way to provide a separate water transport path, or water highway, by electrospaying a second stream with just ionomer and mixing it with the stream with ionomer + catalyst + carbon, and reducing ionomer in the mixed stream to reduce ionomer adsorption on the catalyst.
- The project should set up long-term testing of any electrode using a hydrocarbon carrier polymer as soon as possible to see whether it degrades. Like they say, if it is too good to be true, it probably is not true.
- Plans on how to understand transport properties in electrospun electrodes should be fleshed out more. Consideration should also be given to later studying hybrid electrodes using electrospun fibers mixed with standard catalyst powders and ionomers.
- The researchers should carry out microcomputed tomography studies and/or porosity measurements to characterize the porosity. The team should add in a task for a freeze start test and look at different gas diffusion media materials. The researchers should also report out how cell compression affects the results.
- In situ and ex situ approaches should be used to characterize the electrode microstructures. The PI should also discuss the scalability of the nanofiber approach.
- The team should increase early focus on evaluation of electrodes for support stability. The team should also add some amount of electrode structure characterization to aid development.
- Scale-up impact on mechanistic understanding should be emphasized.

Project #FC-160: ElectroCat (Electrocatalysis Consortium)

Piotr Zelenay; Los Alamos National Laboratory

Brief Summary of Project:

ElectroCat was created as part of the Energy Materials Network in February 2016. The goal of the consortium is to accelerate the deployment of fuel cell systems by eliminating the use of platinum-group-metal (PGM) catalysts. ElectroCat and its member laboratories—Argonne National Laboratory (ANL), Los Alamos National Laboratory (LANL), National Renewable Energy Laboratory (NREL), and Oak Ridge National Laboratory—will develop and implement PGM-free catalysts and electrodes by streamlining access to unique synthesis and characterization tools across national laboratories, developing missing strategic capabilities, and curating a public database of information.



Question 1: Approach to performing the work

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

- The project has a well-defined approach. The key messages taken directly from the presentation are as follows:
 - Durability and cost are the primary challenges to fuel cell commercialization and must be met concurrently.
 - The project's goal is to accelerate the deployment of fuel cell systems by eliminating the use of PGM catalysts.
 - The project's mission is to develop and implement PGM-free catalysts and electrodes by streamlining access to unique synthesis and characterization tools across national laboratories.
 - Demonstrate improved feasibility of segmented cell system for combinatorial PGM-free samples (e.g., Fe-CM-PANI-C catalyst).
 - Extract values for the reaction order with respect to oxygen partial pressure and activation energy as a function of PGM-free catalyst type and/or electrode design.
 - Improved fuel cell performance in both kinetic and mass transport region reaching a current density of 120 mA/cm² at 0.8 V (iR-free).
 - Iron (Fe) dissolution rates for (AD)Fe-N-C are >10×lower than for (CM+PANI)-Fe-C(Zn).
 - Low fuel cell performance caused by dense packing of catalyst layer resulting in uneven ionomer distribution and low porosity.
 - Oxygen mass transport losses are dominated by flooded pores in thick electrodes and film resistance in thin electrodes.
 - High-current-density performance improved by decreasing electrode thickness, tortuosity (m), and size of micropores (rm), and increasing volume fraction (vf) of micropores.
 - Demonstrated hydrogen-air performance of 120 mA/cm² at 0.8 V iR-free with (CM+PANI)-Fe-C(Zn) cathode catalyst, a 25% improvement over the 2016 status.
 - Achieved half-wave potential (E_{1/2}) of 0.83 V with (AD)Fe-N-C in rotating disk electrode (RDE) testing, an increase of 0.02 V over the 2016 status.
 - PGM-free catalyst activity in an MEA: 16 mA/cm² at 0.90 V iR-free and 0.044 A/cm² at 0.87 V.

- The approach being taken with ElectroCat is well designed to address the issues with PGM-free catalysts. It will be important to fundamentally understand the active sites in this class of materials, improve durability, and increase active site density. All of these things are addressed in the project approach. The only minor concern is that some of the initial focus seems to be on improving activity; however, until degradation is well understood, activity and durability should be examined in parallel to prevent development of really good catalysts that may never last more than 100 hours.
- The project should concentrate more on electrode modifications to get thick electrodes to work even in air at high current densities and concentrate less on increasing the volumetric number density of active sites. Extensive high-quality work in recent years appears not to have led to the significant improvement in active site density that is needed if 10-micron electrodes are to suffice. Less reliance should be placed on testing with RDEs and with RDE-similar flow cells; work should be done with membrane electrode assemblies (MEAs). Some of the non-Pt catalysts that have done best in fuel cells have given little or no activity when tested in RDEs. High-throughput methods of synthesis and testing are of questionable value for non-Pt catalysts, the performance of which depends less on chemical composition and more on details of processing. The physical vapor deposition (PVD) generation of thin-layer model catalyst systems is unlikely to be useful, as the methods are too far removed from the complex preparation methods that have given the best non-Pt catalysts, and the number of active sites, already low in high-surface-area catalysts, will likely be too low to give enough activity to allow meaningful mechanistic conclusions to be drawn.
- Comprehensive sets of advanced synthesis and characterization methods are brought to make carbon-based, PGM-free, fuel cell catalysts and MEAs and understand the material behavior. Emphasis on high-throughput approaches should be balanced by fundamental science efforts to address outstanding questions regarding oxygen reduction reaction (ORR) catalysis by PGM-free carbon.
- The project approach appears substantially focused on improving non-PGM catalyst activity and the electrode structure to enable high power capability. While very important, the project does not appear to have a strong apparent focus on addressing the key durability concerns with carbon-based structures.
- The project has a large and expansive approach but is also a highly funded (\$3 million per year).

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 project has demonstrated very good technical progress by incorporating a Zn pore-former in the electrode (slide 13), allowing substantially improved hydrogen–air performance. Use of RDE-inductively coupled plasma mass spectrometry (ICP-MS) can allow for good early detection of stable catalysts and helps assess degradation mechanisms (slide 15). Systematic studies of performance versus electrode properties (thickness, tortuosity, etc., slide 23) appear well executed and should act as a good baseline dataset for the modeling work. Modeling of catalyst active-center durability (slides 24, 25) may be useful if results correlate with experiment. Some work should focus on stability of the catalysts against corrosion relatively early in the project.
- ORR activity of PGM-free ORR catalysts is in continued need of further improvement to reduce cathode thickness and to lower the cost of other stack components. Insufficient long-term stability and performance durability under steady-state and load-cycling conditions, a limited understanding of the ORR mechanism and the nature of the ORR active site, as well as the mechanism of catalyst degradation preventing the rational design of next-generation PGM-free catalysts, requires electrode design and component integration to provide adequate ionic, electronic, and mass transport to and from active sites. It also requires integration with existing automotive fuel cell stack and system technology. The stability of PGM-free catalysts is significantly affected by high-potential excursions (start–stop conditions).
- All aspects of the project appear to be on track. Promising, high-quality results have been generated. There has been good scientific output since the launch of ElectroCat. The atomic resolution and science, technology, engineering, and mathematics (STEM) work is excellent. The durability descriptor calculation based on e-beam damage is unconvincing because it does not reflect the electrochemical processes that occur in a polymer electrolyte membrane fuel cell (PEMFC) cathode.
- Given the size of the project, reasonable incremental improvement in fuel cell performance, mechanistic understanding in activity, and degradation of the catalysts have been demonstrated. All of the project work

is on Fe catalysts. The transition away from Fe is slow. New catalyst approaches are now focusing on a metal–organic framework approach. Fuel cell stability was not discussed in the presentation.

- Modest improvements in performance have been made in the last year. Fluoridation of catalysts (a good idea) has so far led to a loss of Fe and of performance. More work on this is needed to keep Fe levels adequate after fluoridation. While attempts to streamline the national laboratories' intellectual property (IP) procedures are needed, the current approach should be replaced by the labs seeking to get their developments into the public domain as rapidly as possible. The present emphasis on retaining IP for the labs is counterproductive to the goal of getting innovations into the American economy.
- The ElectroCat consortium is clearly off to a good start and making progress towards its planned goals, which are aligned with DOE goals.

Question 3: Collaboration and coordination with other institutions

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

- The team of national laboratories under Zelenay, Myers, Dinh, and More is conducting an excellent project to develop PGM-free catalysts for low-temperature fuel cell applications. The researchers are showing significant progress, which they verify and report formally in a technically sound manner, and they utilize the latest state-of-the-art equipment and methods. They are studying some of the most advanced catalyst materials, and their work is at the leading edge of development for this technology. In addition, this technology is one of the most important for fuel cells at this time.
- ElectroCat appears to be collaborating well between the national laboratories involved. It was not clear if there are other institutions involved or if that will start after the related project awards. An explanation of how there will be industry collaboration would also be beneficial.
- LANL work seems to be rather well integrated into the project as a whole, though closer coordination with outside groups skilled in making high-performing MEAs would be useful. The ANL high-throughput efforts need better coordination with groups experienced in the development and testing of non-Pt catalysts. The NREL personnel planning to use PVD to grow thin-film model non-Pt catalysts should talk long and hard with experienced LANL personnel before committing much time to such an effort.
- The project appears to extensively utilize national laboratory consortia and has a large list of highly capable, no-cost collaborators.
- The project's scientific output so far has been dominated by the lead laboratory (LANL). Collaborative results among the four member laboratories appear to be in the pipeline. External collaborators are listed, but no concrete evidence of collaboration was presented.

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

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

- If successful in developing durable, high-performing non-Pt catalysts, this project could revolutionize fuel cells. Full success in this project is highly unlikely. The most likely benefit from this project would be modest improvements in non-Pt catalyst durability, which would still leave such materials short of the requirements for commercial application.
- This consortium can clearly help achieve DOE goals for fuel cells and have a significant impact. The only uncertainty is whether it is necessary to pursue completely PGM-free catalysts. That may prove to be unnecessary from a cost perspective and technologically too challenging.
- The project aim to develop catalysts with improved activity and performance in electrodes directly addresses key barriers. However, the project does not address carbon durability, a key issue with carbon-based PGM-free electrocatalysts. Achievement of activity and performance parity with PGM catalysts will be for naught if durability cannot be substantially improved.
- Promising results based on completely PGM-free catalysts have been generated, which suggests the carbon-based approach has the potential to meet DOE targets.

- Reducing/removing PGM from a fuel cell is highly relevant. It is unclear how relevant the knowledge learned from the Fe catalyst will be.
- This report acknowledges, but possibly understates, the stability issue with non-PGM catalysts in an acid environment. While this team has shown significant performance improvement and will likely continue to do so, the dependence on RDE leaves the project exposed to some significant surprises. The PEM technical community recognizes RDE as the “baseline test method” for qualifying catalysts based on its proven performance on Pt/C catalysts. However, many publications have shown that there is a significant discrepancy between the stability of non-PGM catalysts as measured by RDE and as measured by subscale cell testing. If RDE is giving unreliable results, alternative methods should be used.

Question 5: Proposed future work

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

- The planned work for 2017 is logical. A high-level Gantt chart beyond 2017 should be added to better understand the full plan. In addition, if the highly active sites being pursued now are inherently unstable, it was not clear how that would be mitigated.
- The project’s future work is focused on development of improved activity catalysts and improved performance electrodes. The project appears to be on track toward these goals. Carbon stability does not appear to be substantially addressed; emphasis on this area should be increased.
- If the current non-PGM catalysts are totally unstable, as shown in the literature, this team has to modify its development plan to increase the focus on stability. Somebody needs to have a task either to modify the RDE test process so it gives valid results for stability or to develop a new, valid procedure for non-PGM catalyst stability testing.
- Long-term durability of the proposed carbon-based catalysts needs to be established. Computation and modeling are not integrated with the experimental efforts. Experimental efforts from catalyst activity improvement to active site identification can all benefit from appropriate modeling work.
- High-throughput methods are unlikely to be productive for these complex non-Pt catalyst systems. Testing should concentrate on single fuel cells and get away from RDE or flow cells as rapidly as possible.

Project strengths:

- The project has an excellent team and is already producing interesting results. The proposed approaches for understanding the active site and degradation mechanism are greatly needed for these materials. If successful, the consortium would enable technology that drastically reduces fuel cell cost at commercial scales.
- LANL experience with non-Pt cathode catalysts should serve to keep the rest of the project within realistic bounds. Consortium characterization skills could elucidate how non-Pt catalysts work and how they degrade, determine the number and location of active sites, and determine why they stop working. The Fuel Cell Consortium for Performance and Durability (FC-PAD) experience should be available for guidance on how to make thicker electrode layers with good performance.
- Most aspects of this project are thoughtfully planned, and the expertise and activities of the member laboratories are nicely interwoven. This project has the opportunity to address urgent scientific questions, including the exact nature of active sites in functionalized carbon cathodes.
- The project has a very strong technical team. Very good improvements in activity and hydrogen–air performance have been demonstrated. High throughput/combinatorial development will be very useful toward accelerating development if/when validated.
- The team provided data for MEAs. The project needs to conduct many more experiments in MEAs and address the ionomer glomeration after testing.

Project weaknesses:

- More emphasis should be placed on getting thick electrodes to work in air at high current density. More realism needs to be applied to the high-throughput effort. It is tough for any electrocatalyst, but very tough for non-Pt catalysts. It seems very strange to have a consortium entitled simply “ElectroCat” and have it

restricted to non-PGM catalysts because for the foreseeable future, practical PEMFC catalysts are likely to contain low levels of PGMs, and much work still needs to be done on PGM catalysts. More effort should be placed on methods of measuring the density of active sites in non-PGM catalysts.

- The consortium should place more emphasis on understanding degradation mechanisms for various possible active sites. Developing a highly active PGM-free catalyst will not be impactful if it is later found to be inherently unstable. Further, some justification for why only a completely PGM-free catalyst is necessary would be beneficial (versus the consortium considering a minimal Pt loading that could still meet long-term cost targets).
- Computation and theory could play a bigger role, but they are not. Catalyst development for ORR and hydrogen oxidation reaction in alkaline metal fuel cells seems to overlap with other projects funded by the Fuel Cells sub-program.
- The project has a small-area MEA. The project uses the anode with 0.3 mg/cm^2 Pt. The compression must be applied, otherwise delamination will occur during the accelerated stress test. The project lacks an understanding of the stability of the catalyst clusters.
- The project's weakness is insufficient effort toward carbon durability.

Recommendations for additions/deletions to project scope:

- The team should redirect a high-throughput effort to a more careful synthesis and fuel cell testing of single samples. Non-Pt catalysts are too complex for shortcut methods to be useful, and compositional variations are less important than details of processing. Non-Pt catalyst precursors are cheap (compared to Pt). Therefore, even initial synthetic work should be done at a scale giving at least 5 g of catalyst to allow testing in fuel cells, as RDE is particularly unreliable for non-Pt catalysts. Scale-up of non-Pt catalysts is also unusually difficult, so one should start as soon as possible with the manufacturability processes. Particular attention must be paid to giving powder surfaces adequate exposure to gases during heat treatments. The team should concentrate more on making thick electrodes work and less on another round of attempts to increase the density of active sites. The team should work harder to develop methods to quantify the number of active sites present in a given catalyst. It is unclear whether an appropriate adsorbate molecule can be found to use to titrate the number of active sites. The team should deemphasize efforts to replace iron, as iron loss does not appear to be the mechanism whereby non-Pt catalysts lose activity (active iron stays in the catalyst and does not hurt the membrane). The project should continue present efforts to remove non-active Fe from the catalyst precursors before MEAs are made.
- In next year's presentation, it would be beneficial to show an explanation for why only a completely PGM-free catalyst is being pursued, a greater emphasis on the degradation mechanisms in the most active catalysts, a high-level Gantt chart for the project, and results from probe molecule studies being conducted.
- The project should strongly accelerate assessment of carbon durability evaluation and, if needed, development of PGM-free catalysts based on more intrinsically stable materials.
- The project should deposit material on the carbon support in order to have a mechanical robustness.

Project #FC-161: Advanced Electrocatalysts through Crystallographic Enhancement

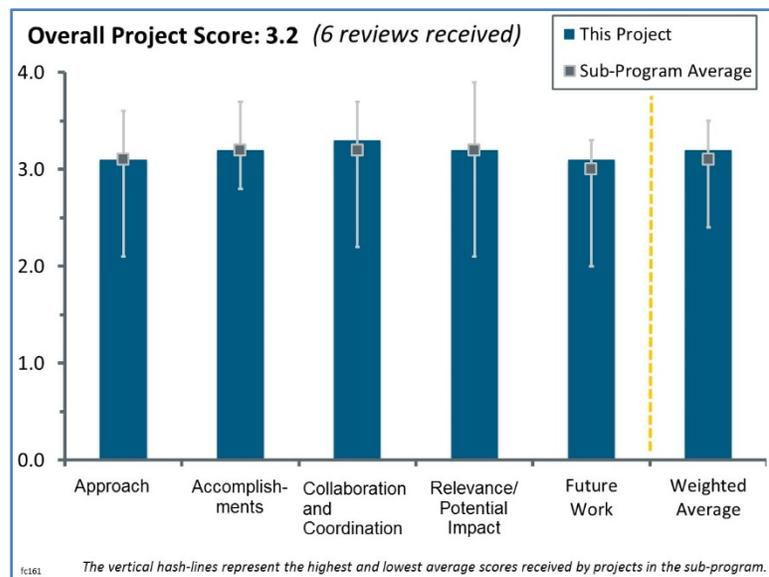
Jacob Spendelow; Los Alamos National Laboratory

Brief Summary of Project:

Los Alamos National Laboratory (LANL) seeks to design active and durable oxygen reduction reaction (ORR) catalysts based on fully ordered intermetallic alloys on highly graphitized nitrogen-doped carbon supports and demonstrate them in high-performance membrane electrode assemblies (MEAs). Synthetic work is guided by computational ORR kinetic studies, and each round of synthetic development is further guided by feedback from MEA testing and characterization studies.

Question 1: Approach to performing the work

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



- The project approach is multifaceted and well designed. Use of highly/fully ordered alloys to enable higher activity and electrocatalyst stability is a reasonably sound approach. The project is also addressing electrocatalyst support durability, another key barrier.
- The approach is appropriate: studying improvements of Pt alloys with Fe, Co, Ni, frequently studied catalysts, involving ordering of these alloys, ordering cores, and forming intermetallic compounds. It opens up the possibility to increase catalyst stability and activity. In particular, stability of the catalyst can be improved, given a better stability of ordered structures and intermetallics compared to alloys. The use of N-doped carbon supports is likely to provide additional improvements of durability and particle dispersion.
- This is a valid approach to addressing the major barrier of catalyst cost and durability. The approach would be enhanced if it was evident that the team has some good concepts to overcome the challenge of making fully ordered alloys without making unacceptably large particles (i.e., more than simply lowering the temperature).
- The approach is well thought out and communicated effectively. The principal investigator (PI) was very clear that the Pt/Fe system serves as a stepping stone to a more realistic alloy that exploits the properties observed in the Pt/Fe system.
- The faceted nanoparticle approach is not new. However, using the face-centered tetragonal (fct) structures may lead to higher activity and stability.
- The LANL-led team attempts to reconcile whether well-ordered nanoparticle alloys can indeed affect catalyst activity.
 - The goals of the project are reasonable, as they follow those crafted by the U.S. Department of Energy (DOE).
 - The approach stems from improving the activity of Pt/C by alloying. There is something wrong with the rotating disk electrode (RDE) results reported by Brown (slide 6), which serves as the unfortunate motivation for the work. The Pt/C standard is clearly low—none of the materials have the expected limiting current of -6 A/cm^2 at 1600 rpm (in fact, they have different limiting currents, most likely due to contamination). The results on slide 15 show an ineffective catalyst, if it is indeed measured at 1600 rpm—and a loading of $5\text{--}20 \mu\text{g Pt/cm}^2$ should be used rather than $60 \mu\text{g Pt/cm}^2$.

- The reporting of these results is quite surprising, as there are now numerous tutorials on how to measure the activity of Pt/C catalysts. Garsany et al. have published a tutorial and then reported the results from a round robin for RDE with the University of Hawaii in 2014 (*Journal of The Electrochemical Society*, 161 [5] F628-F640 [2014]). In this paper, the “benchmark” for Pt/C was moved from around 0.2 A/mg Pt at 0.9 V and 1600 rpm in 0.1 M HClO₄ to 0.4 A/mg Pt. The Japan Automobile Research Institute has also worked on benchmarking papers for Pt/C by RDE. If a laboratory cannot carry out careful measurements on Pt/C and repeat literature results, the results from their other electrocatalysts are also suspect.
- Reporting on mass activities of electrocatalysts in MEAs is likely to be another huge problem, and no clear benchmarks exist in this regime (other than the 2005 Gasteiger paper). Key issues are catalyst and ionomer dispersion in the ink method of making the catalyst-coated membrane, the gas diffusion media, and the cell compression, plus the usual parameters of relative humidity and backpressure. None of this key information is reported in the presentation, although it will have a large bearing on the electrochemical 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 results obtained in the first year of this project are quite good. Improved stability of ordered intermetallics was verified. N-doped carbon supports also indicate improved durability. All catalyst characterizations using several techniques were performed with high skill.
- Accomplishments are reasonable, considering the short time that the project has been active. There was a very surprising result of increasing mass activity of fct-CoPt after 50,000 cycles at 60°C, a substantial loss of Co. While the compositional stability is not a target, the resultant activity gain is intriguing and should be assessed.
- The initial ORR intermediate adsorption strengths show compelling justification for this approach. Initial MEA cycling data show the potential for the desired catalyst stability. This is a good start for a project that is only a few quarters old.
- The project has been active for only a few months and has already shown some new catalysts and some H₂–air performance.
- There were great initial results.
- It is difficult to fairly compare the results because of the issues with the electrochemical evaluation of the materials.

Question 3: Collaboration and coordination with other institutions

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

- This project has excellent collaboration. Collaborators are well-selected experts in the collaboration subject. Coordination of the activities has been carried out very well.
- Collaboration team members provide most or all necessary capabilities to carry out the project work.
- The team appears to be the right mix of expertise in both theoretical and experimental capabilities.
- LANL has put together a strong team involving a wide range of skillsets.
- There are a lot of team members, including some no-cost participants, and all seem to be engaged. However, it is not clear what the University of Pennsylvania will really contribute here.
- LANL should provide more leadership in training its university affiliates in proper electrochemical methodology and reach out to other DOE laboratories for guidance if necessary.

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

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

- The project directly addresses the DOE targets for catalysts. The results, confirming the usefulness of ordered nanoparticles and of intermetallic compounds as catalysts or cores, as well as the illustration of the effects of nitriding carbon supports, will have an impact on future work and the progress of the Hydrogen and Fuel Cells Program.
- The project is directly relevant to key DOE barriers of cost, performance, and durability. Inclusion of integration of the catalyst onto supports with increased durability is necessary to achieve project targets, and it appears that the project is emphasizing this appropriately.
- This project is certainly focused on key barriers. It is probably unlikely to result in a step change in performance improvement, but it could potentially result in substantially improved Pt alloy stability, which is definitely needed.
- This work has the potential to achieve the mass activity and durability targets and further advance the state of the art in low-loading catalysts.
- The potential impact is very good if the team is able to develop catalysts that are <5 nm and have good stability. Initial results of high stability are based on larger particles, hence the true impact cannot be estimated.
- The project could have impact if the researchers took a critical look at the thesis that an ordered nanocrystal is indeed more stable and active, but it is not clear why it should be, as the materials will reach equilibrium while under use and presumably go to a disordered state. The researchers should make sure that they challenge their assumptions carefully.

Question 5: Proposed future work

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

- The proposed future work is planned to complete some initially promising results, complete syntheses, scale up syntheses, and perform MEA tests, all of which is likely to lead to successfully addressing catalyst barriers.
- The proposed future work is generally appropriate. Per slide 20, it is unclear why the team will move away from fct-intermetallics if the DOE mass activity target is not met in MEAs. One year may not be sufficient time for development of new catalysts with high MEA activity. Few catalysts that yield high activity in RDE actually yield high MEA mass activity.
- Proposed future work is clearly laid out and follows a thoughtful plan. Work seems to rely heavily on the ability to substitute Co or Ni in place of the iron. If these metals do not exhibit the same activity or stability, then there may not be very much room to pursue this approach.
- The proposed future work is sound; however, there is no metric on catalyst particle size and MEA-level performance to show <0.125 g Pt/kW.
- This looks to be a good plan.
- The researchers need to rigorously characterize their electrocatalysts with RDE, once they have proven that they can meet the new standard results for Pt/C RDE. LANL should also carefully benchmark their Pt/C MEAs and provide leadership by fully discussing the parameters used in measurements. Without such careful work, the observations made on their well-alloyed electrocatalysts cannot be proven.

Project strengths:

- This team has identified an effect in Pt/Fe systems that has the potential to be exploited in other Pt metal alloys. The project has a good balance of theoretic and experimental approaches. In-cell testing and out-of-cell characterization methods are designed to understand the key variables and aid in the design of these catalysts. Collaborations seem to be well suited for the objectives of this project.

- The project takes a look at some well-loved theories in the U.S. fuel cell community regarding materials with improved d-band structures and well-ordered materials.
- This project has strong fundamentals with durable, high-activity electrocatalysts and integration with improved durability supports.
- There is a strong research team from LANL. Expert collaborators from national laboratories, universities, and industrial laboratories are project strengths.
- This project addresses major barriers. The approach is to maximize the full potential of platinum group metals (PGMs). There is a good potential risk–reward ratio with an excellent team.
- This is a good team and a sound project concept.

Project weaknesses:

- The project appears to have limited options for substituting Fe in the Pt/Fe crystal structure. The balance between activity and durability, as it relates to particle size, may be technically challenging. This challenge was readily stipulated by the PI. Preliminary results show the potential for good stability after cycling in the Pt/Fe system; however, the last points show a large decline. It is unclear whether this is a reproducible effect or experimental error. Nonetheless, the stability as compared to a baseline will need to show regular progress as the project proceeds.
- The timeline to achieve 0.44 A/mg in an MEA within one year is likely too aggressive. It is unclear whether the reasonably high specific surface areas can be achieved. The achievement of high-MEA-rated power near the target PGM may not be feasible.
- Focusing exclusively on Fe, Co, Ni alloying components, even as intermetallic compounds, can dissolve and degrade catalysts' performance after extensive use. This may be a weakness of the project.
- The project is not on track to prove or disprove any electrocatalyst theories because of poor electrochemical methodology.
- It is unclear how the process can be controlled to a small and uniformly sized particle.

Recommendations for additions/deletions to project scope:

- Exploring surface segregation of intermetallics, dissolution of non-noble metal components at long time use, and the Kirkendall effect would address these important aspects of catalyst durability.
- A go/no-go decision should be added for delivering a catalyst that shows both improved activity and stability with particle size ≤ 4 nm.
- In future AMR presentations, it should be made clear what all of the team members are doing (including the University of Pennsylvania), especially the various team members doing catalysts synthesis (i.e., the presentation should explain what each of them does that is unique).
- The project has just started and appears to be off to a good start.
- LANL needs to straighten out its RDE and MEA methods and then start over. The researchers should also take a critical eye to the thesis of this project because having well-ordered nanocrystals in electrochemical cells under cycling loads seems unfeasible in the long term.

Project #FC-162: Vapor Deposition Process for Engineering of Dispersed Polymer Electrolyte Membrane Fuel Cell Oxygen Reduction Reaction Pt/NbOx/C Catalysts

Jim Waldecker; Ford Motor Company

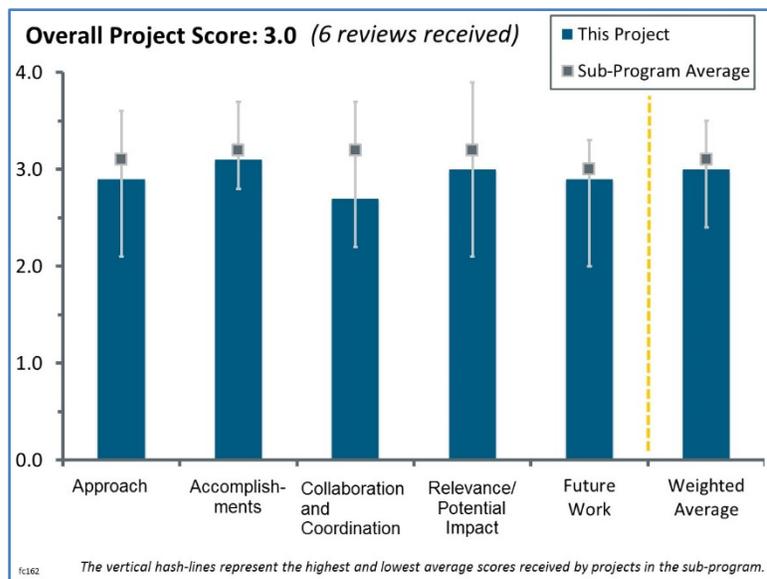
Brief Summary of Project:

The objective of this project is to develop, integrate, and validate a new cathode catalyst material by developing and optimizing a vacuum powder coating physical vapor deposition (PVD) process. Project tasks include (1) development of a new cathode catalyst powder made of titanium, niobium oxide, and carbon; (2) improvement of the PVD process for the manufacture of the catalyst powder; (3) cost-effective scale-up of the PVD process; and (4) integration of the developed cathode catalyst powder into established fuel cell manufacturing processes.

Question 1: Approach to performing the work

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

- The PVD process as described in this project is an interesting process of making a Pt-supported oxygen reduction catalyst. Sputtering assisted vacuum deposition of dry catalysts has been practiced by 3M in its nanostructured thin film (NSTF) catalyst layer fabrication process. Unlike NSTF, which is a multiple-layer deposition process, the present approach is a single-layer deposition process that is expected to be faster. The advantages of a dry PVD process, as described in slide 5, are true. However, the catalyst will be subjected to a solvent-based ink-making process in which most of the solvent-related issues, such as incorporation of contaminants into the electrode layer, will take place. It is not going to have the advantages of the NSTF process, which directly forms the catalyst layer. From some of the early data that the team has presented, it seems that the approach is feasible. The goal of the project is to address the mass transport issue, an area in which Ford has good experience, as explained in the wire-wound bar method in slide 9. It will be interesting to see how the PVD-mediated catalyst gives similar results.
- The project's approach involves using PVD to form Pt/C electrocatalyst particles with niobia stabilizers. The approach is feasible and addresses U.S. Department of Energy (DOE) targets. The project involves a number of university and industry partners that are well placed to ensure success.
- In regard to cost and performance, the inclusion of niobia appears to allow better Pt dispersion and enables lower loadings of Pt without compromising cathode performance. Large-scale PVD on powder is a worthy goal and a promising production method that appears to be within reach. In regard to durability, insufficient information was provided. Establishing the stability of Pt/NbOx/C should have the highest priority.
- This project will develop a Pt/NbOx/C catalyst using a PVD process to overcome the catalyst activity and durability and support durability issues often faced when using conventional Pt/high-surface-area carbon (HSAC) catalysts. Proof of concept for the proposed approach is shown using square wave cycling between 0.1 and 1.05 V in rotating disc electrode (RDE) studies.
- The principal investigator (PI) presented a project on Pt on NbO₂.
 - The premise of the project is that NbO₂ is an electronic conductor and somehow adds to the activity of Pt. Such pursuits of “conductive” supports have never made sense, as an oxide conductor with a conductivity of 1 S cm² has electronic conductivity about four orders of magnitude lower than carbon. Additionally, for thin films and nanomaterials, electronic tunneling is likely, even through insulators. Furthermore, any oxide that contacts the acidic media in



- polymer electrolyte membrane fuel cells (PEMFCs) will immediately convert to an insulating hydrous oxide. If there is one pinhole in the Pt skin, a hydrous oxide surface will break through.
- Supporting Pt on oxides and phosphates is a well-tread area, investigated by many over the last 10 years. Unfortunately, many of the publications on Pt on MO_x-type compounds have not been useful because of poor RDE methodology.
 - Unfortunately, the PI's team continues in this tradition of having poor RDE results. The PI quotes the project's RDE Pt/C activities at 0.2 A/cm²—per Gasteiger's 2005 paper. Since then, Garsany et. al. (*Journal of The Electrochemical Society*, 161 (5) F628-F640 [2014]), with improved methodology, moved the benchmark from around 0.2 A/mg Pt at 0.9 V and 1600 rpm in 0.1 M HClO₄ to 0.4 A/mg Pt. The Japan Automobile Research Institute has also worked on benchmarking papers for Pt/C by RDE. Then DOE national laboratories (the National Renewable Energy Laboratory [NREL] and Argonne National Laboratory) teamed up to write another benchmarking paper on Pt/C this year, published in 2017 in *Electrocatalysis* ("Best Practices and Testing Protocols for Benchmarking [Oxygen Reduction Reaction (ORR)] Activities of Fuel Cell Electrocatalysts Using Rotating Disk Electrode," DOI: 10.1007/s12678-017-0378-6), and supports the Garsany work. The DOE sub-program manager (Papageorgopoulos) is a coauthor; therefore, DOE should be well aware of these results.
 - Because the PI uses an outdated methodology for the Pt/C standard, their results for Pt/NbO₂ are likely skewed. The project's high-specific-activity results are also likely high because the electrochemical surface area of the Pt/NbO₂ is low, skewing the results. This should make more sense once the RDE is improved.
 - The results for fuel cell testing in a membrane electrode assembly (MEA) are poor. The approach is to characterize mass transport losses in the catalyst-coated membranes (CCMs). It is more likely that the hydrophilic NbO₂ is causing water retention in the CCMs and flooding. Others (e.g., 3M) have seen this with oxides. This has also been observed on other Pt/MO_x compounds. How to prevent such flooding is a huge problem and worthy of the team's full attention.
 - The team needs to be realistic about their mechanisms first. The cited papers should be reviewed for accuracy before using them for reference. The PI might try making a Nb₂O₅ support for Pt and testing it to prove whether the premise of requiring an "electronic" NbO₂ support is required. It is not evident that this support is required, and Nb₂O₅ may work just fine.
 - After the researchers have more convincing electrochemical results, they might consider scale-up.
 - Lastly, the heating temperature of the materials may have a large impact on their performance, and this should be investigated at a small scale.
- The approach to performing the technical work is reasonable in terms of the outline suggested for synthesis and evaluation of MEA performance and durability. However, it lacks a solid foundation and baseline for the component materials and, in arguing the usefulness of Pt/NbO_x/C materials, relies on literature data and baseline RDE results that are dubious because of their low values for Pt/C compared to DOE benchmark values. All RDE results presented are low and do not support the notion that the new materials will have higher ORR activity. No basic studies on components such as NbO_x have been conducted to clearly and quantitatively verify that their electronic conductivity and corrosion resistance are better than conventional carbon supports used today. It is unclear what the electronic conductivity of NbO_x is and by what factor its corrosion resistance is higher than conventional carbon blacks.

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

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

- The project is new; it started earlier this year. Within this short period, the team has accomplished all the important elements of the project such as getting the contracts signed, generating some early results, etc. The team has conducted preliminary tasks related to the PVD coating, such as exploring the deposition conditions, sputtering system upgrades for large-batch catalyst production, small-scale PVD deposition at Oak Ridge National Laboratory (ORNL), and testing at the University of Michigan (U-M) and Ford. The team's accomplishments and progress meet the expectations at this point of the project.
- Considering the start date of the project, the accomplishments/initial results presented are impressive and encouraging, although the reason for high mass activity of Pt/NbO_x/C catalysts is unknown.

- Small batch production, testing, and characterization of Pt/NbO_x/C are all underway. The preliminary results are encouraging.
- The project just started; therefore, there are not too many results. Feasibility has been shown in forming conductive niobia and depositing Pt onto niobia-coated C. Further characterization should be performed to determine what the deposited Pt really is. It is unclear whether there is film formation, and it is unclear whether there is an oxide. It is also unclear whether the product is similar to a conventional Pt-supported C in terms of morphology and surface charge.
- Progress made is 0%. Therefore, it is hard to judge accomplishments. The survey of literature and preliminary RDE results presented are inconclusive and do not provide sufficient basis for optimism that the materials suggested have potential use in PEMFCs. Using RDE studies as a screening tool is not in the task list, and jumping to MEAs without any basic affirming studies is questionable. A critical review of literature studies is needed, with weaknesses in them clearly pointed out. Preliminary RDE studies need to be conducted on Pt/C to make sure they hit the DOE best practices and benchmarks reported after verification by several national laboratories.
- The RDE results all need to be improved and repeated. The researchers need to take a critical eye to the MEA results for flooding. Progress has been made on materials scale-up, but it is not clear that this is important yet.

Question 3: Collaboration and coordination with other institutions

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

- The prime (Ford) has arranged a good team of experienced scientists from reputable institutes/ organizations. As the project is very new, the team has not had much opportunity to collaborate; however, the early interactions and results show that the team has good coordination between the members. Work has already started at ORNL, U-M, and Exothermics and has generated some early results (which manifests a good coordination between the stakeholders in the team). Organizations such as Ford, U-M, and ORNL are well versed with DOE projects, and the prime (Ford) is well experienced in running the DOE programs. Overall, the team has shown good interaction and is expected to handle the project well.
- The project has initiated excellent collaboration with two other industrial partners, one national laboratory, and two universities.
- Collaboration and coordination is reasonable for the beginning of the project. Ford should consult with and work with other national laboratories to verify the properties of their novel materials as individual components for electronic conductivity and corrosion resistance. Ford's relationship with the chemical vapor deposition (CVD) company remains mysterious but will probably open up at a later stage in the project. Ford worked on this research for three to five years before this particular project began, and one would expect better control and relationships with collaborators by now.
- A large number of partners are included in the project. The project structure and work breakdown seem appropriate to ensure interaction between lead and subs.
- Partnering with a team to scale up seems premature considering the project is a long way from making a good CCM (and this may not be a solvable problem, per 3M's NSTF oxide-based electrocatalysts, which suffered from flooding). It might help to have NREL contribute to or advise about the RDE results and MEAs.
- There is no evidence of collaboration outside the six partners in this project. Perhaps this is not a fair question since the project has just begun and it should be up to the PIs to determine whether and what additional expertise will be needed for achieving the targets.

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

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

- If indeed the Pt/NbOx/C has improved specific activity, performance, and durability over Pt/C or PtCo/C, it will benefit the Hydrogen and Fuel Cells Program (the Program) significantly. If the CVD manufacturing process is successful for producing large quantities of catalysts without the complications of wet chemistry, it will also be a great advantage and benefit commercialization of PEMFCs.
- The project is relevant to the Multi-Year Research, Development, and Demonstration Plan (MYRDDP). The team's goal is to develop a PVD catalyst, which is aligned with the 2020 technical targets for electrocatalysts and MEAs for transportation. The team is also working on a cost-effective manufacturing process for making such a PVD catalyst, which is aligned with the same 2020 objectives.
- If successful, the proposed research will meet all of the 2020 DOE technical targets for electrocatalyst and catalyst support.
- The project addresses MYRDDP targets and advances Program goals.
- This project will meet DOE needs if indeed the materials are high-performance, but it is hard to tell from the present work and approach.
- The team urgently needs to establish that actual PVD-made Pt/NbOx/C powders have sufficient durability that they can significantly outperform conventional Pt/C catalysts in cost per life cycle.

Question 5: Proposed future work

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

- Future work, the work plan, and milestones are organized toward successfully achieving DOE targets.
- The project is new, and most of the tasks are still not being attempted. The proposed future work is well described and relevant to the project. The work described in slide 19 is logical and important. The team did not discuss possible barriers and relevant mitigation strategies, nor did they provide alternative pathways.
- Since the project is in its early stage, the project proposes to perform catalyst synthesis, characterization, and fuel cell testing, which are necessary to achieve the project goals. It is not clear how the project will achieve the mass activity requirements of a cathode catalyst without using a transition metal alloying element, as shown by most of the previous low-platinum-group-metal (PGM) catalyst development projects funded by the DOE Office of Energy Efficiency & Renewable Energy.
- The future work is consistent with the proposed milestones.
- Little progress has been made at this time, so all work is future work.
- The PI's team needs to be able to characterize Pt/C accurately to the 2014 Garsany *Journal of the Electrochemical Society* work or the 2017 Kocha *Electrocatalysis* work. The researchers will not be able to eliminate the role of mass transfer resistances until they understand the role of flooding. They should also report in detail the specifics for their MEAs, including gas diffusion media and cell compression.
- The fundamental properties of NbOx—as well as those of Pt/NbOx and Pt/NbOx/C—such as electronic conductivity and corrosion resistance are missing and not scheduled to be evaluated. In the team's proposed work, no clear plans are presented on RDE screening and obtaining reasonable baseline data to compare to the results of new materials.

Project strengths:

- The approach of making a PVD-mediated Pt/NbOx/C oxygen reduction catalyst is a somewhat different approach than the traditional wet chemistry method of making catalysts. 3M's NSTF, made by the sputtering process, has demonstrated very high activity and good performance while utilizing very low PGM loadings. The PVD process as proposed in this project has an advantage over the NSTF process since the PVD process is not a layer-by-layer sputtering process. The PVD process is expected to be capable of large-scale catalyst production. The team consists of qualified investigators and organizations with experience in the proposed technology pathways. Regarding technology feasibility, the feasibility of the

PVD process on a small scale has been demonstrated by the team. Therefore, the technical challenge lies in the scale-up, not in the manifestation of the process.

- The team has a good, practical idea, which is strongly focused on producing lower-cost PEMFC catalysts and appears to be achievable within a short timeframe, producing immediate impact on the market.
- There is a potential ease of integration of produced catalyst powders. It is a new approach that could result in a new stable and active catalyst system.
- Niobium oxide is a possible new deposition method for large-volume scale-up, possible higher specific activity, and durability.
- The project should be able to determine whether Pt supported on an oxide has higher activity and durability than standard Pt or Pt alloys on carbon.
- It is too early to make a judgment about the project strengths. Nevertheless, the project has a strong collaboration with two industries, two universities, and one national laboratory.

Project weaknesses:

- In regard to MEA fabrication, the team plans to make powdered catalysts using the PVD process and make MEAs using the wet ink process. However, the solvent-in-wet-ink process can introduce the impurities that the team was trying to avoid. The use of the wet ink process can also introduce catalyst agglomeration and hence lower catalyst activity. The catalyst deposition time, as shown in slide 17 by the small-batch sputtering system at ORNL, is very high. Exothermic's large-batch sputtering system may be more efficient; however, the team needs to address the deposition time of the catalyst.
- A go/no-go decision based on the long-term stability of the Pt/NbO_x/C catalysts and cost savings versus existing Pt/C catalysts should be made in Budget Period 1, not later. No fundamental science on Pt-niobia or niobia-C interaction is planned, despite the project's having university partners.
- The project does not seem to have a backup plan in case any of the components fail, e.g., alternative support materials or alternative catalysts. It would be helpful to have a risk management plan.
- The project has demonstrated weak scientific background work on the evaluation of properties of suggested new materials, poor RDE results, and inconclusive evidence that new materials will be better than Pt/C.
- The premise of the project is questionable, considering the choice of materials is based on poor RDE results.
- It is too early to judge the project weaknesses.

Recommendations for additions/deletions to project scope:

- The project should evaluate with the Brunauer, Emmet, and Teller mathematical modeling method (BET), CO chemisorption, transmission electron microscopy, and x-ray diffraction powder catalysts first, then screen using RDE (after establishing a solid baseline for Pt/C), then prepare MEAs proving that baseline performance has been established using Pt/HSAC, and finally work on catalyst layers with the new materials. If carefully conducted, RDE studies do not show that the new materials show improvement over Pt/C; there should be a go/no-go decision and no need for further studies on MEAs.
- There should be cost analysis to account for capital expenditures or operational expenditures of the sputter system at scale. There should also be greater in-depth characterization of the produced Pt structure and composition, as well as assessment of how it will influence ink integration, catalyst layer functionality, and localized mass transport resistance at high current. In other words, the project needs to determine whether this is a new particle morphology, whether the cathode layer structures are new, and whether they are more prone to mass transport issues.
- The PI's team members should straighten out their electrochemical methods to determine whether there is something special about Pt/NbO₂ (rather than Pt/Nb₂O₅). If they have promising results, perhaps then they should focus on scale-up.
- The team should add go/no-go stages, possible barriers, and mitigation strategies clearly in the project planning.

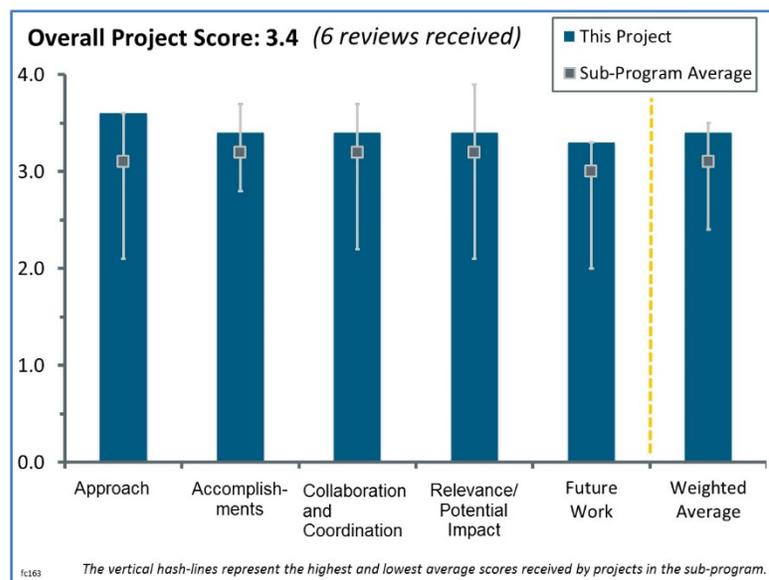
Project #FC-163: Fuel Cell Systems Analysis

Brian James; Strategic Analysis, Inc.

Brief Summary of Project:

This project seeks to estimate current and future costs (for years 2020 and 2025) of automotive, bus, and truck fuel cell systems at high manufacturing rates. Analysis projects the impact of technology improvements on system cost, identifies low-cost pathways to achieve U.S. Department of Energy (DOE) automotive fuel cell cost goals, benchmarks fuel cell systems against production vehicle power systems, and identifies fuel cell system cost drivers to help facilitate Fuel Cell Technologies Office programmatic decisions.

Question 1: Approach to performing the work



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

- The project did very well this year looking at higher-power density for automotive applications. Power densities near 700–800 mW/cm² at rated power no longer represent the state of the art. Even 1095 mW/cm² may be somewhat conservative. The cost decrease (\$53/kW to \$45/kW) was realistic to the extent that it was premised on the decreased power density. The project approach was also enhanced by assuming higher pressure and lower cathode stoichiometry at rated power. The added task to develop cost analyses for medium-duty and heavy-duty applications is consistent with the trend in the industry. As fuel cells show substantial weight advantages versus battery-only systems for these applications, cost analyses will need to be done. The analysis on metal plates went through an exhaustive number of options for forming and coating. However, given the amount of variability that could be caused by 100+ lines running, the approach must consider a constraint on the number of lines.
- The project uses a good approach to provide current and future cost of automotive, bus, and truck fuel cells at high manufacturing rates using the Design for Manufacture and Assembly (DFMA) method. The fact that the model integrates the latest technological advances, combined with the good contacts of Strategic Analysis (SA) with industry, will ensure the validity of the approach. The project aims to identify low-cost pathways to achieve DOE targets and, in general, identify fuel cell system cost drivers. Assessing the impact of technology improvement (e.g., catalysts, bipolar plates [BPPs], membrane electrode assembly, and balance of plant [BOP]) is a very valuable approach to that aim. The project will help DOE to set realistic cost targets. The approach consists of reconsidering different stack components (BPP forming, bipolar coating, and comparison of cathode catalysts) and assessing the impact on system cost. It is a valuable tool for DOE to demonstrate the impact of different technical achievements on the system cost.
- A DFMA approach is sound. The catalyst alloy selection might benefit from past phosphoric acid fuel cell (PAFC) research. PAFC stacks run at steady-state points have achieved over 80,000 hours in commercial service. Admittedly, the operating conditions are different: higher temperature, lower pressure, higher CO content, lower current density, etc. The team should also review what others have fielded. For example, United Technologies Corporation (UTC) fielded rotating component recycle designs starting in the 1990s and pulse width modulation fuel injection around 2010. US Hybrid is still active with pulse width modulated injectors.
- This project shows well-reasoned choices of topics for application of excellent cost-estimation methodologies.

- The project utilizes Toyota's information to verify the cost estimation method, which is a good approach. It is unclear how accurate and insightful the information obtained by Toyota will be. Adding fuel cell cost for heavy- and mid-duty vehicle applications is also relevant. Fuel cell system assumptions should be different from those for fuel cell systems for light-duty passenger vehicles. It is suggested that the project make the fuel cell system assumptions with original equipment manufacturers (OEMs) first.
- The ground-up DFMA approach used by the investigators is commendable. However, validation remains a consideration. For example, the Toyota Mirai analysis is presented as validation, but it is unclear what is being used as the basis for comparison for the model costs. Understandably, this will remain an issue because of OEM and supplier confidentiality. It is unclear whether the researchers can explain what actual methods they use to validate the cost numbers.

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 clear flags placed this year on being able to meet DOE's cost goals for bipolar plates using SS316 are reasonable and should alter DOE's planning a bit. The project has aptly captured the significant cost benefits of the recent developments in catalysts and electrodes under DOE projects. The cost analysis of the Mirai adds an element of realism to the costing expertise and provides a useful check on costs of the current status at low volume.
- The project continues to achieve their annual work product targets. This reviewer appreciates the increased synchronization with the Argonne National Laboratory (ANL) system analysis project and the inclusion this year of a state-of-the-art, non-nanostructure-thin-film-based cathode electrode.
- Considerable progress has been made this year incorporating higher-power density with the higher-pressure and lower-cathode stoichiometry context.
 - The project has been able to deliver results with respect to many different coating options. While the number of lines remains an issue, it is good to see that metal oxide and carbon coatings have been considered.
 - Hydrogen blower calculations for the Mirai appear to contain some small mistakes. The cost per net power at 500,000 systems per year is higher than that at 100,000 systems per year.
 - The project does well to consider the Mirai example. However, there are many assumptions and technology parameters that require refinement and validation, which can be approached with further benchmarking.
- The comment on the change from stainless steel (SS) 316 to SS304 bringing a negligible change is to be expected; the nickel content is about the same, as is the usage. This is an interesting trade study.
 - If the bi-polar plate is being coated, it is unclear whether carbon steel can be used. This is an interesting trade study.
 - Gore's costs did not go down. It was unclear whether the performance increase allows savings on other components.
- The team used results data and feedback from several teams (ANL, Fuel Cell Tech Team, and General Motors [GM]) to reconsider the cathode catalyst choice.
- The project showed good progress by updating the fuel cell cost estimation with a Pt alloy catalyst.

Question 3: Collaboration and coordination with other institutions

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

- The project makes skillful and creative use of information from a wide range of sources in order to give the most accurate cost estimates possible. Interactions with ANL, National Renewable Energy Laboratory (NREL), and the Fuel Cell Tech Team have improved the accuracy of cost predictions and have kept the status more representative of the state of the art than has been true in the past. The project now seems to be getting more complete information from fuel cell developers than has been true in the past.
- The project has good collaboration with ANL and NREL on the results of the Systems Analysis project. Data from GM for high-surface-area carbon Pt/Co has been used. In general, SA has good contacts with

partners in order to get knowledge and feedback and to apply the latest technological advances and system designs.

- SA has shown the ability to collaborate with many different technology partners for many different components.
 - The range of partnership could be expanded on catalyst-coated membrane suppliers beyond Gore. The same could be said for membranes, especially with respect to ionomer cost.
 - The project has expanded the range of supplier collaborations beyond the bounds of DOE-funded projects for metal plate forming and coating. The same level of outside collaborations should be applied for other components as well.
 - Expanding scope into medium- and heavy-duty vehicles will force collaborations with vehicle OEMs. While light-duty vehicles can be assumed to have a low degree of hybridization with battery systems, the same cannot be said for medium- and heavy-duty vehicles. Deeper collaborations will be needed to understand systems for higher-energy vehicles.
- The team's extensive list of collaborators is impressive, but collaborations may be lacking with Japanese companies, academic institutions, and national laboratories. The reviewer inquired as to whether there is any way this project can interact with similar efforts overseas (e.g., the Japan Automobile Research Institute [JARI] and the European Council for Automotive R&D [EUCAR]) to ensure the widest input possible.
- The partners listed have been involved in this work for a while. These partners are not likely to add additional insights. ANL may be of use in upgrading existing computer models. Newer collaborators might enhance thinking for outside the box, which might be helpful.
- The project has good collaboration with industry partners to obtain the materials/process information for cost estimation.

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

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

- This project's relevance is high for DOE program management to present an overall status and indication of progress. The relevance is less so to developers, as they will have their own information to generate their internal assessments. However, having a publicly available project and annual report-out is of general benefit, and the reviewer would go as far as to call it necessary. The team's effort and results are appreciated.
- The project derives relevance based on how it is able to assist DOE in the department's decisions about funding opportunities and the research portfolio. The project does not directly help developers overcome barriers, although there are occasions in which component cost analyses are useful in understanding manufacturing costs for processes that do not presently exist at volume. The project helps to define new targets by exploring existing cost as well as what cost could be with technology advances. Keeping aligned with the system analysis project directions toward reduction or elimination of the humidifier, elimination of active hydrogen recirculation, and the exploration of higher-pressure operating conditions all helps to keep this project relevant.
- This project gives a very professional view of present and anticipated costs for fuel cell systems. Since cost, along with infrastructure, is now the major factor limiting the introduction of fuel cell vehicles, this project is critical to the development of the field.
- The project aims to identify low-cost pathways to achieving DOE targets and, in general, identify fuel cell system cost drivers to help DOE refine future research, development, and demonstration plans.
- The project is relevant if realistic goals and research paths are identified.

Question 5: Proposed future work

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

- The project properly plans to cost out fuel cell systems for a wider range of vehicle types.

- Reduction in the uncertainty in ionomer cost is properly flagged for future study.
- The planned further attention to Pt–alloy-catalyst synthesis costs is definitely needed, as the different experienced catalyst manufacturers differ widely in their estimates. Multiple scenarios of who owns the platinum group metal (PGM), and at what stage of the processing, should be explored, as the cost of the risk of holding a PGM inventory could make a significant difference in the estimate of the synthesis cost.
- Planned further attention to the costs of roll-to-roll processing is sorely needed.
- The project is moving in many directions consistent with the needs of the Fuel Cells sub-program. The mainstream project continues the trend of performing side studies on alternative technologies before incorporating alternative technologies into the main cost analysis for light-duty vehicles. Analyses on medium-/heavy-duty vehicles are consistent with the industry trend because of the advantages fuel cells give these vehicles versus battery systems. Analysis on the Toyota Mirai provides a unique status benchmark.
 - The alternative technology side studies will continue to focus on hydrogen recirculation concepts still being studied in the systems analysis project, which is an appropriate direction. The project is also right to challenge its prior studies with respect to ionomer cost and to look at an alternative membrane reinforcement.
 - The project could use better definition in terms of how to approach cost analyses on medium-/heavy-duty vehicles. Instead of assuming a fuel-cell-dominant system, it may be more interesting to take a few medium-/heavy-duty applications and plot the level of range or energy at which a fuel cell range extender becomes more cost-advantageous than a battery system.
- The reviewer agrees with the plan to perform a deeper dive on membranes and the ionomer, given the high-cost estimates reported here. It is understood that this project also supports various Hydrogen and Fuel Cells sub-programs in their cost study projects. Many projects, and even proposals, would benefit from some earlier technoeconomic studies to help guide their selection and go/no-go decisions. The project should put greater future focus on early support of fuel cell projects to help guide their direction and potential cost impact, if successful.
- In addition to the light-duty vehicle analysis, heavy- and medium-duty vehicle analysis is good, and expected outcomes are interesting.
- The proposed future work listed by the project team is as expected and in line with the results shown.
- Future work is appropriate. Room should be given to explore alternative paths.

Project strengths:

- The project uses a good approach to provide current and future costs of automotive, bus, and truck fuel cells at high manufacturing rates using the DFMA method. One strength is that the model integrated the latest technological advances, combined with the good contacts of SA with industry. The project aims to identify low-cost pathways to achieve DOE targets and, in general, identify fuel cell system cost drivers. Good contact with partners (laboratories and industry) will allow the team to gain knowledge and feedback and to apply the latest technological advances and system designs.
- The investigator has considerable experience in cost analyses over many years of DOE funding.
 - The use of DFMA enables cost analyses to be derived from knowledge of labor, energy, capital equipment, and materials inputs.
 - The project team is collaborating with ANL on system analysis to receive critical performance and BOP component inputs.
 - The project has experience collaborating with many suppliers for both stack and BOP components.
- The project has been creative and successful in gathering input data. The project uses consistent and appropriate costing methodologies. The project updates estimates as new information becomes available. The project flags the few DOE targets that are likely unrealistic. The bases of cost estimates are clearly communicated.
- The project has a strong DFMA background and strong collaborations. The project showed fast response to reviewer feedback, incorporating suggested work into future deliverables.
- Project strengths include a database of accumulated fuel cell materials/process information and the team's cost estimation. The team networks to obtain information from industry.

- The project has detailed cost accounting.

Project weaknesses:

- The project has had difficulty over the years estimating ionomer costs. Project analysis of BPP cost usually can be found to have some unrealistic assumptions buried deep within the analysis. This year, the number of production lines is what appears to be unrealistic. The project sometimes limits itself to what is known through DOE-funded projects. The investigator should more aggressively seek out technology alternatives that are not already related to the Fuel Cells sub-program.
- Not all of the information necessary to accurately estimate costs is released by suppliers and developers, so the project is dependent on the (considerable) skill and (so far, apparently good) judgment of the investigators.
- Validation is always a question mark, but that is the nature of the beast.
- The project lacks validation of the developed model.
- The team needs more “new eyes” to explore less obvious options.

Recommendations for additions/deletions to project scope:

- More attention should be given to what constitutes a reasonable profit for each stage of manufacture. For example, some roll-to-roll processors are used to working with relatively inexpensive materials and expect a high percentage value added for their efforts. Since fuel cell materials are rather expensive, a lower percentage value added is needed, despite the additional costs of handling inventory of such expensive materials as PGMs. Some preliminary costing of non-Pt catalysts at scale might be useful, including such steps as precursor synthesis, pyrolysis, and treatment with hazardous gases (e.g., ammonia and high-energy ball-milling). While processing costs could be high, they likely would still be small versus the material costs of Pt-based catalysts, but this should be checked. Scale-up of non-Pt systems has proven particularly difficult (largely a matter of getting adequate exposure of a powder to gas during heat treatment). Some attention should be given to the costs of recycling PGMs during catalyst synthesis. This could be an unusually large factor in the costs of alternative synthesis techniques, such as physical vapor deposition and atomic-layer deposition. For example, in physical vapor deposition, one would need to recover Pt from non-noble-metal shields that catch Pt overspray.
- It would be good to see the Mirai compared to the SA baseline at 500,000 systems/year. If the study finds that higher ionomer cost is realistic, the higher ionomer cost could lead to a drive for thin membranes, which would compromise drive-cycle efficiency. The project should seek to leverage resources from ANL systems analysis to estimate the impact of lower-cost design choices on drive-cycle efficiency. The project could be used to estimate the “cost” of improved fuel economy. The project should expand its efforts on stack conditioning to determine the additional conditioning costs for catalyst systems (e.g., nanostructured thin films), which would require long conditioning time.
- The team should utilize Toyota’s information to verify that the cost estimation method is a good approach. It is suggested that the team talk with DOE to obtain accurate and insightful Toyota information. Adding fuel cell cost for heavy- and mid-duty vehicle applications is also relevant. Fuel cell system assumptions should be different from those for the fuel cell systems for light-duty passenger vehicles. It is suggested that the fuel cell system assumptions be made with OEMs first.
- It would be good to see an increased focus on supporting DOE fuel cell projects at the earliest stages to ensure that the individual project’s targets and approach, if successful, will result in marked progress on cost (where applicable). Specifically, there should be greater priority on performing cost analysis of DOE projects (even at the proposal stage, if at all feasible).
- The team should reach out to non-fuel-cell researchers for different views and suggestions. For materials, recommendations include organizations such as the American Petroleum Institute, ASME, and the National Association of Corrosion Engineers. For coatings, recommendations include organizations such as Sherwin Williams, 3M, Dow, and DuPont.

Project #FC-164: Development of Corrosion-Resistant Carbon Support for Ultra-Low-Platinum-Group-Metal Catalysts (Small Business Innovation Research Phase I)

Prabhu Ganesan; Greenway Energy, LLC

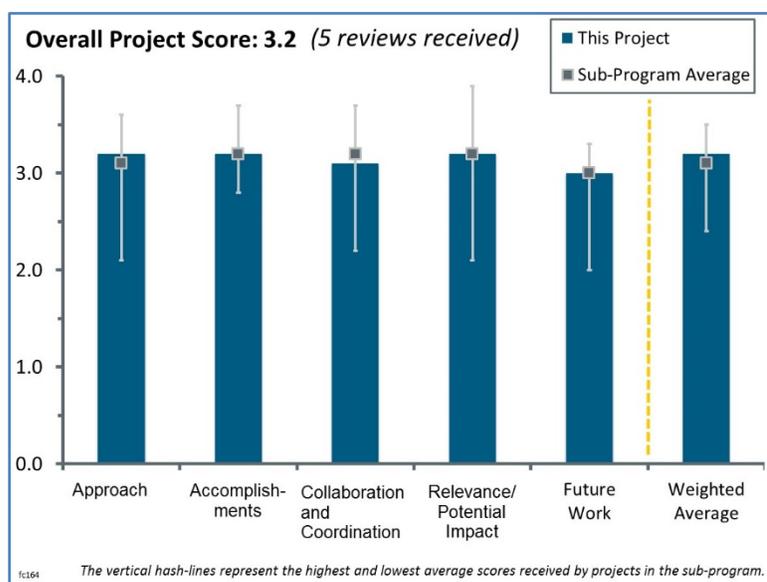
Brief Summary of Project:

This project seeks to demonstrate corrosion-resistant carbon (CRC) support stability in the presence of platinum or platinum-alloy nanoparticles under 1.0–1.5 V potential cycling condition to meet the DOE technical targets for catalyst support. The project will optimize CRC support physical properties, enhance catalyst–support interaction, synthesize platinum/CRC and platinum-alloy/CRC catalysts, and evaluate catalyst activity, support stability, and high-current-density performance.

Question 1: Approach to performing the work

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

- This project uses surface modification of a preexisting carbon to facilitate deposition of Pt with good dispersion. Next, the chemical modifications are largely removed by an annealing step prior to fabrication of the electrode layer. This is a rational approach to making good electrodes from base carbons that are intrinsically corrosion-resistant but, in unmodified form, do not allow proper dispersion of Pt. One can expect incremental improvements of catalyst durability under conditions of incomplete mitigation of start-stop and fuel starvation effects.
- The approach to utilize a functionalized corrosion-resistant carbon support is logical and feasible. The functionalization should increase Pt–support interactions and aid Pt dispersion. The approach addresses Fuel Cell Technologies Office (FCTO) barriers and uses appropriate milestones to guide the work. The work is integrated with other work and appears to build on previous work at the University of South Carolina.
- Carbon corrosion is an important issue that must be addressed. The project’s approach supports identification of carbon supports. The company is focused on ease of tailoring surface area, porosity, pore-size distribution, and hydrophilic/hydrophobic properties. To address corrosion, the project will conduct optimization of metal–support interaction through surface functionalization. Johnson Matthey (JM) will do a stable Pt deposition with uniform particle distribution (3–6 nm).
- The project approach involves surface modification of commercial carbon supports to improve their platinum dispersion properties. The team did not indicate the type of commercial carbon used, but it appears to have a high degree of graphitization. Improving Pt dispersion on graphitized carbons is a good approach to developing better catalysts.
- The project’s general approach is very straightforward and feasible. The description of the approach lacks detail, but this is not uncommon for industrial project teams. The performance metrics are outlined, but the specific steps to meeting those metrics are somewhat unclear.



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.

- X-ray photoelectron spectroscopy (XPS) results show the existence of surface oxygen functionalities. However, XPS of the precursor support should have been shown as well. Some of these surface oxygen species were probably already present in the precursor. The rotating disc electrode (RDE) durability study with cycling between 1 V and 1.6 V showed that most electrochemical surface area (ECSA) was retained, but the comparison vs. the Tanaka Kikinzoku Group (TKK) Pt/C apparently used an inherently unstable carbon. Many commercial carbons could pass this test with ECSA similar to or better than the GreenWay carbon. The membrane electrode assembly (MEA) results are actually much more impressive. The loss of only 23 mV at 1.5 A/cm² is a good result, and the overall MEA performance looks pretty good as well, assuming that the pressure given is absolute and not gauge. For future work, the team should use 150 kPa absolute since this is the specified pressure in DOE targets. The team should also show raw voltages and report high-frequency resistance, rather than iR-free voltages.
- The platinized supports have demonstrated good stability in support stability potential cycling tests in both RDE and MEAs. The data shown indicates the team can increase the micro porosity and surface area using surface modifications and increase Pt dispersion using surface functionalization.
- The project accomplishments to this point are impressive. The team has achieved good scale-up, catalyst-coated material shows significant improvement in durability, and MEA activity is promising.
- This project has started very recently and has made limited but satisfactory progress toward scale-up of the carbon production. The project will need 100 g batches for adequate testing in fuel cells. Preliminary durability testing on rotating disk and small MEAs has provided encouraging results, though comparing to standard Pt/high-surface-area carbon (HSC) cycling to 1.5 V or 1.6 V is like shooting fish in a barrel. Comparisons should also be drawn to the durability of unmodified graphitized carbons. As a durability project, this work should get past RDE studies to MEA studies as soon as possible.
- The presentation highlights include the following:
 - There was a fivefold increase in batch size (400 mg to 2.0 g). This is still a low amount for any industrial processes. The cost and volume that can be manufactured by this technology should be looked into.
 - Highly reproducible (yield, pore size, pore volume, and surface area) multiple 2 g batches are made.
 - The optimization of 5 g and 10 g batches is in progress.

Question 3: Collaboration and coordination with other institutions

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

- Getting 100 g of the carbon to catalyst manufacturer JM should lead to a high-quality evaluation of the potential of the carbon supports generated under this project. It is of some concern that Savannah River National Laboratory (SRNL) reportedly saw little degradation of TKK Pt/HSC when cycled in RDE up to 1.5 V, leading to a change in upper potential limit to 1.6 V. One would expect rapid degradation of that baseline material when cycled to 1.5 V if the testing was working properly.
- The collaboration with JM and SRNL appears to complement the capabilities of GreenWay.
- The project's collaborations appear to be working well. Catalyst supports have already been scaled up to multigram quantity. The integration of JM into the project will be a big step forward.
- The project team makes good use of national laboratory and industrial partners and their resources.
- There is in-kind collaboration with JM.

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

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

- Operational durability of polymer electrolyte membrane fuel cells is strongly related to the stability of the carbon support. Fuel cell development to this point has been optimized for carbon-supported materials. Integration of any other type of electrocatalyst has been proven to be difficult. Therefore, it would be optimal if a durable carbon support could be developed. In this regard, the current work strongly supports the goals of FCTO.
- Developing catalysts with improved performance and durability is arguably the most relevant thing a project can do. This project could be helpful in developing catalysts that can meet both activity and support durability targets.
- Improved catalyst supports can have a large impact on performance and durability. Recent improvements in high-current-density performance have been attributed to having the right pore size, volume, and distribution of Pt within the pores. Therefore, being able to adjust pore size and volume with the modifications proposed here could prove very beneficial.
- This project, if successful, could allow the use of carbon supports more resistant to degradation due to incompletely mitigated start–stop or fuel starvation effects. It is unlikely that any carbon could withstand completely unmitigated start–stop, and it is quite possible that even quite corrodible carbons can withstand operationally fully mitigated start–stop or fuel starvation.
- The project offers an important topic. However, the scale-up of carbon manufacturing is not there. Currently, the project team is focused on process optimization for 5 g and 10 g batch sizes.

Question 5: Proposed future work

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

- The proposed future work addresses the appropriate issues of scale-up and MEA testing and, critically, brings in JM to do MEA testing and optimization. Future work should also look at the impact of pore size and porosity on high-power performance.
- The planned future work constitutes a logical progression of activities to properly evaluate the potential of the innovative modified carbons. Not enough activity, durability, and performance testing will have been completed under Phase I to allow the level of promise of these carbons to be determined. There should be as much MEA testing of durability and performance as possible brought forward in time to see whether further investment in a Phase II project would be justified.
- The proposed future work includes further scale-up, use of Pt-alloy catalysts, and work with industrial partner JM. The project's future work should also include a more fundamental analysis of the mechanism by which stability is enhanced. It is unclear whether the particles are located on the surface or in the interior of the porous carbons. Interior-located catalyst particles could help to limit particle agglomeration but could also result in transport losses.
- The project's future work is appropriate but should include running the DOE start-up/shutdown protocol on the most promising catalysts. While the support accelerated stress test is useful for screening, it does not fully replace actual start-up/shutdown testing.
- The presentation's future work bullets include:
 - Synthesis and performance evaluation of Pt-alloy/CRC catalysts
 - Evaluation of initial mass activities of PtCo/CRC catalyst in rotating ring-disk electrodes (RRDEs) and fuel cell MEAs
 - Support stability studies under accelerated stress test conditions in RRDE (1.0–1.6 V) and MEAs (1.0–1.5 V)
 - Support preparation for industrial partner
 - Agreed to send 100 g CRC support: 25 g support is ready for shipment; 75 g CRC support to be prepared and shipped to JM for Pt and Pt-alloy catalyst synthesis

Project strengths:

- The project has provided dramatic improvements in catalyst stability and the systematic development of all aspects of the formation and scale-up of catalyst-loaded carbon materials. Project collaboration with JM is a good test for the viability of their carbon materials.
- The project's approach is well-thought-out, and the progress so far is impressive.
- The project addresses an important issue, but it is not certain that this company will be able to address it alone. The team should work with companies that know how to scale up carbon production, e.g., Cabot Fuel Cells.
- The stability of the carbon supports being investigated and ability to get well-dispersed Pt on these stable supports are strengths of this project.
- The project gives a rational plan for development of incrementally more durable electrodes through modification of a preexisting carbon black, assuming that the initial carbon black does have significantly better corrosion properties than typical high-surface carbon blacks. The team's very preliminary testing shows somewhat promising results.

Project weaknesses:

- No carbon can withstand totally unmitigated start-stop, and it is not clear that a carbon of modestly improved corrosion resistance would provide significant benefits in a properly designed fuel cell system.
- The project needs to partner with a university for the evaluation of the carbon that the project produces. A larger team is necessary to make faster progress.
- Scale-up seems to be progressing slowly toward the 100 g batches needed to adequately test viability.
- The project should include more fundamental investigation into the mechanism of enhanced durability.

Recommendations for additions/deletions to project scope:

- RDE testing of durability is of questionable relevance to fuel cell operation. Therefore, testing should advance more quickly to MEAs. The feature catalysts should be compared to Pt on unmodified graphitized carbons as well as to Pt on high-surface carbons. The project should include clearer milestones for continuation to a Phase II project.
- The project should include a more fundamental investigation into the mechanism of enhanced durability. The team should perform mass spectrometry measurement of carbon dioxide evolution from fuel cells to quantify carbon corrosion during accelerated durability testing.
- The team should do additional work focusing on impact of the support on high-power performance, in particular, tailoring the support and support porosity to optimize high-power performance.
- The team should partner with others and demonstrate stability of the carbon.
- The project should add start-up/shutdown testing.

Project #FC-165: Mesoporous Non-Carbon Catalyst Supports of Polymer Electrolyte Membrane Fuel Cells (Small Business Innovation Research Phase I)

Jacob Coppage-Gross; Certaintech, Inc.

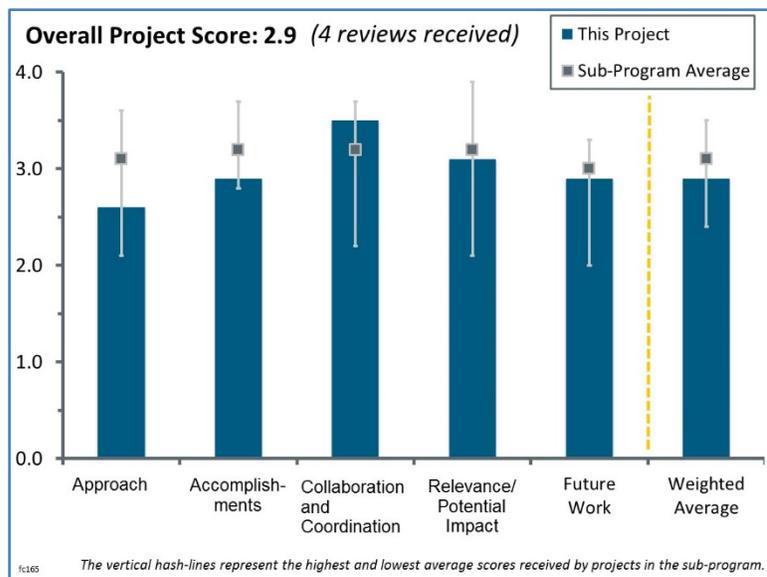
Brief Summary of Project:

The objective of this project is to improve the durability and cost efficiency of fuel cell performance through the development of a mesoporous platinum–metal carbide (Pt-MC) catalyst support material. Pt-MC offers improved corrosion resistance and lower platinum loading compared to traditional carbon-based catalyst supports. The project will synthesize and characterize high-surface-area Pt-MC nanocomposites, and the electrochemical performance of a membrane electrode assembly (MEA) incorporating Pt-MC will be demonstrated.

Question 1: Approach to performing the work

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

- MCs should provide good chemical stability, and the approach of using a sacrificial silica support to impart porosity and increase surface area is logical and feasible. An MC support could provide a corrosion-resistant support. The proposed work addresses the main problem others have had with this approach, obtaining high-surface-area MCs. It is not clear this will provide for lower Pt loadings, as there are currently mesoporous carbons available as supports and it is not clear how well Pt will disperse on these supports. The MCs and MC precursors to be investigated have not been identified, and it is unclear what the materials and processing costs for the support will be. It is unclear what process will be used to convert the precursors to carbides and how that will be done under conditions that retain the structure of the template (temperature is the main concern). There do not appear to be any milestones or targets regarding the MC support, such as a target surface area or electrical conductivity, prior to Pt deposition and testing in a rotating disk electrode (RDE).
- The project's approach is excellent. The plan is to synthesize supports, anchor Pt on them, prove stability and performance in an RDE, and then translate this to an MEA.
- The approach is somewhat novel, but the project team appears to have no idea if it will ever be economical or even if the substrates being proposed will eventually be stable. Removing the templates will require harsh chemicals (hydrofluoric acid solution [HF]), and not much is known about the cost of containing/recycling the HF. A full commercialization plan is not needed at this stage, but a primary effort is expected. The team has a long, hard path to success; the templates have to work, then the materials that the team is generating need to be shown to be stable, then those substrates have to be made catalytically active, and finally the team needs to work them into MEAs. The chances of each of these steps being successful is fairly small.
- The project's process and implementation milestones are well laid out. The project needs to increase focus on fundamental aspects of the supports, especially conductivity, but also including Brunauer–Emmett–Teller (BET) surface area measurement, pore size distribution, and porosity.



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.

- While the project has not been active for long, it appears to be behind schedule for completion of Phase I by November 2017. At this point, the team should have some supports made. However, no preliminary results for any supports were shown, just results showing the project has a pure precursor for the MC and results showing the team can make a mesoporous silica template. There are no results showing the project can make a mesoporous or high-surface-area MC yet.
- The accomplishments are few because the project started only at the end of February. However, this is only a nine-month Phase I project, and the authors have much to do in the remaining six months. The project is still in its first task and needs to actually make high-surface-area supports and functionalize them soon.
- The team is just beginning. The team has begun making templates and the substrates, which is appropriate.
- This project has recently started but has made progress toward first samples.

Question 3: Collaboration and coordination with other institutions

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

- The team's working with Dr. Shimpalee is a good start, as he has much experience in MEA fabrication and characterization. Achieving mass activity loss of <40% for electrocatalyst support stability is a very large task, as catalyst activity in MEAs is a very large undertaking.
- Collaborations have been set up between the partners, and they appear to have the capabilities needed. It is early in the work, and they have not had material to test or characterize yet. Inclusion of a commercial catalyst or MEA manufacturer in Phase II would be beneficial.
- The project has excellent collaborators for electrochemical fabrication and testing.
- The project has good collaboration with two universities and one other company.

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

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

- The project aligns with the Fuel Cell Technologies Office goals to improve the durability of polymer electrolyte membrane fuel cells and could decrease costs if durability is increased, allowing for lower initial Pt loadings.
- A successful project will have a significant impact by producing supports with a high-stability catalyst surface area and conductivity that can replace carbon in highly oxidizing environments.
- The project's topic is certainly relevant to DOE goals. The likelihood of success or even demonstrating feasibility in Phase I will be daunting. The team has not really given enough information about how it will stabilize the supports or why the supports are expected to be stable enough to judge.
- A novel support that can help meet DOE targets is valuable. The project needs to make sure it meets both performance and durability targets.

Question 5: Proposed future work

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

- The project goals are quite ambitious for time and funding, but in general, the team is taking the correct approach. MEA testing should be put off until Phase II. It would be a successful Phase I if the researchers can demonstrate that they can make the substrates, they have the rheology the team is looking for, and the product is stable. Then catalyst generation and MEA fabrication could be the focus of Phase II.

- A good plan is in place for both Phase I and Phase II, but the project needs to directly measure conductivity, BET surface area measurement, etc.
- It is unclear what the Phase I target is. It is unclear what determines success (all project goals are very qualitative). The principal investigator just listed the DOE targets. There seem to be no targets associated with the Phase I and proposed Phase II of this project. There should be clearly defined mass activity, performance, and durability targets that should determine whether the project would go forward. For example, the end-of-life performance after one thousand 1 V to 1.5 V cycles should be better than an electrocatalyst-carbon-based Pt/C catalyst. This should not be too difficult to achieve and should set the stage for better balance-of-life performance in Phase II.
- The proposed future work addresses the broad areas of concern, but the plans are generic and do not address issues specific to the proposed MC-based supports.

Project strengths:

- This project has a good plan for material fabrication. This project has a good plan and collaborators for electrochemical fabrication and characterization.
- This project offers a different approach to try to obtain high-surface-area MCs to use as supports.
- This project has a good team with all relevant capabilities. This project is an interesting idea.
- This project has a proper approach, but it is difficult to gauge without knowing what the composition of the supports is.

Project weaknesses:

- The project lacks focus on important material properties of non-carbon supports: conductivity, BET surface area measurement, pore size distribution, and porosity.
- This project has many hurdles to success. There is uncertainty about whether the supports will be stable. Multiple steps will likely lead to an expensive catalyst.
- The project needs quantitative success/failure criteria for Phase I.
- There is a lack of information on MCs and precursors to be used.

Recommendations for additions/deletions to project scope:

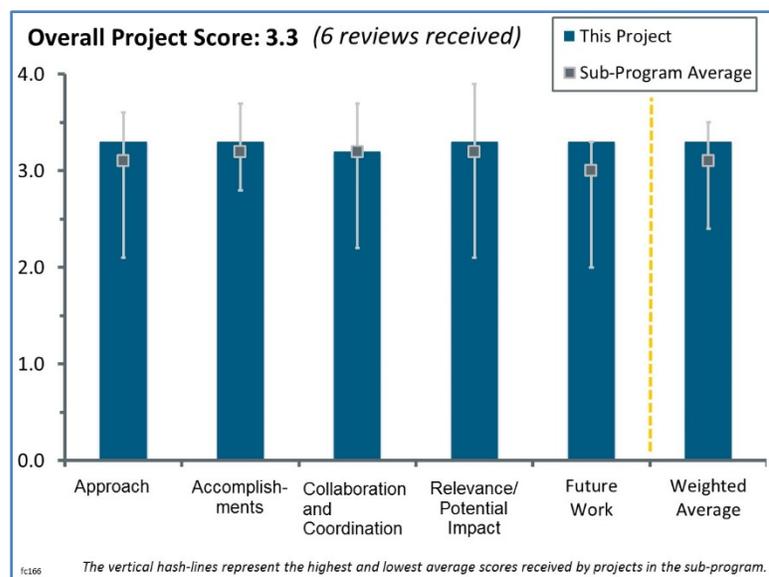
- Some milestones or targets for the MC support prior to Pt deposition and RDE testing may be useful. Minimum electrical conductivity and surface area targets for the support should be met before moving on to platinization and RDE testing.
- The project should focus just on the supports and demonstrating their stability and that they have the structure that the project is seeking in Phase I. If time permits, the team should add the catalyst to the surface, but MEA testing and fabrication should wait until Phase II.
- It is strongly recommended that the project include a detailed plan to assess MC properties.

Project #FC-166: Development of Durable Active Supports for Low-Platinum-Group-Metal Catalysts (Small Business Innovation Research Phase I)

Barr Halevi; Pajarito Powder

Brief Summary of Project:

To enhance the durability of fuel cell electrocatalysts and improve the economics of polymer electrolyte membrane fuel cells (PEMFCs), this project will further develop Pajarito Powder's durable active carbon supports (DACs). This will be done by optimizing the DACs materials characteristics and platinum-support interactions, improving the corrosion resistance of support carbon, and increasing support-platinum activity and durability synergism. To validate DACs performance, platinum-DACs will be deployed in membrane electrode assemblies (MEAs).



Question 1: Approach to performing the work

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

- The project's approach of utilizing different techniques and materials to prepare platinum-group-metal-free (PGM-free) catalysts to prepare active supports for low- Pt catalysts is feasible and addresses fuel cell barriers of cost, durability, and performance. The team has demonstrated good activity with PGM-free catalysts and also the ability to modify the morphology to improve transport and durability in these materials. In addition, N-doped carbons should provide better anchoring for Pt and improve dispersion. The Pt is likely to bond to the PGM-free active site, as the support will likely provide very little to no additional activity for the oxygen reduction reaction (ORR).
- The early application to MEAs and polarization curve testing to higher current densities at low (≤ 0.1 mg/cm²) Pt loadings are appreciated. The project notes the use of iron in the support, an issue known to have negative impacts on PEMFCs. The team should either focus on removing and demonstrating retention of support activity, or demonstrate that the iron does not have a negative impact on performance over life (preferably, the former). Graphitization usually results in reduced surface area and high-current-density performance. While high surface area was mentioned, no data were shown. The project is asked to communicate the electrochemical surface area (ECSA). Tailored pore size is mentioned; it was unclear whether the project considered the findings from FC-144 in pore size impact.
- The team is trying to develop a new corrosion-resistant support for a cathode without PGMs. This is important for start-stop stability.
- This project applies the principal investigator's (PI's) previous experience in PGM-free catalyst development to low-Pt catalysts. The idea is generally good, although the justification on the support stability improvement is not clear.
- It is difficult to see how a combinational approach, non-PGM supporting PGM, is better than just one. Generally, one ends up doing all of the work. It is difficult to see that there will be a synergy between the catalysts. One might say that if the catalyst is going to be supported, why not make it an active support so it is worth pursuing.

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 progress for so early in the project. The early MEA fabrication and polarization curve testing to high current densities with low Pt loadings and cycling results are appreciated.
- The project has been active for only a short time but has already shown improvements in durability from ~80% loss at 0.6 V to ~40% loss at 0.6 V after a start–stop cycling test. However, losses are still much higher than for the DOE target of 30 mV loss at 1.5 A/cm² (data showed >150 mV losses at 1 A/m²).
- Within a short time, the project has made excellent progress.
- It is difficult to rate this because the accomplishments that the project shows are exceptional but were clearly done outside of the project. The team has made many catalysts and tested them in MEAs with continued improved performance, yet the project has spent only \$3,000. There is good improvement on the support durability. The team’s assumptions behind the cost reductions with regard to Pt/C are unclear, especially through 2030.
- The project started only two months before the Annual Merit Review presentation was due. It is too early to assess the full accomplishment.
- The project has just started.

Question 3: Collaboration and coordination with other institutions

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

- While Advent Technologies has experience in electrode development, the team may consider working with Los Alamos National Laboratory (LANL) and/or the Fuel Cell Consortium for Performance and Durability (FC-PAD) for MEA testing (and perhaps MEA development). The team could consider collaborating with Strategic Analysis to validate process costs and overall cost impact.
- The project has only Advent Technologies as a collaborator, which is not surprising for a Phase I effort. Adding Karen More at Idaho National Laboratory would be a good idea for visualizing these catalysts, especially after cycling.
- Collaboration between Advent Technologies and the PI appears to be good, and they have been collaborating successfully in other projects. Collaboration with others outside the project was not apparent.
- The project collaborates with another company for MEA and fuel cell research experience and testing capability.
- The project has just started, and it is hard to judge this aspect, but it does seem to have a good partner in Advent Technologies.

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

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

- Since original equipment manufacturers (OEMs) will employ system mitigation strategies to reduce the impact of carbon-support corrosion, the relevance was rated only as “Good” instead of “High.” As such, OEM focus has moved more to performance at high current densities (at low loadings) with corrosion resistance at low temperatures (relevant to startup) as a secondary priority.
- Lowering PGM loadings while improving support durability is still very relevant and necessary, and this is a viable approach.
- The project advances FCTO goals to reduce Pt loading and increase PEMFC durability. The project objectives are in line with DOE targets and goals.
- This project covers an important component of fuel cells.
- The project addresses fuel cell catalyst support durability, but tests so far were mainly on catalyst/MEA performance. Support-specific tests need to be carried out.

Question 5: Proposed future work

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

- The project focus on MEA development is appreciated. It would be good to see the future plan include:
 - Removal of iron
 - Testing ECSA, and a plan to increase high-surface-area carbon if less than state of the art
 - High-level cost analysis with Strategic Analysis
 The project might also consider testing at LANL, as well as collaboration with LANL/FC-PAD on electrode and MEA development.
- The project's proposed future work is appropriate and logical. The project has appropriate milestones and down-selection points.
- What the project team proposed at the presentation makes sense.
- The team's future focus should be improving support durability and switching to alloy catalysts.
- The Phase I plan is somewhat generic. There should be more specifics on support-stability-related activity.

Project strengths:

- The team and the team's experience with methods to provide porosity and high graphitic content are strengths of this project.
- The project is off to a good start. The team is very well aware of challenges and issues ahead of them and the relevant metrics and obstacles to achieve them.
- There is company-demonstrated capability to make active supports. The project team has an MEA partner for development experience.
- The project has a good team and approach.
- The project approach has a good initial start.

Project weaknesses:

- There must be some sacrifice in making a support active for the ORR, either in materials, cost, structure, or the performance of PGMs. The team will need to demonstrate that these sacrifices are worth it.
- Durability improvement on the catalyst support needs to be demonstrated to justify why the proposed doped materials should have better stability than carbon.
- The project could benefit from further collaborations on MEA development/test and cost analysis.
- The budget is modest; one hopes it is adequate.

Recommendations for additions/deletions to project scope:

- From a stability standpoint, 3 nm particles may be a bit small, as previous work has shown slightly larger particles to be more stable toward dissolution. A study looking at the effect of Pt-support interactions on particle-size stability might be beneficial.
- There are no recommendations. The team is on the correct path to develop and demonstrate these materials. If funding increases, the project will want to partner with an OEM for large-scale fabrication and testing, but that is at least two years away.
- The project should continue focus on MEA development and testing to high current density at low Pt loadings, the removal of iron from supports, and the demonstration of high surface area.

Project #FC-167: Multi-Functional Catalyst Support (Small Business Innovation Research Phase I)

Minette Ocampo; pH Matter LLC

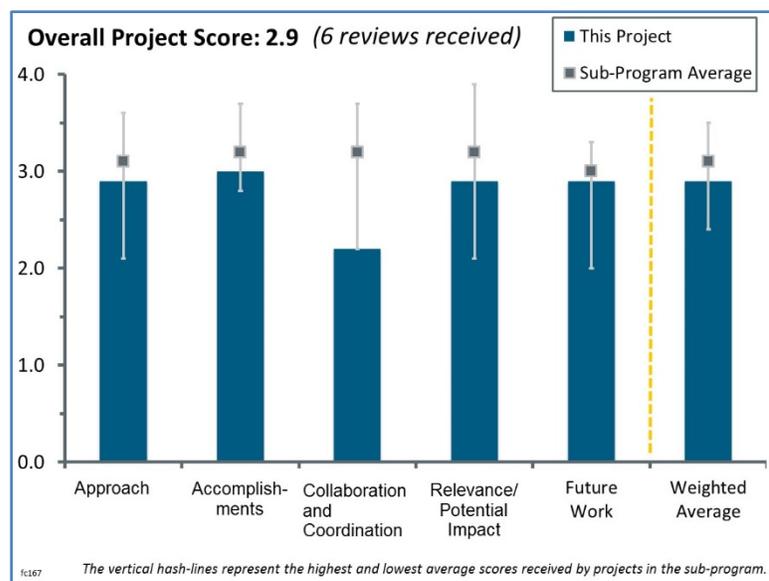
Brief Summary of Project:

This project seeks to develop a multifunctional carbon support engineered to perform better than conventional pure carbon supports used in polymer electrolyte membrane fuel cells (PEMFCs). The support material being developed aims to meet U.S. Department of Energy targets for durability and oxidation resistance and increase hydrophobicity to enable higher current density. Project tasks include (1) material synthesis and characterization, (2) membrane electrode assembly (MEA) fabrication and characterization, and (3) MEA testing.

Question 1: Approach to performing the work

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

- The approach addresses the barriers of durability and cost. The project is well defined and reasonable, and the approach is feasible and integrated with other efforts addressing platinum-group-metal-free (PGM-free) catalysts. The approach to utilize active PGM-free catalysts as supports for low-Pt electrodes is logical, and the N sites should lead to increased Pt dispersion and stronger Pt-support interactions. The support developed by pH Matter demonstrated good durability, even at the high potentials seen in electrolysis as they have been tested in reversible fuel cell mode cycling from 0.6 to 1.7 V. Therefore, the supports should have improved durability for start-stop-type cycling (which goes only to 1.5 V). It is likely the Pt will bond at the active sites for the support, poisoning those sites so the activity of the new structure is not likely to be Pt activity plus the activity of the support without Pt. However, this should lead to stronger Pt-support interactions and more stable Pt.
- This project repurposes a non-PGM carbon-based catalyst as a support for Pt particles. The non-PGM catalyst has demonstrated good stability during oxygen reduction reaction/oxygen evolution reaction (ORR/OER) potential cycling in alkaline conditions. The proposed approach is simply designed, straightforward, and feasible.
- The project's approach is related primarily to using PGM-free ORR catalyst-related materials as a support for Pt to increase Pt mass activity. This approach addresses performance and cost targets. However, it likely does not address the durability-related targets. The approach is likely using materials as a support that are less durable than traditional carbons and especially graphitized carbons.
- Starting with a doped carbon support with intrinsic activity is a good approach if it is well established. It is good that it is not iron-based. However, more detail is necessary to evaluate the approach. Tuning for hydrophobicity is stated, but it is unclear what the target is or why a particular hydrophobicity target should be chosen. It is also unclear what the approach is to determine optimum hydrophobicity (e.g., what the tests and metrics for success are).
- This project applies Pt to pH Matter's iron-free, water-selective, PGM-free catalyst support for improved catalyst stability at low platinum loading. The idea is generally good, although the scientific rationale for the support stability is not clear.



- The objective of this project is to develop a multifunctional carbon support that is engineered to perform better than conventional PEMFC pure carbon supports. It is not clear what kind of multifunctional carbon support will be developed.

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 is a Small Business Innovation Research (SBIR) project that just started and had approximately one month of work prior to having to submit slides for the DOE Hydrogen and Fuel Cells Program (the Program) Annual Merit Review (AMR). It is too early to grade on accomplishments. Pt dispersion has been achieved on the pH Matter support showing good dispersion (by transmission electron microscopy [TEM]). Rotating disk electrode (RDE) results show a similar on-set potential as Pt/C but with widely varying limiting currents. It is unclear why limiting currents vary so much. These RDE measurements should be made in acidic media, if they were not. Slide 8 says 0.1 M HClO₄, but the slide does not indicate media. In terms of results, pH Matter demonstrated some durability in alkaline media but not yet in acidic media, which is the much larger challenge.
- The project started only two months before the AMR slides were due. Therefore, not much has been accomplished. There is some preliminary evidence that the use of the project's carbon supports results in a higher ORR activity than commercial Pt, potentially related to the dispersion of the material on the support. No results related to the durability of the materials have been presented.
- The project has been active only a short time but in this time, pH Matter has shown data indicating the researchers can get good RDE performance with Pt deposited on their active supports. The RDE data showed better performance than a commercial Pt/C, and TEM showed good Pt dispersion.
- The team has shown good dispersion and some activity with ultra-low PGM loadings, so the project has a fair start, given the very limited project duration thus far.
- Through this project, pH Matter developed a catalyst with CNxPy support that demonstrated good ORR activity with low Pt loading.
- The project started only two months before the AMR presentation was due. It is too early to assess the full accomplishment. Initial data from the previous studies look promising.

Question 3: Collaboration and coordination with other institutions

This project was rated **2.2** for its collaboration and coordination.

- There does not appear to be any collaboration at this stage in the work, though this is an early-stage SBIR. Collaboration with an MEA developer is planned in Phase II and would be helpful. Potential collaborators are not identified.
- This is a Phase I SBIR with no partners; collaboration is not expected. The researchers have shown collaboration with Giner and National Renewable Energy Laboratory (NREL) on similar materials.
- There is collaboration with Giner and NREL on another SBIR project. The collaborators on this project are not identified.
- There are no collaborations. If that is not expected in a Phase I SBIR, it is up to DOE to increase the rating or ignore it.
- There are no collaborators in this phase of the project.
- The project at Phase I does not have any collaborators.

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

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

- While carbon corrosion at operational temperature is less of a priority now, because of original equipment manufacturers, system mitigation, high activity, high performance at high current density, and catalyst durability at temperature remain priorities, and thus the project retains relevance.
- The project supports the goals and objectives of the Program and, if successful, would have an impact on reducing Pt loading and increasing durability, thus decreasing costs.
- Stable carbon supports can significantly improve the operational durability of PEMFCs. This project has a direct impact, as it addresses one of the core metrics for PEMFCs provided by the Fuel Cell Technologies Office.
- The project addresses fuel cell catalyst performance and support durability. The team's tests so far were mainly on catalyst performance tested by RDE. Fuel cell data should be forthcoming.
- This project primarily addresses cost and performance. It is unclear what its impact will be on durability.
- It is too early to judge. More progress has to be made.

Question 5: Proposed future work

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

- The project team proposes to address not only the impact of support structure and composition but also the catalyst deposition procedure on the durability of the catalyst material. Additionally, the team proposes to study the degradation mechanism to gain further important insight and guide the further development of the material. This last part is often neglected by other projects, yet it is critical to the development of more durable materials.
- Almost the entire project at this point in time is future work, as the project just was initiated. Initial experiments rely on RDE testing for both performance and durability (support oxidation). After RDE, gas diffusion electrodes (GDEs) will be made for MEA testing and then durability testing. RDE performance can vary widely, and many developers have substantial difficulty in obtaining similar results in MEAs. RDE testing should include ionomer in the catalyst layer. Even initial materials should be moved to GDEs for MEA testing to understand how the RDE tests correlate to real systems. "Alternative" preparation methods are to be explored. These methods should follow on only if RDE and MEA tests show that the carbon support materials have good durability. PGM-free materials based on carbon have not shown good durability to date. Varying the Pt deposition method is not going to affect the support durability; thus, the support durability should be demonstrated before exploring a sequence of Pt deposition methods.
- The proposed future work is logical and addresses the relevant barriers. Partnering with the Fuel Cell Consortium for Performance and Durability (FC-PAD) may be beneficial for electrode characterization and the breakdown of potential losses. Work tailoring structure of the support (carbon porosity and pore structure, agglomerate size, and structure) may be needed and is not apparent in the future work description.
- The focus on quickly moving to MEA fabrication and testing is appreciated. However, the project should set a higher goal than "achieves $>0.8 \text{ A/cm}^2$." State-of-the-art catalysts now perform at $>1.0 \text{ W/cm}^2$ ($>1.5 \text{ A/cm}^2$ with $>0.66 \text{ V}$), a suitable metric for comparison. High-current-density performance should be the goal and will better identify proper property tuning.
- These bullet points are all too general, and the team needs to be more specific in saying who will do what:
 - Explore alternative preparation methods for low-PGM alloy catalysts, such as electrochemical, colloidal, or ion-exchange
 - Further MEA optimization to address mass transport and cathode flooding issues
 - Electrode characterization before and after cycling to better understand degradation mechanisms
 - Partner with MEA manufacturers
- A fuel cell test needs to be carried out soon to verify the RDE data.

Project strengths:

- Use of previously developed material should help shorten development time. The team has significant expertise in catalyst and support development. Support development already shows stability in reversible alkaline fuel cells.

- The use of pH Matter's stable C-NxPy PGM-free catalysts as a support is a strength, as these catalysts have shown excellent stability and good activity for ORR.
- The team has demonstrated the ability to make non-PGM catalysts with some activity and low-loaded-PGM electrodes with reasonable dispersion.
- The project approach has a good initial start.
- The project attempts are promising with functional carbon support.

Project weaknesses:

- There is a lack of evidence of proposed support stability in acid; it could be different from that observed in alkaline electrolyte. The potential for improvement in durability is obvious; however, the mechanism of improvement in activity is less obvious. It is unclear why a carbon support would make low-PGM catalysts viable. At low catalyst loadings, oxygen starvation is a big problem at high current densities, which does not appear to be addressed. It is unclear what makes metal adhesion better on the proposed support materials.
- It is unknown how ionomer in a catalyst layer will interact with these carbon-based support materials; increased hydrophobicity is not necessarily the correct optimization strategy. The authors state that the support will include better durability, but it is unclear what the basis for that is.
- It is unclear whether the team has expertise in fabricating state-of-the-art electrodes and MEAs. The project will benefit from collaboration with FC-PAD in this regard (as stated in the future work).
- The project does not appear to pay sufficient attention to the physical structure and porosity of the support, which will have a large impact on high-power performance.
- For presenting catalyst data, the principal investigator is advised to provide the numerical results such as onset and halfwave potentials for better assessment.
- MEA data needs to be presented as soon as possible.

Recommendations for additions/deletions to project scope:

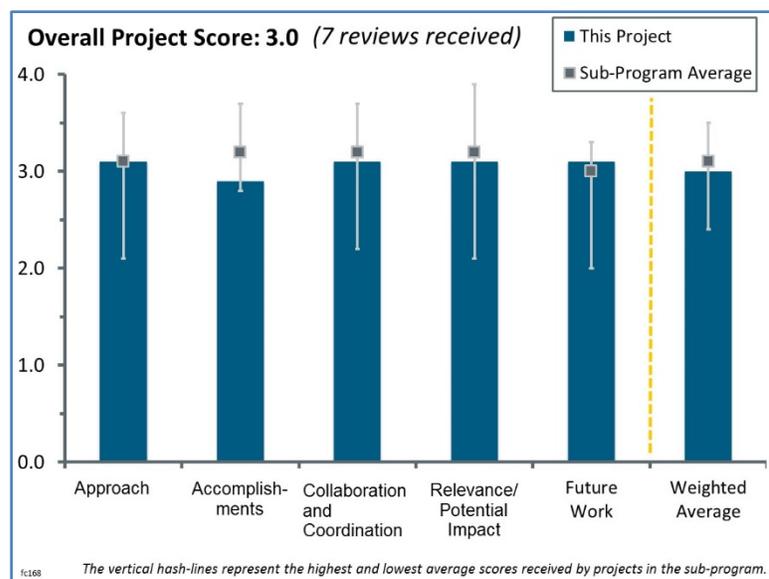
- The project should focus more on high-current-density ($>1.5 \text{ A/cm}^2$) operation under both wet and dry conditions to lead the team's catalyst down-selection.
- "Alternative" preparation methods should be explored only after demonstrating support material durability in acidic media.
- The project should assess the impact of support morphology on oxygen starvation for low-PGM materials in MEAs.
- The project should focus on generating more data.

Project #FC-168: Highly Robust Low-Platinum-Group-Metal Membrane Electrode Assemblies Based upon Composite Supports

Arrelaine Dameron; Forge Nano

Brief Summary of Project:

The objective of this project is to demonstrate a successful overcoat method on commercial low-platinum-group-metal platinum-carbon catalysts by targeting uniform coverage of the carbon support with gas phase access to the platinum catalysts. Project activities include evaluation of the activity, ohmic resistance, and cycling stability of overcoated catalyst materials by rotating disk electrode (RDE) and membrane electrode assembly (MEA) testing; demonstration through MEA testing of improved cycling durability of optimized encapsulated catalysts without significant loss in activity; and down-selection of viable encapsulated platinum-carbon catalyst material based on performance, process scalability, and technoeconomic considerations.



Question 1: Approach to performing the work

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

- The approach directly addresses the barrier for durability. The project appears feasible, but the feasibility is tied to the overcoat materials being investigated, and they are proprietary; so it is not clear whether they will have the appropriate cost, conductivity, or other properties. The overcoat technique has been used to limit noble metal migration and agglomeration on other catalysts and is likely to improve Pt stability and reduce corrosion. The synthetic approach is novel and cost-effective for a deposition technique. The investigators can produce catalysts in large quantities. It would be helpful to have a conductivity target for the overcoat. It may also be helpful to do some deposition on a substrate where the conductivity of the overcoat could easily be measured on a planar surface. It is not clear how deposition of the overcoat on the Pt or deposition blocking access to Pt sites inside the micropores will be prevented or minimized. A target or milestone to demonstrate selective deposition or deposition without blocking access to Pt sites should be included early in the project. Testing should also include cycling over the operating range of interest (at least 0.6–1.0 V vs. standard hydrogen electrode) as well as the start–stop cycle. Phase II studies should go to MEA-level testing as early as possible. Optimum coating thickness and pore size for performance in the RDE liquid environment could be quite different from the optimum for the environment in an MEA.
- This project applies atomic layer deposition (ALD) to overcoat Pt and carbon support as a means to protect catalyst degradation. The idea is generally good, backed up by previous publications. To demonstrate its feasibility, the project must move aggressively in the actual catalyst testing, at least at the RDE level.
- The ALD overcoat approach to addressing electrode durability is relatively novel. It will be interesting to see whether this can be achieved without sacrificing performance, specifically, potential mass transport limitations at higher current densities.
- The project has a good focus on key barriers with mitigation strategies in place. Multiple material approaches and barrier strategies are planned.
- The project approach is to demonstrate an overcoat for the catalyst support to improve durability. It seems that this approach addresses the durability target. However, the approach does not address cost and/or

performance targets and likely makes for a more expensive catalyst. The project has plans to prevent over-coating.

- This project offers a good approach. It is clear how they plan to coat the carbon but not the Pt. The document provided by the project team includes a relevant figure on slide 7 (top right).

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 has just begun. The investigators have prepared 11 different batches of catalysts, which seems reasonable and on schedule for the synthetic portion of the work. However, it is not clear how physical properties of the coating or samples (e.g., thickness, conductivity, hydrophobicity) have been varied. According to the schedule, there should have been some RDE results by this time, but none were available.
- This is a Small Business Innovation Research (SBIR) Phase I project that had approximately only one month after project initiation before slides were due for the Annual Merit Review (AMR). As such, limited progress to date is expected. The ALD process has been demonstrated forming nano-islands.
- Given the early stage of the SBIR, limited progress is to be expected. The project presented some initial ALD results (slide 11), but it was not clear what the target or metric of success was. It was unclear if the team replicated the durability results as shown in the literature in slide 6.
- The project is only a few months old. The project has made good progress for this time. They have made several samples.
- Initial materials have been produced, and electrochemical tests have begun.
- The project began only two months before the AMR presentation was due. Therefore, limited accomplishment is to be expected.
- The project just started.

Question 3: Collaboration and coordination with other institutions

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

- The project offers excellent collaboration. There will be complementary capabilities between the University of Connecticut (UConn) and Forge Nano.
- The project has well-qualified collaborators for electrochemical fabrication and testing.
- Forge Nano is collaborating with UConn for the electrochemical measurements while Forge Nano concentrates on catalyst synthesis.
- The project collaborates with two research groups at UConn with fuel cell catalyst research experience and testing capability.
- It is difficult to determine how well the collaboration is working because the project has just begun. However, there were no testing results yet. It would be helpful if an MEA developer was involved in Phase II because there are integration issues and an optimization of electrode layer properties (e.g., hydrophobicity, ionomer content) will be needed.
- There is adequate collaboration for a Phase I SBIR (UConn for testing/characterization). The project mentions using commercial catalysts, but it is unclear who is fabricating MEAs. The project could consider working with the Fuel Cell Consortium for Performance and Durability and Los Alamos National Laboratory for fabrication purposes.

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

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

- This is an excellent translation of demonstrated technology from gas-phase catalysis to electrocatalysis. There is a high probability of success and impact.
- Catalyst sintering with voltage cycling remains a key area of interest in developing high-performance, low-/ultra-low loaded catalysts. As such, anything that can arrest Pt dissolution is of interest. It is unclear whether this performance can be maintained.
- The project is relevant, supports the Fuel Cell Technologies Office goals and objectives, and directly addresses catalyst durability and performance issues.
- The project addresses fuel cell catalyst durability, which is a key challenge for the current catalyst.
- The relevance and impact will be higher if the team defines concrete targets for both Phase I and Phase II. It is unclear what the targets are for conductivity, activity, and durability.
- The project addresses the durability target. However, it does not address the cost and/or performance targets, which likely makes for a more expensive catalyst.

Question 5: Proposed future work

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

- The inclusion of MEA testing and an economic assessment is appreciated. It is not clear whether high current density and potential mass transport limitations are being tested or addressed. If the process meets the initial metrics, those results are make-or-break and should be included. It is unclear whether the expanded lifetime testing listed on slide 16 follows the high current density demonstration. Durability, conductivity, and potential contamination issues are all of interest. If the project is successful with commercial platinum-carbon catalysts, the demonstration of a state-of-the-art platinum-alloy catalyst would be valuable. It would be especially valuable to explore the impact on pore-size optimized state-of-the-art catalysts as the FC-144 project demonstrates.
- The project's future work and targets are well laid out. A material down-selection early in the project and additional focus on characterizing transport properties through layers would be beneficial.
- An ALD-based approach has not been used in a fuel cell catalyst; an exploratory study can verify its feasibility.
- The proposed future work is appropriate, but it may be too focused on RDE testing. The liquid environment of RDE minimizes some transport issues that can exist in an MEA and could be affected by coating thickness, coating composition, etc.
- This project just started; therefore, the majority of the project is future work.
- The project should define MEA performance and durability targets to define success criteria of Phase I.

Project strengths:

- The project's main strength is Forge Nano's continuous ALD system and the potential for an overcoat to stabilize platinum or platinum-alloy nanoparticles.
- This project offers an excellent concept to build on with a good starting material set. Forge Nano has a track record of scaling up ALD. The property measurements appear complete.
- An overlayer by ALD that is not line-of-sight limited is a novel approach to improving durability.
- This is a novel method addressing an important durability problem for catalysts/supports.
- The project approach is reasonable based on the publications in the field of catalysis.
- The team's expertise in ALD is an apparent strength.

Project weaknesses:

- The use of an overlayer coating by ALD likely makes for a more expensive catalyst. It is also likely the catalyst will have performance limitations. The type of carbon used is potentially crucial to this project. High-surface-area carbons have a large degree of Pt in the interior of the carbon. Other carbons (such as graphitized) have little interior Pt. It is unclear how the ALD process will be at coating the carbon in the interior and whether the pores in the higher-performing carbons will remain open or will be “plugged” by the coating. If they become plugged, that could greatly limit the catalyst performance and make interior Pt inactive. The coating thickness appears critical to this project’s success. If the layer is too thick, the Pt particles will probably be covered, resulting in additional mass transport limitations. To evaluate, UConn should measure electrochemical surface area, mass activity, and high current density performance and impedance.
- It is not clear whether selective deposition on carbon or optimal deposition thickness is targeted. Material down-selection should answer that question as soon as possible. Ballpark resistivity and permeabilities should be known for some ALD layers and may aid in down-selection.
- The project would benefit from an experienced MEA developer in testing at higher current densities that are necessary for investigating potential mass transport limitations.
- Durability improvement should be demonstrated soon, at least at RDE level, which should not be too time-demanding.
- The project weakness is the lack of details available about the overcoat composition.
- The project does not offer any quantitative milestones.

Recommendations for additions/deletions to project scope:

- After initial characterization, if successful, investigating mass transport and high current density performance is suggested, followed by testing with state-of-the-art, porous, high-surface-area platinum–alloy catalysts.
- The type of carbon should be carefully documented and controlled. In addition, various carbons should be evaluated. The project has plans to prevent over-coating when coating the Pt. This needs to be proven as true.
- The addition of microscopy as a morphology descriptor would aid the understanding of resulting performance. This may fit well into Phase II.
- Bringing an MEA developer into Phase II would be beneficial.