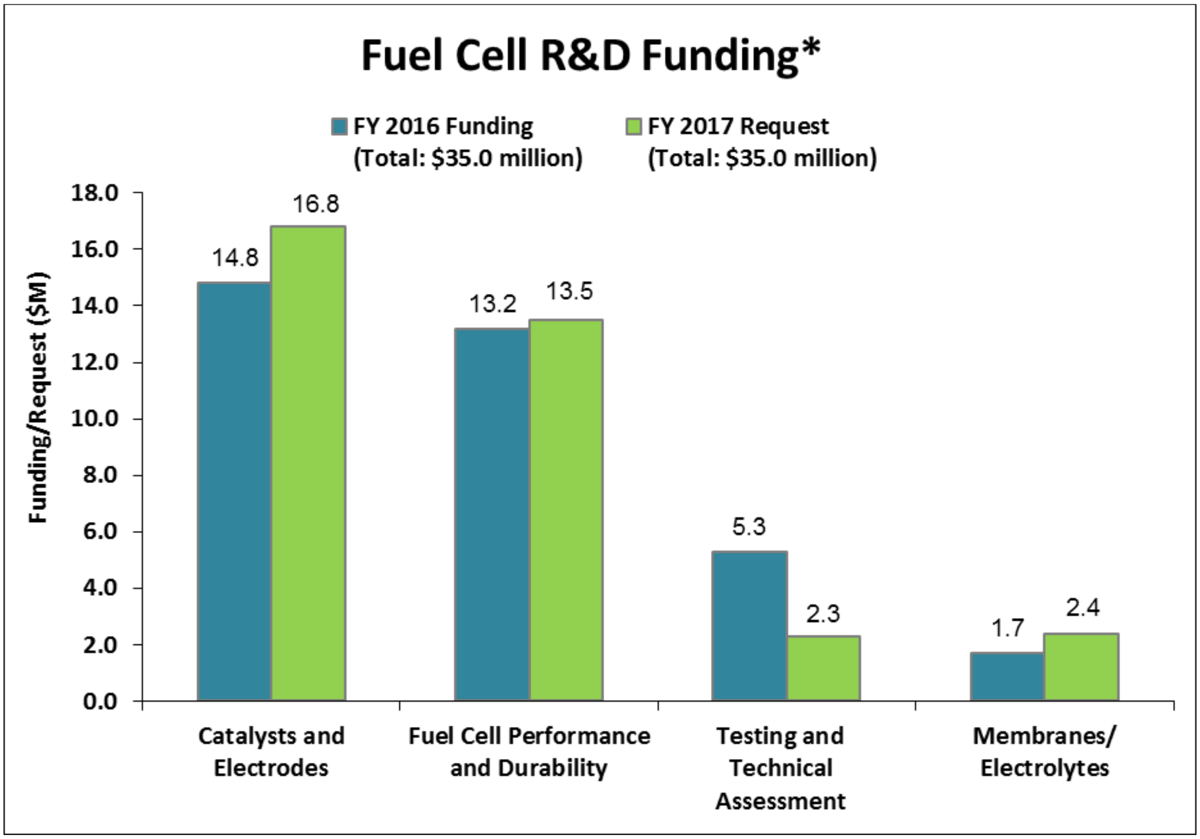
2016 — Fuel Cells
Summary of Annual Merit Review of the Fuel Cells Program

Summary of Reviewer Comments on the Fuel Cells Program:

Reviewers felt that there was a good balance between near-, mid-, and long-term research and development (R&D) in the Fuel Cells program, and they agreed that cost and durability are the major technical challenges. Reviewers praised the program's approach to identifying and addressing these issues and noted its well-structured, focused, and well-managed projects as a strength. In particular, the consortia established by the program were lauded for their potential to transform fuel cell technology. However, some reviewers commented that progress on fuel cell cost reduction has stagnated in the last few years. Key recommendations include increasing focus on technologies that will build on progress achieved thus far, decreasing emphasis on alkaline fuel cell technology, focusing on developing better transport properties for platinum-group-metal (PGM)-free catalysts, and establishing clear and ambitious go/no-go criteria to allow for ending projects not meeting these criteria. Also, one reviewer suggested setting aside a portion of each year's appropriation to support smaller projects, with particular encouragement given to new applicants.

Fuel Cells Funding:

The program received $35 million in fiscal year (FY) 2016. The request for FY 2017 is $35 million. The program focuses on reducing fuel cell costs and improving durability. Efforts include 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 by addressing mass transport and degradation issues. The FY 2016 funding opportunity announcement will result in funding for new fuel cell performance and durability, as well as catalyst and electrode projects. There is no funding in FY 2016 or planned funding in FY 2017 for balance-of-plant (BOP) component projects.



* Subject to appropriations, project go/no-go decisions, and competitive selections. Exact amounts will be determined based on research and development progress in each area.

Majority of Reviewer Comments and Recommendations:

At this year's review, 48 projects funded by the Fuel Cells program were presented, and 33 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 2.6 to 3.5, with an average score of 3.1. This year's highest score of 3.5 and average score of 3.1 were similar to last year's highest and average scores of 3.6 and 3.1, respectively. The lowest score of 2.6 for all projects reviewed in 2016 was a modest increase from 2015's low score of 2.5.

Catalysts and Electrodes: The scores for the nine catalyst projects ranged from 2.7 to 3.2, with an average of 3.0. Reviewers praised the highest-rated project for the progress the project has made in improving durability over its lifetime and the diversity and quality of its team members. However, reviewers commented that the project team had paid inadequate attention to the engineering of thick catalyst layers that resulted in inadequate transport properties. For the lowest-scoring projects, reviewers noted that the project teams had failed to address fundamental barriers in the technology. In one case, this was the over-reliance on PGM catalysts, and in the other, the technical problems with metal supports.

Fuel Cell Performance and Durability: Six projects, all part of the Fuel Cell Performance and Durability (FC-PAD) consortium, including the consortium overview, were reviewed. Three projects received a high score of 3.3, two projects received a low score of 3.1, and the remaining project received a score of 3.2, for an average score of 3.2 for FC-PAD overall. Reviewers praised the highest-rated projects for the relevance of their focus on catalyst and support durability, 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 would face challenges without fostering 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 suggested that the projects shift focus to a foundational understanding of degradation causes and novel fuel cell testing techniques.

Testing and Technical Assessment: Eight projects were reviewed and received scores between 2.7 and 3.4, with an average score of 3.2. Reviewers lauded both the highest-rated projects for their collaborations across the program and with industry, as well as for their focus on addressing specific problems and answering specific questions raised to the program. Reviewers commented that the lowest-rated project's reliance on nanostructured thin film (NSTF) systems was a weakness and recommended that DOE reconsider its focus on NSTF catalyst systems.

MEAs, Cells, and Other Stack Components: Four projects were reviewed in this area, with one project receiving a high score of 3.1, two projects receiving a low score of 2.7, and the remaining project receiving a score of 2.9, for an average score of 2.9. Reviewers felt the highest-rated project's systematic approach and excellent collaboration resulted in an impressive amount of data and helped identify several challenges confronting de-alloyed Pt-based oxygen reduction reaction (ORR) catalysts. For the lowest-scoring projects, reviewers were unimpressed with the accomplishments of each project and with the reasoning behind the approach for each project. They commented that the first project continued to work with a catalyst system, NSTF, that may be reaching a limit of diminishing returns without a major reworking of the system; and that the second project had selected, in perovskites, a class of materials that is well known and had failed to develop the selected materials or to demonstrate a path to improvement.

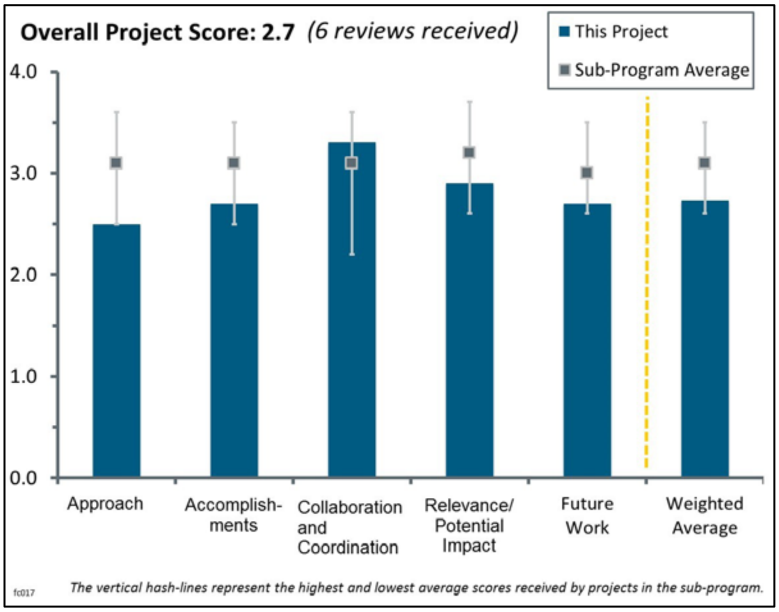
Membranes/Electrolytes: The six membrane projects reviewed received scores between 2.6 and 3.5, with an average score of 3.2. The highest- and lowest-rated projects in this topic area were the highest- and lowest-rated projects in the program. Reviewers were impressed with every aspect of the highest-rated project and found that the excellent team, with good coordination and a valid and novel approach, had led to quantifiable progress toward meeting DOE's membrane targets simultaneously with a single system. They recommended that the project proceed by concentrating on the viability of the perfluoro imide acid (PFIA) side chain. Reviewers were impressed with the potential of stable phosphonium-based anion exchange membranes and the approach the lowest-rated project team had taken, but they were concerned that the degradation tests used are not the most accurate. In addition, reviewers expressed concern about the results achieved, particularly with respect to conductivity and stability.

Project #FC-017: Fuel Cells Systems Analysis

Rajesh Ahluwalia; Argonne National Laboratory

Brief Summary of Project:

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



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Question 1: Approach to performing the work

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

- The approach is good. ANL develops models and applies them to issues of current interest, as advised by the U.S. DRIVE Partnership technical teams and by DOE. ANL also validates the models with laboratory data. The focus in fiscal year (FY) 2016 seems to be very 3M-centric, as outlined by the four quarterly progress measures and milestones.
- ANL uses a balanced and proper approach combining detailed models and data validation. ANL is encouraged to continue with the same modeling/validation approach with the new state-of-the-art materials as they become available through the Fuel Cell Consortium for Performance and Durability (FC-PAD).
- The overall approach was satisfactory. On slide 4, it was not clear which tasks DOE requested for analysis. The way the slide was presented, it could be assumed that DOE requested all of these tasks.
- ANL has reverted to basing stack performance on a 3M nanostructured thin film (NSTF) catalyst, in spite of the lack of any evidence that 3M NSTF has been found applicable for a realistic automotive fuel cell system. The choice of 3M NSTF as the catalyst may have led to a detour into the degradation mechanics of this catalyst that would not have been necessary had a dispersed catalyst system been chosen for the project.
- In general, the project is very dependent on the remainder of the DOE portfolio. While it is understandable that DOE would be fairly selective in choosing catalyst and balance-of-plant (BOP) projects that may provide an opportunity to find breakthroughs in technology, it is not understandable that the system analysis project would be confined to more experimental component sets, especially given the role that the system analysis project plays in establishing the existing status for fuel cell technology. It is not clear that the NSTF catalyst represents the status of the technology, nor that the Roots compressor represents the status of the technology. Regarding the NSTF catalyst, the answer is no, it does not represent the status of the technology. Regarding the Roots compressor, better air machines can probably be found.
- The modeling approach is semi-empirical and may be missing some elements of being predictive for fuel cell performance. Heavy dependence on data for tuning/calibrating the model might be limiting and may not be applicable for novel material sets. Further, the project has been primarily focused on NSTF and limiting the operating window to the optimum for NSTF (e.g., forklift application at a super wet ~80% relative humidity [RH] *in* and 140% RH *out*), thus ignoring most commonly used operating conditions.
- The approach is dominated by the 3M catalyst materials. The latest design is to put a layer of NSTF catalyst on top of a platinum-containing interlayer that could by itself be considered an electrode. Perhaps

this is because of the failure of the NSTF catalyst to adequately promote the removal of product water. It is not clear what the difference is between a de-alloyed catalyst (such as the 3M NSTF Pt_3Ni_7) and a catalyst that is not ordered (such as PtNi). The Fuel Cell Technologies Office (FCTO) should evaluate its commitment to the 3M NSTF catalyst and report what fuel cell organizations (transportation or otherwise) are using the NSTF catalyst after over 10 years of FCTO support.

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

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

- There was a good deal of technical progress in FY 2016. The models are especially useful for predicting performance and durability in stacks, as durability experiments are time-consuming and very costly. Accomplishments and progress were many and range widely, from membranes to catalysis; from air management to fuel management to thermal management to water management; and from stack performance and durability to system cost.
- ANL consistently follows through on its planned work and does a thorough job of capturing the system status. The correlation of NSTF degradation with fluoride emission rate (FER) is an interesting insight, but it is not clear that it has been corroborated. More such evaluations are encouraged, as it is nice to see ANL attempt to move the bar (progress), not just measure the height of the bar (status). However, it would be good to see more focus on dispersed catalysts.
- Accomplishments seem to be primarily focused on understanding the impact of FERs on NSTF irreversible degradation. Such correlations have been previously published, and it looks like a mere correlation rather than FERs being the cause. There has been good progress on the bipolar plate contact resistance coordination activity.
- Data for irreversible degradation of NSTF appear inconsistent. Voltage loss rates for 0.6 V, 90°C in the V Series do not match the rates for 0.6 V, 90°C in the T Series. There is no indication that the model accounts for hydrogen concentration on the anode. To explore why differential cell models do not match load-following cell results, the investigators may want to look at water balances and proton resistivities (perhaps with proton pumping for NSTF, if possible) for both types of cells. Perhaps the differential cell does not allow water generated to escape toward the anode because of the short flow path.
- The principal investigator claims that “Optimal power determined by high frequency resistance and oxygen reduction reaction activity rather than mass transport overpotentials.” However, there is at least a 10 mV difference at 2 A/cm^2 for the model including mass transport versus a model without. It is assumed that the model including mass transport was used in calculating stack and system costs, which would make it strange to think that a mass transport effect at least as large as the cell-to-stack voltage loss does not make a difference. Results from this past year differ from last year in one key respect: system cost now appears to go up at air inlet pressures greater than 2.5 atm. Because stack cost goes down at higher pressure, it appears that the system cost increase directly relates to the cost of the air machine.
    - It is hard to tell what the accomplishments were. During the presentation, it was very difficult to follow the data explanations because of the extensive use of acronyms and abbreviations. SRc was not defined, nor was SR(c)—perhaps they are the same—both were used. There was no statement identifying federal urban driving schedules (FUDS) and federal highway driving schedules (FHDS). It was not clear whether the cost correlations from Strategic Analysis, Inc. (SA) are at production rate of 500,000 units per year or less, or how valid SA’s projected cost correlations are. It is not certain that industry has agreed with these values down to the tenth of a dollar.
    - The Summary of Technical Accomplishments slide does not make clear what these terms refer to: “results #1,” “results #2,” “results #3,” and “results #4.” A search of the presentation for the word “results” revealed it only on the Summary slide or when referring to “modeling results.” It is not clear whether slides 6 through 11 consider only PtCoMn/NSTF on an 850 equivalent weight (EW) perfluorosulfonic acid (PFSA) membrane. The reviewer looked for labels on slides but remains unsure what catalyst was used because Pt_3Ni_7/NSTF was discussed before slide 6.
    - The term TC was identified after being used several times as thermal conditioning, not thermocouple, which was confusing. The thermal conditioning cycles were not defined.
    - It is not clear whether a supported membrane is the same as a reinforced membrane, and if so, why two different terms were used. It is not clear whether PtCoMn/NSTF was operated with a cathode

interlayer (CI) or was tested on a supported membrane (725 EW PFSA). It is not clear how direct comparison is made with Pt_3Ni_7/NSTF and 850 EW PFSA.
    - The hold potential for potentiostatic tests on slide 9 is unknown. Experimental conditions were not fully stated. It is not clear whether the CI replaces the gas diffusion layer or whether both are present, nor what exactly the CI does. It is not clear whether the improvement using d-Pt_3/Ni_7 + CI with 725 EW membrane is due to the 30% reduction in thickness of the membrane (725 compared to 850) or to the change in EW.
- On slide 18, it is not clear whether all of the Pt_3Ni_7 electrodes have a CI while the $Pt_{68}(CoMn)_{32}$ does not. It is not clear how the effect of the CI could be isolated.
- The presentation was much too technical. The information was dense, and many of the acronyms were not defined, which made it very difficult, if not impossible, for a layman to understand. The researchers need to distill out the essence of the results and then explain it in plain English so that a layman who is not an expert in the field can understand.

Question 3: Collaboration and coordination with other institutions

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

- The project has excellent collaboration, partners, and interactions with industry, university, working groups, and technical teams, domestically and internationally.
- There was a high degree of collaboration with multiple suppliers in this project. The work appears to be exclusively focused on transportation applications, although the overall objective includes stationary applications.
- There are many good interactions.
- ANL has had to collaborate with other DOE investigators (e.g., 3M and Eaton) to gather information for modeling stack and air machine performance. The project relies on these collaborations as well as those with SA for cost modeling. This project plays a key role in defining what DOE recognizes as the status for automotive and bus fuel cell technology.
- Ideally, ANL should be able to explore technology outside what DOE has funded. There is very little connection thus far between the system being analyzed in this project and the systems that are being deployed in vehicles. It would make sense to expand collaboration to understand systems that are on the road. It would be good to see more than one collaborator for certain components, similar to what is already being done with 3M and Johnson Matthey on membrane electrode assemblies (MEAs). It would be preferred to see more than one air machine collaborator, more than one anode subsystem collaborator, and multiple collaborators on other components.
- The project maintains a strong collaboration effort with outside partners, though Oak Ridge National Laboratory's diagnostic/characterization laboratory is not mentioned. Perhaps this project's modeling efforts can shed light on results coming from that laboratory (Karren More's project) or from National Institute of Standards and Technology data. Project integration with the FC-PAD projects and deliverables should be a key focus in the upcoming years.
- ANL is working with several collaborators, and the interaction is obviously generating useful information. It is not clear whether the interaction with SA is a collaboration or ANL is just receiving data from SA. It is not clear how SA contributes to a life-cycle cost—whether SA estimates a beginning-of-life cost or calculates the recovery cost at end of life.

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

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

- The project's annual status determination is a key output of this (and SA's) project and the Hydrogen and Fuel Cells Program (the Program) and, as such, rates an "excellent." The project meets its purpose as an independent assessment well. However, it is of lesser benefit to original equipment manufacturers/

developers. The Program should continue to seek out areas where it can do more of the latter (durability studies, mass transport limitations, etc.).
- This is the primary source of performance modeling for fuel cells. The project should publish the experimental test matrix and the corresponding NSTF results through the FC-PAD website to increase impact.
- Funding of $550,000 seems high for modeling work. The project impact is broad, ranging from quantifying the source of decrease in power density and increase in cost due to heat rejection constraint, identifying the dominant NSTF catalyst degradation mechanism and determining the upper limit of cumulative fluoride release (CFR) to meet the durability target, projecting increase in power density and decrease in cost by reducing the anode Pt loading and replacing cathode catalyst type, and using a thinner membrane with a lower EW.
- There are so many fuel cell designs that are evolving that it is difficult to keep abreast of all of them. If the areas of evaluation were specified by DOE, then the project is well focused on those specific tasks.
- Automakers that are seeking to commercialize fuel cell electric vehicles will not rely on this study to do so. This study is most relevant to DOE's own ability to assess the status of fuel cell technology and to be able to assign a research portfolio accordingly. The project might be more relevant to automakers if it were able to begin answering questions that SA still has regarding bipolar plate welding time and length, inspection rates, and other plate-related issues.
- It would be good to be able to say the relevance/potential was outstanding; however, a high confidence level in this project remains in question based on the information provided in the presentation.

Question 5: Proposed future work

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

- The field is so diverse that it is very difficult to determine what the follow-on priorities should be. It would be interesting to understand *why* the new tasks are being undertaken. The proposed future work on bus systems is important and welcome.
- Future work on catalysts should include PtCo. This is especially true given the usage of PtCo in the Toyota Mirai and the new General Motors project focusing on PtCo. Air machine studies need to be expanded beyond just Eaton. Future work needs a much greater emphasis on bipolar plates. If the NSTF work is to go forward, the future work should focus on the gaps between the differential cell model and the load-following cell data.
- Proposed future work seems too broad. The value of continuing to work on NSTF is uncertain, given the problems with scaling up this solution. Near-term focus (2016 toward the Program's 2017 Annual Merit Review) should be d-PtNi/C and also any other state-of-the-art MEAs through FC-PAD. Also, the project should ensure the entire dataset is available through the FC-PAD website.
- The project should place a higher priority/emphasis on dispersed (non-NSTF) catalysts. Project integration with the FC-PAD projects and deliverables should be a key focus in the upcoming years.
- The proposed future work should improve understanding and development of advanced fuel cell systems.

Project strengths:

- ANL has been responsive in the past to feedback to add parametric studies, to add BOP equipment most relevant at low current density, and to add thermal restrictions. ANL does use a performance map to look at voltage response over a wide range of operating conditions. ANL has improved its coordination with the SA cost model over the past year.
- Modeling work is relevant and impactful, and progress seems to cover a wide range of topics. Modeling can be a cost-effective way to predict stack and system performance and durability, if done correctly, and can accurately predict these phenomena.
- The technical resources and expertise that have been brought to bear on evaluating the various tasks are a significant strength of this project. The key is to select tasks that advance the overall progress of the industry.
- Strengths include deep modeling capabilities and integration with DOE projects (with good access to those data).

- There is a solid model and a good team.
- The presenter and the ANL team are all well established, experienced researchers. The quality of the presentation was very disappointing.

Project weaknesses:

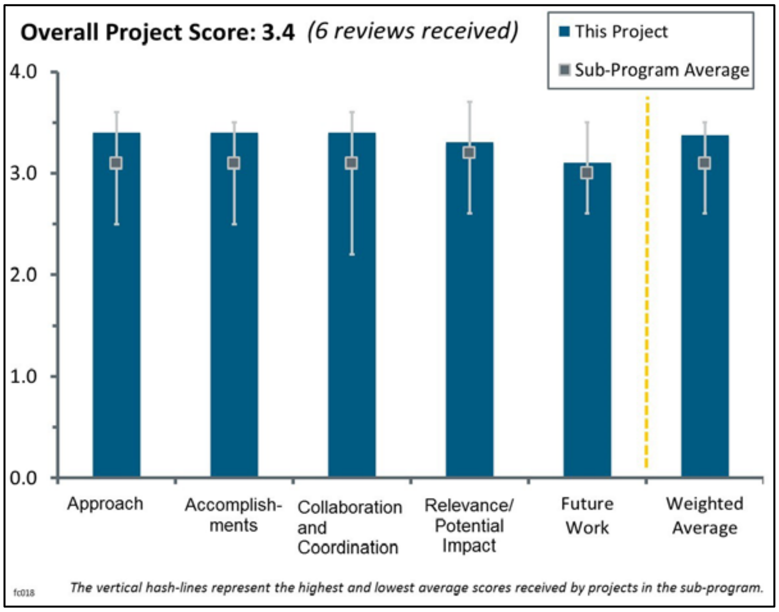
- In the past, access to state-of-the-art materials not associated with other DOE projects has been an issue (as it has been with the other DOE projects). It is hoped that this improves with the formation of FC-PAD and its emphasis on state-of-the-art materials access.
- The project lacks manufacturing details that would assist in understanding cost minimization for many BOP components and for plates. The project is basing its analysis on a catalyst system that is not known to be used in existing vehicles. ANL rarely seeks out more than one supplier for a given component. This means that the project misses numerous opportunities either to explore the actual status of the technology or to learn how cost can be minimized versus the existing status. ANL can sometimes miss on critical system assumptions. One example is the assumption of constant pressure drop from compressor outlet to expander inlet.
- The FCTO commitment to 3M’s NSTF should be questioned. The commitment was valid 10 years ago; however, more recent data generated by FCTO projects suggest a re-evaluation of the benefits of NSTF should be done. It is hard to think of any other catalyst system for PEM that has received as much funding over a long period with so little industry acceptance.
- The results that were presented were not presented in a form that was understandable to a layman. The report needs to extract the essence of the results and reduce them to simple English so that a person who is not an expert in the field can understand what is being reported. A single slide is needed that identifies all the acronyms used. The lack of acronym definitions was very frustrating.
- The choice of NSTF is a weakness.
- The project cost seems high.

Recommendations for additions/deletions to project scope:

- The scope needs to be expanded to include suppliers outside of what DOE is funding or has funded. It would be good to survey multiple suppliers per component. The scope needs to expand greatly on understanding bipolar plates. This includes how plates are made as well as how plates affect performance. The project needs to focus on what system status actually is, so long as status is being used for cost modeling. A dispersed catalyst layer needs to be the basis for the analysis, regardless of the cost consequences. Analysis of NSTF durability belongs in a 3M project, not in a system analysis project. If the particular failure modes associated with NSTF are not an issue for catalyst layers that are actually in practice, then having a system analysis project dive into this detail is counterproductive.
- The project should resolve whether the performance enhancement of d-Pt_3Ni_7 + CI over $Pt_{68}(CoMn)_{32}$ is the result of the CI. The project should resolve whether the improvement in power density of d-Pt_3Ni_7 + CI with the 725 EW membrane compared to the d-PT_3Ni_7 + CI with the 850 EW membrane is due to the difference in membrane thickness (~40%) or difference in EW.
- There should be greater emphasis on modeling and understanding a dispersed catalyst, with less priority on NSTF. Regarding the anode gas recirculation, the project should investigate the physics and impact of pulsed ejector flow.
- NSTF should be deleted from future work (2016 and 2017). The project should figure out a way to make the test matrix and datasets available online.
- It is not clear how tasks are defined. Perhaps they are a result of a formal meeting between DOE and the laboratory to develop the next set of tasks based on the needs of industry. An annual review should take place to clearly define priorities.

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

Brian James; Strategic Analysis, Inc.



Brief Summary of Project:

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

Question 1: Approach to performing the work

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

- The project takes a good approach to providing a cost analysis using the Design for Manufacture and Assembly (DFMA) method. The benchmarking against the Toyota Mirai was very useful. The feedback from the stakeholders is very valuable and helps with the validity of this approach.
- The project has a well-documented and reasonably wide scope of possible alternate methods for lower cost fabrication, particularly with respect to catalyst and bipolar plates (for the former, the project shifted to a lower cost approach and for the latter, the team recognized that prior estimates may have been low). The project team displayed a reasonable recognition of practical limitations in some instances.
- The project takes a solid approach in cost modeling and is very good at reaching out to collaborators and seeking the most relevant information. Some cost models seem way too optimistic (e.g., for the gas diffusion layer [GDL], but new results to understand sensitivity are encouraging to see). The addition of the anode recirculation pump has added cost, which may not be the most optimal use of money.
- The project continues to adjust to changes in the market and needs as in the case of the benchmarking against the new production vehicles.
- Strategic Analysis, Inc. (SA) uses established costing and scale-up methods to provide annual updates. SA is intimately in contact with stakeholders—original equipment manufacturers (OEMs), raw materials and component suppliers, equipment suppliers, laboratories, end users, systems analyzers, stack and systems integrators—to ensure the validity and timeliness of materials, processing, and operating conditions assumptions. SA uses DFMA techniques, vendor quotes, supplier estimates, and user experience to estimate costs bottom-up including raw materials and manufacturing equipment (both type and quantity).
- SA's DFMA approach to the cost status evaluation and their interaction with the fuel cell stakeholders is proper and adequate in addressing the project goals. It is unclear how the low-volume cost efforts were approached. Specifically, it is unclear if the processes were appropriately modified or optimized for each volume. The final report should list detailed assumptions for processes and their respective costs for each major operation and component.

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.

- This project has made good progress with collaborators in seeking to understand the impact of dealloyed catalysts on total system cost. The cost increases from reconsideration of various stack components show how frequent evaluations of each component is important. The benchmarking against the Mirai was very interesting although the cost analysis appears to be more subjective than objective due to a lack of publicly available information.
- The project has made good progress in modeling the cost of competitive vehicles (e.g., Mirai); however, near-term solutions for commercialization cannot be used to change the long-term goals (e.g., ARP, cell voltage monitor).
- The project was well-organized and presented a summary of alternatives. Slide 13 is particularly useful in explaining the basis for change. Note that rate of change of costs is lower than needed to achieve DOE goals.
- An updated cost analysis has been provided for the car and bus fuel cell systems. The benchmarking exercise against the Mirai showed very good results in terms of the accuracy of their projection. The risk identification exercise in terms of industry manufacturing readiness level (MRL) and fuel cell MRL is also very valuable for DOE.
- The team has been responsive to DOE’s requested deliverables. The timely incorporation of Argonne National Laboratory’s (ANL’s) power density model results, which are key to the annual cost status projection, is appreciated. The sensor cost update (listed in 2015 future work) does not seem to have been covered. This may be an area where cost estimates are too high.
- SA is responsive to DOE and industry needs as the technology evolves. Alternative technology scenarios are run in a timely manner. Although SA does not develop or demonstrate technology, they identify where costs need to be addressed thus helping to guide technology development priorities.

Question 3: Collaboration and coordination with other institutions

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

- The partners have done an excellent job in engaging the industry (over 30 companies) and receiving manufacturing inputs, which is no easy task. The engagement of Toyota was a very positive feature in this project.
- SA is in constant contact with stakeholders (especially systems analysis stakeholders) to ensure that the latest materials and process technologies are incorporated into the analysis. SA performs many “side jobs” and trade-off studies to evaluate specific component costs and the impact of novel technologies.
- The project has achieved strong, ongoing engagement from several industrial collaborators.
- The accuracy of the cost model depends heavily on the performance model from ANL (FC-017) and hence, there needs to be a close collaboration between these two teams. There is a need for collaboration with non-U.S. suppliers (e.g., for GDL, plate coatings) to understand best-in-class processes for cost reduction.
- SA appropriately works with ANL’s systems analysis project and collaborates with many of the stakeholders to attempt to evaluate a representative status of the technology.
- Contact Edison Welding Institute in Columbus, Ohio, for additional input on the bipolar plate welding process. Contact Oak Ridge National Laboratory for development work on carbon fibers that may be applicable to GDL. GDL remains a significant cost item for all sizes of polymer electrolyte membrane fuel cells—any progress in cost reduction 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 **3.3** for its relevance/potential impact.

- The project is very relevant to DOE Hydrogen and Fuel Cells Program (the Program) and addresses the cost related barriers very well. The project can be very helpful to decide how to prioritize research areas and to highlight the areas in need of attention.
- The relevance and impact of the project are high. SA's costing work helps guide the Program by identifying high-cost areas and evaluating technology options.
- This project is the single source of information for projected cost for fuel cells. Therefore, the project is highly relevant.
- This well-documented study provides the basis for understanding cost drivers at high production rates. The path to get from current production rates to high production rates is a bit unclear and the confidence level that high production rate costs can be achieved is not clearly defined.
- By its nature and charter, the project does not advance the progress towards the goal, but the project is intended to give a cost status. In that effort, the project does a good job. Adding a section highlighting areas, components, and processes that can be improved (in a pre-competitive environment) to enhance cost-affordability would be helpful. To that end, the presentations should incorporate a listing of major component and operation (e.g., stack assembly) costs (in dollars, not just dollars per kilowatt) in table form so that the project end-users and reviewers can more clearly see what items are major cost-contributors, and how they change from year to year.
- More targeted direction or feedback, such as the suggestion for a roll-to-roll bipolar plate manufacturing process, will further increase the impact of this work.

Question 5: Proposed future work

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

- The proposed future work is good. It might be good to add details about which balance-of-plant (BOP) costs are driving the overall system cost, and how what type of work at the stack level can bring the BOP costs to less than $\$15/kW_{net}$.
- The future work is dependent on changes in the technology status. SA quickly responds to alternative scenarios. Trade-off analyses of alternatives will continue (e.g., ejector versus hydrogen recirculation blower). An annual update will be performed.
- The future work is not ambitious but it is in line with project priorities. The final report should detail the following: detailed costs versus components and operations; main system parameters (as shown on slide 37) versus year; and for the different volumes, key process assumptions.
- The proposed future work is very relevant and will be useful for the project's results.
- The proposed future work appears to be appropriate.
- The project ends in three months; the future work is primarily documentation.

Project strengths:

- The project did very well to reduce the wide range in estimates of GDL and bipolar plate costs, identify forces driving cost up, and make suggestions for directions of future efforts for further cost savings.
- The team has a long history with the project and has demonstrated the capability to perform the task. The team also works well with ANL (Systems Analysis) and OEMs to provide the input assumptions, which are vital to a fair estimation of the cost status.
- The solid analysis using the DFMA framework is a project strength. The project has very good collaborations with industry.
- The project has excellent communication with and responsiveness to stakeholders, especially DOE.
- The project has well documented and refined cost estimates as well as good analysis.

Project weaknesses:

- There are no notable weaknesses. The project benefits from a solid history allowing improvements each year as new technology and fabrication methods develop or are evaluated.
- There are no strong weaknesses.
- The project is limited to what information is public and what information the developers are willing to contribute (confidential or otherwise) and thus, one must accept that there are significant uncertainties in any such cost status projection. But that is to be expected.
- Overcoming the inherent subjective nature of some of the analyses continues to be a challenge of the project.
- The project relies too much on nanostructured thin-film catalysts.

Recommendations for additions/deletions to project scope:

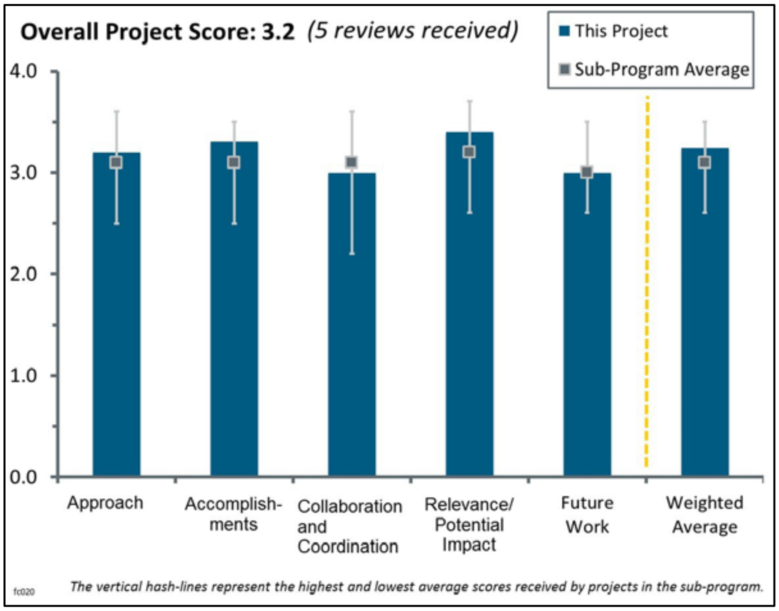
- Continue to monitor developments including the Mirai and other fuel cell electric vehicles.
- Consider the cost of state-of-the-art membrane electrode assemblies (MEAs) at low volume production (around 1,000 systems per year). The components in the imaginary system are all using low cost materials without consideration for any interactions due to integrating these components. It would be beneficial to model the cost of state-of-the-art MEAs along with some cell configurations as tested.
- If continued evaluations of cost at low volume are planned, it would be of benefit to show where processes were volume-optimized including the impacts of capital, labor, quality control, etc.
- DOE should note that it might be informative to compare all of the costing projects and identify major differences.

Project #FC-020: New Fuel Cell Materials: Characterization and Method Development

Karren More; Oak Ridge National Laboratory

Brief Summary of Project:

The objectives of this project are to (1) identify and develop novel high-resolution imaging and compositional/chemical analysis techniques and unique specimen preparation methodologies for the micro- to Angstrom-scale characterization of materials comprising fuel cell membrane electrode assemblies (MEAs);
(2) optimize imaging/spectroscopy methodologies toward specific fuel cell materials, including electrocatalyst atomic-scale structure and chemistry, ionomer mapping in catalyst layers, and three-dimensional (3-D) electron tomography; and (3) share unique capabilities and expertise with fuel cell researchers outside of Oak Ridge National Laboratory (ORNL).



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Question 1: Approach to performing the work

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

- The use of advanced electron microscopy and four-dimensional tomography to elucidate the atomic-scale structure of electrocatalysts and for mapping the ionomer contents/coatings on electrocatalysts is a great approach, which the team has taken to help increase understanding of the FC-PAD (Fuel Cell Consortium for Performance and Durability) MEAs sourced from different suppliers. The team's approach of collaborating with the fuel cell community is nice and will help the community to use newly developed advanced analytical techniques and implement these techniques across the industry, hence achieving an actual comparison between the samples.
- This project continues to develop useful advanced characterization techniques and approaches for identifying critical fuel cell MEA materials issues. The principal investigator (PI) identifies a general focus on standards before and after use that is effective for suggesting materials changes that can be connected with changes in performance.
- The extent to which the characterization methods and their advancement are pushing the state of the art (SOA) for fuel cell characterization is difficult to assess. That said, the PI has clearly been in this community for some time and demonstrates an overall knowledge of the current relevant issues for fuel cell development. Evidence for this comes, in part, from continued high ratings at Hydrogen and Fuel Cells Program Annual Merit Reviews (AMRs), as was the case last year. This represents high value for DOE investments in national laboratory capabilities and (importantly) expertise.
- The approach is excellent and contributes to overcoming some key barriers related to having a better fundamental understanding of the MEAs and MEA components.
- The approach to develop new techniques using advanced microscopy is critical for better understanding key MEA components and their role in performance and durability.
- The use of scanning transmission electron microscopy (STEM), scanning transmission x-ray microscopy (STXM), and spectral analysis to distinguish elements in the resulting images has become more widely known over the past few years. Other researchers have been able to make use of the Advanced Light Source at Lawrence Berkeley National Laboratory or the Canadian Light Source to accomplish similar

results in two dimensions. The PI in this project has been able to stay a step ahead by performing analysis in three dimensions. The approach would be enhanced if it were clear that the ultimate goal is to provide similar analysis with a wet sample. Perhaps even better would be to provide an in situ technique, allowing imaging on materials in an extremely small-scale fuel cell.

  - The project occasionally diverges into making presumptions about fuel cell catalyst layers, although the sample being discussed may be just one result from a wide range of possible parameters, including ionomer-to-carbon ratio, ionomer equivalent weight, deposition technique, and ink solvent and composition ratios. With so much work in the system and cost analyses premised on 3M nanostructured thin film (NSTF), it is refreshing (although inconsistent) to see one DOE project that focuses on dispersed catalyst layers.

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.

- Outstanding progress has been made on 3-D imaging of ionomer dispersion in catalyst layers and electrocatalyst distribution in 3-D. These techniques can be very helpful to correlate the MEA performance to catalyst layer (CL) design and optimization.
- As usual, the project has produced new and interesting results that continue to provide valuable insights.
- Because the project started in 2016, not much in the way of technical accomplishments is expected during this short period before the AMR. However, the team has taken a great approach of utilizing its collaborators and past contacts to obtain new and baseline MEA samples for analysis. The team should try to engage some of the mainstream MEA suppliers and automotive companies to obtain their SOA MEAs for FC-PAD analysis. This will help the team to correlate the structure of the commercial MEA to other MEAs.
- Research accomplishments and progress, as assessed by the presentation highlights and publication numbers, are good. Publication numbers of five to six are good (but not outstanding), and the PI is regularly presenting work at national and international meetings (including one prestigious invited presentation). This looks similar to last year’s “quantitative” productivity.
  - The results described during the presentation represent substantial new information, and the approaches to characterizing ionomer distributions in catalyst layers seem to be a real advance. On the other hand, the catalyst metal 3-D imaging is applying SOA methods to synthesized fuel cell catalyst materials.
  - It is difficult to assess how the “split” in this project will affect progress in the future. Developing advancements in the methods should be inextricably linked to addressing critical issues in fuel cell catalyst development. It appears this project has effectively done this in the past. Connecting part of the activity directly to the FC-PAD activity is outstanding; it is not clear why the whole activity is not part of FC-PAD. That is, it has been a goal of this project to use the advancing methods on realistic materials via collaborations. Those have been occurring, and, understandably, many of these are with other national laboratory activities where materials are more readily obtained and where the materials issues are clearly identified via various performance changes and/or synthesis processing procedure changes. These types of important collaborations for an activity aimed at advancing SOA characterization methods would be optimum for the community, so, again, FC-PAD seems a most appropriate “umbrella” for the project to ensure that these collaborations become even more productive. Furthermore, FC-PAD, as a consortium, may help with some of the difficulties in getting SOA materials, rather than just having this ORNL characterization project ask for them from industry partners. Therefore, reconnecting this activity with the “split-out” FC-PAD one is recommended. Also, in this way, it would avoid potential confusion in trying to assess which part of the methods development and application work should be attributed to which part of the activity.
- What has been accomplished has been the development of 3-D imaging and spectral analysis for catalyst layers under ex situ, dry conditions in order to understand where ionomer and platinum agglomerates reside. This has been good and can be helpful toward assisting developers in understanding how catalyst layers should appear to extend performance and durability. While the images are impressive, a proper evaluation of the project must keep the barriers to fuel cell commercialization in perspective. Over the past

few years, it has been apparent to nearly all developers that more graphitized carbons can compromise performance as well as resistance to Pt agglomeration. Furthermore, it has also been apparent that high-surface-area carbons allow for Pt deposition within agglomerates, as opposed to Vulcan and graphitized carbons. Repeating these findings does not advance the technology. The project must focus on learning something new about catalyst layers that has not already been uncovered. The results of the ionomer study are very interesting but must be advanced to understand what happens with the ionomer under wet conditions, or under some conditions that might be described as in situ.

Question 3: Collaboration and coordination with other institutions

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

- Obviously the project is well coordinated with the FC-PAD team, and it also includes a decent list of external materials suppliers. DOE’s 50% cost share offer should help to increase the level of external collaborations.
- Collaborations have been good ones and are poised to be even more productive via the FC-PAD umbrella. That said, it will be interesting to see if the goal of attracting many (perhaps 10) new industry collaborations can be achieved. In many ways, the collaborations via FC-PAD might well be more productive ones. Certainly, critical new information about materials properties should be of interest to industry, but research activities in the consortium may make the most use of these advances, rather than industry directly.
- The collaborators in the project are very broad, encompassing a university, national laboratories, and industry. The cooperative research and development agreement (CRADA) is expected to result in more collaborators and sample suppliers. It seems that more time is needed to observe the benefit of the CRADA and others who may join the team for evaluating their MEA samples.
- Collaboration with partners is not evident from progress shown. Many strong partners are mentioned, but details of collaboration are missing, so it is hard to judge the extent of collaboration. More industry partners and strong collaboration with FC-PAD Thrust Area 2 for CL integration will be helpful to get the meaningful information about various CL designs using these new 3-D techniques.
- The project has historically relied on collaboration. Indeed, the project is limited by the samples provided to it by outside collaborators. However, the list of collaborations this year appears much smaller. The list of partnerships established in the past year is confined to three national laboratories (which are now a given, especially with the emergence of FC-PAD), the nearby University of Tennessee, and two small companies (Ion Power and IRD Fuel Cells). The question then remains as to whether the project is actually engaging developers that are actively trying to move the technology forward with a linkage to vehicle or product application. It would be interesting to know the identity of the partners involved in the CRADAs. The 50% cost share rule may have discouraged some partnerships. It will be interesting to see whether FC-PAD restores ORNL’s access to a wider range of developer materials.

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 microscopy techniques developed and demonstrated by ORNL are very relevant to understanding MEA performance and durability. The microscopy procedures developed by ORNL are aligned with DOE’s goal to address the barriers of the commercialization of fuel cell technology. The focus of the activities is on better understanding the interaction between different components in the MEA (e.g., catalyst, membrane, catalyst support, and gas diffusion layer) that affects the performance and durability of the MEA. Understanding this interaction is one of the critical challenges in successful commercialization of fuel cells.
- The understanding that can be gained regarding the ionomer dispersion and electrocatalyst distribution can guide the catalyst layer development and optimization using the SOA catalysts.
- Clearly, this work is focused on issues of primary concern to the development of fuel cell MEAs.

- The project certainly is relevant; the only criticism is the PI seems to have opinions about what the community thinks the catalyst layer looks like, when these simple representations (e.g., sphere-shaped pores) are just convenient geometries to utilize for modeling efforts.
- The relevance of atomic-scale characterization to the future commercialization of fuel cell electric vehicles is without question. At present, much of what happens to create catalyst layers is a black art—ionomer, platinum, and pore sizes find themselves distributed in an almost random fashion, given the indirect methods of control. Therefore, advanced materials characterization is necessary to provide feedback as to how both materials and processes have affected the resulting catalyst layer. The big question with regard to relevance is whether it is still necessary for DOE to fund a distinct characterization project in light of (1) the emergence of FC-PAD and (2) the wider availability of techniques through other U.S. national laboratories or through laboratories in Canada, Japan, Korea, or Europe. Unless the project demonstrates an ability to stay a few steps ahead of the work that is being done elsewhere, it is difficult to say that a distinct characterization project is relevant to eliminating the barriers to fuel cell commercialization.

Question 5: Proposed future work

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

- The proposed research by the team is logical and fits with the nature of the project. As proposed, the team should work with academia and industry to optimize the catalyst-support-ionomer structure and establish correlations between them. The team should also try to determine how this correlation impacts the performance and durability of the MEA when the catalyst is changed while keeping the support (e.g., carbon) and ionomer (e.g., Nafion®) constant in the construction of the MEA.
- Future work plans are most appropriate, although it is unclear whether the goal to establish many new collaborations is practically achievable, or even advisable. A strong connection of this activity within the scope of a productive FC-PAD umbrella, in which this project’s goals are highly connected to other research goals of the consortium, seems like an optimum approach.
- Future work highlights the challenge of getting SOA materials from the industry, which is a very valid concern. However, many more studies can still be done on catalyst ink characterization and correlating that to catalyst layers. Catalyst ink is still considered as black art, so these new techniques can surely help ink optimization.
- The future work slide lacks any discussion of how materials characterization techniques will be improved to obtain even more sophisticated quantification of catalyst layers than what already exists. While the National Institute of Standards and Technology neutron-imaging project focuses heavily on what needs to be done to obtain better spatial and temporal resolution, this project is not as focused on what could be done to make its own characterization techniques better in the future. The optimization of fuel cell catalyst layers should be understood as something that catalyst-coated membrane suppliers or even automakers are attempting to accomplish. This project provides the feedback loop between performance and durability results and the processing improvements needed to make improved catalyst layers. However, this project should not assume the optimization responsibility itself.

Project strengths:

- The team’s instrumental capability, established scientific network, and vast experience in MEA analysis are certainly assets. With such a combination, the team is well positioned in the forefront of the technology and capable of conducting world-class research.
- The project has been able to facilitate 3-D imaging of a dry catalyst layer ex situ. The project has access to considerable resources for characterization within the national laboratory system. The project has been responsive to past requests to image different types of carbon, as well as to image ionomers in the catalyst layer.
- ORNL and the PI’s capability are the main strengths of the project.

Project weaknesses:

- Lack of SOA materials is a weakness, but that is highlighted by the PI, and one hopes the team will get good support from industry stakeholders.
- The team needs to find a suitable way to obtain MEA samples from automotive and commercial MEA companies for comparison. It is understandable that most of these companies are hesitant to share their SOA MEAs for outside evaluation and publication of those results. With all its resources and knowledge base, the team should encourage these companies to be a part of this endeavor and determine a pathway to share the analytical results with the fuel cell community while respecting the intellectual property sensitivity of the companies.
- A potential weakness may be the split of this project into an ORNL-only activity and an activity carried out as part of FC-PAD. This split might create an artificial and inefficient “barrier” between the actions carried out by these two activities. It will probably also make it very difficult to assess progress in future AMR reviews (in fact, it already has; it is not clear whether this project represented all of the activities carried out this past year or how the proposed future work might be different and distinct from the PI’s FC-PAD activities).
- The project needs to direct its focus toward improvement of characterization techniques, not just application, to stay ahead of other facilities worldwide. Being able to go further toward an understanding of how catalyst layers behave under wet conditions, or even in environments that would represent in situ conditions, may be beyond this project’s capabilities. The collaborations appear to be limited to the national laboratory network, a nearby university, and a few small companies. The project needs to re-expand the collaboration network to include entities directly involved with commercializing fuel cell technology.

Recommendations for additions/deletions to project scope:

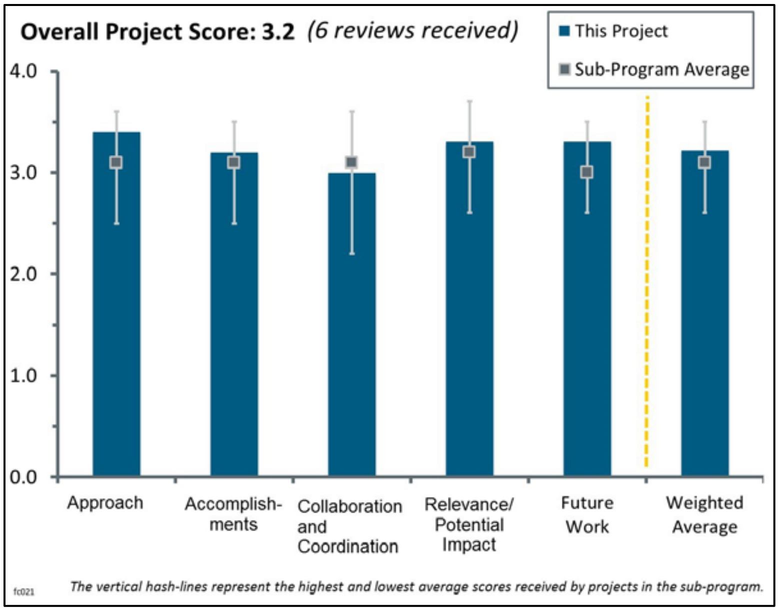
- Overall, the project looks good. No further additions/deletions are needed.
- Some focus on ink-to-catalyst-layer correlation would be great.
- To ensure productive and efficient progress in the future, the Program and ORNL might want to reconsider the decision to create two separate activities out of this project.
- The project scope should be directed toward imaging catalyst layers under wet conditions or in situ conditions. There may be limitations with existing equipment; nevertheless, the goal should be to develop the world’s best materials characterization for fuel cell materials. As STEM, STXM, high-angle annular dark-field detection, etc., become more commonplace, this project must stay ahead of other efforts. As carbons have become well understood, the emphasis on different carbon types and how they affect performance and durability can be lessened. In general, this part of the project should be more of a concern to developers. The emphasis here should be on developing new and improved microscopy techniques.

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 **3.4** for its approach.

- The National Institute of Standards and Technology (NIST) is continually innovating to meet the water-imaging needs of the fuel cell community by pushing to improve spatial resolution (ultimately to 1 µm) to allow studies of electrode layers, reducing imaging time for faster, more dynamic studies, and incorporating new capabilities (such as complementary x-ray tomography). This approach is sound and has excellent near-term and long-term vision.
- NIST maintains a national user facility for neutron imaging of fuel cells. It consults with the fuel cell community to plan facility improvements that would be useful in future studies. The facility 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 progress achieved to increase the geometric resolution was clearly highlighted. It is recommended that NIST equally highlight progress in relation to the time-scale resolution and compare progress with time scales associated with water processes such as a water drop traveling through the cell, membrane wetting and dehydration, and water accumulation in and removal from the gas diffusion electrode.
- The approach of using neutron imaging to study the water transport in single cells and stack has proven to be very good and very insightful. Every year, the NIST team works hard to add more capabilities and to increase the spatial resolution.
- NIST is attempting a wide variety of ways to advance both spatial and temporal resolution for neutron imaging. Furthermore, the team is attempting to satisfy customers both in the short term (grating, centroiding) and in the long term (cold neutrons). The approach focuses mostly on improving the analytical technique itself, as it should. NIST has wisely not confused the approach of this project with the approaches of fuel cell projects that seek to develop new material or optimize material design.
- The one criticism of the approach is that the efforts to improve resolution appear to be taking a while. Perhaps more resources could be spared to help. Until then, much of the fuel cell community appears to have lost interest.
- The overall approach is sound, although progress seems to have stalled compared to previous years.

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

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

- In September 2015, NIST commissioned a new cold imaging facility with higher resolution than previously available and with potential to resolve ice and water. Last year, NIST commissioned a complementary x-ray imaging system and made it available to all users. NIST is also making the beamline available for hydrogen storage experiments. NIST continues to develop methods to improve spatial resolution. The ongoing improvements include an image intensifier, centroiding with detector microscope resolution <9 µm, 4 µm grating resolution, and a neutron microscope with 1–20 µm spatial resolution with 10-second to 10-minute time resolution.
- Most of the accomplishments and progress this year seem focused on technique and hardware development. The new cold neutron instrument, complementary x-ray tomography system, and slit and centroid imaging are all examples of ongoing development aimed at meeting the requirements of the fuel cell community. The application of these new capabilities to fuel cell systems has been limited, but slow and steady progress might be expected when pushing the boundaries of spatial and temporal resolution.
- Progress made toward the spatial resolution is excellent, and it looks like resolution is on its way to 1 µm by 2018. New cold neutron imaging is commissioned and ready, which will help researchers understand and, one hopes, resolve cold startup issues.
- Installation of the cold neutron imaging facility has been a very positive development and represents a possible future of high spatial and temporal resolution. The possibility of separating ice and liquid water provides hope. Although the slit imaging can provide resolution down to 4 µm, the 17-hour collection time limits what can be done within the course of assigned beam time. Many researchers will probably not be interested in 17-hour collection periods. The centroiding imaging is much better for collection time (four times) versus the slit imaging, but the gain in resolution is small versus the incumbent techniques. It is difficult to say whether advancing from 9 to 5 µm resolution will increase interest. Combined neutron and x-ray imaging is a good idea, although it is confined to cells of just 0.6 cm^2.
- Three milestones were completed, and work is ongoing for another milestone.
- Progress toward lower spatial resolution is good, but one must be cognizant of time resolution as well. The overall facility upgrades are quite interesting, but their use in experiments for understanding transport is not as compelling.

Question 3: Collaboration and coordination with other institutions

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

- Collaboration is excellent across the board, including many industries and academia. Close collaboration with General Motors is great to study fuel cells in operation.
- NIST listed a number of partners, users, and collaborators from academia, national laboratories, and industry. The presentation also highlighted results from a user program with University of California, Merced.
- Twenty percent of the beam time is allocated to fuel cell and hydrogen storage experiments suggested or requested by the community. Exemplary data for non-precious-group-metal catalysts were provided.
- The project is very collaborative and dependent in terms of getting materials. It would be good to see more coordination with new consortia and with state-of-the-art materials and designs.
- Collaborations have been focused on method and hardware development, with a very limited number of user collaborations reported, the one example being liquid water saturation studies in diffusion media with University of California, Merced. The reported allocated beam time was down from 43% last year to 20% this year. It is important that the team better balance its efforts between user work and instrument development in the upcoming year.
- The way in which collaboration with partners has been expressed in the slides is somewhat casual; nearly all partners throughout the course of the project are listed early in the presentation, but it is difficult to see which collaborations have been ongoing in the past year. It would be useful to understand which collaborations pertain to the true work of this project, which is the advancement of the neutron imaging

technique to improved spatial and temporal resolution. In this regard, collaborations are more useful to note than the collaborations with fuel cell customers (e.g., General Motors). It would be interesting to understand the depth at which collaborations exist with Commissariat àl'énergie atomique (CEA), the National Aeronautics and Space Administration (NASA), and others that have expertise in detectors and beam line technology.

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.

- NIST is actively engaged in developing state-of-the-art neutron imaging capability for fuel cell hardware and making it available to the community of fuel cell researchers and developers. The capability has been successfully applied to studying the dynamics of water transport in flow fields and manifolds. Further improvements in spatial resolution are needed to provide more details of the water content in catalyst layers.
- The project is very relevant in terms of understanding where the water is in the cell. The overall impact depends on others and the samples and experiments provided. It is not clear how much time is proprietary versus nonproprietary.
- With a series of new capabilities and techniques under development, the NIST project is well positioned to have a substantial impact on the Hydrogen and Fuel Cells Program in the upcoming years.
- The project addresses several barriers: durability, performance, and water transport within a stack.
- This project has hit an interesting juncture, which has a direct impact on its relevance. Much of the knowledge that could be gained from imaging fuel cells at resolutions down to 10 µm has already been gained, as evidenced by the decrease in fuel cell customers using neutron imaging. The future relevance of the project actually depends upon the project's ability to deliver higher resolution so that water can be imaged within catalyst layers, membranes, and other thin components. As x-ray techniques develop that are able to image water at higher resolution and at more widespread locations than neutron imaging, neutron imaging will have to provide unique advantages such as operation on a relatively large cell. Another inherent advantage is the lack of neutron cross-sectioning with iron and other materials of construction. Trying to merge both neutrons and x-rays together as complementary techniques is a good idea.

Question 5: Proposed future work

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

- The proposed future work is very similar to last year's, with aims to further drive resolution to 1 µm and improve detection limits and imaging time. Progress has been slow but steady, as might be expected for these challenging endeavors. There continues to be strong multiyear vision. In the upcoming year, demonstrating the application of these new systems, such as cold neutrons and complementary X-ray tomography, to fuel cell studies should be an area of strong focus.
- The future work in terms of both new resolution and techniques and new capabilities is compelling, although there seem to be multiple pathways. It is not clear how much is feasible or supported by the Fuel Cell Technologies Office rather than other programs. Segmented cell and similar capabilities would be good.
- Future work describes the efforts to increase the spatial resolution, continue refining current methods, continue with the neutron microscope to improve the spatial and temporal resolution, and combine X-rays with neutron imaging. It would be nice to see how the new cold neutron imaging will be used in collaboration with industry to understand and separate water and ice formation and management in fuel cells.
- NIST outlined the ongoing three-year project on a neutron microscope to improve the spatial resolution to 20 µm in 2017 and to 1 µm in 2018. It would be useful to understand how NIST decides the direction of the future work, what specific recommendations have been received from the users, and the directions from DOE and from NIST.

- The possibility of combining centroid imaging and neutron microscopy to further increase geometric resolution should be considered.
- Establishing 5 µm centroiding at 4-hour (or 1-hour) temporal resolution may be valuable to some. The future work involving the neutron microscope is a large part of where this project now is. Many developers are awaiting the 2017 milestone of 10-second/20 µm resolution, and especially the 2018 milestone of 20-minute/1 um resolution. NIST may need to begin contemplating how to handle the pent-up demand for testing when these capabilities become ready. It would be good to hear whether collaborations with other laboratories or with NASA are contributing to the development of the neutron microscope. It is not clear whether NASA actively participates. Perhaps there is some way that the development could be accelerated.

Project strengths:

- NIST has been very responsive to user needs and understands that its scope is the development of a technique, not the optimization of fuel cell materials. The project leverages a fairly large facility for neutron beams at NIST. NIST has personnel that proactively seek out ways to make neutron imaging better with advanced detectors and other equipment. Neutron imaging can be done on large cells with little cross-section with cell structural materials.
- Neutron imaging capabilities are impressive at NIST. Strengths include the team’s efforts to keep improving, refining current methods, and also developing new methods to add more tools for water management understanding in fuel cells.
- The project has a balanced approach combining method development to improve geometric and time-scale resolutions, and there are multiple ways for users to access equipment, solve issues, and study water transport phenomena.
- This represents the best technique for imaging fuel cells in a nondestructive fashion, especially the water. Progress and plans toward better resolution are also strengths.
- There is a very good multiyear vision and good near-term progress in development of new techniques and instrumentation.

Project weaknesses:

- The experiments that researchers wanted at the existing resolution have slowed down before higher resolution can be developed, which has diminished the interest in neutron imaging for now. Neutron facilities are difficult to access. Making use of the neutron scattering technique will always require travel to a beam line. There will still be a wait until 1 µm resolution can be obtained. In the meantime, fuel cell technology has advanced to a place where 1 µm resolution is necessary to extract information about catalyst layer performance and durability.
- User work seems to be down this year. While the focus on future work is important, the current capabilities of the facility seem to be underutilized.
- The operating principle of the opaque gratings (Gadolinium oxysulfide) for slit imaging should be given.
- The project is dependent on others for experiments and guidance.

Recommendations for additions/deletions to project scope:

- The progress achieved to increase the geometric resolution was clearly highlighted. It is recommended that the project equally highlight progress in relation to the time-scale resolution and compare progress with time scales associated with water processes such as a water drop traveling through the cell, membrane wetting and dehydration, and water accumulation in and removal from the gas diffusion electrode. The possibility of combining centroid imaging and neutron microscopy to further increase geometric resolution should be considered.
- It may be useful to add resources to accelerate the pace at which 1 µm resolution is being developed. The continuation of work on x-ray–neutron combined experiments should be predicated on interest. If users do not show interest, the work stream should be removed. Similar principles should be applied to slit and centroid imaging.
- NIST should reach out for guidance about what the critical techniques and information required are.

Project #FC-052: Technical Assistance to Developers

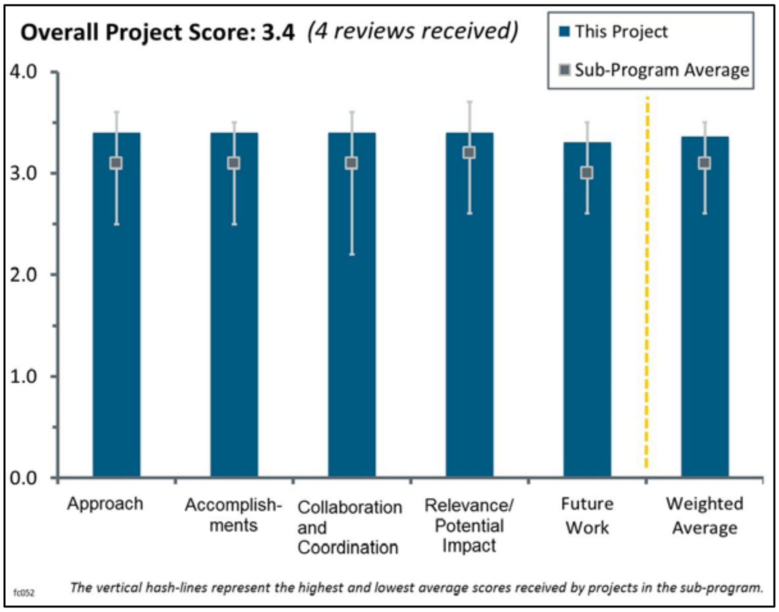
Tommy Rockward; Los Alamos National Laboratory

Brief Summary of Project:

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

Question 1: Approach to performing the work

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



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

- LANL applies its broad and deep fuel cell knowledge and facilities to a wide array of specific fuel cell materials, operating, and processing issues as requested by stakeholders and approved by the U.S. Department of Energy (DOE). Examples include bipolar plates, platinum-group-metal (PGM)-free catalysts, membrane electrode assemblies (MEAs), and stack testing. LANL applies accepted industry practices and procedures and innovative approaches to obtain meaningful results for stakeholders.
- Use of LANL’s infrastructure and expertise for evaluation and diagnostics of commercial stacks/MEAs/catalysts is uniquely appropriate for this project.
- This is an excellent use of national laboratory resources to help solve specific problems submitted by industry and other laboratories.

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.

- LANL provided technical support in the areas of bipolar plate coatings and their application, PGM-free catalyst assessment, powder properties, stack testing, cell architecture, and catalyst testing for a wide range of entities. This support enhanced progress toward DOE goals. LANL provided not only data but also technical insights based on years of fuel cell experience.
- Each task was well planned and well executed. The project fully supports DOE goals. Results were clearly presented in a form that a layman could understand.
- This is a very good array of investigations.

Question 3: Collaboration and coordination with other institutions

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

- LANL has provided technical support for many stakeholders, including powder manufacturers, catalyst and membrane developers (conventional and alternative), plate producers, automotive original equipment manufacturers, and stack integrators.

- This whole project is all about collaboration with organizations that brought a specific problem for LANL to investigate.
- LANL needs more collaborators on the project or needs to do better job reaching out for user facility service.

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.

- LANL's work is relevant to development of several of the cell components that comprise polymer electrolyte membrane (PEM) stacks. The project work expedites PEM technology development by entities that do not have the necessary equipment and facilities or expertise. This approach reduces development cost and time to DOE.
- All of the sub-projects addressed a DOE research and development objective.

Question 5: Proposed future work

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

- Future work continues previous and existing collaborations and recognizes the unknown character of future tasks.
- Future work is a continuation of the effort. No doubt new tasks will be introduced.

Project strengths:

- Deep and broad knowledge of PEM technology is a strength.
- This project is an excellent use of LANL facilities and expertise. It is a focused effort on solving specific problems that progress DOE objectives. The description of the activities and results was clear and concise.
- Collaboration with a variety of companies on focused research that LANL is uniquely qualified to conduct is a major plus. LANL participation in the Fuel Cell Technology Team is a strength.

Project weaknesses:

- The narrow focus on PEM technology is a weakness.

Recommendations for additions/deletions to project scope:

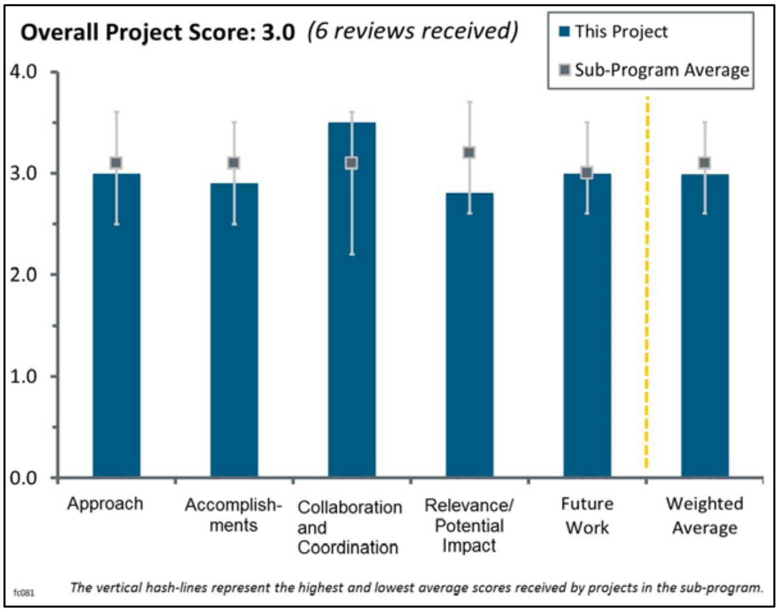
- This project should be kept going and expanded, if possible. The approach should be applied to other national laboratories.

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

Jennifer Kurtz; National Renewable Energy Laboratory

Brief Summary of Project:

The fiscal year 2016 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.



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Question 1: Approach to performing the work

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

- Given the constraints required to protect proprietary information and the reliance on voluntary submission of data from industry, the project has managed to achieve a good level of participation. The whole project relies on achieving this balance.
- The approach in terms of statistical analysis is solid and provides valuable information. The approach based on receiving voluntary information, although very difficult, is starting to pay off, and incentives through the provision of feedback were very interesting. The feedback provided in terms of ranking will also be very valuable to the industry.
- The project aims to gather data from voluntary data suppliers (mostly industrial) and analyze the data to produce both detailed data products (DDPs) and composite data products (CDPs). Receiving such a large number of data from partners is a real challenge, especially for durability data. The project employs a statistical approach that analyzes data with many heterogeneities (different technologies, suppliers, operating conditions, testing protocols, etc.). Considerable work in preprocessing, pre-selection, and standardization of data is done prior to analysis. The analysis of data is based on statistical analysis/fitting. The degradation fitting is based on segment linear fitting, which is not always appropriate. Using more physics and applying some adequate degradation models may give more accurate values of the projected voltage at 10% nominal voltage and therefore more accurate durability value (20% is more compatible with stationary applications in the Multi-Year Research, Development, and Demonstration Plan [MYRDDP]).
- Collecting real-life operational data from the developer is the best way to measure the industry technology status. On the other hand, as stated in the presentation, industry is looking not only for the highest performance but rather for the balance between performance and cost. It would be more useful to develop a combined standard to measure the progress.
- This is a far more difficult and uncertain task than most people recognize. The approach needs to be bolstered to ensure validity and accuracy.
- This is a generally well-constructed and ambitious project. However, the volume of data collected and (in many cases) the lack of detailed information makes data analysis extremely difficult.

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 does not address any objectives set in the MYRDDP, such as increasing durability or lowering cost, but it is meant to provide an assessment of the status of fuel cell and electrolyzer durability and cost by gathering valuable information from industry. The project is a valuable tool for DOE to assess the evolution of the technology regarding the targets. Data about electrolyzers have been integrated and would be a good support for DOE to set targets for this technology.
- The project does not address any of the barriers on its own but it does so by indirectly providing feedback to DOE. The datasets collected so far are an impressive accomplishment, given that industries are often very reluctant to provide such confidential information. The addition of the set of electrolyzer information is also a positive feature.
- While team members may have extracted as much as possible out of the data they were given, it appears that there is limited NREL evaluation of the data. The analysis is almost a meta-analysis to show general directions rather than a set of conclusions drawn from careful examination. Ideally, there would be enough knowledge about the systems to make observations and insightful conclusions and categorizations. It is concerning that the degree of battery hybridization may be a major factor in (helping or hurting) the longevity of the stacks in ways not captured by the project’s methodology. The results are interesting and worthwhile but ultimately they are of limited usefulness because of the complexity and variations of designs considered (and lack of knowledge about each system). The data inappropriately lumps all fuel cell technologies (solid oxide fuel cells, direct methanol fuel cells, polymer electrolyte membrane fuel cells, etc.) together in one data file. It is hard to envision meaningful or nuanced conclusions coming out of such co-mingled data.
- Given the diversity of technologies and system applications, it is very difficult to analyze the data when all the different systems in a specific group (e.g., automotive) are lumped together rather than looking at a specific supplier and application to see the trend over time. Therefore, it is important to question the value of the analysis to DOE in making decisions on goals and investments in research and development (R&D). However, that is for the end-use customer, DOE, to decide.
- The data from testing need to be standardized to a common set of conditions when possible. If this is not possible, models could be used to project received data to a common basis.

Question 3: Collaboration and coordination with other institutions

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

- The project has done an excellent job in interacting with the industry for data collection. It is evident that participants are very active in data collection and their outreach includes international developers.
- Owing to the voluntary requirement, a high level of collaboration with industry is required to obtain data. The project has done well to get as much information as it has.
- There was good participation from a variety of suppliers.
- Up to now, the project team has been very successful in convincing several partners (data providers) to share data (22 new data sets have been gathered since 2015, and 174 have been analyzed from the start of the project in 2009). However, the fact that the data are provided on a voluntary basis makes the project very dependent on the good will of other institutions (leading to issues with data quality; incomplete information about the testing conditions, incidents, and testing history; etc.). This can lead to unreliable results despite the good analysis of the team. Some partners have provided data for several successive years, which means they are satisfied with the resulting analysis quality of the DDPs. The project certainly has good collaboration with other partners, given the facts cited above.
- U.S. and international fuel cell developers who will supply data voluntarily and review published results are the collaborators. It is unclear what the response rate was and whether statistical evaluation is needed. It is also unclear how vendor veracity is checked.

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

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

- The impact of the project on DOE is indirect but present. It has been useful to see a comparison between different areas of the world given that there are international fuel cell developers in the datasets. The comparison between application areas can provide feedback to DOE as to where to focus its efforts.
- Tracking the evolutions of durability and cost will allow DOE to assess the technologies' advancement and correlate their current status with the objectives and means that have been put to achieve them (funding). This work can support DOE in more efficiently identifying the topics in which to put more effort.
- Given the constraints, the analysis has produced the best possible results. However, it is still unclear whether the results are good enough for DOE to make informed decisions about its R&D program. That is DOE's call. Based on the outputs presented, it seems that the trends will be difficult to identify.
- The project provides useful insight into the overall fuel cell status but the value is severely limited by the complexity of the data, lack of knowledge concerning the circumstances of each data set, and the co-mingling of data from multiple fuel cell types.
- The impact depends on many factors, and these need to be discussed. The workers do recognize some limitations.

Question 5: Proposed future work

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

- The proposed work is reasonable and expected. The cost analysis will also be useful in seeing the relationship between cost reduction and durability.
- Given the constraints on the project, the future work is satisfactory.
- The proposed future work is in line with the results shown. However, it is strongly dependent on whether data are supplied by partners and on the quality of the supplied data (availability of all needed information for analysis).
- The approach should be reviewed to determine merits and limitations.

Project strengths:

- There was good industry engagement in the collection of confidential data and useful analysis showing the trend for the year in each application area.
- There is very good collaboration with industry to obtain voluntary data, which is the core requirement for this project to be in existence.
- This is an ambitious project undertaken by an NREL team uniquely qualified to perform the task. There is a logical approach and competent execution, given the complexity and volume of data involved.
- This project's approach is an independent and uniform analysis of valuable data from key stakeholders. The project offers access to the data without jeopardizing confidentiality, which is a big issue, especially with data linked to durability. Independent and uniform analysis is an important tool for DOE to assess the status and progress of the current technologies regarding the objectives set in the MYRDDP and the funding involved. Analysis is also an important tool for the data suppliers to assess the evolution of their technologies (DDPs) and compare it objectively with the market evolution (through CDPs).

Project weaknesses:

- The fact that the analysis is applied uniformly (no matter what the technology is) makes interpretation and comparisons difficult. For instance, the degradation functions of different technologies are not necessarily the same. In the current analysis, the projected value for stationary applications seems to be underestimated: a value of 20% voltage degradation seems to be a more adapted metric to assess voltage degradation (MYRDDP). Even if the project team submitted a detailed metafile to the data suppliers, data suppliers cannot guarantee what the data “experienced” during operation. For instance, several faults could occur during operation and affect the durability. If the task of gathering technical data has been solved by setting a clear metadata template (though all needed technical data are not supplied), it should also try to include the history of the data (incidents, recovered faults, etc.). However, this kind of information is not easy to transfer.

- The data are too scattered for meaningful data analysis. Insights and conclusions regarding the data are not made in sufficient quantity. The fuel cell technologies are all lumped together. It is possible that “old” stack data are combined with “new” stack data, thereby merely averaging the values, whereas conclusions discerning the performance differences would be preferable.
- The measurement of the yearly progress is not clean. Industry does not look only into performance, so the analysis should include other factors to show industry trend. This trend could also guide technology development in the Hydrogen and Fuel Cells Program.
- The “vanilla” approach to analyzing data required to preserve supplier proprietary information makes it difficult to clearly identify progress and where resources need to be applied.
- It is disappointing that there is no breakdown by technology for both fuel cells and electrolyzers.

Recommendations for additions/deletions to project scope:

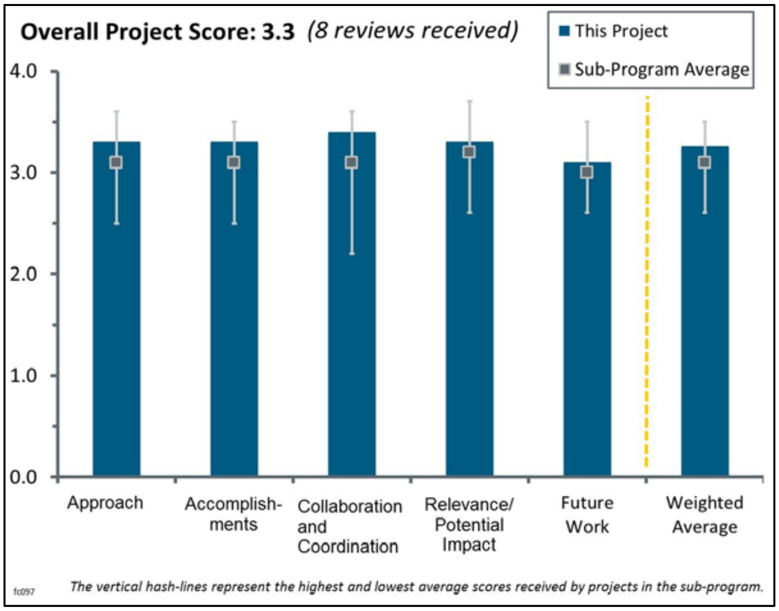
- Grouping the data by technology for different applications and slightly adapting the analysis to each technology/application could help with interpreting the results (20% is more compatible with stationary applications [MYRDDP]). Instead of a basic linear fit for the degradation models, adding more physics behind the fitting function would give a more precise value of the durability. Approaches of data-based prognostics and health management are very good tools for useful lifetime estimation. The DOE-funded projects could be highly encouraged to share a part of the generated data with the project team. The metadata template should include information about incidents that may have an impact on the durability. This information should be taken into account in the analysis. Data about electrolyzers have been integrated and would be of great help for DOE to set targets for this technology. It would be interesting to create categories of electrolyzer technologies so the analysis is run by technology category.
- Given the presence of international fuel cell developers in the datasets, it would have been nice to see a comparison between these regions (e.g., United States vs. European Union vs. Japan) to see the competencies in each application area. Outreach to other funding entities for collaboration would be extremely useful as more participants could be urged to join the data collection exercise.
- The project should collect real-life data from industrial developers. These data are more representative than laboratory testing data. The project should cover multiple types of fuel cells.
- The project team needs to dig deeper into the data to remove the scattered and blended nature of the voluminous datasets. This may/will require additional data from the suppliers—data the suppliers may not be willing to give.
- DOE should analyze the value of the project as currently constituted.

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

Vincent Contini; Battelle

Brief Summary of Project:

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



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Question 1: Approach to performing the work

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

- The objectives of the project are well aligned at addressing the barriers of cost reduction of fuel cell components and materials. The methodology that has been followed so far is well planned, and the system design for other technologies is well broken down and detailed.
- The project approach appears to be well organized and focused around estimating the manufacturing cost of small to medium-sized FCSs for stationary and backup power. The study takes an agnostic approach to specific fuel cell chemistry, providing a useful point of comparison for polymer electrolyte membranes (PEMs)and solid oxide fuel cells (SOFCs). Addressing a couple minor issues would improve this project. First, the parasitic load is pegged at 20% of the gross power. This is too high for a well-designed system and leads to balance-of-plant (BOP) components that are oversized in addition to an oversized stack. Second, one of the stated objectives is to identify areas of manufacturing improvement. One of the strengths of cost estimation is that it highlights existing manufacturing practices that are inefficient. The other strength is that it can be used to identify components and materials that drive cost. Neither of these discussion points was addressed at the level or depth they deserve. Maybe there was not enough time in the presentation, but this is the kind of insight that is needed for this work to have its maximum benefit.
- The project does an adequate job of identifying the main contributions to the cost of the two FCSs. Connecting with additional commercial suppliers currently selling combined heat and power (CHP) systems should be a priority for the approach. While not directly contributing technical solutions to the barriers, this project is helping answer questions about acceptance and focus areas moving forward.
- The project as conceived—independently evaluating costs of low-temperature PEM (LTPEM) and SOFC systems—would provide additional insight into which technologies are best suited for stationary CHP applications.
- The manufacturing cost methodology is well developed for the task at hand.
- The project has a well-organized approach. The cost modeling does not identify Design for Manufacture and Assembly (DFMA); however, DFMA is reported in the presentation.
- Battelle’s methodology involves market assessment, system design, and costing supported and guided by stakeholders from most aspects of the technology. It is not clear what entities provide input into market assessment and system design. Market assessment does not seem to involve end users. Battelle uses

established DFMA techniques for cost estimates where appropriate. Parametric analysis by system size and manufacturing volume is informative. Technology Readiness Level 9 for stacks and fuel processors is optimistic.
- The decision to use a venturi approach to the SOFC anode recirculation instead of a blower had the design impact of strongly limiting the turndown ratio. This in turn reinforced the decision that the target market was urban and always on the grid or almost always on the grid. This unnecessarily limited the target market. Non-urban non-grid adopters were excluded, but these customers might be early adopters and willing to pay a premium. Examples include the oil exploration/fracking industry. Not using a recirculation blower also removed an expensive and poorly developed piece of equipment from the cost/development equation. High-temperature SOFC anode recirculation blowers are difficult to come by and have a low mean time between failures. The team repeatedly received input that potential customers would expect SOFC CHP systems to be able to provide backup power should the grid go down. The system that this effort developed therefore may be targeting an unnecessarily small niche—urban users who rarely expect the grid to go down. The customers looking for backup power would have to look elsewhere.

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 following important specific conclusions have been obtained: electronics and power conversion dominate system cost, particularly as system size increases; an attractive value proposition exists under specific utility rate conditions; Manufacturing Readiness Level (MRL) for many BOP components not ready for mass production could be a significant cost driver; and DFMA performed on specific components (fuel processing, stack) assumes technology at greater than MRL 9.
- The primary goal was quantifying cost reduction likelihood as production levels increased. For the system chosen, this was very well done. The analysis is sound.
- The breakdown of costs that have been provided can be very useful for DOE, as it can identify the most important cost contributors. It is evident power electronics for both applications are expected to be the biggest contributor to BOP costs for both technologies, and therefore future DOE efforts should involve projects dealing with this issue. It is strange that, in the forecasts shown, there is no labor cost decrease with the increasing number of units.
- The project appears to have made good progress toward surveying and summarizing the size, application, and chemistry space. It would be useful for understanding the system designs if the authors were to include equivalent systems for comparison and to highlight where assumptions have been made.
- Cost analyses were completed for 100- and 250-kW PEM and SOFC systems for CHP and primary power. Tornado and waterfall charts and identification of high-impact R&D would be informative.
- It is not clear why the fuel processor for the PEM system requires two high-temperature shift reactors. Commercial SOFC (Bloom Energy) systems use stack heat to improve electrical efficiency. It is not clear that this design loses electrical efficiency by having a micro-CHP application or that other proposed commercial SOFCs have a CHP component, e.g., the LG Fuel Cell System. System life is projected to be 50,000 hours. It is not clear whether the PEM or SOFC stacks will last 50,000 hours and, if so, what experimental evidence exists for such long stack life. Overall efficiency of 80% for a PEM system is very high considering the low quality of heat from a PEM fuel cell; it is not clear how this is justified. Electrical efficiency of 40% for an SOFC system is low when compared to the reported Bloom Energy SOFC electrical efficiency (50% or greater). Most SOFC grid-connected systems are for base load with peak power supplemented by other systems. The justification for not identifying the SOFC as a base load system is not clear. The use of PEM systems for off-grid operation as a critical load or backup power is justified. The SOFC system as a backup power source would be difficult to justify based on operating at hot standby waiting for backup power applications. For PEM applications, the gas diffusion layer does not have a microporous layer added to the paper, the feasibility of which seems questionable. Silicone is poison to PEM fuel cells and is normally not used in PEM fuel cell seals. The use of silicone should be discussed with fuel cell original equipment manufacturers (OEMs) and fuel cell seal manufacturers. DFMA methodology is reported on slide 13 but is not identified in the Approach. It would be good to know how the cost per kilowatt-electric for PEM compares with the Ballard or Altergy cost (slide 14). PEM BOP

costs are much greater than PEM stack costs, perhaps implying that the Fuel Cell Technologies Office should redirect efforts to reduce BOP cost. Total system cost per kilowatt with markup is considerably below the Bloom Energy reported cost of $10,000 per kilowatt. How Battelle rationalizes its cost numbers when comparing them to Bloom Energy's is unclear. The reviewer agrees with the overall summary.
- Lack of data on commercial systems in the power range presented as well as limited customer acceptance data left the project lacking real informative data for the last barrier. Updating and revising the same methodology used over the last few years does provide interesting cost and system-level information, but the missing current CHP data and customer usage requirements leaves room for progress next year.
- The deemphasizing of HTPEM systems is regrettable.
- While the cost models were very detailed and the methodology sound, there were few bill-of-material or manufacturing cost reduction suggestions, and team did not suggest R&D areas to improve cost. These would significantly increase the value of the project to DOE.

Question 3: Collaboration and coordination with other institutions

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

- The stakeholder input is done through 19 companies that represent the sector well. They have been consulted for design inputs, cost inputs, and reviewing the results. Dissemination of the results should be encouraged.
- The team had significant participation from industry in the areas of LTPEMs and SOFCs.
- Collaboration is very extensive.
- The list of collaborators is appropriate for the systems being analyzed.
- There are good interactions. A SOFC OEM should be included.
- A large cross-section of stakeholders is listed to provide design and cost input and to assess validity of the results. Involvement of raw materials suppliers and stack/system component providers seems weak.
- The long list of collaborators proves that data are being requested and used to fill in the model, but a lack of mature system data is noticeable. Looking to Europe or Japan for current CHP data and performance metrics is suggested to help increase the fidelity of the project's customer acceptance portion.
- It would have been better if potential customers had played a larger role in the early part of this effort, when the performance parameters of the CHP system were being determined.

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 has very high relevance and potential impact by helping to identify the biggest contributors to system cost. Though more data are needed for this project, the cost and system data provided can help DOE focus investment in future years.
- The work helps answer questions about opportunities for cost reduction to penetrate non-automotive applications.
- Cost estimates provide insight into key areas for more R&D aimed at reducing cost.
- The primary goal was cost reduction. That analysis was excellent—a 4.0. The secondary goal was manufacturing capability. That was also well explored. The third goal was customer acceptance. The approach chosen was to win customer acceptance by driving down unit cost. That was flawed. Customer acceptance would have been enhanced by better understanding customer requirements.
- The project has a strategic impact, as it can affect and adjust DOE's plans regarding funding cost-reducing projects and regarding projects that focus on increasing the MRL of the components that are identified here. It would have been good if, through the project's analysis and the feedback received from the stakeholders, suggestions for further cost reductions could be made.
- The project highlights the importance of BOP to the overall cost. Some of the analyses need to be compared to real-world costs. The results reported here should be compared with costs for a state-of-the-art

gas turbine. It is unknown whether a state-of-the-art gas turbine would be less costly and how emissions would compare to a gas turbine.
- While the authors have broken down component cost contributions, it is not clear that the authors have addressed areas in which current manufacturing approaches need improvement. This is the kind of insight that is needed for this work to have its maximum benefit.

Question 5: Proposed future work

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

- Reviewing and updating all of the work done over the duration of the project with the most up-to-date information will be a huge benefit to this project. There has been much progress over the years, and making improvements to past simulations will be very useful.
- Future work involves straightforward updating of previous cost estimates. The project is ending in 2017. Applications and reports will be revisited and revised as appropriate.
- Work appears to be concluding. Battelle will revisit all applications in the previous four budget periods and update all reports.
- The project is all but complete.
- The proposed future work is not detailed and was mentioned only briefly.
- This effort is near its conclusion, so there is neither a great deal of proposed future work nor much room to change. One thing that might possibly be done would be to take the finished system design, predicted performance, and predicted costs, and “shop it around” to potential customers to gauge the product’s effectiveness in attracting market interest. The results of that could have an impact on future cost studies and system designs.
- Future work is not specific, but a review is necessary.
- The proposed future work leaves out significant detail. Because this is the final year of the project, it makes sense to review the analyses of the previous four years. However, it would have been helpful to know what specific weaknesses of the previous analyses the authors expect to address, and maybe some global trends that they would like to explore. At this point, the authors should be in a position to address what the key cost drivers are.

Project strengths:

- Many valuable data have been created through this project. The system design and cost analysis have been very detailed. The project has done a good job on receiving input for the system design and manufacturing costs from a large number of stakeholders.
- The project does a great job of identifying the cost drivers for the CHP market. The project highlights the need to address BOP, which seemed to be a common theme in the review this year.
- The project provides a broad survey of fuel cell applications including primary power, backup power, auxiliary power units, and material handling equipment. Business cases were explored.
- There is a broad team from across the LTPEM and SOFC industry. The cost modeling methodology is sound and detailed.
- The system design is well developed. The project uses a systematic approach.
- The cost analysis performed was excellent and educational.

Project weaknesses:

- The decision to not use an anode recirculation blower for the SOFC may have unnecessarily limited the potential market, which means the analysis is weakened because of its applicability. The effort might have benefited from having representatives from the customer base involved as collaborators.
- The project lacks real CHP data from current market leaders. There is also a lack of an international baseline because CHP is more common overseas.
- The project did not include HTPEM. Information from companies such as Advent or Serenergy could have been used.
- Several sizes and types of systems were analyzed without fully exploring manufacturing issues.

- There is a large discrepancy between the analyses and costs for commercial FCSs.
- There is a lack of solutions proposed that address the cost reduction issues.

Recommendations for additions/deletions to project scope:

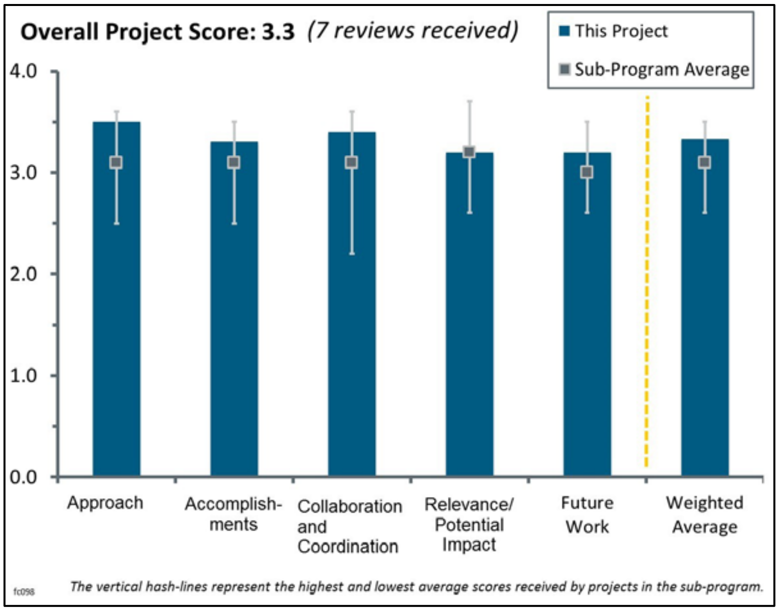
- This is a good project.
- DOE should take note that if we rely on all types of PEM cells and applications to build the manufacturing base and move down the learning curve, the technologies should use “the same” cell materials and processes. It is not clear that such a comparison has been made between the cost programs of Battelle, Lawrence Berkley National Laboratory, and Systems Analysis, Inc.
- If possible, in the months left and with the small amount of funding left, “shopping the design around” to potential customers would help assess how germane the research is to the potential customers who are interested in CHP.
- The project should rationalize the differences between the cost analyses presented here and the cost of commercial FCSs and the cost/properties of a state-of-the-art gas turbine system.
- The project should find commercial data for commercial acceptance and cost savings.

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

Max Wei; Lawrence Berkeley National Laboratory

Brief Summary of Project:

The overall objective of this project is to develop a total cost of ownership (TCO) modeling tool for design and manufacture of fuel cells in stationary and materials handling systems in emerging markets. Project goals include (1) expanding the modeling framework to include life cycle analysis and possible ancillary financial benefits, including carbon credits, health/environmental externalities, end-of-life recycling, and reduced costs for building operation; (2) identifying system designs that meet lowest manufacturing cost and TCO goals as a function of application requirements, power capacity, and production volume; and (3) providing the capability for sensitivity analysis to key assumptions.



Question 1: Approach to performing the work

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

- An independent TCO model of low-temperature polymer electrolyte membranes (LTPEMs), high-temperature polymer electrolyte membranes (HTPEMs), and solid oxide fuel cells (SOFCs) with recommendations to the U.S. Department of Energy on the most effective areas for continued research and development (R&D) is an excellent project.
- Lawrence Berkeley National Laboratory involves stakeholders from most aspects of the technology to provide input to materials, processing, stack and system design, and operations and to review results for validity. Parametric analyses on the basis of production volume and system output is instructive. More information on the Air Pollution Emission Experiments and Policy 2 analysis model (APEEP2) is needed. Focusing on externalities and applying findings to other fuel cell costing projects would be interesting.
- The life-cycle impact assessment is a good addition to the Fuel Cell Technologies Office.
- The approach appeared sound and complete.
- The approach for the project is credible, but inputting data from multiple stack producers and using current density from one vendor and power density from another does not seem realistic. The cost information updates compared to last year’s data were appreciated. Presenting combined heat and power (CHP) usage examples in various markets is an excellent approach to identify acceptance criteria.
- The attempt to value environmental externalities is carefully presented and included in the overall life-cycle assessment. However, externalities may be overvalued. HTPEMs would be attractive for CHP; however, industry consensus is that durability is inadequate. There was no mention of HTPEMs in the discussion; the project just needs to justify ignoring them.

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 accomplishments over the last year have been excellent. Highlighting cost benefits, health benefits, and potential future regulations all help paint a more accurate picture of what CHP systems need to do and cost to be competitive.
- Extensive revisions to the SOFC CHP systems life-cycle cost model were completed, including updated financial quantification of externalities (e.g., environmental benefits). Tornado charts provide insight into the areas needing the most cost-reduction R&D. Perhaps tornado charts could be generated for sensitivity of externality inputs and assumptions, if that is possible.
- The effort is near its end, and the results are almost finalized. The accomplishments and work progress are excellent and essentially complete.
- The de-emphasis of HTPEM is regrettable. The team’s approach was very detailed, and the TCO model is impressive and appears sound in most areas. As to overcoming barriers, there appeared to be few recommendations for improving the TCO models. Leasing and platinum recycling are two areas that could significantly improve the cost of electricity (COE).
- The focus on SOFC for CHP is good. System temperatures seem a little low for current SOFC technology. The discussion of turndown and other system operating factors that influence effectiveness as CHP were not included in the presentation (it is not clear if operating factors were included in the analysis but just not presented). PEM systems may also be relevant for CHP when heat available from fuel processing is included. Specifics of PEM hotel evaluation are lacking (whether waste heat recovery is counted). It is not clear whether either system is considered capable of operating off-grid in backup power mode. Based on SOFC schematics, off-grid operation seems not to have been considered. Grid-outage operation may be an important consideration for end-user value—saved business and saved product during grid outage translates into real dollars, unlike environmental externalities, which typically do not have real cash value in most locations.
- There was no sulfur clean-up in the 50 kW SOFC CHP system; this should be added. It is not clear where heat comes from in the second heat exchanger that increases the air temperature to 650°C. A 59% average system net electrical efficiency is high compared to Bloom Energy’s ~57% electrical efficiency with no CHP component. Stack yield numbers on slide 16 appear to be very high. The scrap rate was not provided. The system cost is very low compared to Bloom Energy’s cost. There was no definition of the marginal emission factors (MEF). It was difficult to follow the greenhouse gas (GHG) benefits. Units change between slides 23 and 24.
- It was not clear how the costs determined here compare to a state-of-the-art gas turbine system. The state-of-the-art gas turbine system has GHG emissions better than or equivalent to the Bloom Energy systems; it is not clear how this system compares.

Question 3: Collaboration and coordination with other institutions

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

- Stakeholders from most aspects of the technology provide input and results assessment. Involvement of Strategic Analysis, Inc., is encouraging because of the company’s long experience in estimating costs of fuel cell systems.
- The project has great input from multiple collaborators. It is clear the project has reached out and communicated with experts across the United States and international communities.
- The nature of this project does not lend itself well to collaboration in carrying out the work, but it is good to see that industry has been consulted as a reality check on assumptions.
- The diversity of the collaborator group seemed well thought out and comprehensive.
- The list of subcontractors and contacts is extensive.
- Teaming arrangements are very good.

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.

- Insights into high-cost areas (tornado charts) are crucial to identifying R&D needs. Quantification of externality benefits (emissions) helps assess the market potential and initial geographical and application penetration and identify early market opportunities from an environmental standpoint.
- The work is highly relevant. The regional emissions data and potential impacts that fuel cells can offer to those highly affected regions are huge. The coupling of the health impacts with the cost data and TCO keeps this project highly relevant.
- There is one aspect to how this effort was performed that is noted here rather than in Accomplishments and Progress, and that is the unknown confidence interval in the calculations used to determine the health/environmental impact in dollar per kilowatt-hour. The results of the study are clear: even at the highest production levels studied, from a private entity's perspective, adoption of this technology is not cost-effective. Only when the societal impact of NO_X/SO_X/PM is factored in does the technology appear cost-effective. To calculate that environmental externality, it was necessary to depart from standard Design for Manufacture and Assembly (DFMA) analysis and extrapolate from a university study to estimate the health/environmental impacts of NO_X/SO_X/PM reductions in monetary terms. (The analysis showed that health was the driving externality by far over GHG emissions, even when the CO_2 reduction was measured at a high GHG credit rate of $40/ton of CO_2.) The problem is that there is no confidence interval in the calculation of the health/environmental impact. The danger is that the results of this study might be used to argue for mandated use of these systems to displace diesel systems when the calculated value is used without knowing a true confidence interval. Without the confidence interval, making a policy recommendation based on the results of this study implies an analytical underpinning that is stronger than what actually exists.
- The team highlights progress in cost reductions, cost of energy increases, and high spark spread areas and accurately predicts geographies where there is a strong existing COE. Including "escalating social cost" as recommendations for improving the COE of commercial CHP systems will be difficult.
- The relevance/potential is not as high based on differences between the analyses presented here and actual costs of commercial fuel cell systems. There is a disconnect somewhere. It is not clear how the GHG emissions compare to state-of-the-art gas turbine systems or whether the project is making an improvement.
- The focus on environmental externalities is valuable but probably insufficient to assist with market penetration in most localities under most current regulations.

Question 5: Proposed future work

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

- This project is near its end, and there may not be much room to make any changes. But if it were possible, taking a stab at determining the confidence interval of the health/environmental externality impact on notional cash flow would advance the overall project's worth.
- The project is in the final phase. Future work involves final estimation revisions and preparation of the final report. The report should include adequate discussion of externality concepts and benefits.
- The project ends in three months; future work is primarily documentation and reporting.
- This is the end of the project; no future work was addressed.
- The project is near completion.
- No future work was proposed.

Project strengths:

- Studying TCO and health (societal) impacts of using fuel cells in a CHP is definitely a project strength. Adding future impacts of the clean power regulations shows the project is looking into the future to ensure the data provided are accurate for the near future and further out.

- The analysis is detailed and rigorous. The COE model is impressive. There has been extensive vetting with industry and collaborators.
- The project is well envisioned, has good analytical rigor, and is well performed.
- Quantification of externalities is very informative.
- The project attempts to value environmental externalities.
- The project benefits from the inclusion of life cycle impact assessment.

Project weaknesses:

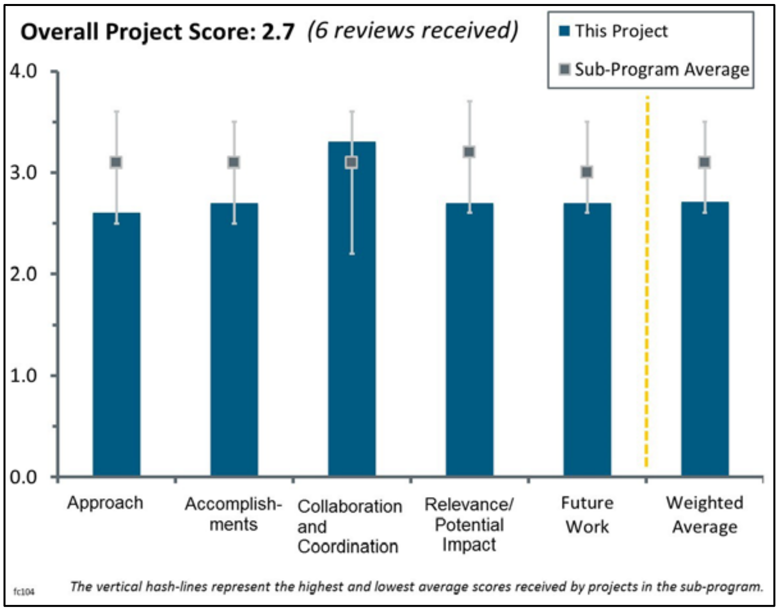
- This is not as much a weakness as a result of the effort: the advisability of using these systems rests upon a societal benefit because the cost analysis, when looking at just the impact for the system owner, indicates that there would not be a cost benefit to adopting it. The societal benefit is determined by the one element of the analysis outside the standard DFMA framework: the health/environmental externality. Without a confidence interval for that calculation, is it difficult to know how to weight any decisions to adopt such systems.
- The data can be very complex when presented; the only weakness would be in the pace of the presentation.
- It is hard to tell if off-grid (grid outage) operation is included—probably not. If not, this is a significant oversight in evaluating the TCO.
- Lack of HTPEM data is a weakness. Detailed information could be gathered from companies such as Advent and Serenergy.
- The project needs to compare its results to state-of-the-art gas turbine systems in terms of cost and GHG.

Recommendations for additions/deletions to project scope:

- The project should include grid-outage evaluation. Operation off-grid will require additional hardware and changes in design compared to what was presented and will therefore increase cost. However, the value of avoided losses in business and/or product (think frozen food at the grocery store) will more than offset the additional cost.
- Slide 34 shows future stack durability as 40,000 hours. A better number would be 60,000–80,000 hours. It would be good to know whether this changes the results and conclusions.
- The project should attempt to calculate the health/environmental externality financial cost confidence interval.
- The SOFC cost analyses need to be rationalized to the commercial SOFC cost data.

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

Andrew Steinbach; 3M



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Brief Summary of Project:

The overall objective of this project is to develop a durable, low-cost, robust, and high-performance membrane electrode assembly (MEA) for transportation applications able to meet or exceed the U.S. Department of Energy (DOE) 2020 MEA targets. Objectives for fiscal year 2016 are to (1) produce project best-of-class components and catalyst-coated membranes (CCMs) via continuous pilot manufacturing processes; (2) validate performance and operational robustness of MEAs in short stack; and (3) evaluate MEAs for performance/cost modeling and durability under accelerated stress tests (ASTs) and load cycling.

Question 1: Approach to performing the work

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

- The project aims to overcome issues with the nanostructured thin film (NSTF) structure in order to take advantage of its inherent benefits. While the benefits in terms of corrosion resistance and high specific activity are considerable, several years of funding have now been expended in an effort to overcome the limitations. There has been considerable progress, but reaching the project goals seems unlikely at this point. There has been extensive testing and a reasonable level of characterization and diagnosis. Although the presentation lists an approach to identify mechanisms of unanticipated component interactions through advanced diagnostics, it is not entirely clear what was done here aside from operational studies and linkages to membrane degradation. These are good but not entirely sufficient. The performance drops off as the cathode oxygen reduction reaction (ORR) decreases below 10 mA/cm^2. This behavior looks similar to conventional supported catalyst losses at low roughness factors/low activity. Losses were stated as being due to intrinsic specific activity loss (structure, composition); extrinsic (contamination); and coarsening/dissolution. These seem to be all the same factors that affect more conventional designs but with the added challenge of the lower surface area. The NSTF catalyst seems to be converging with conventional catalysts at these low loadings with little inherent benefit and with reduced design space levers. However, there should still be a benefit of no ionomer required, which may alleviate the thin ionomer transport losses but the extent of which is not clear. Since the challenges of NSTF catalysts and more conventional Pt/C catalyst designs at low loadings are converging, understanding gained under one system may be applied to the other system with the differences helping to elucidate effects. More use of models and more fundamental understanding on the limitations with additional diagnostic approaches may have been helpful. In terms of stated mitigation approaches, the approach to decrease 3M perfluorosulfonic acid (PFSA) polymer electrolyte membrane (PEM) decomposition rates should be relatively easy to test through the use of PEM stabilization additives. It is not clear if this has been done. While the stated mitigation approaches address the kinetic activity losses, these losses affect only 30% of the performance loss. It is not clear what approaches will address the over-70% of mass transport loss observed. The external contamination effect remains a real risk for the NSTF catalyst. An assessment of the level of risk for this effect compared to a high-surface-area catalyst should be established. Improved approaches to understand proton transport in the NSTF layer would have been useful to potentially design a better layer or to leverage the understanding to other catalyst layer designs.

- The researchers have provided General Motors (GM) with MEAs for independent testing of the NSTF 3M MEAs. This is a much-needed independent validation test of the materials. The researchers were unable to reconcile the differences in MEA performance.
- The effort let by 3M to address fuel cell cost has been well designed and proven to be feasible. NSTF-based catalyst supports have been very well characterized. The implementation of this technique is extremely effective at addressing the issues with catalyst loading and power density. The issues of durability with this system have been in question for some time; the system may not be able to address the durability targets.
- This project continues incremental improvements to NSTF MEAs relying on dispersed-catalyst interlayers to improve robustness, giving the anticipated decrease in durability against voltage cycling versus NSTF alone. Apparently the growth of longer support whiskers, which could increase the electrode thickness for better operational robustness while maintaining the durability of NSTF alone, has not proven feasible.
- Within the limits of the NSTF MEAs that 3M has been evaluating stubbornly for the last 17 years, the approach of fine-tuning and juggling is fine in the attempt to find some progress that makes the materials competitive with traditional MEAs using supported catalysts. Testing in short stacks is a good way to show the progress or lack thereof very clearly.
- Status of each performance metric is clearly identified against the DOE targets. Identification of mechanisms for unanticipated component interactions through advanced diagnostics is not matured or demonstrated sufficiently to ensure all MEA targets will be achieved by the end of the project.

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

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

- The accomplishments and progress on addressing fuel cell cost are very good. This is only addressing a few barriers to fuel cell implementation.
- A good deal of work was accomplished; however, it was not sufficient to fully resolve major issues with the NSTF platform: load-cycle durability at rated power density as well as robustness in transients at ambient temperatures. Work over the past year focused mostly on mitigating solutions via modifying individual components outside the NSTF electrodes, which still did not prove sufficient to meet robustness or durability targets. It is quite unfortunate that the researchers found fundamental root causes of low polarization performance in stack due to elevated overpotential in the anode quite late in the project. Using alloys with transitional metals, especially at the anode side, may change reference potentials and promote leaching of ions. Activation of both electrodes should have been addressed in greater detail early in the project.
- Overall, progress continues on the use of the NSTF catalyst-layer design, but over the past year, progress appears to have slowed with no real new advancements. Accomplishments include the following: 12 MEAs tested at pilot scale with good reproducibility; increased mass activity and specific area achieved at pilot versus laboratory scale; significant improvements in low-temperature performance with the reduced hydrophobic backing treatment (X3); and the interlayer concept's enablement of the ability to achieve load transients—Type B selected with 16 µg/cm^2. This year saw down-selection of designs and increased characterization and testing; robustness targets have not been met, but improvements have been made. The design with M catalyst has further small improvements and is getting closer to targets, designs with interlayers pass DOE AST targets, single-cell testing load transient data provides similar results between GM and 3M, and improved robustness for best-of-class (BOC) MEA is confirmed. A number of issues remain including the following: durability of less than 800 hours during load/relative humidity (RH) cycle for 10% degradation in performance (30% kinetic losses and 70% mass transport losses); lower performance in single-cell testing at GM (60 mV), even though 3M testing shows expected performance; and disappointing short stack evaluation results with much lower polarization curve performance. The stack transient performance was also much lower than expected, and cells failed at 70°C, 100% RH. However, the NSTF baseline CCMs passed, though there was still some instability observed. Therefore, more optimization and understanding are needed; conditioning ineffectiveness may have been a contributing factor, and the project is actively working on and making progress on improved activation procedures.
- The results were a bit disappointing. GM was not able to validate the best performance results. The dealloying catalysts do not seem to provide benefits over conventional catalysts. It still remains to be seen

whether the researchers can translate their single-cell results into stacks. The activation protocol appears to be difficult to reproduce and may not work in stacks.
- The accomplishments are minimal but acceptable considering very few knobs left to tweak after a decade of work on very gradual improvements to arrive at the state in which the project finds itself today. The negative results in stack studies can be blamed, as usual, on poor break-in/conditioning/thermal cycles in GM's test stands. It is not surprising, and 3M MEAs have never worked in a PEM fuel cell stack under practical PEM fuel cell conditions. If they had, automakers would be using them in their fuel cell electric vehicles (FCEVs). Although this year's project is a freebie, getting GM or anyone else to evaluate 3M MEAs in PEM fuel cells could be a distraction.
- The new BOC MEA has given mixed results, performing poorly at high current density at GM (perhaps owing to problems with proper break-in technique) and not yet meeting DOE 2020 loading, specific power, and load cycle durability tests. The 30,000 load cycle test could probably be passed with a slightly higher loading of interlayer, but MEAs of interlayers will likely never match the durability of base NSTF in ASTs. The new BOC MEA with interlayer also failed short stack transient testing at GM while, to the surprise of all, the NSTF baseline, without interlayer, passed. Based on the project results, it is not clear that continued investigation of NSTF is going to get to the ultimate DOE targets and automotive targets for operational robustness.

Question 3: Collaboration and coordination with other institutions

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

- It is always an excellent idea that 3M gets its NSTF MEAs tested in automotive stack environments by independent parties. It exposes the severe shortcomings of their NSTF catalyst that is well known in the automotive fuel cell industry in a fair and transparent manner. Owing to a variety of folks providing data, the units used within each slide vary from atmosphere absolute and kilopascal absolute, depending on the scientist plotting the data. The SI system was designed to be used as absolute units, and kilopascal is more accurate. Kilopascal gauge and kilopascal absolute are not real, valid units. Atmosphere absolute is also obsolete. Gauge and absolute pressure are used only in the English system units such as pounds per square inch gage and pounds per square inch absolute.
- 3M has strong collaborations with various universities and DOE national laboratories and a good strong partnership with GM. The effort is well coordinated. It is not clear whether cathode catalyst alternatives to Pt-Ni are being evaluated.
- The work shown in this presentation appeared to have been done at only 3M and GM, with modeling done at ANL. Perhaps the other subcontractors completed their work prior to this last (extended) year of the project. Completion of stack testing at GM has required a year's no-cost extension of the project.
- The reported year collaborations, as presented, were focused on the GDL partner and modeling efforts and stack testing at GM.
- GM data was a bit limited compared to what 3M provided.
- The project has a strong team.

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 **2.7** for its relevance/potential impact.

- The research effort was needed, as the NSTF MEA performance was difficult to reproduce between investigators. It remains to be seen if this type of MEA will meet 2020 performance targets.
- Although early on the technology showed exceptional promise to meet the low-loading and durability targets, the continued problems with operational robustness and the durability results under load/RH cycling are reducing the probability of success with this design. At this point, it is considered unlikely that the NSTF will be the design of choice for future automotive stacks. However, the value of the work could still be reasonable if increased modeling and diagnostics were incorporated to learn from the design.

- The base NSTF system has sufficient durability virtues that attempts to patch its shortcomings in operational robustness have been justifiable. The addition of an interlayer to improve transient and cold-start conditions raises sufficient durability issues that extended testing was appropriate. The modest (if any) net gains over the past year suggest that further work along these lines might not have much impact on the industry.
- If DOE asks all the FCEV companies for their input on whether these 3M NSTF MEAs are suitable for automotive stacks, DOE will receive a close-to-unanimous vote. 3M stacks have been evaluated in a number of automotive companies with no success; the MEAs used today in stacks of cars that are on the road or about to be on the road are typically PtCo/C, as openly reported.
- While the relevance and potential impact of the project remains high, the speed of the progress toward achieving the goals and, therefore, being accepted for commercial distribution is slowing down.
- The project aligns with the goals and objectives of the DOE Hydrogen and Fuel Cells Program (the Program). Insufficient information was presented to assess whether the approach to address durability in the NSTF-based catalysts will have the impact to address the Program’s goals.

Question 5: Proposed future work

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

- The proposed future work focuses on stack operation and a down-selection of MEA. This project will meet the MEA cost goals.
- The project has essentially ended with only a few months left. The stated future work is appropriate.
- This project is almost complete. Future DOE-sponsored work would be on catalyst development under a new project reviewed elsewhere.
- It is really difficult to assess the effect of the project’s future work on progress, but all indications are negative unless they drastically change the NSTF catalyst layer to a hybrid of some sort. Only modest modification of the NSTF structure was allowed, according to 3M (on slide 18). The likelihood of success with this constraint is low.
- The mitigation strategy to develop ionomers to minimize contaminant generation proposed for future work would again shift the focus away from the NSTF electrodes into different component development and should be avoided. Focusing on maturing activation procedures for both electrodes as well as transferring this technology and its early diagnostics to the stack project partner is essential to further narrow discrepancies between 3M and GM testing.
- There is much future work needed to replicate the previously reported data.

Project strengths:

- The project has a strong team with excellent industrial partners and testing under relevant conditions. The design provides an opportunity for additional understanding around issues with low catalyst loading at high current densities.
- The project develops significant insights on the NSTF-based electrodes for application in the MEAs in PEM fuel cells. The status of all project metrics is clearly identified against DOE targets, and if successful, this project has a high potential impact on the automotive fuel cell industry. Findings in the project have high and synergistic values narrating issues with thin film electrodes for performance at mass-transport-limited power densities and transient behavior.
- It is a strength to have a project where NSTF catalysts are evaluated by an independent laboratory. The candidness of the current 3M researchers in reporting less-than-spectacular results is appreciated.
- The project continues diligent and well-thought-out work on incremental improvements to NSTF MEAs towards meeting DOE’s ultimate targets and automotive requirements on operational robustness.

Project weaknesses:

- NSTF-based MEAs remain highly sensitive to practical operational aspects of PEM fuel cells in load-following applications specific to thermal and load transients, start-up, and ability to demonstrate required power density, which limits the industry appetite to test these MEAs in stacks. Several aspects addressed by

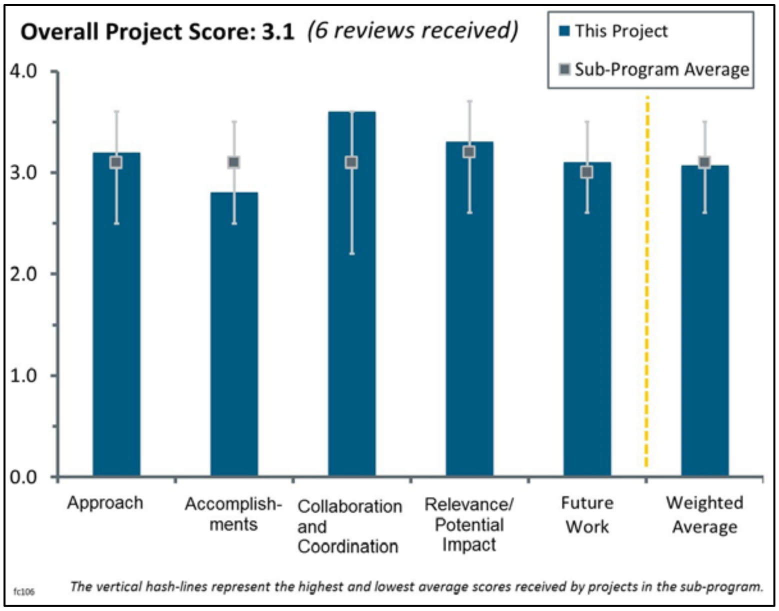
3M for robustness to ambient temperature and operating power density at the beginning of life were quite derated in durability testing, showing insufficient mitigation adopted to resolve the fundamental issues.
- The project approach relies heavily on addressing each challenge incrementally and does not appear to do sufficient analysis of underlying issues. Inclusion of modeling with increased predictive and mechanistic exploration would have been useful.
- The gains of the project over the past year have been marginal, at best, suggesting that NSTF may be reaching a limit of diminishing returns without a major reworking of the system.
- The inability to translate single-cell break-in protocols to stacks was a major weakness.
- The focus only on the Pt-Ni cathode catalyst was a weakness.
- Only modest modification of the NSTF structure was allowed, according to 3M on slide 18. The likelihood of success with this constraint is low to negligible.
- Collaboration with an FCEV auto company shows clear evidence that this project, which has lasted a long time, has run its course.

Recommendations for additions/deletions to project scope:

- The team should be refocused on polishing activation procedure and commissioning diagnostics for both electrodes as well as technology transfer to the project partner. Synchronization of the short stack size and flow fields between 3M and GM testing is desirable to ensure similar test results. The project should avoid shifting focus on the additional components, such as development of new ionomers for the MEAs, but rather investigate the model systems for sensitivity of electrodes to contamination.
- The project should continue studies on why the break-in procedures do not work in stacks.
- The team should complete the project and move on to changes to the basic support structure of NSTF or to non-NSTF MEAs.
- Small modifications to the scope will not be of any help after 17 years of development.
- The project is essentially complete.

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

Deborah Myers; Argonne National Laboratory



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Brief Summary of Project:

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

Question 1: Approach to performing the work

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

- The approach used in this (by now complete) project has been very thorough, from catalyst pre-treatment to MEA processing. Several key factors for the cathode/MEA performance were investigated, with the focus on improvements to the high current response of dealloyed PtNi catalysts. The project generated considerable output, which best attests to the value of the approach taken.
- The approach is perfectly adjusted for improvement of MEA performance with utilization of a complex dealloyed PtNi/KB catalyst. The characterization methods were selected based on determination of main parameters of MEA fabrication that will affect the overall performance.
- The approach is a good combination of careful analyses of a relevant materials set.
- Taking on the issues of catalyst cost, performance, and durability is highly relevant for fuel cell systems. The specific approach of this project has three components: determine electrodes/catalyst properties that limit high current density performance (not particularly compelling as conveyed), use characterization to determine performance-limiting properties (good science but unclear how it relates back to improved performance and durability), and design catalyst layers to mitigate performance limitations (good science in some areas but, like the inks, less compelling in the area of catalyst supports). These are all highly relevant pursuits, but they are difficult to accomplish, and it is unclear how effective any of the proposed approaches have been at advancing the state of understanding or performance. The approaches applied are fairly empirical and have limited impact potential, but by starting with Johnson–Matthey Fuel Cells Inc. (JMFC) state-of-the-art materials, the approaches have good performance as a starting point. Much of the work seems to be focused on lower loading of materials developed/demonstrated on earlier projects.
- The project focuses on important factors that limit the performance of dealloyed PtNi catalyst, namely the ionomer distribution, carbon/ionomer agglomerate structure, and leached Ni effect. The uses of in-cell diagnostics and advanced ex situ techniques such as ultra-small-angle X-ray scattering (USAXS), X-ray absorption spectroscopy (XAS), and transmission electron microscopy (TEM) were interesting. This is mainly a characterization project with limited material development effort. Electrode development appears

to be a shotgun approach with little depth into each path. The electrode/ink optimization study in this project has little value to experienced MEA integrators. Target/milestone settings are somewhat arbitrary and did not align with the DOE targets or heat rejection criteria. Overall, the targets were quite modest.

- The approach is reasonable; however, dealloying is a bit questionable for improving the performance of Pt. The baseline annealed Pt is a questionable experimental control, as it may not be the optimal 5 nm Pt catalyst. It would have been preferable for the researchers to use an as-prepared 5 nm Pt catalyst with a narrow particle size distribution. Annealed samples tend to display log normal particle size distributions that may grow faster than monotonic dispersions.

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.

- During the last year of the project, final experiments on integration of PtNi/KB catalysts into MEA structures were performed via optimization of ink composition, modification of support, etc. These experiments allowed the project to complete the matrix of proposed tasks and obtain the crucial information on MEA performance with a new generation of catalysts.
- The project was needed to validate the value of the dealloyed catalysts. The performance gains observed in rotating ring-disk electrode (RRDE) measurements have not translated to improved fuel cell performance. The development of advanced characterization and modeling techniques within this project will benefit future Fuel Cell Technologies Office (FCTO) investigations.
- Good progress was made, with all reviewer comments addressed.
- This is the last year of this three-year project, the objective of which was to "to realize the ORR mass activity benefits of advanced Pt-based catalysts." The team has met both fiscal year 2016 performance milestones, except for the durability using trapezoid cycling up to 0.95 V (a 22% performance loss in peak power, greater than the 10% target). The benefits of dealloyed PtNi catalysts are unclear; at the beginning of life, the catalysts already suffer from significant high-current-density losses (catastrophic at 30% relative humidity [RH]) and trail An-Pt/C in cycling durability testing, not meeting the targets for electrochemically-active surface area and mass activity (unless a less-demanding General Motors [GM] cycling protocol is used). There are no conclusions regarding the viability of cathodes based on the dealloyed PtNi catalysts, which one would expect at this point. There are apparent differences in the durability testing at the United Technologies Research Center (UTRC) and JMFC, with the results of the latter attesting to a noticeably better d-PtNi/C performance than the UTRC test data, even after cycling up to 1.0 V. This is confusing. Sharp improvements in mass activity and "negative" cell voltage loss in UTRC durability testing of the An-Pt/C catalyst are puzzling. They ultimately result in better mass activity and higher cell voltage at end of life than at beginning of life, which needs explanation. Comparison with similar data for dealloyed catalysts strongly favors the Ni-free system. The question of the PtNi catalyst viability thus remains unanswered.
- In comparing the summary of results from the 2015 presentation to the 2016 presentation, it seems that the new additions for this review period are the use of lower equivalent weight ionomer inks and low-loaded performance status. (Slide 19, which is a summary of results, is essentially unchanged from the 2015 presentation, and this overlap is reflected in some of the content presented for this review period.) There are certainly new data in the presentation that go beyond this, including areas of microstructural analysis and limiting current measurements. It would have been preferable to have more conclusions from this work with more information on mechanisms and what can be applied in future systems from what was investigated here. In general, there was too much focus on data instead of increasing knowledge of the systems.
- Broadly speaking, it is not obvious what was learned from this project that was not already known prior to the project. The project further illustrated the effect of leached Ni in many ways but failed to quantify the effect, propose a solution/criteria, or develop a solution. USAXS on the inks and TEM on the electrodes were quite interesting. However, these samples were not prepared from a process that an experienced MEA integrator would carry out. It is dubious how useful these learnings are. MEA testing appears to be the bottleneck of this project. Only a handful of tests were carried out throughout the project. Most data have

only one data point, making interpretation of data quality very challenging. The project largely meets the milestones, but the targets seem modest.

Question 3: Collaboration and coordination with other institutions

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

- The project is a fine example of coordinating multiple laboratories toward a common goal. Characterization work is excellent.
- The team has excellent participants well known in precious metal catalyst design, MEA fabrication, and characterization.
- This project has involved many partner organizations with complementary skills and well-defined roles. It appears to have been very well coordinated, too.
- The team arrangement is good, with JMFC providing the catalyst and MEA, UTRC testing the MEA, and the University of Texas at Austin doing TEM. However, with the involvement of JMFC falling short of expectation and with the loss of UTC Power, securing resources and know-how must have been quite challenging. It is good to see some continuation with GM as a consultant.
- The integration of work is excellent and seems to be due to good collaboration. The modeling could be better integrated.
- The team is strong. Including GM as an in-kind contributor is a strong addition, although GM’s role is not completely clear.

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

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

- The presented research on catalyst layers is highly relevant to FCTO goals and objectives. It focuses on the issues of utmost importance to fuel cell systems for automotive applications, directly targeting improvements in performance and cost reductions.
- The major value of the project was showing that the dealloyed Pt-Ni did not show major performance improvement over Pt.
- The project addresses an important challenge in achieving high power on highly active catalysts.
- The project allowed for a very promising material to be examined in more detail and at lower loading. Some progress was demonstrated toward achieving a number of the DOE targets.
- The project aims to benchmark and understand relevant issues in electrode construction using technical catalysts.
- The project was ended successfully, meeting of all the major milestones and go/no-go.

Question 5: Proposed future work

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

- As the project has essentially ended, this is less of a concern than other areas. Work focused on d-PtNi/C is highly desirable (in particular, Ni-ion issues, and mass-transport and low-RH issues). Next steps in characterization/understanding are of some interest but are less compelling (X-ray tomography, USAXS, electrochemical impedance spectroscopy [EIS], and cell performance data).
- Proposed future work is relevant to initially proposed targets (and will be done during a no-cost extension).
- The project has ended but proposed some continuation to the Fuel Cell Consortium for Performance and Durability (FC-PAD) and a new GM project. It would be ideal to use the newly developed techniques on other relevant materials under the new projects.
- The project has ended; remaining work is aligned with FC-PAD.
- Proposed “next steps” (rather than “future work,” as the project effectively ended in March 2016) are logical, stemming from the work performed to date. There is some doubt whether simple measures, such as

development of catalysts with "more uniform morphology," are going to be effective in rectifying severe issues facing dealloyed PtNi/C catalysts and possibly other dealloyed catalysts as well. There seems to be little understanding of the causes for poor performance of such catalysts, especially at low RH. Future efforts in the field should concentrate more on understanding, even if less routine testing were to be performed.

Project strengths:

- The project has generated an impressive number of data and helped identify several challenges confronting dealloyed Pt-based ORR catalysts. This should help to better focus future efforts in the field and ultimately result in improved stability of such binary catalysts.
- This is a systematic study with good cross-functional collaboration. The uses of advanced characterization (USAXS, XAS, and TEM) have added visibility to this important challenge.
- The project has good teaming with a technical catalyst manufacturer. Characterization and analysis are excellent. The emphasis on MEA-based testing is a strength.
- The team is strong. Materials are highly developed and highly performing.
- This is an excellent collaboration for performance testing and materials characterization.

Project weaknesses:

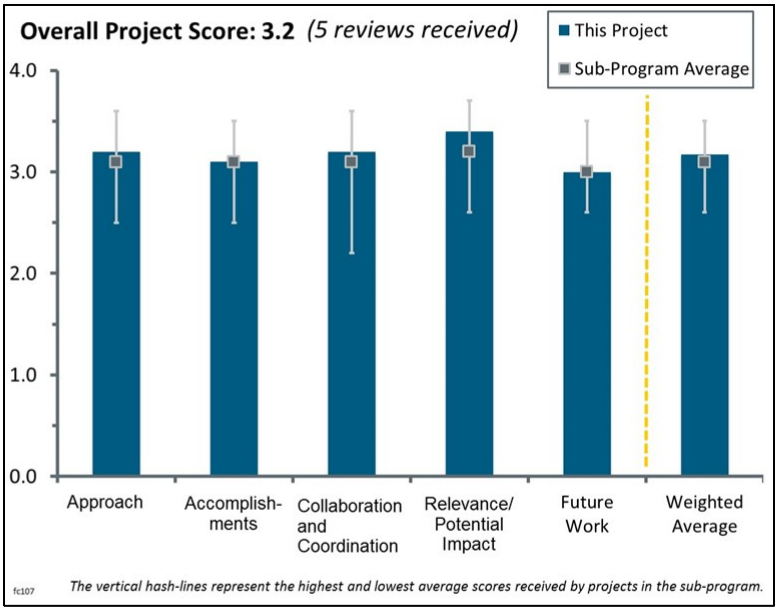
- The MEA testing constraint is a weakness. It is unclear whether the studied materials set is relevant to the state-of-the-art process/materials. Details on the inks and electrodes are not disclosed, making the findings mostly meaningless to anyone else. Material development is limited; this is mostly a characterization project. The project is unlikely to provide any solution.
- Lack of error estimates on some of the summary data makes it hard to validate conclusions, for example, in slide 10. There is no automotive partner for testing and cutting-edge MEA, but this seems to be corrected in the rolling into FC-PAD. Acid washing of electrodes is likely not a viable approach, and in addition, result interpretation is highly questionable because of convoluting effects.
- The main project weakness is relatively little insight into the reasons for observed phenomena, such as losses in the d-PtNi/C cathode upon cycling and poor performance at low RH. At this stage, one would also expect a more definite statement about the feasibility of the d-PtNi catalyst, given its performance limitations identified in the project.
- There are limitations in the knowledge gained from this project. It is more a data-mining activity than one that provides insight into mechanisms or alternative approaches for improved performance. Little was presented on the "rational design" of electrodes.
- The fundamental hypothesis that dealloyed Pt-Ni would be a better catalyst than Pt was probably incorrect.

Recommendations for additions/deletions to project scope:

- The cells used were of triple serpentine flow channels, which are known to have back pressure and other issues. Other designs, such as the Ballard system developed under DOE funding, should be used.
- Research of dealloyed catalysts is worth continuing with greater focus on understanding, knowledge-based interpretation of the test data, and direct feedback into the design of next-generation catalysts.

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

Piotr Zelenay; Los Alamos National Laboratory



Brief Summary of Project:

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

Question 1: Approach to performing the work

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

- The approach is relevant to U.S. Department of Energy (DOE) targets and objectives and directly addresses the barrier of high fuel cell cost by pursuing potentially low-cost catalysts to replace PGM catalysts. The approach addresses the barrier of durability by addressing PGM-free catalyst durability. Los Alamos National Laboratory (LANL) combines a practical approach to improve activity and performance with computational effort and some experimental studies to try to help determine what species are responsible for the ORR activity. The recent shift to increase the level of work on non-Fe PGM-free catalysts is in line with automotive original equipment manufacturer recommendations and concerns about Fe leaching's leading to membrane degradation and short lifetimes. The approach is multifaceted, with effort directed at trying to identify and characterize the active sites, efforts at improving mass transport, and efforts to improve activity through synthesis efforts at what appear to be three different organizations. It is not clear how much the characterization and modeling are influencing the synthetic approaches. Last year's modeling effort seemed to imply a bimetallic site (e.g., Fe_2N_5 or Mn_2N_5 or $CoMnN_5$) would be more active, but the synthetic approaches this past year did not seem to be directed at trying to obtain a bimetallic site.
- The multipronged approach provides significant value, including the focus on the following:
  - Catalyst activity – new synthesis routes with promising improvements in activity, porosity tuning, and good analysis on active site characterization and understanding
  - Durability – reasonable effort on using alternate transition metals to Fe, and evaluation of corrosion and fluoride release effects
  - Membrane electrode assembly (MEA) performance analysis – imaging with a computerized tomography (CT) scanner, which is an excellent approach, along with the linkages to models to optimize structure

  The stated approach to improve water management is important, but although there was good model-based investigation, there was not much evidence of experimental results or model validation in this area.
- The principal investigator's approach is quite effective in addressing all critical barriers in PGM-free catalysts, as evidenced by the publication and presentation record as well as by listed accomplishments in the Annual Merit Review presentation.
- The approach to improving kinetic activity is good. The quality of durability testing has improved, with more attention to higher voltages (e.g., 0.7 V RHE) than were used in earlier work out of Los Alamos National Laboratory. More attention needs to be paid to the engineering of thick (~100 µm) catalyst layers

with good transport properties while operating on air. Intermediate targets may be achievable with thinner layers, but to meet ultimate practical requirements, thick catalyst layers will be needed for non-Pt catalysts in acid. If it proves impossible to get good transport properties in such thick layers, work on this class of non-Pt cathode catalysts should stop. The 2018 target of 0.88 V at 0.044 A/cm^2 (in oxygen and iR-free) sets the bar way too low for practical applications and distorts the research effort away from what is really needed. More attention should be paid to higher current densities in air. Zn evaporation is an ingenious way of introducing porosity. Further data should be shown on whether this porosity improved the high-current-density performance in air.

- Modeling describes why Co- and Mn-based catalysts have somewhat lower activity than Fe-based catalysts. However, it would be hoped that LANL could also add ideas about how to enhance activity of Fe-free catalysts. Some of the catalyst layer modeling points toward a need to achieve at least five times greater active site density to reach 0.5 W/cm^2 at 0.6 V. The modeling is good, but what needs to happen at some point is for this to be translated into a high-current-density target, even if it is defined only within the project. A power density of 0.5 W/cm^2 is still not high enough. A better approach to this project than what has been taken would be to start with high current density in mind, not low. Although high open circuit voltages are important to gauge if the catalyst has any chance at all for application, the next step should be to figure out how to get the active site density that is sufficient for 1 W/cm^2 at 0.6 V. This would likely filter out a number of ideas and focus the project on the best ideas. Furthermore, it would create urgency to generate electrodes with low thickness. Data taken with thick catalyst layers (>70 µm) may not be representative of ohmic losses. It appears that acid leaching the catalyst has helped create greater durability, but this is not clear.

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.

- Good progress has been made, and high-value work has been completed. The catalyst performance activities have met project goals and are on track for the 2018 targets. Good work has been done on the non-Fe catalysts and model exploration to determine the relative activity of Fe vs. Mn, Co, and Ni. Preliminary tests on the effect of the Fe on the membrane degradation mechanism indicate that this may not be an issue. Further elucidation of this result is required to determine the importance of eliminating the Fe from the catalyst. Specific highlights and comments on the project’s accomplishments include:
    - The Zn-induced microporosity work showed an impressive three-times increase in Brunauer–Emmett–Teller surface area.
    - The work to understand Fe atomic dispersion is good.
    - The improvement in durability was good.
    - The work to understand the carbon corrosion rate and fluoride evolution is good, but some of the conditions chosen to be reported are of low relevance. Although the corrosion rate at 0.6 V is similar to Pt/C, the rates are relatively low anyway and appear to be of baseline values, so it is not clear why presentation space was taken up with the bar charts on slide 16. However, the rate increases significantly in the region of concern at 0.9 V and is significantly higher than that of Pt/C.
    - Regarding the fluoride loss, a similar comment can be made. Membrane degradation is typically accelerated under open-circuit voltage conditions, whereas the plot on slide 16 shows not much difference in degradation between this condition and 0.6 V. Further explanation or work is required to understand this result and confirm the relative effects of the catalyst system on degradation. No information was supplied on repeatability of the results or measurement error.
    - The high-angle annular dark-field imaging scanning transmission electron microscopy and electron energy loss spectroscopy work provide the insight into atomic-level FeN$_x$ sites, which is excellent work, and should yield good information for further understanding and development.
    - There has been some reasonable progress in the area of the CT scanner characterization, including analysis of the ionomer density and actual layer morphology. This technique appears to be very powerful.
    - The parametric model studies are useful, and it will be interesting to see whether experimental parametric studies will match model trends. Based on the tornado plot, the hydrophobicity effect

  - seems to be most critical to address for performance. However, this result does not appear to have any validation, and the probability of achieving this performance gain would be low.
  - The work on catalyst-to-solvent ratios provides some nice studies and analysis with useful results that should help to optimize the catalyst layer structure. There is still quite a mix of conditions shown, as well as a variety of membrane thicknesses. There have been improvements in this area, but it has not been completely addressed.
- LANL has made significant progress, increasing the performance of PGM-free catalysts, reaching activity of 0.044 A/cm^2 at 0.87 V, and making good progress toward the DOE 2020 target of 0.9 V at 0.044A/cm^2. LANL has made significant progress increasing performance in oxygen (increasing current density at 0.6 V from ~0.85 A/cm^2 to over 1.2 A/cm^2 in 1 bar O_2) and performance in air, where measurements were rarely made prior to the start of this project. Development of a magnetic separation method to remove Fe particles has been beneficial. The electrode voltage loss study and mass transport studies, including the ionomer loading modeling, have been beneficial
- This project is one of the most successful DOE-funded efforts in terms of novelty, achievements, and accomplishments.
- There has been good progress using side chains and Zn to enhance activity. However, the site density targets are still far from being reached. There is no line-of-sight described to reach active site density targets, and it is still very difficult to see how this will be anywhere close to being part of a commercial vehicle in the next 20 years. Durability of PGM-free catalysts is shown to be similar to Pt/C catalysts, but the reasons are unclear. Durability was shown on the basis of CO_2 and F^- emission rates, but it is not clear what this might mean for surface area, catalyst layer thickness, and the resulting performance. The slides do not make clear which catalyst sample was used for many of the data shown. Modeling work needs to be validated—in particular, the contention that increased hydrophobicity would increase power density needs to be validated through experiments. The same can be said for decreased ionomer tortuosity. Catalyst layer thickness should be reported to understand to what extent mass transfer limitations are playing a role in the data reported.
- The project appears to have achieved a modest increase in kinetic activity toward the self-stated 2018 target. LANL has been far too slow in proceeding to testing in decent-sized (5–50 cm^2) MEAs, even given the delays in implementation of subcontractor contracts. LANL appears to have made some progress in estimating active site densities through comparisons of experiment and model calculations, but the presentation did not explicitly emphasize this point. One of the major problems in PGM-free catalyst research has been the lack of methods to quantify the number of catalytically active sites. LANL has developed new ways to image, if not necessarily to quantify, the apparent FeN_4 active site. Significant improvements in durability at meaningfully high potential (0.7 V RHE) have been achieved by removing iron that is not properly coordinated with nitrogen. The presentation should have paid more attention to whether peroxide was produced during ORR, particularly for non-Fe catalysts.

Question 3: Collaboration and coordination with other institutions

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

- Collaboration between the partners appears to be good. The characterization efforts and modeling efforts appear to be working well together to help inform the electrode preparation and help define loading for MEAs. Some collaborations outside the team are evident, with publications including some authors in the field outside of the current team.
- This project can be used as an example of how collaboration should be organized, coordinated, and executed.
- Los Alamos National Laboratory collaborations with the University of New Mexico, Argonne National Laboratory (ANL), and Oak Ridge National Laboratory (ORNL) appear to have been good in the past year. The presentation did not make it clear whether interactions with other subcontractors have been effective.
- The General Motors (GM) aspect of the collaboration was not clearly shown this year. The same can be said for the University of Waterloo and the University of Rochester. Most recent collaborations appear focused on ORNL and Carnegie Mellon University for characterization of powders and layers. The FeN_4 site found by ORNL is an interesting contribution. Some collaboration between Carnegie Mellon and IRD Fuel Cells (IRD) appeared to help thin out the ionomer and improve performance. It would be interesting to

know more about the University of Buffalo catalyst. It is not clear what the structure of the “Fe-MOF” is, how durability could be improved, or how higher power density (higher active site density) could be generated.

- This is a large project team with what appears to be good coordination between groups.

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 has high relevance and potential to significantly reduce the cost of polymer electrolyte membrane fuel cells (PEMFCs). Replacing PGM catalysts with PGM-free catalysts could be a potential game changer, as the catalyst costs are currently estimated to account for approximately 50% of the stack costs at high volume because of the high cost of the PGM. PGM-free catalysts are a key strategy to meet DOE’s ultimate cost targets. The project also addresses the durability barrier, addressing durability issues with PGM-free catalysts.
- It is unclear whether PGM-free catalysts will have a role in automotive fuel cells, but advances achieved in the last few years are improving the possibility. The durability of the catalysts remains a major concern. The work is relevant, as the goal to minimize Pt from the PEMFC is a worthwhile one.
- From the very beginning, this effort can be considered as cutting-edge research and development of PGM-free materials. Progress made and achievements are striking, and DOE funding is perfectly justified by achieved targets.
- A project on PGM-free catalysts is relevant because precious metals have been shown to contribute a high percentage of cost to conventional PEMFC systems at high production volume. Technical targets that focus on low current density in an equivalent manner to PGM catalysts are appropriate. Throughout a polarization curve, similar power densities should be expected because of cost and vehicle packaging constraints. As with any PGM-free catalyst, durability targets are appropriate. One target is noticeably missing from the relevance slide is performance at high current density. Owing to the need to restrain the expense of membranes, gas diffusion media, plates, and other repeating parts, there should not be an expectation to increase stack active area to accommodate a PGM-free catalyst. Therefore, a high current density performance target is needed.
- This project has been tightly focused on the holy grail of fuel cell research: an effective ORR catalyst that is free of precious metals and is effective in acid. The impact of the project would have been greater had LANL paid adequate attention to the engineering of thick catalyst layers with effective mass transport. The use of Fe in such catalysts has been challenged on the basis of Fenton’s degradation of membranes. Because LANL appeared to have generated catalysts with Fe only in the active FeN_4 sites, it should have addressed the question of whether Fenton degradation of membranes is avoided when Fe is restricted to only those active sites.

Question 5: Proposed future work

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

- The team has laid out the important future work to be addressed, with some promising approaches for activity and durability. Increased focus on understanding the durability mechanism is important.
- This is the last year of the project, which will continue in differently formatted funding. For that reason, the proposed future work, which will rely on full utilization of research capabilities across national laboratories, is a logical continuation in strategy to further advance PGM-free systems.
- The project is complete, except for a few subcontracts. Future work to complete the subcontracts is logical and should accomplish the remaining project milestones and goals.
- The project is almost complete, and therefore, the presentation did not concentrate on details of future work. The listing of remaining challenges and barriers is comprehensive but differs little from a similar list that would have been given at the start of the project. The listing of proposed future work gives little in the

way of specific innovations besides trying to induce strain in bimetallic catalysts as a means of increasing the activity of Fe-free catalysts.

- The project ended in March, with the exception of some tasks for IRD and GM. IRD is delivering MEAs to be tested at GM. Based on the results of the project, it is unlikely the MEA test will reveal a catalyst capable of meeting high-power-density performance required to displace precious metals. LANL would have done well to spend some remaining time attempting to validate the modeling. In particular, any validation of the options that might improve power density would have been worthwhile.

Project strengths:

- This is a large, multi-talented team addressing the work from various angles: modeling and characterization to elucidate the activity and durability mechanisms, use of novel characterization methods to link models to actual structures, and an industrial partner to provide relevant MEA preparation and testing.
- The project demonstrated progress on durability, apparently through avoidance of Fe in forms other than the FeN_4 active site. The project showed some progress on estimation of active site densities through comparison of experiment-based calculations with model calculations.
- LANL is aware of many of the fundamentals of fuel cell testing and in situ diagnostic breakdowns (e.g., high-frequency resistance and limiting currents). LANL has access to many universities, suppliers, and developers for collaboration. LANL has made progress compared to where the technology was five to ten years ago. This does not mean LANL is on a trajectory toward meeting application targets, but it does have some ability to make improved materials. LANL has access to advanced characterization equipment through both universities and national laboratories.
- All aspects of this project have been executed in the most effective manner, as evidenced by the publication list and project goals achieved.
- Over its lifetime, LANL made significant improvements in PGM-free catalyst activity and durability.

Project weaknesses:

- LANL has focused too much on low current density rather than high current density. LANL has focused too much on short-term goals and objectives rather than attempting to think seriously about what a catalyst would need to be to be suitable for an automotive fuel cell stack. There is still too much reliance on Fe for activity. LANL should have attempted to expand upon the activity found with Co and Mn. There is not enough explanation behind why durability has been improved. From the perspective of the presentation, the work may have been empirical in its understanding of durability. Models need to be validated to understand what the future paths are toward improving power density.
- The project paid inadequate attention to the engineering of thick catalyst layers with adequate transport properties. The project demonstrated modest, if any, net kinetic activity gains. The project set too-low intermediate targets that could be achieved without the transport-challenged thick catalyst layers that would be needed for practical applications.
- There is some inconsistency in data and conditions. The project has broad scope, with promising results in a number of areas. Further in-depth studies in each of these areas could have been useful, indicating the resources may be spread too broadly.

Recommendations for additions/deletions to project scope:

- For any succeeding projects, it will be critically important to look at the power density that can be generated with a precious-metal-free catalyst. This project looked perhaps too hard at low current density and not enough at current densities that exist around 0.6 V. Catalyst layer thicknesses should be reported, and they should ideally be minimized. The project was probably not aggressive enough in attempting to reduce catalyst layer thicknesses. High-voltage cycling experiments should be done in addition to steady state holds. Durability of the materials is still largely unproven. Analysis to understand whether hydrogen peroxide is being produced is recommended.
- Any future projects on PGM-free catalysts should keep the nose to the grindstone explicitly in engineering 100-micron-thick electrodes with good transport properties. Because of the low density of active sites in

non-Pt catalysts, high activities per active site would be of little practical utility unless a large thickness of electrode can be effectively utilized.

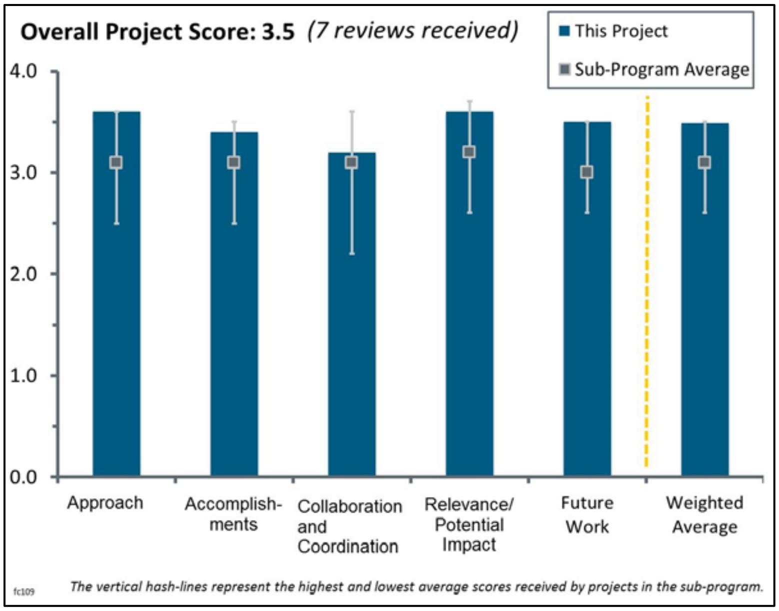
- The team followed last year’s recommendations to run parametric analysis with the models to understand opportunities for catalyst layer optimization. Further validation of some of these effects is required. Much more can be done to understand and achieve catalyst layer optimization.
- The computational efforts should be continued into the Electrocatalysis Consortium (ElectroCat) and should be used more to help drive the synthetic approach and help determine ways to increase active site density.
- Recommendations include a well-justified approach that would tackle both fundamental principles that are behind the mechanism of operation and applied aspects of implementation of PGM-free systems.

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

Michael Yandrasits; 3M

Brief Summary of Project:

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



Question 1: Approach to performing the work

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

- The project is very well conceived and well executed, focused on creating improved fluoroionomers for polymer electrolyte membrane fuel cell (PEMFC) applications. The approach of using monomer units with two or more protonic groups is now well established as a means of reducing ionomer ion exchange capacity while not sacrificing other materials properties. The approach is sound and has been logically and ably pursued.
- The approach to performing the work is excellent. 3M has laid out a clear and cogent plan that aims to simultaneously achieve all DOE membrane targets. By building on the multi-acid side chain approach developed in a previous 3M project, along with incorporation of robust supports and radical scavenging additives, 3M has met most targets.
- The project is aligned with DOE targets and goals. The project approach utilizing multi-acid side chains allows one to decrease equivalent weight (EW) while maintaining more mechanical strength than standard perfluorosulfonic acid (PFSA) materials with the same EW. 3M is focused on meeting all the targets at the same time.
- The approach is good. The project is investigating new lower EW PFSA materials. Performance appears to show good improvements.
- This project has an excellent approach that is making polymer electrolyte membrane (PEM) materials that have not been synthesized elsewhere.
- The approach is excellent for developing a low-relative-humidity (RH), high-temperature membrane. However, the ionomer in the catalyst layer should be addressed in parallel.
- The approach might well be better than the score, but these types of presentations are the hardest to judge; so much of the data is “Ionomer A, Support B, Additive C,” etc., and the reviewer really has no method of judging. The approach is incrementally improving the best PFSAs, which is definitely worth pursuing. It would be good to see more fundamental work on whether the nitrogen linkage in the perfluoroimide acid (PFIA) is viable. The project has some data that show it may not be and may poison the catalysts. This should be demonstrated as soon as possible; if it is not, further incremental improvements are a waste of time.

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.

- Accomplishments so far are excellent and show increased progress over the previous year.
- The project continues to make excellent progress. The principle achievement in the last year was the successful completion of milestone 8, which yielded a membrane that meets most of the DOE targets. The milestone 8 membrane could not quite meet the most aggressive area-specific resistance (ASR) target at 120°C, but 3M has done an excellent job of mapping out a path forward to meet this target, with projections showing that further reduction in ionomer EW could yield membranes of 10- to 14-micron thickness that meet all ASR targets. Notably, while 3M has done an excellent job of attacking this target, automotive original equipment manufacturers (OEMs) have indicated that high performance at 120°C is not strictly necessary because most OEMs would be reasonably satisfied with high performance at 95°C. A bigger concern is the possible durability impacts of the 3M membrane. While the membrane itself has successfully achieved DOE durability targets, membrane degradation products could negatively affect catalyst and electrode durability. These degradation products appear to play a role in the low observed open circuit voltage, which is probably due to adsorption of degradation products on the catalyst surface. As discussed in the presentation, the existence of multiple acid groups per side chain may make adsorption of degradation products from the 3M membrane a bigger issue than with conventional PFSAs. Going forward, this is an issue that 3M will need to address more thoroughly, but based on limited results reported so far, it appears that 3M has some ideas of how to attack the problem. The stack testing component of the project is not necessary at this stage. From a membrane development standpoint, it is not clear what stack testing would reveal that cannot be revealed in single-cell testing. Given the known issues with membrane degradation products, it would have made more sense to skip the stack testing and devote more resources to ionomer and membrane development. Furthermore, stack testing results will be strongly dependent on MEA performance, and little work has been done on MEA integration to date.
- The team has consistently met its marks in terms of project milestones and has generated useful insights into the advantages and limitations of its approach. The systematic approach to simultaneously increasing ion exchange capacity and reducing film thickness has led to incremental but impressive improvements in membrane properties. The project came up a little short on oxygen permeability and on ASR under the most aggressive conditions of 120°C and 40 kPa water vapor pressure (20% RH), but other than this, the team has provided excellent advances.
- 3M has made significant progress toward meeting the DOE membrane targets simultaneously. Most of the targets have been met, except for conductivity at 120°C and low RH. Recent work on supported membranes has allowed reduction in thickness to 10 microns, which improves water management and reduces ASR. The fiber distribution work should have a durability component (at least RH cycling) to show the effect of fiber distribution on mechanical durability. The fiber diameter study is interesting, but the larger-diameter fiber sample also had a higher fiber content (40% higher), which could account for the differences. This study needs to be repeated with more samples and closer control of fiber content.
- There has been very strong incremental improvement on the best PFSAs. There is a very systematic, well-thought-out approach on what it will take to meet targets using these systems. Some results, notably water uptake, were clearly just wrong, and the presenter seemed to be aware of this and should have looked more deeply.
- This project has promising results on a new membrane material. There are some questions about the relative stability of the membrane and the effect that the material has on catalyst activity that should be addressed quickly.
- It appears much of the progress toward the targets is a result of thinning the membrane from 14 microns to 10 microns. Thus it is not clear how much of the improvement in performance is due to a fundamentally improved material or simply the same-performing material at a thinner thickness.

Question 3: Collaboration and coordination with other institutions

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

- Project coordination with a key stakeholder OEM (General Motors [GM]) and a university providing novel nanofiber support structures is excellent and gives good guidance to the project.
- Collaboration with the university and GM are good. It appears GM is being used simply to test 3M materials and check whether the materials meet targets.
- The collaborations with Vanderbilt University and GM appear to be going well. GM durability testing is ongoing, and stack testing with the 3M membrane has been initiated.
- 3M really does not need to collaborate; the team is fully capable of characterizing, developing, and taking these products to market. It is really important to know what the different supports are in order to judge them against each other.
- The collaborations with GM and Vanderbilt add value to the project. GM's role in performing advanced durability studies, including peroxide vapor degradation and blister strength tests, is particularly valuable. The Vanderbilt nanofiber support studies are valuable in terms of improving understanding of support properties, but despite some interesting and informative results from Vanderbilt, it is not clear to what extent these results are actually feeding into 3M's nanofiber development.
- The collaboration between 3M and Vanderbilt appears to be valuable. However, the primary development rests solely on 3M, with GM conducting primarily validation and Vanderbilt working on nanofiber development. It seems like they have not been widely incorporated into the project.
- The work with Vanderbilt seems like an add-on; it was not clear what the work added or whether it was necessary. The role of fibers in controlling swelling and providing mechanical strength was clear, but it was not clear which fiber approach was best, or whether it mattered what fibers were used or how the fibers were used. Mention was made of polyvinylidene fluoride in fibers, which could create problems with peroxide stability—problems that were not addressed.

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

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

- The project is highly relevant to the Hydrogen and Fuel Cells Program (the Program) goals and has potential to have a high impact. The 3M membranes have the best chance of providing suitable high-temperature (95°C–120°C) performance of any membrane currently under development. The 3M project also provides DOE with the most likely path to meet the aggressive ASR targets without sacrificing durability and mechanical properties. The biggest unknown in terms of the potential impact at this stage is how the membrane will impact MEA durability. 3M will need to demonstrate that the membrane can be incorporated into a MEA that meets MEA durability targets, but this work is beyond the scope of the current project.
- The project is highly relevant to DOE and to fuel cell development. Membranes with better performance at low RH can reduce system costs by eliminating the need for humidifiers and increasing power density. Membrane costs are a significant portion of stack cost at low production volumes.
- This project has consistently provided new materials that meet or exceed DOE/FCTO milestones. The team has been generous in providing materials to other DOE-supported workers. The project has had high impact.
- Membrane developments in this project are showing steady progress toward the DOE targets.
- This is an important and relevant project.
- An improved membrane and ionomer that works at low RH and high temperature would be enabling for the commercialization of automotive PEMFCs. However, the proprietary nature of the materials and additives is of great concern, especially as the project is using taxpayer funds.
- These are incremental tweaks and improvements. Having said that, they are improvements on the best PFSAs we have, so the work is definitely worthwhile. The project team members do not present a convincing case as to why their supports are needed/better compared to expanded polytetrafluoroethylene

(ePTFE). Ultimately, the team needs to address costs, especially if the team members think that is an advantage.

Question 5: Proposed future work

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

- Stack testing plans seem well planned and likely to succeed.
- Ionomer and nanofiber development are excellent next steps in addition to stack testing.
- The proposed future work represents a logical path forward. Some areas that should have been included on the future work slide are the further development of lower-EW ionomers with three or more acid groups per side chain, and studies of the ionomer degradation products. Both these issues were mentioned during the presentation, but it is not clear how much they will be addressed in the remainder of the project.
- The future work seems relatively straightforward; however, 3M could get more from its partners (especially GM) and also leverage others to help with understanding the material properties better.
- The work is very difficult to judge because so many of the materials are a “black box.” The project is nearly done with project goals, however, and progress has been very good.
- The future fiber distribution work should have a durability component (at least RH cycling) to see the effect of fiber distribution on mechanical durability. The peroxide vapor chemical stability study should look into the membrane degradation route to determine the level of chemical suppression needed. PFIA side chain fragments are likely catalyst poisons. The degradation mechanism also has implications for the applicability of perfluoroionene chain extended 3 (PFICE 3) or PFICE 4 derivatives.
- When going to membranes as thin as 10 microns, more focus on membrane lifetime should be undertaken. It is not clear whether a “rainbow” stack is the best platform to study the lifetime of different membranes.

Project strengths:

- This is the best membrane project that the Program has funded to date. The project was thoughtfully designed and has an excellent chance of meeting most DOE membrane targets. While the most aggressive ASR targets probably will not be met, there is a clear path forward to meet them in future work.
- The approach is valid and novel in the area, and results seem to indicate that there is promise for the material. The strengths of this project seem to be in the synthesis of the materials.
- The path, metrics, and methods are very clear. All the right tests are being done with the proper controls and targets.
- 3M has strong polymer background, GM has testing capability, and Vanderbilt has expertise in electrospun fibers.
- The project has excellent materials, has a good team, and has made excellent progress.
- The project has an excellent team, which, with good coordination, leads to quantifiable progress toward DOE goals.

Project weaknesses:

- The PFIA membrane degradation story is as yet incomplete. It is difficult to understand how valuable this membrane/ionomer is or could be without a better understanding of the effect that it seems to be having on the oxygen reduction reaction (ORR). It is not clear whether it is three times that of Nafion (because it has three times the SO_x groups) or whether something else is going on. Advanced characterization of the membranes—such as determining membrane crystallinity through small-angle x-ray scattering, wide-angle x-ray scattering, or other techniques—is missing to date and could potentially provide valuable information.
- Simply relying on reducing membrane thickness to reach the DOE goals is a risky approach.
- The proprietary nature of materials and additives renders the work not so useful to the U.S. taxpayer.
- The project needs a stronger argument on the support side. The advantages in performance or ultimate cost should be made clear.
- The stack testing task adds little value to the project at this stage.

Recommendations for additions/deletions to project scope:

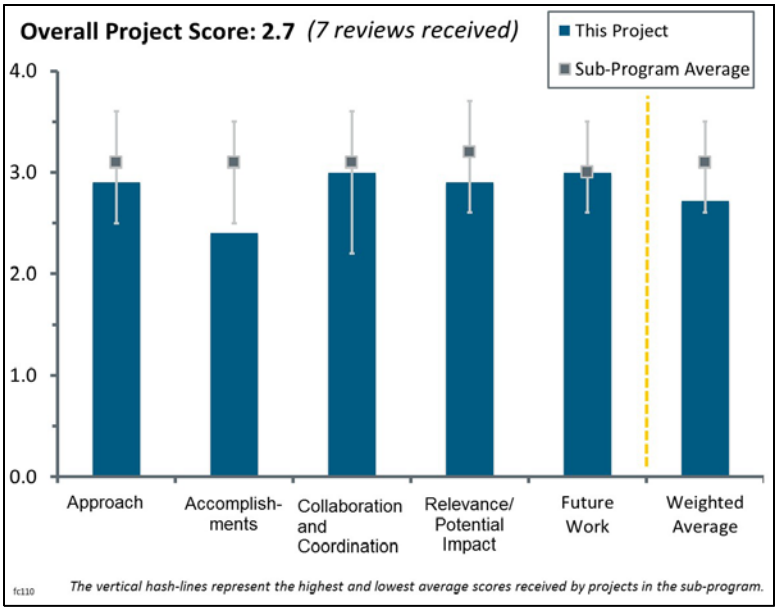
- This year, 3M has addressed the question about the effect on catalyst activity, which is good; however, questions remain. There are multiple paths that 3M could pursue to help understand whether this quick open circuit voltage decay is a problem. Suggestions include providing some of the PFIA as an ionomer to conduct rotating disk electrode evaluation and comparing this to Nafion's effect on reduction in ORR kinetics (e.g., the National Renewable Energy Laboratory). PFIA could be provided to an organization that regularly conducts membrane accelerated stress tests, including degradation fragment analysis such as F and SO_x with water analysis and H_2 crossover with time (e.g., Los Alamos National Laboratory).
- The project should do more work on membrane lifetime in the 10-micron thickness range. It is not clear whether this is a feasible membrane thickness.
- The concentration has to be on the viability of the PFIA side chain. This needs to be cleared up before more work is done; it is the most important thing to demonstrate for acceptance of these materials.
- The project is scheduled to end in December, so changes to scope should not be made at this point.

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 Ω 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.



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Question 1: Approach to performing the work

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

- The principal investigator (PI) is testing multiple approaches to incorporating heteropoly acid (HPA) protogenic groups into membranes for polymer electrolyte membrane fuel cell (PEMFC) applications. This is a long-standing line of investigation from this PI. Most work from the past year focused on adding aryl phosphonate groups onto fluoroelastomer polymers via an unusual coupling reaction between phenol groups and a specific carbon in the vinylidene difluoride-hexafluoropropylene (VDF–HFP) linkage in the fluoroelastomer. This reaction has previously been used to provide crosslinking in fluorelastomers. HPA groups have multiple attractive properties, including high conductivity at low water activity and an ability to mitigate chemical damage from peroxide. The approach of attaching HPA groups to fluoroelastomers seems sensible, though the fluoroelastomer stability may need to be studied to understand how it compares to other materials in response to fuel-cell-like challenges.
- The multi-directional approaches taken by the team for the completion of all tasks are adequate. All the analytical techniques have been thought through appropriately. The study on material synthesis based on functionalized superacidic inorganic moieties is the personable approach. Stabilization of HPA in polymer matrix may be a great challenge. However, learning from first-generation membrane work seems to have helped the team in tackling the stability issue.
- Barriers are clearly addressed, with new performing polymers that show progress over state-of-the-art perfluorosulfonic acid (PFSA) ionomers.
- The approach of using anchored HPAs in a polymeric matrix has the potential for high conductivity under low RH conditions and could overcome barriers to high conductivity under high-temperature, low-humidity conditions. High-conductivity membranes can help overcome cost barriers by enabling simplification of balance of plant. Leaching tests still show loss of HPA (reduction from 70 wt.% to 60 wt.%). More needs to be done to determine whether additional leaching occurs, whether leaching is controlled by the equilibrium concentration in the leachate, or whether it is controlled by the fraction of doubly attached or singly attached HPA. ASR measurements should be measured directly through plane rather than calculated from in-plane conductivity. It is not clear why ASR is decreasing and then increasing with increasing temperature at 95% RH for some samples.

- Using polymers functionalized with HPAs is a promising approach to achieve high conductivity at low RH. There does not seem to be a systematic approach to meeting the requirements especially to reduce swelling and improve mechanical stability.
- The focus of this work has been on improving synthetic techniques to achieve DOE ASR targets with HPA-based membranes. However, given how much work the PI has already performed on HPA-based membranes without demonstration of a membrane that can actually be incorporated into an MEA, this approach does not seem promising.
- This group has been working on this approach for a very long time, and the members' thoughts and direction are not yet mature; they appear to still be stumbling through the dark to find something that will work. Each year, there are unsubstantiated, last-minute results with incomplete thoughts behind them. At this point, the polymer and HPA to be used should be clear and the team members should be perfecting and iterating. The fact that they are considering a polymer that splits on its own when thin demonstrates lack of background data; if the polymer can drive itself apart while drying, the source of hope to survive RH cycling is unclear. Beyond mechanical stability, the team does not give a very good justification for why the base, hydrocarbon-based polymer might survive chemically, other than that the HPA acts as a radical scavenger. If the team members have shown this, they did not show it here. Again, this is an extremely easy thing to demonstrate with traditional membranes.

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

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

- Recent progress with the HPA/FC-2178 system is excellent. The synthesis is refined and is yielding large amounts of high-quality material in good yield. HPA-loaded materials lose some HPA upon exposure to boiling water but retain much more and show high conductivity and low ASR even at 50% RH. The discovery that thin films have low ASR due to cracks is significant insofar as the crack problem may be relatively easy to fix by switching to a supported membrane, e.g., an expanded polytetrafluoroethylene (ePTFE), and/or by blending. An area that needs more attention is the film durability; e.g., the stability of FC-2178 upon challenge by peroxide before and after phenol addition and before and after HPA addition is not established and needs to be rigorously tested. Mechanical testing will also be important at some point.
- There has been good recent progress on polymer synthesis/membrane preparation, but overall progress has been delayed. Recent experiments have shown high conductivity and achieved the intermediate target.
- This project makes and characterizes new membranes that show improvements.
- It is understandable that the team could not achieve much this year because of the mishap that occurred in the laboratory. Despite this, the team seems to be on target to accomplish the project's 2016 goals. The HPA seems not to be very stable in the polymer matrix. In slide 12, the author mentioned that with improved processing (cross-linking), the loss of HPA is decreasing and is presently at about 60%. This means the HPA is not completely stabilized in the polymer matrix. The swelling property of the membrane is also not very good. From slide 14, it seems that there is still room for improvement in the membrane's swelling property by further cross-linking. The team needs to focus on the membrane's optimization to the extent that it gives desirable conductivity. It is not clear why the team chose to compare the project's conductivity data with N117, which is much thicker and does not respond to humidity change quickly. The team should use a standard 2-mil Nafion® membrane for conductivity comparison.
- Progress to date has been limited. Some promising results were reported in terms of possible low ASR from 70°C–110°C at 50% RH, but the questionable approach to ASR testing undermines these results. Colorado School of Mines (CSM) needs to test ASR in a through-plane setup. Calculation of ASR using in-plane conductivity is inappropriate, which was clearly demonstrated in the project's own results in which the existence of cracks resulted in an unrealistically high ASR estimate at 50% RH. Comparisons with N117 were made on several slides, but such a thick membrane is not relevant to the Hydrogen and Fuel Cells Program (the Program). A more relevant membrane with a thickness of 25 microns or less should be used. Properties such as chemical and mechanical durability and gas crossover have not been robustly addressed to date (aside from the Fenton testing), and they cannot really be addressed until CSM produces a membrane that can be incorporated into an MEA. Most of the HPA appears to be water-stable at 60°C, but the membrane HPA's ability to be retained in an operating MEA will need to be demonstrated. CSM

indicates that a viable membrane will be produced soon, but the project does not have a good history of living up to claims of future performance.
- Some progress has been made toward meeting the conductivity target. The conductivity data are inconsistent. After much effort, the project has been able to make films without cracks, but it does not leave much encouragement that the team will be able to make mechanically stable films with this chemistry. There are no ASR data; reported ASRs were calculated from in-plane conductivity measurements. There are no conductivity data at the targeted temperature of 120°C. There are no chemical or mechanical durability data.
- Despite the project being in its third year in the Program and ~10 years on these systems, the project team members are not even close to MEA testing let alone RH cycling, chemical stability testing, or open-circuit voltage testing. All they have done is some conductivity testing, and the presenter admitted that they do this poorly (bad ovens, not reaching temperature, etc.), and it is done only in-plane. The thought process on HPA containment is either not mature or was not well presented.

Question 3: Collaboration and coordination with other institutions

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

- The team consists of a good mix of national laboratory and industrial partners. Collaboration with 3M, Nissan, and the National Renewable Energy Laboratory (NREL) will be beneficial to the team.
- So far, this project is an excellent collaboration between CSM, 3M, and Steve Hamrock, formerly of 3M. Other partners are less involved at this stage.
- 3M seems to be providing valuable support with polymer synthesis and film formation. Neither NREL nor Nissan appears to have contributed to the project as of yet.
- There is good collaboration with 3M and a good approach to base work on available polymers.
- Collaboration has been limited to date because acceptable membranes were not being made. Now that a method for preparing acceptable membranes has been found, testing at the partner organizations can be initiated.
- Participation by 3M has been valuable in providing materials and in consulting on synthetic technique. NREL and Nissan involvement to date has been limited by the lack of a membrane with which they can work. Plans for their involvement going forward appear reasonable, though.
- Work with Dr. Hamrock is very good and should steer things for the better; however, it is uncertain how this technology will be eventually transferred or brought to market. Other partners are appropriate, but it was not made clear, for instance, what Nissan’s role is.

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

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

- This project provides a unique approach to solving the problem of poor membrane ionic conductivity at low water activity. The behavior of HPA proton donors is unique and quite different from that of organic acids, which are the usual protogenic groups in polymers. The quite high conductivities and low ASRs from these materials at low RH are difficult to achieve from other materials, and as such, the materials from this project offer much to forward the goals of DOE and the Fuel Cell Technologies Office (FCTO) in hydrogen energy conversion. There is also a high potential for producing materials at low cost.
- The project is relevant to the objectives of the FCTO Multi-Year Research, Development, and Demonstration Plan (MYRDDP). The activities are aligned with DOE’s goal to address the commercial barriers such as performance, cost, and durability. The focus of the project is to demonstrate a low-cost hybrid inorganic/polymer from superacidic inorganic functionalized monomers. This is an alternative low-cost approach to develop a low-RH (inlet <50%), high-temperature membrane (95°C–120ºC).
- The project supports the MYRDDP and advances progress toward DOE goals and objectives. Membranes that can operate under low-RH conditions and at high temperature can reduce system costs and improve system performance.

- The project has the ability to improve the state-of-the-art membrane performance; the project is well aligned with DOE program goals.
- Developing durable, high-performing membranes is highly relevant to the DOE program goals and objectives. The project did not receive a 4.0 because cost considerations are not included.
- The relevance of this work to the Program is questionable. Reasonably robust and durable membrane technologies are already currently available, and other DOE projects have further advanced the state of commercial or near-commercial technology. The technical maturity of the ionomers being developed in this project is so low that it is difficult to predict whether they will ever be commercially relevant at this stage, but the amount of work already performed in this project and previous projects, with little progress to show for it, suggests that the HPA-based approach to providing membrane conductivity is not likely to make it into real fuel cells in the foreseeable future.
- It is extremely hard to see how this project can be turned around in the last year with over half of all funding spent. The team does not yet have a viable membrane. Worse is that at this point there is still no evidence that HPA is a good and viable approach. Dr. Herring did an exceptional job years ago educating the community about the possibility of these materials, but we still do not know how to utilize these materials or whether their conductivity enhances the ionomer in which they are imbedded.

Question 5: Proposed future work

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

- Studies on material and membrane durability are needed. Future work with supported membranes is planned and is needed. The prior observations of membrane cracks suggest that even with supported membranes, mechanical durability could be an issue (e.g., in response to an RH cycling test). Such tests should be pursued.
- The proposed future research is aligned with the goals of the project; however, there is a significant amount of work to be done. The team should get some extra time because of the time lost this year due to the mishap in the laboratory.
- The future work is directed at characterizing the other relevant membrane properties. Work has begun on incorporating support materials to improve mechanical properties. Future conductivity measurements should extend to 120°C and to lower water content (water pressure [pH_2O] down to 40 kPa).
- The project is still near the very beginning of a polymer program, having just decided on the base materials. The team recognizes the shortcomings and has shown the two most important things, HPA retention and conductivity. There is a good deal of work just to get to mechanical viability—the project will look at supports and blends—but this is just an idea at this time. Demonstrating through-plane conductivity, chemical durability, and mechanical durability needs to be the focus.
- While CSM has done a good job of reporting on its synthetic method development thus far, future work in this area was not adequately described. The future work for the current year includes extensive testing at NREL, 3M, and Nissan, but it is still not clear that CSM will actually be able to provide a membrane for them to test.
- Scale-up and electrode optimization should not be done until there is some proof of durability using the DOE accelerated stress tests. The primary focus should be mechanical stability.
- More work is needed on demonstrating key improvements of the new polymer over the incumbent PFSA polymer—in particular on cell lifetime and cost.

Project strengths:

- The team is composed of respectable research organizations with adequate expertise. Overall, the team is equipped with the necessary knowledge base, resources, and industry/academia/national laboratory mix that is required for the success of this project.
- The HPA membranes made to date have had very high conductivity. HPA membranes should have benefits for chemical stability, as HPAs have been added as a chemical stabilizing agent for PFSA membranes.
- HPAs bound to polymers is a promising approach to meet membrane performance and durability targets.
- The strength is the unique properties of HPA protogenic groups.

- There has been good progress on developing new materials that are converted to membranes that can be characterized.
- Chasing the potential of HPA in ionomers is still an interesting endeavor.

Project weaknesses:

- More focus is needed to prepare a polymer HPA composite in which the HPA is permanently bound to the polymer groups. The team should make thinner membranes with the desired conductivity. The conductivity of the HPA/polymer membrane seems to vary with membrane thickness.
- CSM has been pursuing a similar approach for years now with little success. There is little reason to think the project will be successful at this point and even if a membrane for testing is produced within this project, it seems unlikely that the membrane will make it into commercial fuel cell products in the foreseeable future.
- There is a lack of focus and direction. Progress to date is very poor. The project is still searching for its polymer system. The project team needs to better understand the nature of the conductivity of these systems through a clear design of experiments in which investigators vary the HPA loading and break down conductivity to the ionomer and the HPA.
- There is no proof the membranes can be made mechanically or hydrolytically stable. There is very limited data—primarily just some fixed RH conductivity data. It was nice to see repeat measurements, but the data is highly variable.
- There is no fuel cell data and no indication of near-term or long-term cost benefits over PFSA.
- Membrane mechanical properties and potentially membrane durability are weaknesses.
- Project progress has been slow to date. The properties of this new class of membranes are not known.

Recommendations for additions/deletions to project scope:

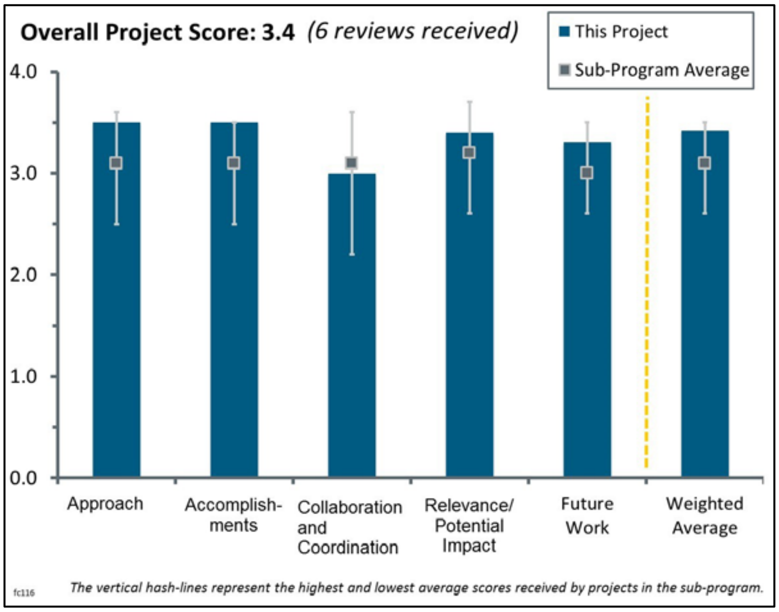
- ASR measurements should be conducted. The project should dig into the root cause of the weight loss after washing; it is unclear whether the weight loss was due to non-bonded HPA or whether bonds were broken. The project should confirm whether weight loss is due to HPA. The temperature and time dependence of the mass loss are unknown as is the impact of annealing on conductivity. The project should measure mechanical properties (e.g., through tensile tests).
- The focus should be on showing viability of these systems and doing a fundamental study so the HPA approach can be judged. A systematic loading study should be done with believable RH conductivity testing in a robust system. The project needs to be able to measure conductivity well, including through-plane. A systematic study of HPA loading with water uptake and conductivity as a function of temperature and RH needs to be done.
- Less work should be done on new polymer investigation. More work should be done on converting existing project polymers into membranes that can be demonstrated in cells. There should be more demonstration of costs and a go/no-go analysis. New materials offer no benefit to Program goals if manufacturing costs are a non-starter.
- The project should conduct more studies on membrane durability (e.g., in response to challenges from peroxide and to RH cycling).
- The electrode development portion of the future work should be deemphasized or removed. Given the low maturity of the membrane technology, any MEA testing performed should be done with standard electrodes or even platinum black electrodes so that the focus remains on the membrane.

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

Chao-yi Yuh; FuelCell Energy, Inc.

Brief Summary of Project:

The overall objective of this project is to develop an innovative, durable, molten carbonate fuel cell (MCFC) electrolyte matrix (Smart Matrix) to enable >420 kW rated stack power and 10-year (80,000-hour) stack service life. Compared to current-generation MCFC commercial technology, these correlate to a >20% increase in cell power density and ~100% increase in stack service life. The objectives for the current project year are to scale up manufacturing of the Smart Matrix and prepare for stack evaluation.



Question 1: Approach to performing the work

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

- The FuelCell Energy (FCE) team has done an excellent job in planning a promising route to achieve U.S. Department of Energy (DOE) durability targets for stationary fuel cell durability. The approach addresses all aspects of molten carbonate fuel cell (MCFC) matrix performance and durability and, given the central role of the matrix in this technology, the project appears likely to provide significant improvements in overall MCFC performance and durability.
- The project’s approach is very clear and perfectly addresses barriers A and B. The project is very focused and well-structured. The timeline set out is very logical and well-thought-out.
- The approach is well-structured, -organized, and -conceived. It has milestones and go/no-go decision points.
- The approach based on accelerated testing to evaluate matrix stability is excellent.
- FCE methodically develops an understanding of degradation mechanisms and moves on to defining and verifying mitigation approaches. Accelerated testing is part of the approach to defining and mitigating degradation processes, as is out-of-cell analyses and testing. Acceleration parameters need more discussion in terms of justification and in their not creating different degradation mechanisms. Coarsening studies without electrolytes may underestimate degradation severity. More electrochemical screening could be informative.
- The approach is generally sound. Accelerated aging tests are necessary for improving durability of MCFCs. The accelerated aging studies strongly support the hypothesis that the CO_2 partial pressure has a major effect on the aging of the $LiALO_2$. It would be very useful to determine that the presence of oxide ions accelerates the decomposition of the lithium aluminate. This could be determined by electrochemical measurements of the oxide ion activity within the melt as a function of the CO_2 partial pressure. An yttria-stabilized zirconia (YSZ) probe may be used for these determinations. Overall, the research approach has contributed to improving the lifetime and durability of MCFCs.

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

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

- The FCE project has made excellent progress in the last year. The principle achievement this year was the demonstration of 80,000-hour durability (projected) through accelerated testing procedures. By reporting the accelerating conditions (i.e., higher temperature, humidity, and fuel utilization), and by showing the correlation between degradation metrics for single-cell accelerated testing and real-world stack operation, FCE has provided ample evidence for the validity of their accelerated testing and the accuracy of their durability projections. Other significant progress this year includes the demonstration of more than a 40% increase in matrix snap strength and improvement in Ohmic resistance relative to the baseline matrix.
- The accomplishments shown as a result of the accelerated testing are impressive. Understanding of the degradation mechanisms has been improved. Material stability of the new Smart Matrix has been confirmed. What remains is to show that the Smart Matrix can, indeed, be the main life-extending factor, and that can only be done by long-term stack testing.
- FCE has made substantial progress and is preparing a stack test to verify durability improvements; technical milestones have been met so far.
- The project is on track in achieving its milestones. The status is well-identified.
- The project team has achieved all the milestones as planned.
- The research team demonstrated substantial improvement in the durability of the electrolyte separator and met the 5,000-hour test milestone. The team predicts an 80,000-hour lifetime; however, the slope of the degradation curve is so flat that the errors in this estimate have to be very large. The average pore size was reduced, as previously presented, and the particle size distribution was significantly narrowed. The narrowing of the particle size distribution from the mean value is known to decrease the ripening of the ceramic particles.

Question 3: Collaboration and coordination with other institutions

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

- There are very few MCFC developers/experts worldwide. FCE collaborates with key experienced players such as the Illinois Institute of Technology (IIT) and the University of Connecticut (UConn). FCE works closely with raw materials suppliers.
- FCE collaborates well with subcontractors UConn and IIT to develop fundamental understandings of matrix coarsening and wettability and to help design mitigation approaches.
- Project strengths include the collaborations with UConn and IIT.
- Collaboration between partners is good, and there is good complementarity between them. However, there is no evident interaction or collaborations outside the sphere of the project in terms of knowledge exchange and dissemination activities.
- The collaboration with UConn is satisfactory; however, better engagement of the university resources would foster a better scientific understanding of the degradation mechanisms. Perhaps involvement with other universities with experience in high-temperature molten salt electrochemistry would be beneficial.
- Most of the work is being performed by FCE, but mechanistic studies of coarsening mechanisms at UConn and matrix wetting at IIT are contributing to the matrix design. Since much of the work is of a proprietary nature, this task distribution makes sense.

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

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

- The project is highly relevant to the Hydrogen and Fuel Cells Program's efforts on stationary fuel cells representing the most promising route to achieve stationary fuel cell performance and durability targets.

FCE is a leader in fuel cell commercialization and has produced and deployed more fuel cells (on a megawatt basis) than any other U.S. company, but further technical improvements in FCE's MCFC technology could broaden the market and accelerate fuel cell commercialization, so the impact of the proposed work could be significant.

- The relevance is good. MCFC technology is relatively mature; however, increased lifetime would substantially reduce cost of ownership. The potential impact is also good in that it facilitates the entry of fuel cell power sources for stationary generation.
- Success by FCE would enable meeting the durability/life target for stationary combined heat and power (CHP) and distributed generation (DG) fuel cell systems. Ownership costs (e.g., cost of electricity [COE] payback period) would be greatly reduced.
- The impact the project can have with the increase of lifetime is substantial. The fact that FCE is leading the project and is one of the industry leaders ensures that the impact will have an immediate effect on the fuel cell industry.
- The project aims at improving life and cost of MCFCs, thus fully supporting FCTO's research, development, and demonstration objectives.
- Doubling stack life to 10 years can result in a substantial COE reduction.

Question 5: Proposed future work

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

- Proposed future work is good. The new electrolyte separator technologies need to be scaled up and incorporated into full-sized power stacks.
- Future work logically follows progress to date. Parameters and processing conditions will be optimized and scaled up. A stack will be built from the new matrix and tested for 5,000 hours.
- Appropriate future work has been proposed.
- The future work is appropriate, but more detail on the future tasks would be helpful. Also, all future tasks appear to be FCE tasks, and it is not clear whether the university partners will still have a role going forward.
- Controlled release of Smart Matrix in direct fuel cell (DFC) products is planned to enhance DFC market penetration and clean energy job creation, enable a cost-effective distributed hydrogen-production DFC-H_2 system, and enable DFC-CO_2 capture for reducing CO_2 emissions.
- The proposed future is absolutely necessary for the validation of the results and to make sure that the barriers are addressed. The additional 30 kW technology stack endurance testing that will extend beyond this project is very valuable. It is very good to see that, outside this project, FCE is focusing on other life-limiting factors apart from the Smart Matrix. More details could have been provided on the plans to improve the manufacturing process and yield and on the cost-reduction approach.

Project strengths:

- The focus is very clear, excellently combining the expertise of the industrial and academic partners. If successful, the project will have an immediate impact on the fuel cell industry, as the results will be integrated in FCE products.
- The milestones for this project year were clearly met. The 5,000-hour test was completed with excellent results. The research should result in a substantial improvement in the durability of MCFCs.
- Approaches to evaluate the proposed Smart Matrix are excellent. Future work should focus on obtaining a better understanding of degradation mechanisms and proposed solutions.
- The project has met all milestones and succeeded in producing a new matrix that will enable improved performance and durability relative to the baseline.
- The project has a strong industrial participant.
- The project has a methodical approach.

Project weaknesses:

- While FCE has clearly achieved major improvements in the matrix design, the technical details of how these improvements have been achieved are mostly unknown. This may be unavoidable given the proprietary nature of the research, but FCE should give more detail about the materials and processes used, where possible.
- Some details on key technical work were not described, e.g., how to estimate life based on accelerated data. There were no details given on partner work, especially the work at UConn.
- Input from customers/end users would lend credibility to the project.
- The degradation model may be insufficient for the prediction of the cell lifetime. The x-ray photoelectron spectroscopy data show a decrease in sodium concentration within the matrix after testing. The impact of this loss was not discussed during the presentation. The project would benefit from high-temperature thermochemical and electrochemical data collection and analysis. This would provide a better understanding of the degradation mechanisms at play.
- In terms of publications and presentations, there are only four outside the sphere of DOE; more would have been expected, given the positive results.

Recommendations for additions/deletions to project scope:

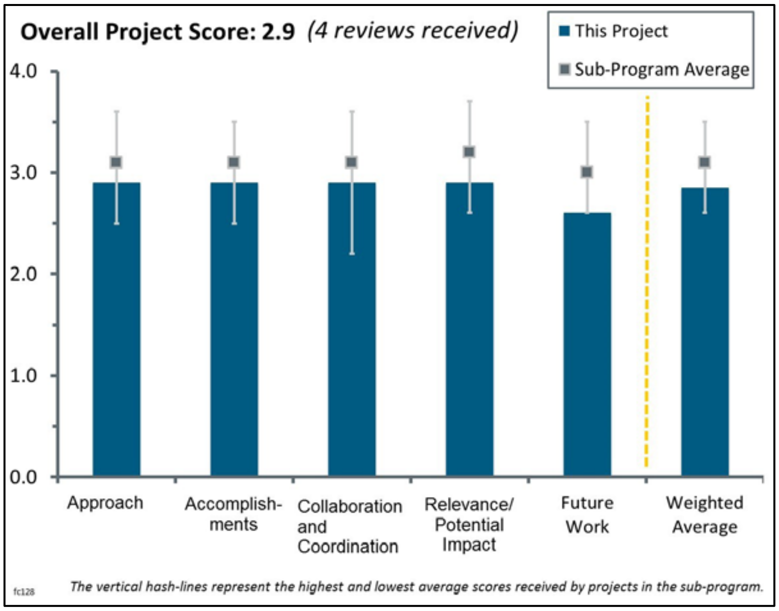
- More information should be provided in terms of the planned cost reductions. Manufacturing improvements could be described in more detail (e.g., present yield and how will it be improved).
- The project should add high-temperature electrochemical measurements of oxide ion activity as a function of CO_2 and water vapor pressure.

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 (PBI and TPS), and optimize structures and reaction conditions.



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Question 1: Approach to performing the work

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

- The project started with very strong preliminary data, and the proposed approach to use high temperature membranes/MEAs is a natural one.
- The project approach addresses the project targets to increase maximum power of a direct liquid fuel cell and decrease Pt loading from that of a current DMFC by switching to a more energy-dense fuel and utilizing a high temperature polymer electrolyte membrane (PEM). The approach utilizing the higher temperature of a high temperature-PEM should prove beneficial for a DME-based fuel cell. It is not clear whether there is any steam reforming or hydrolysis of the DME prior to the fuel cell at the temperatures of operation. The project should perform some calculations to look at the stability of DME in steam at 160–200°C. It is not clear that the targets for the project would provide high enough power density for applications. Power density targets are fairly low for the applications mentioned on slide 3.
- Other than using DME as fuel, no new catalyst or membrane is to be developed by this project. It is essentially a system integration project. The principal investigator (PI) repeatedly emphasizes the importance of electrode structure, yet no scientific rationale or hypothesis was given on what structural improvement will be pursued.
- While the DME-based fuel cells are intriguing from a scientific standpoint, their performance (and loadings) to date seems so far away from commercial relevance that targeting small incremental advances in the technology, as has been proposed in this project, seems insufficient for an Office of Energy Efficiency and Renewable Energy (EERE)-funded project. From a science standpoint, developing improved membranes and catalysts for DME fuel cells can be done, and the proposed approach does this (although there is no real synergy in trying to do both; ideally, it would have been a catalyst-only project, as the current PBI membrane is not really the critical limiting element—rather, it is the poor cell performance and high catalytic overpotentials). The rationale for PtRuPd is reasonable based on the question-and-answer session (Pd is used to cleave ether linkage) but was not clear from the presentation. There is not currently a commercially viable high temperature DMFC, and just being as good as DMFCs is not compelling for this technology. Finally, DMFCs operate effectively by recycling the water in the system, and this is

accomplished by the natural phase separation of CO_2 gas from aqueous methanol solutions. It is unclear that such separation could ever be done effectively in the proposed DME system.

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 made good progress to date with substantial improvements over performance with Pt/C.
- The PI perhaps should have delineated clearly whether each/every single milestone in the project has been met. The data on slide 8 seem to suggest that the milestone is well met.
- The project is only six months old. Only limited experimental results are reported, mostly in test condition definition and benchmarking.
- From a scientific standpoint, the results are interesting, but too often a relevant baseline comparison is not included. It is not clear how the performance of DME with the Pd catalysts compares to PtRu (Pd-free catalysts) with either DME or methanol. More relevant comparisons of these families would improve the ability to judge performance improvements. Still, the performances reported are low (in terms of both current density and voltage). These represent modest improvement over previous performance but remain far from what is required for anything resembling commercial relevance. Some techno-economic analysis and market analysis could help define what is required for commercial viability. The project goals seem to be incremental improvements in today's performance without concern over what would be required to make commercially viable systems. The catalyst work is the most interesting. The membrane may offer incremental improvements, but it is not the critically limiting factor.

Question 3: Collaboration and coordination with other institutions

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

- Collaborations have been limited to LANL to date, as the project is fairly new. Reasonable agreement has been achieved between tests run at LANL and Advent Technologies, Inc. (Advent). Plans to collaborate with the University of South Carolina are in the future.
- The PI has identified possible relevant partners for incorporating non-precious metal catalysts.
- The team relies on the catalyst expertise from a national laboratory and membranes from the supplier/collaborator, making it difficult for the PI to control the project development pace.
- This really seems like it is just Advent with LANL playing a supporting role in catalyst development. It is not clear where the value proposition is for this with Advent.

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.

- Auxiliary power for transportation and for backup power is an important application for high temperature fuel cells.
- The project is relevant to portable power, backup power, and distributed generation fuel cell markets. The technology is different enough from low-temperature PEMs that market development with high temperature -PEM direct DME fuel cells will not be beneficial to the PEM supply chain.
- The project is relevant to DOE Fuel Cell Technologies Office goals.
- Performance is so far away from any targets in the DOE Multi-Year Research, Development, and Demonstration Plan that it is hard to imagine this project having an impact on any of them.

Question 5: Proposed future work

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

- Future work focused on reducing platinum-group-metal (PGM) loading and optimizing the electrode layer is appropriate and will lower costs and improve power density.
- Research targets are clearly identified. However, the paths are generic and lack specifics.
- More details are needed.
- The proposed future work is primarily associated with targets that are not compelling. They focus on incremental improvements in performance from poor baseline performance or in scaling up and reducing loading to a very high loading level: 4.5 mg PGM/cm^2. The remaining challenges and barriers are also not compelling. For optimizing DME, especially when performance is so poor at high loadings, efforts to improve water ratio seem like a poor area of focus.

Project strengths:

- The novel approach to utilizing a direct liquid fuel cell for the backup/distributed power market is a project strength.
- The team has a good collaboration between industry, a laboratory, and academia.
- The strong performance of the DME MEA is a project strength.
- Catalyst work is interesting from a fundamental science standpoint.

Project weaknesses:

- More details are needed for the annual report.
- There is a significant lack of innovation in this project.
- Performance of these systems and possibilities of commercial relevance are project weaknesses.
- The project does not align with the transportation focus of the EERE Fuel Cell program, and learnings from this will not really cross over to or have an impact on PEM fuel cells for transportation.

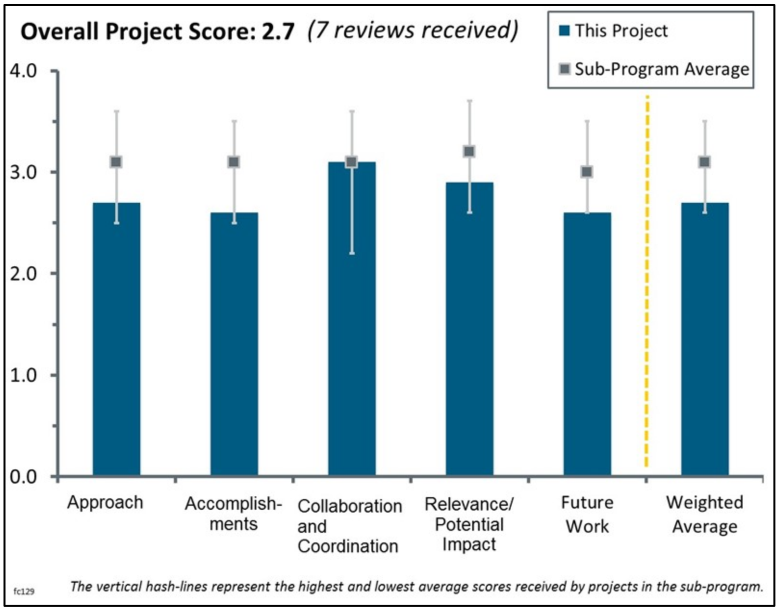
Recommendations for additions/deletions to project scope:

- Techno-economic analysis should be added to make a case for what performance and costs would enable the technology to be competitive in specific markets. Without this, the project targets are relatively meaningless.
- The PI should provide a clear description if a milestone has been met and what exactly will be done next year.

Project #FC-129: Advanced Catalysts and Membrane Electrode Assemblies for Reversible Alkaline Membrane Fuel Cells

Hui Xuastro; Giner, Inc.

Brief Summary of Project:



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

The project aims to combine an anion-exchange membrane (AEM) water electrolyzer with a fuel cell in a single stack to develop a reversible AEM fuel cell for energy storage and conversion. A water electrolyzer is an ideal device for storing energy as hydrogen from wind turbines and solar farms. The stored hydrogen can later be used in fuel cells to generate low-cost electricity during peak times. Use of catalysts based on non-platinum-group metals (non-PGMs) drives down capital costs. The project also contributes to maturing AEM technology and developing new concepts for oxide catalyst design. Tasks include (1) designing and developing oxygen reduction reaction and oxygen evolution reaction (ORR/OER) bi-functional oxide PGM-free catalysts and (2) integrating ORR/OER bi-functional oxide catalysts and alkaline membranes to develop highly efficient, reversible alkaline membrane fuel cells for stationary energy storage.

Question 1: Approach to performing the work

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

- The selection of the catalyst materials is sensible. Carbon nanotubes (CNTs) represent a good direction for more robust catalysts. The important issue is whether these catalysts can ultimately overcome the oxidation deactivation issue. There is no clear role for the National Renewable Energy Laboratory (NREL) in this project. NREL is leveraging their contribution from another U.S. Department of Energy (DOE)-funded project, although they have not demonstrated a clear path to reduce Pt usage.
- The use of a unitized electrolyzer/fuel cell in the alkaline domain has been the goal of electrochemists for decades. Use of perovskites as bifunctional oxygen electrodes is also known, albeit never showing stellar performance in a practical system. The approach, therefore, though sound, is not tremendously innovative. In addition, the project's approach using rotating disk electrodes (RDEs) as the sole indicator of effectiveness in an operating cell is very ambiguous. However, as per project metrics, this effort is on target.
- Despite the fact that several classes of metal oxides, spinels, and perovskites have reasonably high activity in both ORR and OER, the approach selected in this project based on usage of carbon supports cannot be scientifically justified. All types of carbons with no dependence on the level of graphitization will be oxidized to CO_2 at potentials higher than 1.23 V versus RHE. Slide 7 shows the stability of different types of carbons during the cycling between 0 V and 1.9 V. Taking scan rate into account, the whole experiment duration should be around 30 minutes, and dramatic degradation of material is seen. Electrodes fabricated with PGM-free oxides will operate in AEM electrolyzers at realistic potentials of 2–2.1 V (in deionized water), and the expected life-time should be 50,000+ hours. Thermodynamically, there is no carbon material that can withstand these conditions.
- The project addresses multiple reversible fuel cell barriers associated with the catalysts for reversible alkaline membrane fuel cells. However, the challenges of achieving sufficient activity and durability with a

single bifunctional catalyst are high enough that, despite significant progress, the project has a slim chance of leading to a commercially viable technology.

- The proposed approach has limited potential in addressing aggressive barriers in reversible fuel cells. Besides the proposed methods, investigators should be focused on examination of catalysts' structures after electrochemical cycling.
- While the original plan assumed a comprehensive approach, targeting development of cathode and anode catalysts, as well as the membrane electrode assembly (MEA), so far, this project has been dominated by the development of non-precious metal ORR/OER catalysts. Much less attention has been devoted to the hydrogen oxidation and evolution reaction (HOR/HER) catalysts and virtually none to MEA integration. This evident imbalance needs to be corrected to enhance the odds for success of this project. The use of carbon supports for OER catalysts is highly questionable.
- There are serious weaknesses in evaluating results, both internally and externally. The two goals for the project are to develop and then test bifunctional catalysts. For this period, only the development of materials took place, and this development seems to ignore most background work that has been done to date.

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

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

- The team has successfully met the first go/no-go milestone. Compared to other incubator projects, this one is more on track and on performance. The current perovskite-/spinel-based catalysts' performance remains significantly inferior to Pt/C for ORR in an alkaline medium. This should be addressed as the project continues.
- The overall project metrics and goals, including the go/no-go decision points, have been met using the RDE technique. All other tasks are well under way.
- The activity of two types of catalysts—Co_3O_4/CNTs and graphene tubes in OER and ORR—is very confusing. On slide 10, ORR performance at 0.9 V was 1.24 mA/mg and the limiting current was 90 mA/mg, while on slide 16, the second catalyst had a limiting current of just 2.5 mA/mg. The same confusing results were obtained for OER catalysts. The electrochemical performance was reported partially in mA/mg and partially in mA/cm^2 with no information on catalyst loading, which makes correct interpretation intrinsic activity and stability extremely complicated.
- Giner, Inc., and Giner's coworkers have made significant progress toward the project objectives. The main achievement this year was demonstration of 1 mA/mg ORR performance at 0.9 V and 15 mA/mg OER performance at 1.6 V in RDE testing, meeting go/no-go criteria. This milestone was satisfied with two different types of catalysts synthesized within the project. However, both ORR and OER activity are still rather low compared to the activity of dedicated monofunctional catalysts, and the round-trip efficiency of a reversible fuel cell based on the catalysts developed in this project would be rather low. Some progress has been made on development of hydrogen catalysts as well, but the catalysts developed to date are PGM-based. The possibility of eliminating PGMs is the main reason for interest in alkaline membrane fuel cell and electrolyzer technology, so the inclusion of PGMs on the hydrogen electrode is undesirable. Catalyst results to date come from RDE testing; MEA performance is not reported yet. DOE goals for this technology are not well defined, so it is not clear that this project is contributing to meeting the overall objectives of the Fuel Cell Technologies Office (FCTO).
- The oxygen catalyst development has followed two approaches. The results are interesting, and progress is evident. Performance targets, inexplicably defined in terms of mass activity (not justified for non-PGM catalysts), have been met, though they were not very challenging. In reality, the ORR activities are generally low—below the state of the art—and also indicate possibly high peroxide generation (which was not determined). There is no convincing evidence in the presented results that OER currents are carbon-corrosion-free (a realistic possibility, based on some presented results). Relative to the effort invested in the development of the oxygen catalyst, the team has paid little attention to the development of the hydrogen catalyst. No polarization plots for PtNi nanowires have been shown, which makes true activity evaluation impossible. There is some evidence of a relatively poor stability of the nanowires, not surprising for a PtNi alloy. No catalyst developed in this project has been MEA-tested, which is disappointing.

- While some progress has been reported, the status of perovskite-based materials remains unclear. There is an obvious discrepancy between what was proposed in the technical milestone table, what is in slide 23, and what has been reported.
- Reported progress is not supported by the data presented. All the perovskite goals—phase purity, crystallite size, and performance—are not demonstrated in data, and the data that are presented contradict the stated success. Further, no identification or quantification of the oxygen vacancies is presented, and while the selected materials set (Co-based nanoparticles on graphene oxide tubes) does appear to demonstrate performance, there are issues with this approach as well. It is unclear how these are graphene nanotubes. There are no characterization data for these. Further, there is no explanation or description of the path toward improved performance in this poorly defined and described materials set.

Question 3: Collaboration and coordination with other institutions

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

- The collaboration with State University of New York–Buffalo (SUNY-Buffalo) on development of CNT-supported oxides appears to have contributed significantly to the project. The NREL collaboration is critical since NREL is supplying the hydrogen catalysts and the membranes.
- This project features excellent partnerships between various entities, including a university (SUNY-Buffalo), a national laboratory (NREL), and a commercial partner (Giner, Inc.), with clear goals and objectives.
- The team has excellent participants that are well known in PGM-free materials design, electrodes fabrication, and characterization.
- The team includes a university and a national laboratory and is well balanced.
- The three partner organizations in the project appear to collaborate with one another. NREL’s contribution has been less than that of the two other partners, but the laboratory’s role may increase in the future when the focus shifts more toward the MEA and hydrogen catalyst development. No external collaborations were listed. An addition of potential future customers in the second year of this project could be helpful.
- Interactions between participants can be improved by engaging methods to investigate catalyst structural properties during and after electrochemical cycling. The PI should also consider stability screening of carbon-based catalyst supports through additional in situ techniques.

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

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

- The focus of this research is on the development of better materials for “reversible” alkaline fuel cells/electrolyzers, including catalysts for both electrodes and MEAs capable of competing with the state of the art in the field, e.g., systems based on Tokuyama membranes. While viability of the alkaline system of this type is not certain and the effort lies on the peripheries of FCTO, the project is interesting and overall relevant to the FCTO objectives.
- The project is relevant to the FCTO and DOE research, development, and demonstration objectives.
- The project is relevant to FCTO’s goals.
- While reversible fuel cells are not likely to be vehicle-deployable, they promise additional future hydrogen generation and storage options.
- A unitized fuel cell electrolyzer is a great objective toward which to strive. Being in the alkaline domain, the principle advantage is the freedom from noble metal catalysts. However, challenges include severe overpotential losses for both HOR/HER. No clear strategy is mentioned for overcoming these losses.
- The project’s main achievements at the moment of presentation are made with oxides supported on carbon or carbon-based electrocatalysts, which intrinsically cannot be stable at electrolyzer conditions.
- The project is not very relevant to FCTO goals. The substantial technical challenges faced by unitized reversible fuel cells make them unlikely to be commercialized in the foreseeable future. Furthermore,

technology for reversible alkaline membrane fuel cells is sufficiently different from conventional fuel cell technology that improvements made by this project will not be relevant to other fuel cell projects.

Question 5: Proposed future work

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

- The research plan is rational. The technology barrier remains significantly high.
- Future work follows in line with the milestones and metrics for this effort.
- If the proposed future work is based on Co_3O_4/CNTs and graphene catalysts, it may result in achievement of initial performance in unitized regenerative fuel cells; however, the cell will degrade substantially in the first hundred hours of operation.
- The future work is largely dedicated to MEA integration of the novel catalysts and testing in MEAs, which is appropriate at this stage, but further technical detail on the integration and testing plans is needed.
- In addition to MEA fabrication, future work should be more focused on the link between measured electrochemical properties and structural features of the catalyst. That would be helpful in overcoming barriers and would guide the synthesis of robust catalysts.
- There was very little substance in the future plans slide in this presentation. MEA fabrication and optimization of test conditions have little to do with the development of materials, on which the team should continue to be focusing. The same is true of modifications to “fuel cell configuration” and the test station for intermittent operation.
- Future work was unclear, nor was it clarified when the presenter was asked.

Project strengths:

- Strengths of the project include its focus to achieve performance that would meet targets and overcome barriers, implementation of both electrochemical methods RDE and MEA, and reliance on non-precious-metal-based catalysts for ORR/OER.
- The project strength can be in design of unsupported oxides and perovskites with high electrical conductivity.
- The project is doing a good job of leveraging previous work and outside work to produce novel catalyst structures and is on track in terms of meeting milestones.
- The oxygen catalyst development is by far the strongest part of the effort.
- The project is well on track.
- The concept is interesting. Giner has demonstrated systems experience.
- The project aims at enabling a unitary fuel cell operating with an AEM. Partners in this effort are well placed to effectively meet the goals and objectives of this effort. ORR/OER catalysts have met the project go/no-go decision point. However, this has been obtained using the RDE technique, which is significantly distant from obtaining the same result in an MEA half or single cell.

Project weaknesses:

- Hydrogen catalyst development has trailed the oxygen catalyst effort with no polarization data presented. There seems to be lack of understanding of the causes of performance differences between materials; for example, various carbon supports and carbon tubes have been derived from different transition metals. CNTs have been selected as a stable nanocarbon support for the OER in spite of exhibiting what appears to be persistent corrosion at potentials higher than 1.7 V.
- Weaknesses include apparent discrepancies between the technical milestones and systems in this report; need for careful investigation of stability for carbon-based supports; lack of detailed insight into catalyst structure before, during, and after electrochemical cycling; and lack of in situ methods for structural characterizations of catalysts. Quantitative analysis of the metal oxide catalysts before and after electrochemical cycling is needed, as well as strong proof that observed currents are not associated with dissolution of employed materials. Improved coordination between participants is also needed. Existence of iron in the most active catalyst could induce damage of the membrane. Evaluation for perovskite- and spinel-based materials is needed.

- The project is not very relevant to FCTO. Even if the project meets its goals, it is unlikely to substantially increase the odds of unitized reversible fuel cell technology becoming commercially viable.
- The project does not examine any innovative class of materials for ORR/OER; perovskites are well known. There is very little attempt at performing detailed structure property relationship studies in concert with catalyst activity. Important details of the kinds of metal oxides and their conductivity should be included. Taking into consideration that the AEM ionomer has a significant effect on catalyst activity, the ionomer's role should have been a part of the RDE studies. No details were provided about the nature of graphene oxide tubes, including surface areas and corrosion analysis.
- A project weakness is the completely wrong selection of materials (carbon-based supports or catalysts) for ORR and OER.
- The materials developed are poorly developed, and a path to improvement is not presented.

Recommendations for additions/deletions to project scope:

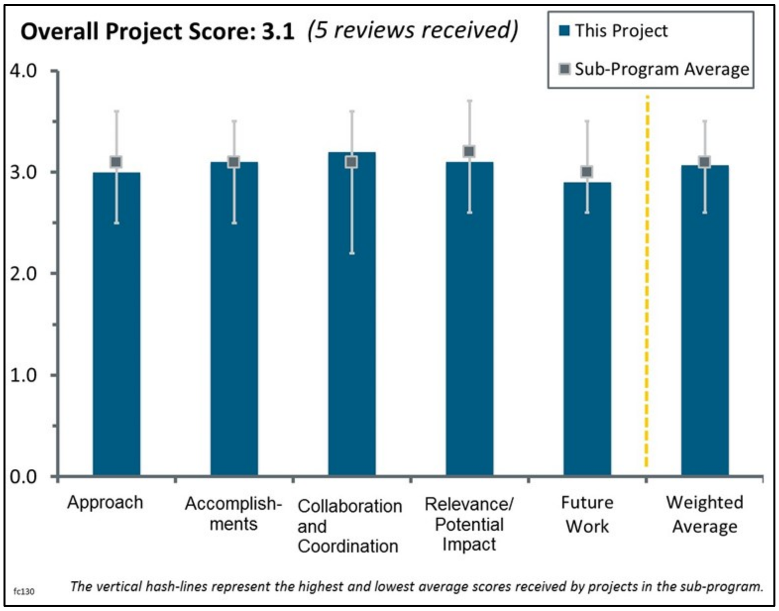
- More details on the ORR/OER metal oxide catalysts, such as electronic conductivity and/or structure, should be provided. It is not clear what the active site is for ORR and OER or whether there is a correlation with defects. It is not known whether the cycling tests were conducted under inert gas flow or under a blanket of oxygen—or, in the case of the former, how it would be reconciled with actual cell operation. Economic analysis should include the comparison of operating a system with AEM membranes but with the electrolyzer and fuel cell separate.
- This project would benefit from a better understanding of the reasons behind observed differences in catalyst performance and from early discarding of materials that do not perform/show promise. For example, the durability testing of the Co_3O_4-oCNT catalyst should not have been carried out, given the low ORR activity of that catalyst. The levels of peroxide generation rates need to be evaluated and used as one of the down-selection criteria for ORR/OER catalysts. Fuel cell testing is necessary. Non-PGM catalyst performance targets should be given in terms of surface-specific activity, not mass activity.
- Better proof of the materials development claims and performance is needed to demonstrate the viability of the approach.
- The project would benefit from additional screening of the catalyst's structure using in situ methods (carbon-based supports and metals) and compositional analysis of the catalyst and quantitative comparison of the metal content before and after electrochemical cycling.
- The reviewer recommends a no-go decision regarding carbon-supported materials, and additional concentration on passing a go/no-go design point with unsupported conductive oxides.

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/cm^2). Objectives include developing PGM-free electrocatalysts for hydrogen oxidation reactions 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 **3.0** for its approach.

- The approach generally appears sound, with a multi-faceted approach that addresses the major aspects of developing a PGM-free hydrogen-oxidation-reaction (HOR) catalyst for alkaline media. Recommendations include the following: provide more information on the cost and target analysis; benchmark data against other systems; discuss stability issues and whether any evaluation will be done; justify the scale-up activities; establish MEA evaluation criteria; increase ionomer integration work; and develop plans to establish the structure-to-properties relationships.
- The approach taken by the project team is generally effective; however, it could be improved. For example, catalyst performance and material preparation processes could be evaluated/optimized with parametric studies/experiments.
- The approach is good, with two exceptions that are related to potential durability issues. First, the MEA testing does not appear to include testing on air. It was unclear how the team will determine whether there are issues with CO_2. This is an anode catalyst, but CO_2 will cross over from the cathode and may cause other issues that may have an impact on the performance (e.g., carbonates in the catalyst layers). Second, the approach does not appear to assess stability over the full electrochemical potential range to which an anode catalyst will be exposed.
- Using a nickel-based catalyst for HOR in alkaline medium is a relatively well-known approach. The team uses other metals as additives with some encouraging development. The progress appears rather slow, given that the synthesis and characterizing methods used are relatively straightforward. The result is not particularly encouraging.
- The authors are attempting to fabricate, characterize, and scale up a new catalyst for the HOR in alkaline solution, and much of the work being done in this project is new. The approach is to characterize electrocatalysts with a rotating disk electrode (RDE). The catalyst loading is very high, and very thick electrodes are being used versus the standard thin-film RDE methodology. As a result, it is very hard to evaluate the data. It would have been helpful if the principal investigator (PI) had provided references to the proven methods employed because they may not be as well-known as the PI assumes. On slide 7, it appears that Los Alamos National Laboratory (LANL) is doing the baseline voltammetry for ionomer evaluation in 0.1 M $HClO_4$. The purpose of these studies is not clear because it is well known that RDE is

inadequate for HOR measurements of Pt/C in acid because of mass transport limitations (see Julien Durst, Christoph Simon, Frédéric Hasché, and Hubert A. Gasteiger, “Hydrogen Oxidation and Evolution Reaction Kinetics on Carbon Supported Pt, Ir, Rh, and Pd Electrocatalysts in Acidic Media,” *Journal of The Electrochemical Society* 162 no. 1 (2015):F190-F203, doi:10.1149/2.0981501jes, and references therein). Also, no details are provided on the catalyst coated membrane (CCM) manufacture and assembly. A simple error in over-compression of a gas diffusion layer can cause the results seen between the two different ionomers. More information on the CCM manufacture (provided in the back-up slides) would have been helpful.

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 Ni-Mo-Cu sample has a current density of 0.055 mA/cm^2 at a voltage of 0.01 V, which is approaching the target of 0.085 mA/cm^2, which is the first go/no-go design point. It is not clear what the timeline to achieve this target should be. Regarding ionomer development, work has been done to select the ionomer, with density functional theory (DFT) modeling to support. The experimental data and DFT modeling are in agreement, and the team was able to rank the ionomers and downselect. Although the work on the ionomer testing on slide 16 shows various systems tested, no conclusions are presented, and it is not clear what the team has learned from the ionomer testing. The scale-up process work has been initiated, and the MEA design and testing has been initiated, but no results have been shown for the target catalyst systems. Baseline Pt/C MEA testing data are shown.
- The project team has made significant progress toward achieving the go/no-go decision point.
- The team appears to be on track midway through the project. The MEA performance is disappointing, but other results are good so far.
- According to its own metrics, the project has made significant progress. However, good benchmarks for HOR catalyst performance in alkaline solution by RDE or in CCMs do not exist. It is also not clear that RDE voltammetry has been validated as a useful tool for predicting electrocatalyst performance in a functional cell. It is difficult to evaluate whether accurate kinetics can be measured in such a way. It would have been helpful if the PI had provided the reasoning behind the “benchmark” of 0.085 mA/cm^2 at 0.01 V. It is not clear if this target is iR-corrected, nor is it clear what is expected for Pt. Overall, not enough information was provided to evaluate the project’s accomplishments. In the project’s defense, this is probably a ten-year project, so the one-year accomplishments are reasonable, especially with the focus on transition imposed by the DOE Office of Energy Efficiency and Renewable Energy.
- The project is clearly behind schedule.

Question 3: Collaboration and coordination with other institutions

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

- Collaboration and coordination with other institutions is excellent; the team seems to be working well together, and it is a nice mix of industry, national laboratory, and universities.
- Good collaboration exists between university, national laboratory, and industry.
- A good, effective team has been assembled, each with clear expertise and contributions to the project. However, it is difficult to tell how much interaction the team has on a regular basis, and given the nature of the work, it is recommended that regular meetings are held to improve the effectiveness of the integration activities.
- The collaborations are good, although in hindsight, the project partners should have pursued collaborations that would yield more fundamental information about their electrocatalysts. Given the applied nature of the project and the two-year timeline, the collaborations are acceptable.
- Although the project involves multiple partners, the contribution from other members seems very minor at this stage. For example, there seems to be a great deal of needed MEA development, which is supposed to be done by IRD Fuel Cells (IRD). It is unclear whether IRD has adequate knowledge and expertise in handling the metallic-material-based ink.

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.

- Slow HOR kinetics in alkaline solution is a known problem, and seeking a non-precious-metal replacement to Pt/C would be beneficial. The development of an inexpensive but effective electrocatalyst for the HOR in alkaline solution could be very important.
- The alkaline fuel cell anode is an important area for cost reduction.
- The project aims to develop PGM-free catalysts for HOR, thus fully supporting DOE research, development, and demonstration objectives.
- The work is relevant to developing the AEMFC, which is an important and potentially disruptive technology that may achieve Fuel Cell Technologies Office objectives at a lower cost. However, the project team has not provided analysis to clearly show the potential positive impacts.
- This project is good to include as part of an alternative to conventional proton exchange fuel cells (PEFCs), but it does not address all of the major issues with alkaline fuel cells. Since it is well accepted that PGM-free catalysts can be used in alkaline fuel cells (AFCs), it may be more useful for DOE to focus on the more serious AFC barriers, such as membrane stability and carbonates, which may not precipitate in AEMFCs but still have a major impact on ionic conductivity of membrane and catalyst layers.

Question 5: Proposed future work

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

- The activities outlined in the path forward appear suitable: decrease particle size of nickel-molybdenum-metal (Ni-Mo-M) catalysts; optimize RDE working electrode preparation to establish kinetics parameters; integrate a PGM-free anode into an MEA structure; design an MEA fabrication and testing protocol. Further work is required in the areas of catalyst system development and ionomer integration work to understand optimization opportunities. Increased information should be provided on what work will be conducted on establishing the structure-to-properties relationships. This will be important work.
- The proposed future work is good, but the team should assess stability in the presence of air.
- The project partners should focus less on DFT and powder scale-up and more on electrochemical methods (RDE and CCMs). Although the project partners probably do not have time, in situ spectroscopy measurements would have seemed more important than DFT work.
- Although future challenges are identified, no clear solutions or approaches are identified.
- It is not clear what impact the proposed near-term activities will have on overcoming the barriers.

Project strengths:

- The concept of the project is very good, and it is positive that there are commercial partners and interactions with numerous collaborators.
- There is a strong team addressing project objectives with a multi-faceted, integrated approach.
- Project strengths include the focus on key activities to support the project objectives and sound technical approaches, the ability to make 50 g batches of catalysts, full MEA testing, and no C supports.
- The team achieved some promising initial results.

Project weaknesses:

- This is a very complicated project. It is not clear that the project has an active electrocatalyst because of limitations and challenges with the test methods. The authors are using “boilerplate” methods for advanced catalysts—scale-up, DFT, etc. More thought is needed on the fundamental electrocatalysis and methodology.
- The RDE testing is a primary evaluation tool, and it is not clear whether the kinetic data being reported will translate to MEA data, or how they compare against any benchmark data.

- Potential stability issues are a weakness with the project. The team should assess stability in the presence of air (1.23 V RHE) since anodes in a real-world fuel cell system will be periodically exposed to air (it is practically impossible to keep hydrogen on at all times). The lack of MEA testing in air is a weakness.
- The lack of identification of key impact/optimization factors, for example, for catalyst preparation processes, is a weakness.
- The progress is significantly behind schedule, particularly given the high level of funding for the project.

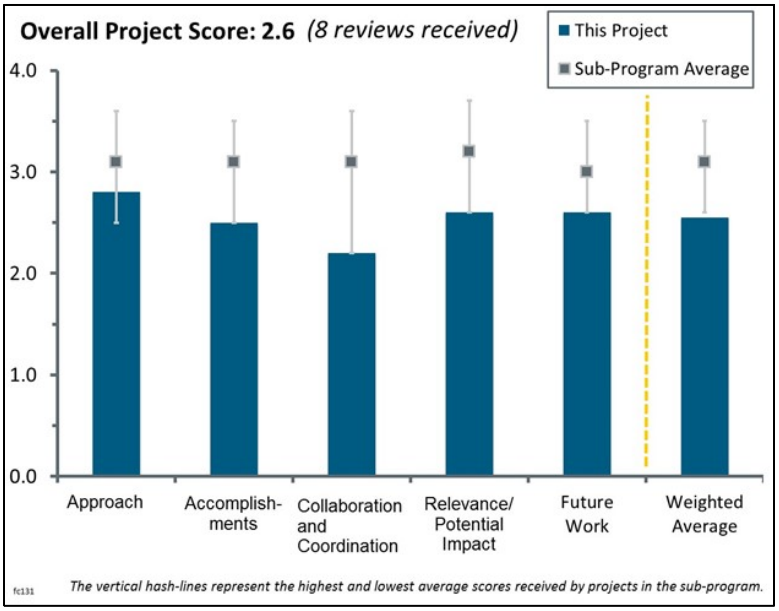
Recommendations for additions/deletions to project scope:

- More information regarding relevance of the work and the potential impact should be included. The University of New Mexico (UNM) should identify whether cost and scenario analysis has been done to help set targets, and if not, what information would be required to get to the point when the analysis can be done. Stability of PGM-free HOR catalyst is a primary concern, and the manner in which this will be addressed should be identified. Benchmarking data of catalysts for relevant systems should be provided. It is not clear whether the scale-up work is required to support the other project objectives.
- LANL work should focus on systems more directly related to the UNM work. Perhaps the project could deemphasize scale-up and emphasize more work on method development.
- Specifications for materials preparation processes should be developed. Electrode/material performance stability studies should be performed.
- The project should add cyclic voltammetry testing that includes cycles up to 1.23 V RHE to assess whether there are potential stability issues. Hydrogen/air testing with MEAs should be added.

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 **2.8** for its approach.

- This project seeks to extend the principal investigator's (PI's) prior work on alkali-stable aryl phosphonium ionomer AEMs for use in double-membrane RFBs having Ce(4+) as an active component. The approach is to identify new phosphonium organocations that are stable to extended exposure to Ce(4+), and incorporate them into ionomers from which oxidatively stable AEMs may be fabricated and tested. This is a generally good approach.
- The project has a nice systematic approach to developing stable phosphonium cation-based AEMs. It is unclear why phosphonium is preferred over ammonium cations. The cost projections based on the high-voltage redox chemistry show the value in pursuing this approach. The project highlighted a specific redox chemistry using a bipolar membrane or a pair of polymer electrolyte membranes (PEMs)/AEMs, but the bulk of the presentation outlined the synthesis of the AEM ionomer. It is unclear whether the project is to develop the RFB chemistry and membrane, or just the membrane.
- 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 synthetic approach appears solid. It would be beneficial to see at least some focus on conductivity/resistance measurements in addition to stability. This project would greatly benefit from some analysis as to what is required for specific applications. It is not clear why milestones are limited to 40°C. It is not clear how the ex situ stability tests correlate with in situ degradation. Methods for backbone and functional group down-selection are unclear.
- The approach to make stable AEMs using non-conventional cationic groups is good. The stability study for the candidate cationic groups before putting in the polymer structure is reasonable. However, the rationale for using phosphonium versus ammonium is not fully justified. The cation exchange membrane and AEM

approach for redox flow cells to increase the potential window is innovative. The approach to develop such systems should include other requirements such as compatibility with redox active species, conductivity, and crossover. The synthetic approach to make AEMs is satisfactory but not innovative.

- The project appears to be systematically addressing oxidative stability issues. While durability is promising, conductivity of the materials was not reported, suggesting it is not being characterized. Both durability and conductivity must be co-optimized.
- One element that is lacking is a consideration of cation crossover, including Ce(4+) and, as the PI mentioned in his comments, H+. This seems like something that should be given attention for an RFB because very high concentrations of redox agents are going to be desired.
- The stability test methodologies (looking for weight change and color change) are not the most accurate methods to determine membrane degradation. An analysis of the leach solution for residual organic components would be more sensitive and provide some information about the degradation site. The 9MeTTP+ cation should be quite stable if it can be attached to a polymer backbone; however, the steric factors, which help the stability, will make it difficult to link this cation to the ionomer backbone.
- The Program’s technical targets are not mentioned in the presentation. It is unclear how this work is advancing toward meeting any membrane performance goals. It is not clear whether this is a battery project or a fuel cell project.

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

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

- There is nice work improving the stability of the linking group between the cationic group and backbone.
- The PI has identified a phosphonium cation and screened several polymer backbones that are stable to extended exposure to Ce(4+). T
- Timely progress was made against project milestones. Durability and synthesis is improving. Conductivity of materials is unclear.
- The project is working toward meeting durability targets on ionomers, but it is having difficulty in the synthesis in the transition from small-molecule salts to ionomers. The PI’s initial coupling strategy to make ionomers did not give high enough coupling yields, so he switched to another coupling strategy, which worked better but was not durable on exposure to Ce(4+). He has identified a third approach that he expects will succeed, but he has not yet tested it. Conductivity measurements have not yet been pursued because adequate ionomers have not been made. He is doing the right things to get the synthetic chemistry to work; he just has to work through the difficulties that inevitably come up in chemical synthesis.
- Some progress was made on making stable polymers; however, it is not clear how the weight loss measurements related to fuel cell stability. For example, in perfluorosulfonic acid materials, a weight loss measurement is not typically used to forecast membrane lifetime; only in-cell accelerated tests accurately replicate operational stability. The technical readiness of this project is far from this point.
- The project is narrowly focused on ionomer development for alkaline membranes, and it appears that good progress is being made in developing the proposed synthetic pathway, but a clear justification of how these results represent progress toward the overall project goals and DOE goals is lacking.
- Stable polymer backbones have been identified. 9MeTTP+ has been connected to a commercial polymer backbone with high yield. There is minimal stability data on functionalized polymers. Quality membrane films have not been prepared. There are no conductivity or performance data.
- The project has run into difficulties preparing a 9MeTTP+ derivative of a polysulfone backbone, has not been able to produce a membrane to begin tests, and is behind schedule. An alternate approach utilizing one of the methyl groups of the 9MeTTP+ to link to the backbone through an amine linking group has been proposed. There appear to be problems with this approach as well, as bromination at multiple methyl groups is possible. Bi- or tribrominated cations would lead to crosslinking of the membranes and poor membrane properties.
- Justification of the claims that the project has met the milestones is insufficient. Significant technical detail on the synthetic pathways being pursued was reported, but no data were presented to address specific milestones.

- Polymer backbone stability of unfunctionalized PSF, PEEK, and 6F-PBI for 1,000 hours at 40°C (slide 8) should have been done in more rigorous conditions for the sake of time. Also, measuring only weight after the stability test may not be good enough for ensuring stability. Mechanical properties and/or gas permeation chromatography (GPC) measurements should have been performed before synthesizing AEMs. There is relatively good progress on synthesizing polymer membranes. However, more membrane characterizations such as conductivity, titration, and stability measurements need to be done or have been planned, so overall progress on this project looks to be slow considering that this is two-year project. Much of the polymer degradation can happen with the combination of polymer backbone and cationic group. As the stability test for the target polymer is incomplete, it is doubtful that the PI can complete the membrane development and testing within the remaining project time.

Question 3: Collaboration and coordination with other institutions

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

- It appears that no work has been done outside the University of Delaware yet. The National Renewable Energy Laboratory's (NREL's) role is to test membrane durability and make and test MEAs. Because no membranes have been prepared yet, there has been no work for NREL to do.
- It appears that there has been no collaboration/coordination because synthesis difficulties are causing the PI not to have materials to give to NREL for testing.
- Collaboration with NREL is stated; however, the project is far from the ability to make testable membranes.
- There is no evidence of collaboration with NREL, the only partner on this project.
- It is unclear whether the single project partner, NREL, is engaged.
- NREL's role is unclear.
- So far, the project does not seem to have meaningful collaboration. The reported results all appear to have come from the University of Delaware. More collaboration may come in the future, with planned membrane and MEA testing at NREL, but this work was not discussed in a meaningful way.
- This is a project sharing resources between University of Delaware and NREL. No NREL work has been identified. NREL's work plan is not well defined. It may not be realistic for NREL to perform all membrane durability, fuel cell testing, and flow cell testing with $100,000 budgets. No interactions are specified besides NREL, and there is no clear pathway to get stable polymers from Rensselaer Polytechnic Institute (RPI). The project is about developing new AEMs, and it is not clear how the PI wants to transfer the major task to RPI.

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

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

- Developing stable AEMs for flow/fuel cells is relevant to the Program. Adopting phosphonium into polymer structures may be valuable to study and give potential impact on flow/fuel cell developments. Developing redox-flow cells is also relevant to developing advanced fuel cells because both systems share some similar requirements, even though the current project does not have much system study.
- Development of stable AEMs is a key need for AEM fuel cells. It is unclear whether AEM fuel cells have the potential to achieve commercial relevance against PEM fuel cells because of their poor hydrogen oxidation reaction kinetics and lower intrinsic conductivity.
- The project goals are aligned toward making a membrane stable in a high-voltage flow cell. The flow cell battery environment is different from that for a fuel cell, but it is thought that a membrane stable in that environment should be stable in an alkaline fuel cell.
- Making anion-exchange ionomers that are stable to Ce(4+) is a good goal, but that goal is not so closely tied to the Program; it is more closely tied to energy storage, e.g., with a flow battery. The project is fine for what it is, but the focus seemed a bit mismatched with Fuel Cell Technologies Office (FCTO) goals from the start.

- The basic concept of high-voltage flow batteries is appealing. It is unclear whether all the materials are stable to the potentials and will have the required durability.
- The project seeks to advance RFB technology, which is mentioned in the FCTO Multi-Year Research, Development, and Demonstration Plan but does not represent a major part of the Program. The Program does not appear to be working toward any specific goals or milestones with respect to RFBs. Therefore, the relevance of this project is questionable.
- This project only partially supports Program goals. It is more a fundamental material research project.
- It is not clear how reversible flow batteries address U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program (the Program) goals. While the approach is directed at a membrane for high-voltage flow cells, the membrane should be applicable to AEM fuel cells and address stability issues of alkaline membranes.
- It is unclear to what application the double-membrane RFBs would be applied and what the technical requirements for such membranes should be. Even if the project reaches all of its milestones (high oxidative stability in an alkaline media), without any performance criteria (or even measurements) there is no expectation that the project will advance progress toward the Program goals and objectives. Perhaps it will demonstrate something about the fundamental stability of AEMs.

Question 5: Proposed future work

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

- The PI correctly notes that his problems have to do with synthesis, and he is working as best as he can to correct the problems.
- Future work does put emphasis on preparing membranes that could be placed in fuel cell or flow battery test fixtures. More emphasis on this type of work would be appropriate for the Program.
- The proposed future work addresses the failure in attempting to directly link the 9MeTTP+ to the polymer backbone and has some potential. Attachment to the backbone through one of the methyl groups should avoid the problems caused by steric crowding at the P atom and should prove feasible. It may be difficult to limit substitution/bromination to one of the methyl groups. Methylation of the P and then attachment through a phenyl methyl group could lead to decreased stability of the phosphonium cation, as the steric crowding at the P is less.
- Specified future work includes improvements to several synthetic steps, and a brief mention of testing in flow batteries or fuel cells, but more detailed discussion of the future work is needed.
- Future work to improve the ionomer stability seems well focused. More emphasis needs to be placed on ionomer characterization such as ion-exchange capacity or conductivity measurements. Clearer plans on testing in flow battery or fuel cell devices would be helpful.
- Conductivity evaluations are necessary.
- The project should put some emphasis on conductivity and performance. The project should also focus on mechanical and higher-temperature stability.
- Proposed work is not specific, and there are no clear plans for device testing. No milestones have been established based on the future plans listed on slide 17.

Project strengths:

- The project strengths are the new and novel material research work; the project does have the potential to develop game-changing materials for fuel cells and flow batteries.
- Using AEMs for innovative RFBs is of great interest. Model studies using different phosphonium cations give valuable insight for advanced AEM development.
- The aryl phosphonium organocations with which this PI works are very stable and could be the basis of a new generation of oxidatively stable AEMs.
- Development of a stable posphonium-based AEM may have applications beyond flow batteries. High-voltage flow batteries may have potential in high-power-density devices. Another strength is the systematic approach to identify stable polymer backbones and cation attachment chemistry.
- The project builds off earlier work performed by the PI in an ARPA-E project and successfully leverages that earlier work.

- Solid polymer chemistry approach and polymer characterization (infrared and nuclear magnetic resonance) are project strengths.
- The phosphonium cations chosen have high stability.

Project weaknesses:

- The synthetic strategies have been difficult to implement.
- It is unclear whether the focus of the project is to simultaneously develop an ionomer and the Zn-Ce RFB chemistry. The roll of PEM and the related requirements are not discussed. More details on International Electrotechnical Committee standards testing or conductivity would be helpful. A good explanation of why ammonium cation AEMs are not suitable would also be helpful.
- So far, the synthetic routes from small molecules to ionomers have been difficult. Also, the connection to hydrogen technologies is not clear.
- The project is narrowly focused on alkaline membrane development for flow batteries and other devices, and relevance to the broader Program is limited. While the synthetic strategy was well presented, a lack of clear metrics or goals makes it difficult to assess progress.
- AEM property characterization was planned with only a stability perspective. Current polymer structure is somewhat deviated from the original proposed structure because of synthetic difficulties. This is acceptable; however, it is unprovable that the current structure can satisfy all requirements for redox battery AEM requirements. If the AEM is proposed in the use of RFBs, the project should have device performance targets rather than just ex situ stability targets.
- It is not clear how this project fits in the Program. No technical targets for performance can be measured.
- The project is not clearly tied to the Program objectives. The scope is limited to oxidative stability at low temperature. There is a lack of collaboration and no clear approach for concept down-selection.

Recommendations for additions/deletions to project scope:

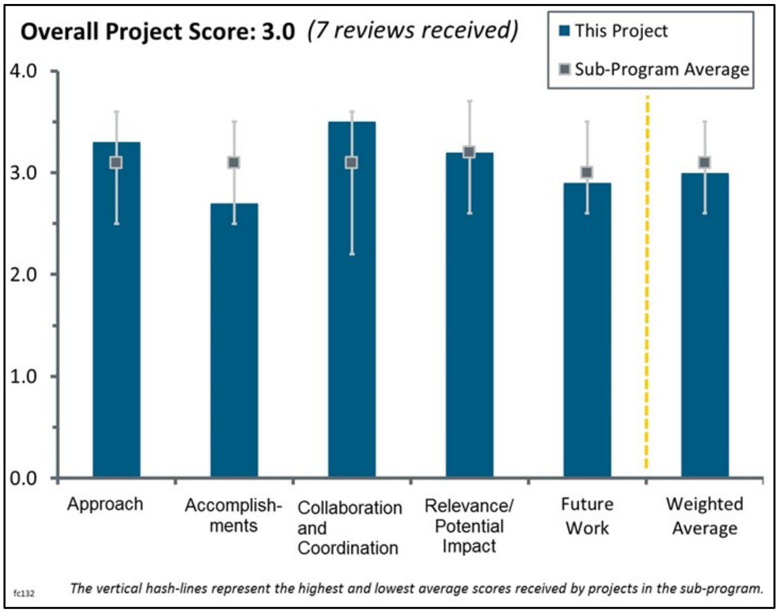
- This project probably needs a go/no-go decision point to decide whether the synthetic approaches are giving ionomers that can be made into durable and ionically conductive membranes. It appears that the PI does not yet have that. His initial synthesis proposals are not giving what he needs, and the proposed new routes might give suitable materials, but they are yet untested. This is okay, but at some point, he has to meet a milestone for the ionomer, not just the parts.
- Stability tests should entail some more sophisticated diagnostics, such as testing leachate for organic residue or for P. Color changes can be misleading, and weight changes can be difficult with substituted materials because water adsorption can change and drying to the same state of hydration is not always easy.
- The project should add focus on conductivity, mechanical stability, and higher-temperature stability. The project should collaborate with a systems modeler to provide the technical targets required to enable successful commercial application.
- The project should focus more efforts on making membranes of any kind that can be built into fuel cells, even if very small. Also, this will allow other membrane characterization tests, such as conductivity. This will be a way to judge the technical readiness of this project for the Program.
- An anion conductivity (ClO_4 or sulfate) target should be added to the milestone. A mechanical milestone (or target) should also be set. Fuel cell testing is irrelevant and may be deleted. All stability assessments should be based on spectroscopic data and mechanical data in addition to internal combustion engine change. This may require third-party evaluation.
- The project should increase focus on characterization.

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 non-platinum-group-metal (non-PGM) electrocatalysts that would allow for high performance in high-temperature polymer electrolyte membrane (HT-PEM) fuel cells. A successful outcome will enable HT-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, reduced carbon footprint, and improved U.S. energy security.



Question 1: Approach to performing the work

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

- Northeastern University (NEU) project is pursuing a promising approach to develop PGM-free catalysts for HT-PEM fuel cells. 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-based fuel cells. By further developing several related PGM-free catalysts based on carbon- and nitrogen-coordinated iron centers developed for low-temperature PEM fuel cells in a previous project, this project is leveraging previous work toward a new application.
- This work directly addresses the cost barriers for fuel cell technology implementation. The approach to catalyst development is effective. MEAs developed with these catalysts may have a very different implementation from those developed on traditional carbon or nanostructured thin film supports. Issues with MEA development have already been seen with IV characterization of the novel catalysts in the early phases of the Hydrogen and Fuel Cells Program (the Program).
- The approach to developing a variety of non-PGM catalysts for HT- PEM fuel cells is great and includes NEU’s metal–organic framework (MOF) catalyst and the University of New Mexico’s (UNM’s) catalysts. These catalysts have great potential to mitigate anion (phosphate) adsorption. This is very significant. In addition, their synthesis a simple and facile.
- The project addresses the barrier of cost by attempting to reduce Pt loading in HT-PEM fuel cells by utilizing PGM-free catalysts. The project specifically addresses cost of stationary systems as HT-PEM is not applicable to automotive transportation systems because of the lower power density. The approach addresses phosphate poisoning, which decreases performance in HT-PEM fuel cells, and is addressing mass transport losses through the use of the sacrificial support method to create porous structures. The recent breakdown of potential losses indicates that transportation losses are the main issue. Work should focus more on electrode structure and reducing mass transport losses.
- The ball-milling approach and the use of sacrificial support (to generate porosity) and MOF materials are good for generating PGM-free cathode catalysts.
- Using non-PGM materials to replace Pt is critically important in reducing fuel cell cost. The non-PGM catalysts reported in this project represent the extension of materials developed from a project previously funded by the Fuel Cell Technologies Office. The stability of carbon-based non-PGM catalysts proposed by the project investigators is a major concern during high-temperature operation. These catalysts could be

oxidized rapidly in the presence of oxygen. The investigators need to develop a strategy to improve the oxidation resistance of their catalysts. The non-PGM catalysts of this project have different active site distribution from that of Pt-based catalysts. The humidification plays an important role in the proton transfer to the highly distributed active sites. Therefore, high-temperature, low-humidity operation seems to work against the usage of such materials, which may be a major cause of high overpotential.
- The replacement of Pt-based catalysts with non-PGM catalysts represents an enormous challenge. However, if successful, the approach could provide a solution to phosphate contamination of the cathode. Claimed improvement to mass transport and especially corrosion resistance characteristics of catalysts, thanks to the use of the sacrificial support approach, is not obvious. The team spared no effort in studying the active site in catalysts that showed poor activity in the fuel cell cathode. The benefit to the project objectives is not clear.

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

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

- (1) The progress of this project seems to be a little bit behind schedule. The original targeted performance was for hydrogen/air but the demonstrated performance from hydrogen/oxygen is good and the hydrogen/air performance is poor. The principal investigator (PI) claims that further MEA optimization, with Advent's help, could help to improve the performance and it is hoped that better hydrogen/air performance can be achieved in the second year. (2) It is unknown whether UNM's two catalysts, the Fe-AApyr catalyst and Fe-Nicarbazin catalyst, were tested for fuel cell performance. (3) In situ x-ray absorption spectroscopy (XAS) is unique, helping to identify the active site of the catalysts. (4) RDE data comparison was very systematic, disclosing a good deal of valuable information.
- The project has made progress scaling up PGM-free catalysts. Current studies indicate phosphate poisoning is reduced in these PGM-free catalysts. Rotating disk electrode (RDE) experiments indicate PGM-free catalysts have higher activity than Pt in 10 mM phosphoric acid; MEA experiments indicate iR-corrected losses and transport losses are higher for PGM-free catalysts. It is not clear what differences there are between the catalyst preparation and scale-up here versus that already developed under the DOE-funded project studying non-PGM catalysts for low-temperature PEMs. The same techniques are used with the same descriptions.
- The team has made good progress toward meeting the milestones set in the project. As of now, MEA performance is not meeting the targets as set but ways to improve have been identified.
- The first milestone, which specifies good reproducibility of catalyst activity in RDE testing, was apparently met but was not adequately described in the presentation. The project has yet to meet the Year 1 fuel cell performance milestones for operation on oxygen and on air. Improvements in electrode structure have been proposed to meet these milestones but the justification for these improvements is rather weak. For both oxygen and air testing, the reported voltage loss breakdowns indicate that mass transfer losses are relatively small though the presenter indicated that the reported breakdowns may not be accurate. Electrode flooding was proposed as a significant loss mechanism but given the high temperatures and low humidities used, liquid water should not be present. More detailed reporting on the electrode structures used would be required to clarify the relevant loss mechanisms. Given the early stage of the project, there is still time to address these issues and a good chance for significant performance improvements.
- Progress is steady; however, one target (go/no-go 1) may not have been hit at the time of the presentation. It was not clear from the presentation or the supporting material whether the work presented included data to be evaluated 12 months into the project. There may have been a performance loss from what had been previously reported (2015 package) and what was presented at the Annual Merit Review. This may have been due to MEA fabrication; this was not discussed.
- Performance targets on oxygen and air were not met in this project. A pressure 1.7 times higher than the target value was required for ultimately meeting the milestone performance on oxygen at 0.7 V. The results point to an even larger gap between the demonstrated and target performances when the fuel cell is operated on air, with the demonstrated current density of ca. 40 mA/cm^2 five times lower than the milestone current density of 200 mA/cm^2. Switching to new MEA formulation has not helped so far. Much better tolerance of non-PGM catalysts than Pt catalysts to anions, including phosphates, has been known for

many years. Reassessment of that property in this project tied up the resources unnecessarily. High CO tolerance is promising.

- The project progress report is inadequate. The PI provided a milestone table but failed to include the most important column, i.e., the status update. The project is nearly at its mid-point. The status updates for milestone 1.1a, 1.1b, 2.1, and 1.2 should be reported and compared to the goal. The project is behind schedule.

Question 3: Collaboration and coordination with other institutions

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

- The team includes two universities and three industrial partners. The role of each team member appears well defined.
- The collaboration is great as the assembled team includes two industrial partners and two universities, all of which have good experience in catalyst development and fuel cell component design, respectively.
- The team members all provide useful know-how and have been collaborating well.
- The partners appear to be collaborating well. Scale-up is progressing, indicating collaboration between Pajarito Powder and the synthetic efforts of UNM and NEU is going well. Most of the partners have a history of collaborating and have worked well together in previous projects. It is not clear how much FuelCell Energy (FCE) has been involved in the project to date.
- The project features an excellent collaboration between NEU and UNM on catalyst development. Advent is a critical partner in supplying materials and helping with MEA integration. Pajarito Powder will participate in scaling up the catalyst but does not seem to have been significantly involved as of yet. The role of FCE is presumably in fuel cell testing, which has not yet commenced but this role should have been described more explicitly.
- NEU has excellent interaction with its technical collaborators. Integrating the technology developed in this effort with that of others in the fuel cell program may be difficult. The compatibility of the catalysts being developed with Nafion-type electrolytes seems uncertain (it is noted that this is not a focus of the present work).
- The project involves several organizations with complementary skills. The role of FCE is unclear, though.

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 to the Program as it is following a promising pathway to improved performance and decreased cost of HT-PEM fuel cells. Furthermore, advancements in PGM-free catalysts developed through this project may prove helpful in developing PGM-free catalysts for low-temperature PEM fuel cells as well. If successful, the project could have a large impact in accelerating commercialization of stationary fuel cell technology.
- The project could have a substantial impact on the cost of HT-PEM fuel cells and the cost of stationary fuel cells. The project impact on low-temperature PEM fuel cells or fuel cell vehicles is expected to be minimal. The efforts focused on phosphate poisoning are specific to phosphoric-acid-based fuel cells. The MEA issues and low oxygen and proton transport are also related to phosphoric acid and are unlikely to transfer to low-temperature PEM fuel cells.
- High-temperature fuel cells will have significant impact on the combined heat and power (CHP) application, as proposed.
- By addressing the HT-PEM cathode catalyst challenge, this project is well aligned with the Program goals for CHP systems.
- If successful, the project results would be very meaningful for the Program mission because of the following advantages: 1) the application of non-PGM would enable cost reduction of fuel cell components and 2) HT-PEM fuel cells would alleviate the CO positioning and heat management.
- If successful, the project will align well with the goals and objectives of the Program.

- The project is relevant to the Fuel Cell Technologies Office's goal.

Question 5: Proposed future work

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

- The proposed future work includes optimizing the MEA fabrication. This is very important and will address some issues.
- The proposed future plan is great as the PI prioritized the MEA design to further improve the catalyst performance in a fuel cell. The approaches to improving the MEA performance are very detailed. The other plan the PI should also discuss is the modification of the project's original targets to DOE targets.
- Future work is focused on improving MEA and electrode design, attempting to address the mass transport and iR losses and address flooding issues. The durability of these PGM-free catalysts in HT-PEM systems is a question. PGM-free catalysts have not had the durability required for low-temperature PEMs. It is not clear that the higher temperature involved in HT-PEMs will not accelerate corrosion of these systems. The project should address durability of these catalysts.
- The chief problem seems to be mass-transport-related and the team has identified ways to address this. It is also important to improve the activity of the catalysts.
- The future work presented included a good discussion of the planned electrode development work but discussion of durability studies was surprisingly absent. The milestones table indicates significant work that will be performed on characterizing and validating durability so it is not clear why this was not described in the future work. Further improvements to the catalyst and scale-up efforts were also not discussed though it appears that these are intended to be part of the Year 2 effort.
- This project needs radical solutions to the challenges identified to date. The performance is much below the interim targets. Proposed MEA optimization (Teflon content, MEA annealing conditions, tweaks to hot-pressing) is not likely to ensure significant progress. There seems to be no Plan B.
- The PI identified no clear research path to overcome the major gap between the current catalyst performance and the project goal.

Project strengths:

- The project brings together an excellent team with significant experience and expertise in PGM-free catalysts and HT-PEM fuel cell development. The approach and the planned work represent a promising path to achieving project goals and accelerating the commercialization of stationary fuel cells.
- This is an innovative approach to eliminating PGM from fuel cells. The use of HT-PEM fuel cells is also beneficial toward addressing the implementation of batteries into fuel cell technology, especially the transportation sector.
- This is a great team with excellent experience in individual areas. Other strengths include the diversified non-PGM catalyst development and the in situ XAS characterization and systematic RDE design.
- The project partners have a strong collaboration. The project team has unique expertise in PGM-free catalysts and catalyst characterization techniques.
- The team members are all well-established researchers and bring to the project complementary skills that are essential to the project's success.
- A strong team and skillful catalyst characterization have been this project's biggest strengths.
- This is a good characterization effort providing interesting insight on the active site.

Project weaknesses:

- Catalyst compatibility with traditional PEM fuel cell electrolytes is a weakness (but is not a focus of this project).
- The project is relatively new but it has already missed two milestones and the proposed path for delayed completion of these milestones is not convincing. While characterization of transport losses is supposed to be a significant part of the project, the effort in this area seems weak so far, and the reported mass transport overpotentials and loss mechanisms do not seem accurate or realistic.

- Given the high level of funding for universities, project achievement has fallen far behind schedule. This is particularly true since the project is based on previous DOE-funded work. The reported catalytic activity is well below the expectation.
- The project should focus on the major deliverables: MEA performance, especially when these targets are not yet met. In this context, the basic characterizations, such as Mossbauer and x-ray absorption near edge structure (XANES)/x-ray photoelectron spectroscopy (XPS) should be less emphasized.
- The fuel cell performance, particularly for hydrogen/air, was not satisfactory. It is hoped that the performance can be improved in the second year following the “future plan.”
- There is little flexibility in the approach, especially in confrontation with lower-than-expected MEA performance, calling for sweeping solutions.

Recommendations for additions/deletions to project scope:

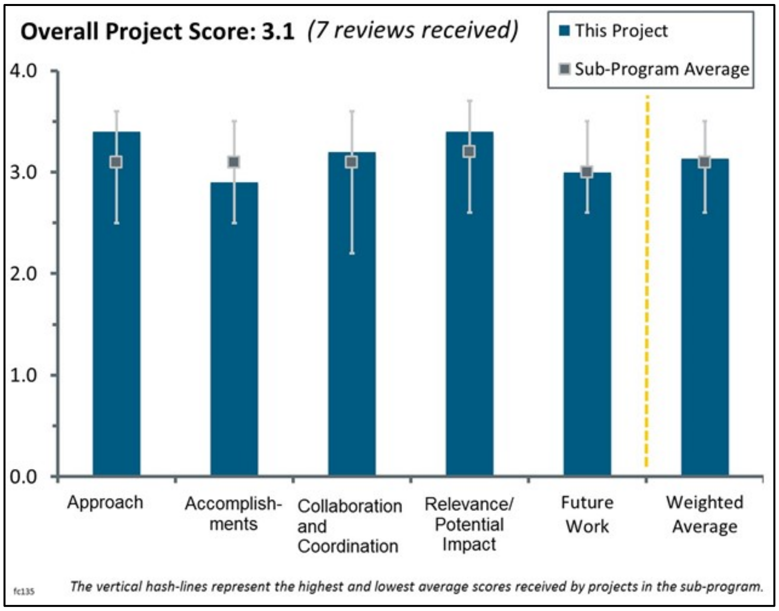
- The team needs to assess the origins of poor cathode performance. It is not clear whether flooding or simply insufficient activity of non-PGM catalysts is the cause. There is growing evidence in the field of non-PGM electrocatalysis that encapsulated metal salts and other metal-rich phases in non-PGM catalysts play no role in the oxygen reduction reaction. Such catalysts may actually be less active than formulations with highly dispersed iron or another transition metal. The team should identify the most promising formulation and focus on it in the second year of the project.
- The technical status, as of now, is significantly below the project targets. At this point, the PI should focus on addressing catalyst performance improvement.
- The report should clearly name the different types of catalysts and use consistent labels so that it is easier to follow the characterization data and MEA performance of each catalyst. The non-MEA (synthesis/scale-up) milestones were not specifically mentioned as being met.
- The PI should demonstrate the performance of alternative catalysts from UNM.

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

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.



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Question 1: Approach to performing the work

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

- The approach of bringing in the resources and talents of five national laboratories and many different suppliers and academia in a consortium to harmonize the fuel cell activities and share/understand the merits/limitations of available state-of-the-art (SOA) materials/technologies is a well-thought-out approach. This approach will certainly help with synergy in research and development (R&D) activities in different organizations and with developing a common understanding of all SOA materials/technologies, which may help the fuel cell community to have a consensus in the status of the SOA materials/technologies available from different suppliers. The overall structure of the consortium seems to be logical and includes furthering the scope by integrating new members into the consortium.
- The FC-PAD approach with six thrust areas is excellent and designated to national laboratories with the appropriate core capabilities. Coordination between these thrust areas (as shown in the presentation as an example) is well structured and thoughtful. Barriers are very relevant and clearly focused on catalyst layers—the current industry need—as a key component.
- The overall approach of the project consists of modeling and characterization of fuel cell components to improve their durability and performance. The fact that the objectives are split between the different thrust areas (characterization, performance testing, and modeling of different cell components) is a very efficient way to fully understand the related loss of performance and to address the durability issues.
- The proposed approach is excellent for advancing the performance and durability of PEMFCs, developing the knowledge base, and for optimizing structures for more durable and high-performance PEMFC components. The approach is excellent because it aims to benefit the fuel cell community by providing a better understanding of materials evolution in the membrane electrode assembly (MEA) components.
- The FC-PAD approach appears to be a good method to get even more collaboration and interaction between various fuel cell researchers at the national laboratories (although it is impressive that most of the laboratory researchers have been reasonably collaborative lately). What is missing is even more engagement with industry and universities (although new awards should help), but the amount of outside participation is already good.
- FC-PAD is a strong effort to coordinate/bring focus to a wide array of researchers to address the technical barriers to fuel cell development.

- Of the techniques shown on the capabilities highlights slide, the following may duplicate efforts/capabilities of some component developers: catalyst activity measurement, advanced MEA fabrication (especially with a bench-scale ultrasonic spray system), electrode simulations, multiphysics multiscale models, advanced MEA diagnostics, and transport property measurements. Of the techniques shown on the capabilities highlights slide, the following likely complement efforts/capabilities of component developers: analytical electron microscopy (beyond conventional SEM scanning and transmission electron microscopy [TEM]), advanced x-ray techniques (beyond x-ray diffraction [XRD], x-ray fluorescence [XRF], x-ray photoelectron spectroscopy [XPS]), x-ray tomography, and bulk and thin film morphology and properties. A high fraction of the effort should be devoted to techniques that may not be immediately available within to a stack developer. Certainly x-ray and neutron beams are part of this, but there could also be some electrochemical or fuel cell techniques that veer considerably off the beaten path. The combinatorial cell at Argonne National Laboratory serves as one example. Different types of segmented cells also come to mind, especially if the segmentation includes a diagnostic technique analyzed by segment (e.g., gas crossover, cell resistance, limiting current, etc.).

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 first- and second-quarter deliverables of all five national laboratories in FC-PAD have been completed, and some of the publications have been submitted or are in the process of submission. This shows good technical progress and accomplishments, and the team is on track to meet the deliverables for fiscal year 2016. It is good to see that the team successfully launched the website in time, which is a great accomplishment as this helps external customers (would-be members) to get information about the consortium and its activities.
- This is the beginning period of FC-PAD, and overall, all the national laboratories have made very good progress in the given time. The focus is clearly aligned with research needs. Most of the quarterly milestones have been met.
- Since the project start, the team has been very productive between the various thrust areas. A huge amount of data has been collected, and the project team seems to be highly integrated and collaborative.
- Overall, the FC-PAD team has already done an impressive amount of work. However, it is recommended that the FC-PAD overview presentation should *not* attempt to present any of these results in any detail (especially because most of the different thrust areas are presented individually). Instead, this overview should focus on (1) what the team has decided to focus on over the past year (or appropriate period), (2) why the team focused on these topics (vs. other options), (3) how the work was assigned to different thrust areas and how the different groups are interacting, (4) high-level key learnings and how they affect what will be focused on next, and (5) the future focus of FC-PAD.
- The project clearly addresses the DOE targets in terms of durability, performance, and cost (at a lower level). The durability studies concern the components (gas diffusion layer [GDL], cathode catalyst layer [CCL], MEA, membrane) and not (or perhaps not yet) the stack/system. Therefore, to assess the durability target set by DOE, validation at system level should be undertaken. Several consortium milestones have been completed on time.
- FC-PAD has a great start. The progress of vetting new consortium collaborators is an issue; this may have taken too long to implement (about six months).
- Results with regard to FC-PAD thrusts apply to other projects and should be evaluated in those project reviews. It appears the progress in this project accounts for the establishment of a website as well as numerous meetings and presentations. There are some operational points of progress that should be noted, however. No mention is made as to whether the non-disclosure agreement for working with all five laboratories has been established. It is not clear how data are to be managed other than by using the website for reporting data. It is unknown whether FC-PAD will have proper data security when needed. File transfer websites are not trivial and need to be set up. No progress is shown in this respect. The model that derives from FC-APOLLO (or other past modeling efforts) should be incorporated. It would be good to see what the strategy might be with regard to model inventory.

Question 3: Collaboration and coordination with other institutions

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

- Because the consortium brings in all the important research organizations that are involved in fuel cell R&D, there will be ample opportunities for the consortium members to conduct collaborative work and technology/knowledge transfer for mutual benefit of the member organizations and hence toward the advancement of fuel cell technology. The list of institutions involved in the consortium (slides 19 and 20) is impressive and given the fact they are all connected to one another, the consortium is expected to foster many new collaborative activities that otherwise would not have been possible.
- The main purpose of FC-PAD seems to be to increase national laboratories' interaction with industry and academia and use national laboratory core competencies to support industry and academic technology development. Current FC-PAD members (national laboratories) are working with many collaborators and will work with DOE-funded project teams.
- The project seems well organized with strong coordination (a director and deputy director) of the overall consortium and a coordinator for each thrust area. The consortium seems to have good collaboration with external partners, including laboratories, universities, and international institutions with complementary expertise—and there is a future plan to integrate new organizations.
- The project has collaborated with many partners. This project is highly collaborative with the overall aim of servicing the fuel cell community.
- FC-PAD is a vehicle for collaboration; it will have great interaction.
- Collaboration among the FC-PAD members appears to be excellent. However, there are a couple of minor areas in which more coordination between different thrust areas could be improved, e.g., ionomer studies and electrodes. Interactions with outside groups are very good at this phase without any more formal engagements in place.
- Obviously, collaboration is everything to FC-PAD, but for this particular part (FC-135), the evaluation should be done based on the collaborations relevant to the mechanics of setting up FC-PAD. What is relevant to consider is how well the national laboratories are collaborating with each other to establish legal frameworks, data management, and objectives. There appear to be some difficulties in setting up data management and non-disclosure agreements, which points to a lack of collaboration between laboratories. It would also be good to see the FC-PAD laboratories able to collaborate with DOE user facilities that may be administered by the Office of Science, NNSA, and other agencies.

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 creation of the FC-PAD consortium is a great Fuel Cell Technologies Office initiative with a goal to provide technical expertise and harmonize activities of national laboratories with industrial developers. This initiative is relevant to the objectives of the Multi-Year Research, Development, and Demonstration Plan (MYRDDP). The activities are aligned with DOE's goal to address the commercial barriers, such as performance, cost, and durability.
- The key technical barriers are performance and durability, especially (1) beginning of life performance with SOA catalyst loadings at high current densities and (2) high-activity alloy catalyst durability, which is not meeting targets. The FC-PAD team is certainly focused on addressing these key issues and more.
- The FC-PAD consortium and project is critical to the Hydrogen and Fuel Cells Program and has the potential to significantly advance progress toward DOE research, development, and demonstration goals and objectives with respect to improving the performance and durability of PEMFC systems.
- The main objectives are improving fuel cell durability and performance while lowering cost. These are the most important targets set by DOE in the MYRDDP. The main mechanisms of component degradation are addressed in the different thrust areas for a better understanding leading to a better mitigation solution.
- If successful, this project will make important strides toward addressing issues of cost and durability for fuel cell implementation.

- The objectives of FC-PAD align well with the DOE R&D objectives.
- There are some serious difficulties with understanding the relevance of FC-PAD. The key question is who the customer is, i.e., who is benefiting from this effort. Of the partners that are listed so far (outside the FC-PAD laboratories themselves), there are nine catalyst or MEA suppliers, five universities, four international research institutes, one U.S. government laboratory, and General Motors (GM). The suppliers are providing materials, but to prevent public disclosures of confidential technology developments, the materials are not likely to be the suppliers' latest and most cutting-edge. Except for GM, none of these partners will be designing unit cells and stacks. It is the designers of unit cells and stacks that would be thought to have the most to gain from the knowledge acquired by the project, but that does not appear to be developing. GM has had a long relationship with DOE fuel cell efforts, so its partnership could be expected, but automakers that have recently introduced vehicles are not part of this effort. Membrane stabilization is often obtained through additive packages that are highly confidential. It is unclear how a public project will be able to build upon supplier efforts that have already contributed to SOA membranes. Optimizing structures apart from the use of a fairly high-volume catalyst-coated membrane, GDL, or bipolar plate production line would appear to be, at best, an academic exercise. Developing a knowledge base about materials and structures, performing fuel cell tests, and modeling performance and durability appear to be what happens inside an automotive stack developer. This project must avoid being a duplication of effort with automakers.

Question 5: Proposed future work

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

- The overall proposed research for the consortium and for individual thrust areas are well-thought-out, thorough, and aligned with DOE goals. The project covers individual goals and necessary activities required to address the challenges related to components (catalyst, electrode, ionomer, GDL) and methods (characterization, modeling, operational conditions).
- The proposed future work to define mechanisms for collaboration is appropriate, as is identifying supporting roles for the FC-PAD core national laboratories.
- The proposed future work for project organization corresponds to the immediate needs of the project.
- The project has planned its future in a logical manner. There are issues with the timeframe of adding collaborators and the duration for incorporating new collaborators; incorporation should be streamlined.
- FC-PAD is only few months old, so more time may be needed to make the future work planning concrete.
- The future work for the Consortium should not be very brief summaries of the future focus of the various thrust areas (because this is a repeat). Instead, future work should be a high-level explanation of the overall strategy of FC-PAD, specifically what the key remaining technical barriers are, how the Consortium is going to prioritize these barriers, and what new capabilities FC-PAD may require to better address these barriers.
- The Future Work slide addresses the future work of the thrusts but does not adequately address the future work associated with setting up FC-PAD. There are obvious mechanics of collaboration that are still missing at this stage. The future work should focus on what the individual laboratories are going to do to make sure they work better with each other and with all the partners. The focus should be on streamlined processes for working with national laboratories as well as data sharing and security. There should also be some consideration of how to involve more stack developers in the work.

Project strengths:

- The project has access to a considerable amount of equipment and resources. The project has managed to generate interest from at least nine MEA or MEA component suppliers. The investigators have combined years of experience in fuel cells that can probably be measured in centuries.
- The main strength of the project is the team, which constitutes the ensemble of all subject matter experts required for such broad activities. The team has all necessary technical expertise and equipment resources needed to conduct the proposed R&D for FC-PAD.
- FC-PAD is utilizing each of the participating national laboratory's core competencies to meet the DOE R&D goals and addressing the current research barriers. Dividing into six thrust areas to cover all the required fields/needs to advance fuel cell technology is well received.
- The project appears to be further improving collaboration between the national laboratories. The investigators of the Consortium and the thrust areas are outstanding. The new website is a strength.
- Having strong national laboratories with complementary expertise is a good guarantee for success. Splitting the tasks into different thrust areas allows the project to treat each one of the components and its degradation mechanisms in a rather complete and deep way.
- Collaboration with researchers with varied backgrounds is a strength.
- The project's strengths include the excellent research and the unique capabilities of the core team.

Project weaknesses:

- No weaknesses were specifically identified; however, the mechanisms for collaboration and cooperation with industrial partners and data management can be a challenge in terms of impact as the FC-PAD activities are coordinated across six different thrust areas.
- Although engaging different commercial entities in the consortium and getting their SOA materials/ technologies for evaluation is a very ambitious initiative, it will be very difficult to manage such activities unless a robust intellectual property/non-disclosure/confidential disclosure agreement is in place. The team should have clear understanding of the intellectual property ownership and legal pitfalls that often come with such a broad coalition of R&D entities.
- The project will likely have difficulty accessing SOA materials sets. The project is lacking a clearly defined customer. The probability of overlapping stack developer efforts is very high. The project cannot guarantee access to user facilities other than those of the Office of Energy Efficiency and Renewable Energy. There are still some matters to sort out with regard to data-sharing and streamlining the interactions between a given party and all five core laboratories.
- Integration of new partners and coordination of the whole consortium could be a weakness if strong communication means are not clearly set.
- The path forward to work with DOE-funded project teams is not clear. The extent to which the collaboration with new partners will be made is not clear.

Recommendations for additions/deletions to project scope:

- It will be interesting to see how the project milestones for the FC-PAD national laboratories related to newly awarded projects evolve and how outreach and impact evolve.
- The work focuses on components and the related degradation mechanisms and studies each one of them in a rather deep and complete way. The question is whether the methods developed (models and characterizations) are useful in dissociating some close degradation mechanisms (for instance, catalyst growth/agglomeration mechanism and carbon corrosion). Otherwise, the results do not enable a better understanding of these degradation mechanisms and how they affect each other. Validation at system level with real-world operation does not seem to be part of the future work. This could be helpful in validating the improvements done on different components within a complete stack/system. The project should investigate the effect of simultaneous occurrence of degradation mechanisms (for instance, at catalyst and membrane level) vs. the impact of each of them individually on cell durability and the potential consequential impact on the accelerated stress test.
- Extra effort should be directed toward secure data management for all partners. Emphasis should be put on finding ways to streamline the process of working with all five national laboratories. The project should

attempt to remove all activities that overlap with what stack developers are doing. The project should seek fuel cell testing techniques that are novel and go far beyond just taking polarization curves and running diagnostics. The project should also seek running diagnostics in a segmented fashion, in situ material characterization, and other things that only a national laboratory might be able to devise. If stack developers are doing something, national laboratories should seek to go deeper and further to go beyond that "something" and add to it.

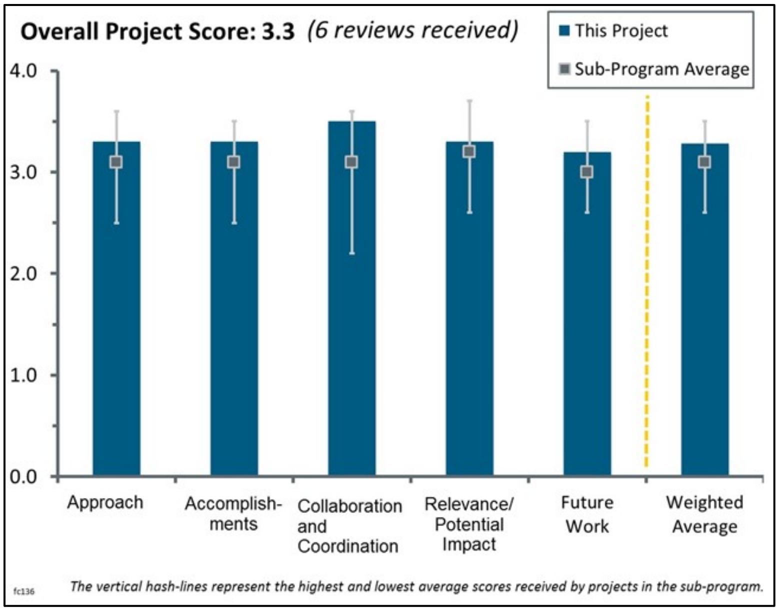
- The project should allow new collaborations on a one-year project effort with an optional one-year follow-on based on performance review. If an activity does not make it to the second year, others would be given an opportunity to contribute.
- The presentation on the Consortium should be more of a high-level strategy explanation.

Project #FC-136: Fuel Cell Consortium for Performance and Durability – Electrocatalysts and Supports

Debbie Meyers; Argonne 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 **3.3** for its approach.

- The team has correctly approached the problem of understanding the catalyst and support degradation mechanisms, understanding mutual interactions between the catalyst and support, and quantifying the impact of catalyst degradation on cell performance using high-resolution transmission electron microscopy (TEM) and x-ray diffraction (XRD) techniques supported by modeling inputs. The team is equipped with subject matter experts and state-of-the-art (SOA) equipment to conduct these studies.
- Catalyst stability and interaction with other electrode components are not fully understood and research on these topics is key to achieving performance, cost, and durability targets. The approach is well designed: elucidating catalyst and support degradation mechanisms as a function of catalyst and support physicochemical properties and cell operating conditions and quantifying the impact of catalyst degradation.
- FC-PAD is a strong effort to coordinate/bring research focus to address the technical barriers to fuel cell development. The focus of this work is on electrocatalysts and catalyst supports. This work could possibly have the highest impact toward achieving cost and durability targets of the U.S. Department of Energy (DOE) Hydrogen and Fuel Cell Program (the Program).
- This is Thrust Area 1 of FC-PAD. The objectives and approach are very clear.
- The overall approach is excellent since it emphasizes focusing on fundamentals and understanding of mechanisms. However, one aspect that appears to be missing is a subsequent goal to distill these learnings into recommendations to the community on how to improve performance and durability of these key components. For example, after determining the key issues with PtCo and PtNi, the team should provide recommendations on how to improve these materials. The project should determine whether the recommendations for these two different alloys would be the same or different.
- Most of the project focuses on taking a roster of commercial catalysts, as well as catalysts developed in DOE-funded projects, and using them to develop fundamental degradation relationships with respect to voltage or voltage cycling. This presents a few problems. First, while the study has been very high-quality and the results are very systematically organized, the results are very familiar to stack developers. The project needs to stay away from doing a very good job studying what stack developers already know. Second, most of the results (not all) are carried out in glass-cell or ex situ environments, which are different from a fuel cell environment in terms of water activity, proton activity, and many other factors. Third, the

project needs to show validation that the relationships derived from ex situ techniques are relevant to what occurs in a fuel cell. To some extent, this is being pursued for carbon corrosion although factors such as temperature and relative humidity also need variation to provide information that developers can use. While the attempt is made to generate systematic degradation data, the different ways in which suppliers treat their catalysts will introduce noise factors to what otherwise would be fairly clean trends. Some accounting needs to be made for this in any modeling efforts.

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 team has made significant accomplishments toward the objective of identifying Pt alloys that meet or exceed the DOE 2020 ORR activity target (>440 mA/mg Pt). The results from potential cycling to determine the Pt dissolution rate are also very interesting. The Pt dissolution rate was found to be lower for IRD Fuel Cells (IRD) “spongy” Pt_3Co compared to TKK “solid” Pt_3Co, whereas the Co dissolution rate is much higher. This is a great piece of information and clearly shows the merit and limitations of spongy and solid catalysts. The results from the study of thermodynamic and kinetic rates of PtOx formation are relevant to the understanding of the long-term durability/stability of individual catalysts. The measurement of carbon corrosion during drive cycle on three types of carbon supports is also relevant for the goal of understanding the stability of catalyst supports.
- Excellent progress has been made in the given time and with a given set of catalysts under DOE projects. Much work has been done on the PtCo catalyst system and carbon supports.
- The accomplishment of the team is solid and well-coordinated, and is taking methods and results from other projects into account.
- The FC-PAD electrocatalyst and catalyst support thrust already has a wide group of partners/collaborators contributing to baseline work. This is/was an important step for evaluating the new collaborators. The crossover between work from other thrust areas (electrode layer and ionomer/gas diffusion layer) may not be completely defined.
- The number of accomplishments to date is impressive. However, with respect to the presentation, a summary of the oxidation and dissolution mechanisms would be very helpful (there are seven slides on this topic but no summary). The summary on carbon corrosion (slide 23) is great and a good example of what should have been included on the catalyst loss results.
- The systematic study of degradation with voltage or upper voltage limits is very good but there are many other parameters associated with practical fuel cell operation: humidity, temperature, pressure (which translates to oxygen activity), and flow rates. If the information generated in this project is to be of any practical use, it must be incorporated into a model with some ability to predict lifetime. Such a model will prompt questions as to how degradation rates change, not just with voltage and different particle sizes but also with temperature, humidity, ionomer content, Pt weight percent, and other operating conditions or design factors. For what the study has attempted to do, it is very good. The dissolution rates of Pt versus voltage are well plotted and agreeable with other data. The carbon corrosion information also corroborates with other data. However, the unfortunate part of this is that for a publicly funded project, saying the results agree with other data means that others have already studied the same phenomena. With the results as they are, it may be possible for a developer to incorporate some trends into a model that presumes a very limited set of catalyst layer design parameters and operating conditions but that may be the full extent of the usefulness of the data. The project needs to think deeply about what it is that developers still need to predict stack lifetime.

Question 3: Collaboration and coordination with other institutions

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

- Since the consortium brings in all the important research organizations that are involved in fuel cell research and development (R&D), there will be ample opportunities for the Consortium members to conduct collaborative work and technology/knowledge transfer for the mutual benefit of the member

organizations and hence toward the advancement of fuel cell technology. The list of institutions involved in the Consortium is impressive and given the fact they are all connected to one another, this project is expected to foster many new collaborative activities that otherwise would not have been possible.
- A positive thing is that the project has engaged most of the most likely catalyst suppliers, but others should also be considered. Automakers and other stack developers need to be better engaged so that the project can understand what data should be delivered to enhance models for predicting stack lifetime. The project should determine, for example, whether there are temperatures or temperature cycles that need to be considered, how humidity should be varied, and how catalyst layer design has an influence. It is good to see a wider representation of the other DOE-funded project materials. The University of South Carolina materials have needed to be included in a project like this for a while.
- There is good collaboration with industry and other academic partners. Collaboration with other thrust areas is mentioned but not evident from results. Collaboration should not be limited to catalysts and supports developed under DOE projects only.
- There are good, expanded collaborations between material and component developers and existing collaborations are well-maintained.
- FC-PAD is a vehicle for collaboration.

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

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

- The creation of FC-PAD is a great Fuel Cell Technologies Office initiative with a goal to provide technical expertise and harmonize activities of national laboratories with industrial developers. This initiative is relevant to the objectives of the Multi-Year Research, Development, and Demonstration Plan (MYRDDP). The activities are aligned with DOE's goal to address the commercial barriers, such as performance, cost, and durability. This project is in the thrust area "Electrocatalysts and Supports," whose objective is to realize the ORR mass activity benefits of advanced Pt-based cathode electrocatalysts in high current density, with air performance for over 5,000 operating hours, and with low0PGM loading (<0.1 mg Pt/cm^2). The objective is aligned with the MYRDDP.
- This thrust area is very well focused and well executed to overcome major obstacles in fuel cell performance and durability.
- If successful, this program will make important strides toward addressing issues of cost and durability for fuel cell implementation.
- The team's focus on durability issues is warranted based on the current status of SOA PEMFCs.
- The project's aspects align well with DOE R&D objectives.
- It is difficult to see how the thrust area becomes relevant to advancing the Program. Suppliers deliver catalysts that are not SOA for testing and then trends that may have already been familiar to developers years ago. This is the fundamental problem with being able to say that the project ultimately supports Program objectives. As with other areas of FC-PAD, the project suffers from not doing enough up front to identify customers and deliverables. It is not clear who the customers are or what they need. If the customers are identified to be stack developers, many of whom are automakers, the unfortunate truth is that many of these developers are already very familiar with the potentials at which various modes of Pt oxidation and dissolution occur; developers are familiar with surface oxides on carbon and have models to describe not only CO_2 evolution but also how it varies with Pt weight percent, carbon types, temperature, humidity, and other factors. One example of what a developer might need is a quick screening tool that is ex situ and can predict catalyst lifetime. However, this project is not presently driving toward such a goal or objective.

Question 5: Proposed future work

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

- The team's proposed future research is aligned with the overall objectives of the Consortium. The study of IRD and Umicore Pt_3Co catalysts using TEM-EDAX, x-ray fluorescence (XRF), extended x-ray absorption fine structure (EXAFS), Delta-μ x-ray absorption near edge structure (XANES), oxygen permeability, carbon corrosion, and Pt dissolution studies will provide better understanding of how these catalysts behave under different cell operational and accelerated stress test conditions.
- Future work is well-thought-out and clearly focused on addressing the challenges for PtCo and understanding the degradation mechanism. A PtNi or catalyst system will not be very different from the PtCo system, but other newly developed high-ORR catalysts should be added to the future work. Future work is technically focused but does address the way the project will work with DOE-funded project teams.
- The project has planned its future in a logical manner. It is not clear whether specific decision points will be made with respect to collaborator performance. This effort should allow for great research toward achieving Program objectives.
- Studies of effects such as Pt dissolution as a function of carbon type and correlation with changes in particle size distribution and electrochemically active surface area could be expanded to include studies of a broader range of Pt alloys.
- More work on PtNi, especially dealloyed PtNi, would appear to be warranted based on results shown with these materials.
- There are many experiments listed that are functions of voltage but not of temperature or water activity (with the exception of oxygen permeability in catalyst layers). Many of the experiments listed drive toward understanding degradation rates of Pt, carbon, or Pt in the presence of different carbon types. However, the rates themselves must be validated through insertion into a cell model and then through subsequent cell testing. The feedback loop is not well represented here, which calls into question what the overall deliverable might be. If the overall deliverable is to measure degradation rates only, this may be good for generating publications but not for assisting developers that may already have insight on degradation rates. The claim is that Delta-μ analysis on XANES will be done in collaboration with General Motors (GM). It is not clear whether the project has the capability to perform Delta-μ without GM's assistance.

Project strengths:

- The project is able to carry out degradation testing in a very systematic fashion, perhaps even better than most other DOE-funded projects. The project personnel are very skilled in analysis of catalysts and have numerous resources available to them. Numerous catalyst suppliers are listed as partners. Data presented for Pt and carbon degradation are corroborated elsewhere.
- The overall proposed research for the Consortium and for individual thrust areas is well-thought-out, thorough, and aligned with DOE goals. The project covers individual goals and necessary activities required to address the challenges related to catalysts and catalyst supports used in different commercially available and developmental catalysts.
- The approach to addressing the key challenges to understand the durability issues is very clear. Academic and industry collaboration is good, with most of the high-ORR catalysts under study or planned for study.
- This project has a strong team that seeks input from collaborators to remain relevant and to effectively achieve advanced fundamental understanding.
- A project strength is the major focus on durability (instead of performance) of catalysts and supports, which is good since this is a metric that is not being met, especially with high-activity Pt-alloy catalysts.
- Collaboration is a strength.

Project weaknesses:

- The project lacks an overall direction as to who the customer is and what is to be delivered. There needs to be further direction to understand how data will be used. Suppliers will likely not be compelled to deliver their most SOA catalysts. Many of the data generated will show trends already familiar to developers.

Experimental design needs to be more open to variations in operating conditions and catalyst layer design factors.

- Although engaging different commercial entities in the Consortium and getting their SOA catalysts for evaluation is a very ambitious initiative, it will be very difficult to manage such activities unless a robust intellectual property/non-disclosure/confidential disclosure agreement is in place. The team should have clear understanding of the intellectual property ownership and legal pitfalls that often come with such a broad coalition of catalyst manufacturing and user companies.
- There are no criteria or mechanism set for future collaboration with DOE-funded projects. Newly developed facet- and/or shape-controlled catalysts should be included in the study. The collaboration mechanism or extent of collaboration with other FC-PAD thrust area members is not well defined.
- The project should be cautious in the work to apply a catalyst corrosion model to membrane electrode assembly (MEA) data. The interaction and interplay between other MEA components should be considered.
- There is a lack of simple summaries of learnings to date, which should ideally also include recommendations on knowledge to date (e.g., whether one should utilize solid or spongy alloys).

Recommendations for additions/deletions to project scope:

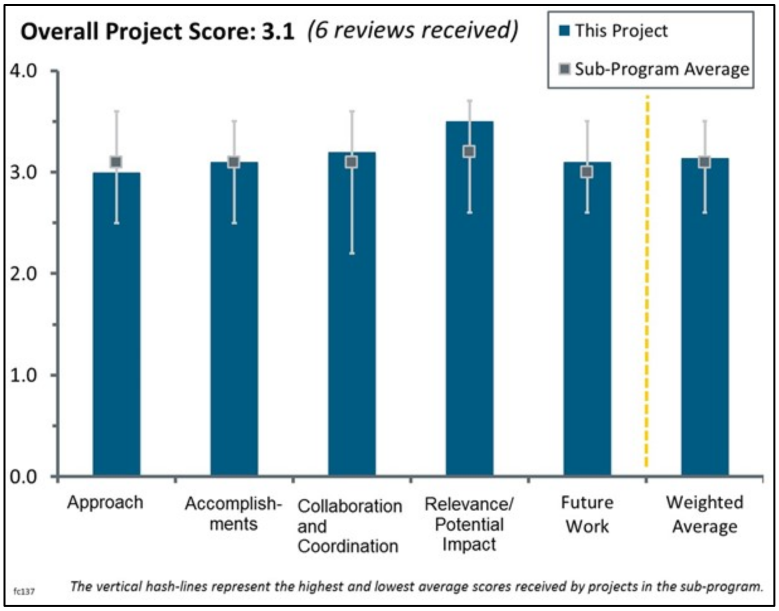
- Stronger communication and collaboration with other FC-PAD members (national laboratories) on methods such as in situ TEM to understand the degradation mechanism will be very helpful.
- It would be interesting to include mitigation strategies in the future work on applying a catalyst corrosion model to cell data using TEM-EDAX and XRF quantification of Pt and Co in cell components. Overall, this is well-executed and very interesting work.
- The project should remove tasks that appear to overlap with what developers are already doing or that do not contribute to an overall deliverable. Customers should be identified and the project should find an overall objective or vision of how customers will use data. The project should collaborate with other FC-PAD thrusts to understand how data could be used and validated versus cell testing. Emphasis should be added on other cell operating conditions such as temperature and humidity.
- In future presentations, the project should include more brief summaries of key learnings (and less detail). In scope, the project should be sure to strive to make recommendations to the community on how to make future improvements.

Project #FC-137: Fuel Cell Consortium for Performance and Durability – Electrode Layer Integration

Shyam Kocha; National Renewable Energy 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.



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Question 1: Approach to performing the work

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

- The team has correctly approached the thrust-specific objectives to understand and mitigate the issues with electrode layer integration by identifying state-of-the-art (SOA) catalysts, optimizing them in catalyst layers, developing diagnostics to help resolve the high-current-density/low-loading problem, and mitigating the problem through the use of novel electrode design, novel components, and novel diagnostics techniques, all complemented with modeling.
- This is one of the key thrust areas of FC-PAD, focusing on catalyst layers. The approach outlined to address the barrier is clear and excellent. The overall approach to take learnings from rotating disk electrode (RDE) study to membrane electrode assembly (MEA)-catalyst layer optimization of SOA catalysts with the help of modeling is very good idea.
- FC-PAD is a strong effort coordinating/focusing research to address the technical barriers to fuel cell development. The focus on electrode layers and integration may address issues with durability. Through FC-PAD, this focus area should be integrated with electrocatalysts and catalyst supports.
- The proposed approach in the thrust area is appropriate because it engages numerous partners and models novel electrode designs and diagnostics.
- The focus on the cathode catalyst layer is definitely warranted since catalyst activity targets are being met but high-power-density targets are not being met. The approach, as outlined on slide 7, is good; however, it does not appear that this approach is really being executed. In particular, #1, #2, and #4 on slide 7 are being pursued but not so much #3. In particular, it appears that the team has already decided that the high-current-transport issue is due to the ionomer film. There is no clear evidence presented supporting this hypothesis. The first priority should be to investigate the different hypotheses that have been proposed. The team should first focus on #3 before moving into #4.
- From an overview, the approach sounds logical, but there appear to be many details missing. Yes, the overall power density needs to be met, and there are losses at lower loading that do not appear at higher loading. The project intends to address these by developing new structures, which is good. But what would be interesting to know is how the project will approach developing new structures, especially during ink

processing. The project should define particular ink properties that are of interest and techniques that will be developed to look at how particles aggregate in ink and how this might affect the resulting structure. There is a deeper level of detail that would enhance what has been identified as the approach. The use of two phases for proton transport is interesting and was well-explored in a prior project (FC-125). However, there is a question here as to whether this will become a dominant theme in this project or whether there are other ideas that might provide a more facile change with existing ink processing and application techniques.

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 prepared and evaluated all three SOA catalyst layers. All three have met the DOE mass activity oxygen reduction reaction (ORR) target of 440 mA/mg-Pt. However, the rated power target was not met. Progress has been made on understanding transport through the layer using diagnostic tools and modeling. The team has also demonstrated that the ORR kinetics can be separated from the ionomer effect and true ORR can be measured. The comparison of SOA catalyst performance of three commercial catalysts in slide 17 and other analytical results is a great way to help catalyst developers to improve performance of their catalysts.
- Much progress has been made in the previous similar projects from industry; the national laboratories, academic partners, and principal investigator acknowledge that. Many of the SAO catalysts have been identified, and some very good progress has been made in fundamental studies regarding hydrogen contaminant detector (HCD) diagnostics, oxide dependency.
- Interesting work is presented, and progress has been made on understanding transport through the layer using diagnostic tools and modeling.
- Most of the work shown, particularly that with in situ cell testing, overlaps with what stack developers will do. The inability of high-mass-activity catalysts to achieve high-current-density targets is well understood. Stack developers frequently analyze catalyst layers using limiting current and proton pump techniques. More would need to be known about the nanofibers and the balance of the catalyst layer used for the nanofiber experiments to extract information that could be generalized for all catalyst layers and the fundamental limitations of performance. The microstructural modeling, in conjunction with x-ray tomography, might be a step ahead of developers, particularly in the modeling of liquid water movement in a catalyst layer. Dry imaging and mapping of catalyst layers is slowly becoming familiar, but a good model that can predict movement of condensed water would constitute significant assistance for developers. No accomplishments were shown with respect to how catalyst layers get made and how processing affects structure.
- The FC-PAD Electrode Layer Integration thrust has already shown baseline data that address electrode layer design. The crossover between work from other thrusts (e.g., electrocatalysts and ionomer/gas diffusion layer) may not be completely defined.
- There is too much focus on possible mitigation strategies before establishing the actual limiting mechanisms.

Question 3: Collaboration and coordination with other institutions

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

- Since the Consortium brings in all the important research organizations involved in fuel cell research and development, there will be ample opportunities for Consortium members to conduct collaborative work and technology/knowledge transfer for mutual benefit and to advance fuel cell technology. The list of institutions involved in the Consortium is impressive, and given the fact they are all connected to one another, it is expected to foster many new collaborative activities that otherwise would not be possible.
- This is one of the important thrust areas in FC-PAD, which addresses the key barriers for catalyst layer integration and optimization; close collaboration with other FC-PAD members and industry stake holders is very necessary. The project approach outlines and also exhibits this close collaboration.

- The collaborations are very good and should continue.
- FC-PAD is a vehicle for collaboration.
- The team has done a good job interacting with industry suppliers and has obtained a good variety of materials. However, it is not clear how much the team is interacting with the rest of the FC-PAD team. For example, Lawrence Berkeley National Laboratory (LBNL) shows that MEA ink solvent has a major impact on ionomer and catalyst distribution; however, the National Renewable Energy Laboratory is not studying how this affects the performance of the catalyst layer. Conversely, LBNL should focus on ionomer interfaces.
- Collaboration appears to be a weak point for this aspect of FC-PAD: four catalyst suppliers and General Motors are noted for supplying materials, but it is not clear that the collaboration is deeper than that. While there is collaboration with the other FC-PAD laboratories, collaboration with the other FC-PAD laboratories is a baseline for the project. The project needs to collaborate better with outside developers to understand what needs to be delivered.

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

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

- The creation of the FC-PAD Consortium is a great initiative by the Fuel Cell Technologies Office, with a goal to provide technical expertise and harmonize activities of national laboratories with industrial developers. This initiative is relevant to the objectives of DOE’s Multi-Year Research, Development, and Demonstration Plan (MYRDDP). The activities are aligned to DOE’s goal to address the commercial barriers such as performance, cost, and durability. This project is on the thrust area “Electrode Layer Integration,” whose primary objective is to integrate SOA electrocatalysts that meet or exceed the DOE mass activity targets of 440 mA/mg Pt and optimize the catalyst layer to attain the DOE peak power density requirements of 1W/cm^2 and 0.125 g-Pt/kW while simultaneously meeting durability targets. The objective is aligned with the MYRDDP.
- While FC-PAD suffers overall from a need to identify customers and deliverables, this particular thrust addresses an area that is highly relevant to lowering cost: enhancing performance at high current density with improved catalyst layers. The relevance slide speaks to activities that have a chance to go beyond what stack developers already do. There is mention of developing new diagnostics and capabilities that perhaps developers have not yet done.
- Potential impact from realizing the activities and durability of SOA catalysts in MEA by integrating them in MEA and optimizing the catalyst layer is very high. The project is clearly focused on key challenges for catalyst layers.
- The project is targeting the main challenges in fuel cell development: improving performance and durability. Diagnostic tools and modeling can be useful to understand transport through the layer and have an impact on designing or optimizing catalyst layers.
- If successful, this project will make important strides to addressing issues of cost and durability for fuel cell implementation.
- The team is definitely focused on a key barrier and has the potential to make significant impact, but the approach could be improved.

Question 5: Proposed future work

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

- The proposed future research aligns with the overall objectives of the Consortium. The MEA screening of remaining SOA catalyst materials is logical. Optimization of catalyst layers to achieve peak beginning-of-life performance for promising candidates is necessary for identifying true high-performing SOA catalysts.
- Key future work already included—catalyst layer (CL) optimization with SOA catalyst to meet performance, alternative CL designs, relevance of kinetics to HCD, and alternative ionomers—is very important.

- Model performance diagnostics data at high current densities and identifying and implementing alternative ionomers in catalyst layers and examining effects on performance will improve understanding.
- The project should define how the team will “optimize catalyst layer” without first obtaining rigorous fundamental understanding of what mechanisms are limiting the performance. The team should consider doing some systematic fundamental studies to prove or disprove possible hypotheses for the HCD limitation. For example, to probe the impact of ionomer or ionomer film, the team could make a series of MEAs with identical parameters except the equivalent weight of the ionomer in the CL (or, alternatively, different solvents in the MEA ink) and see if these parameters have a dramatic impact, as one might expect if the ionomer is the key. This should, of course, be compared with modeling results.
- The project team needs to define how the catalyst layers will be optimized in fiscal year 2016, the design principles, how layers will be optimized if processing/structure relationships are unknown, and tasks associated with understanding how ink properties and processing parameters affect the resulting catalyst layer structure. In principle, identifying alternative designs for catalyst layers is a good idea, but there needs to be understanding of how to achieve those designs. Future work should include examination of inks and processing parameters.
- The project has planned its future in a logical manner. The decision points between other focus areas as well as evaluating collaborator performance needs to be better defined.

Project strengths:

- The overall proposed research for the Consortium and for individual thrust areas is well conceived, thorough, and aligned with DOE goals. The proposed research covers individual goals and necessary activities required to address the challenges related to electrode layer integration of catalysts available from different commercial and developmental catalyst sources.
- Project strengths include close collaboration with remaining FC-PAD members and industry partners; clear understanding of the current status of the technology, not reinventing the wheel; and consideration of most of the key parameters needed to improve the CL to meet the targets.
- Project strengths include the strong team and well-thought-out work plan and its approach.
- The team has the capability and materials required to make SOA MEAs.
- Collaboration is a project strength.
- The project understands that high-current-density performance is lacking with highly active catalysts. The project’s access to equipment and facilities within the national laboratories is excellent. Another strength is the use of microstructural characterization to begin to understand—at least on a dry, ex situ basis—how a catalyst layer is structured.

Project weaknesses:

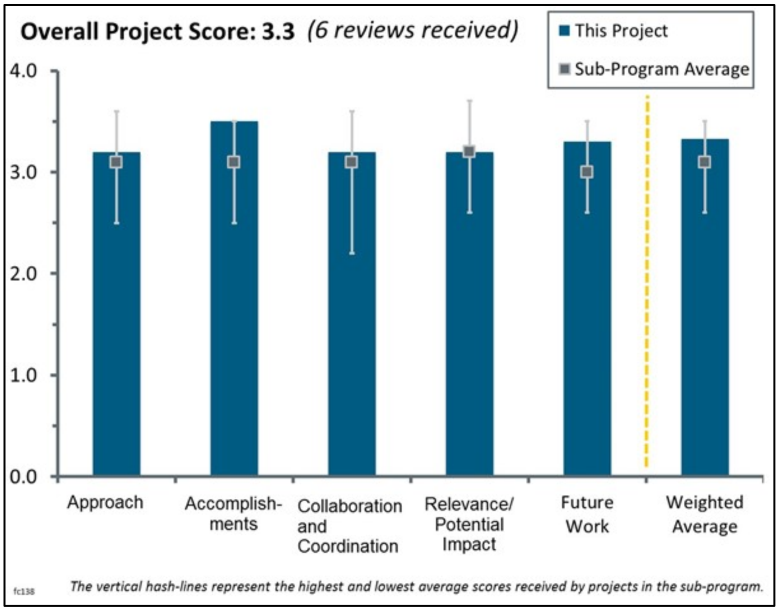
- To this point, no weaknesses were specifically identified for this project.
- A concern is that while the project recognizes the overall problem, there may be too much emphasis on in situ analysis and fairly novel ways of approaching the problem, especially when so much about conventional catalyst layers still remains unknown—other than what can be measured in situ. The project needs to think more broadly about what developers do not have access to, or what still remains a mystery about conventional catalyst layers.
- Although engaging different commercial entities in the Consortium and getting their SOA catalysts for evaluation is a very ambitious initiative, it will be very difficult to manage such activities unless a robust intellectual property/non-disclosure/confidential disclosure agreement is in place. The team should have clear understanding of the intellectual property ownership and legal pitfalls that often come with such a broad coalition of catalyst and MEA manufacturing companies.
- There is no evidence presented that the team can actually make SOA MEAs. There is an overemphasis on mitigation strategies instead of first obtaining a fundamental understanding of the root cause for what is limiting the performance at high current densities. The project is presenting unvalidated modeling results (slide 23) as a motivation for a certain approach.

Recommendations for additions/deletions to project scope:

- In general, the project needs to shift from a heavy focus on evaluation toward a better understanding of relationships between processing and structure, structure and layer properties, and layer properties and performance. There is still much that is unknown about what happens in the ink, what happens as ink is applied and dried, and what happens to generate the resulting catalyst layer structure. The increased use of microstructural characterization and modeling to understand the movement and phase changes associated with condensed water in a catalyst layer would benefit the project.
- It would be interesting to see whether kinetics actually come into play at high current densities; the project should examine effects on performance and relationship to durability studies/accelerated stress tests on catalysts (FC-136) coupled to the catalyst/ionomers in electrode layers.
- There should be more focus on fundamentals, which should be used to demonstrate SOA performance and potential mitigations or improvements.

Project #FC-138: Fuel Cell Consortium for Performance and Durability – Ionomers, Gas Diffusion Layers, Interfaces

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 (PEMFCs). FC-PAD efforts include six complementary thrust areas, three of which are involved in this project, which entails modeling, evaluation, and characterization of fuel cell components for performance and durability improvements. The components include catalysts, electrodes, and ionomers/gas diffusion layers (GDLs).

Question 1: Approach to performing the work

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

- The project is using operando, modeling, and ex situ diagnostics to elucidate governing behavior and optimize performance and durability in ionomer membranes, ionomer thin films, GDLs, and respective interfaces. This is the correct approach to understand how these components communicate and, hence, perform at their best. Identification of any obstacle to these communications (interactions) is bound to have an impact on performance/durability. The team has taken the correct approach of identifying and mitigating issues involved in individual components and their interfacial junctions.
- FC-PAD is a strong effort to coordinate/bring research focus to address the technical barriers to fuel cell development. This is the third thrust area, which focuses on ionomers, GDLs, and interfaces. This thrust area can have direct impact on both performance and durability barriers to fuel cell development. As part of the Consortium, this project integrates well with other efforts.
- This approach is focusing on several sub-component systems, such as catalysts, electrodes, and ionomers/GDLs, using operando, modeling, and ex situ diagnostics to elucidate governing behavior and optimize performance and durability. The approach is critical and important.
- This is one of the key thrust areas in FC-PAD’s focus on addressing challenges that are overlooked or that have not been prioritized in the past.
- The approach is excellent, with focus on multiple key components and an emphasis on many different types of studies and tools. However, one aspect that appears to be missing is a greater focus on ionomers under the “Interface” category, as these interfaces appear to be hypothesized by some (including other FC-PAD members, such as the National Renewable Energy Laboratory [NREL]) to be important contributors to the transport losses at high current densities with ultra-low catalyst loadings.
- The approach described in the presentation is very general—optimizing performance and durability can lead down many different pathways. What the approach lacks is discernment about which tasks developers need and which tasks have already been covered by fuel cell stack developers. The approach also lacks a defined purpose. For example, all of this is unclear: why transport properties for ionomer thin films are being measured, whether they will be used in a model, whether a developer will be using them in a model, or whether they will be validated in some sense to be relevant for fuel cell operating conditions.

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

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

- Significant progress has been made considering the short time span. Though most of the work comes from previous or ongoing projects, the understanding provided for membrane interfacial resistances, water behavior, cerium migration, and ionomer thin films is commendable.
- The overall progress and accomplishments are good. Good progress has been made toward model transport and of microporous layer–catalyst layer and GDL–channel interfaces.
- This FC-PAD thrust area has already shown very good progress with collaborators. This work should be enhanced when the next round of funded collaborators are announced.
- There have been impressive results in a short amount of time.
- The ultra-small-angle x-ray scattering technique demonstrates a good method for characterizing the size of ionomer particles in inks. This may be of assistance to developers in understanding processing–structure relationships. The elemental mapping of a catalyst layer is good, but it is unclear whether this was developed in this project. The weak phase separation of ionomer thin films at very low thickness is useful to understand. It would be useful to developers to have confirmation that this weak phase separation results in thin film properties that affect performance. State-of-the-art (SOA) membranes are fairly complex; therefore, an analysis of changes in mechanical properties of Nafion® with aging may or may not be relevant. Trends with water uptake and alpha relaxations with aging are qualitatively understood by developers. Cerium migration from the membrane into catalyst layers has already been observed by stack developers. For a thorough study, cerium content in effluent water should be checked. The membrane interfacial resistance study could be very useful if combined with a model that would cover one of the more difficult things about fuel cells: predicting hydration throughout a unit cell. Such a goal would be worthwhile, but the objectives of the project do not clearly state this. Nevertheless, there is the beginning of something useful here. Some questions should be asked as to whether the use of a humidity sensor is the best way to go about some of the resistance measurements here, or whether a segmented high-frequency resistance (HFR) technique might be good for validation.
- The experimental evidence of solvent effect and processing conditions for different ionomers is interesting. The separation of different particle sizes with increasing temperature for water dispersion of Nafion® (slide 6) is an interesting phenomenon. It will be interesting to see whether Nafion® dispersion behaves in the same way in alcohol and water or whether the presence of alcohol helps molecules of different sizes to come closer to make larger agglomerates. The d-spacing results for 3M and Nafion® ionomers in slide 8 seems to suggest some effect of side chain size, especially for the bulk membrane d-spacing. It will be nice to see the effect in Solvay's Aquivion® membrane, in which the side chain is much smaller. The aged membrane effect is understandable; however, it is not clear whether this is a true representation of the aging process that the membrane suffers under fuel cell operational conditions. It is very unlikely that the membrane can undergo sulfonic acid dimerization under fuel cell operational conditions unless the membrane is very thick and the cell is running very dry. For a thin membrane (Gore® type), it is very unlikely that the membrane will undergo conductivity loss due to sulfonic site loss or dimerization mediated loss. The cerium washout depends on how the cerium has been imbibed into the membrane. From the slides, it is not clear how the cerium was imbibed into the membrane.

Question 3: Collaboration and coordination with other institutions

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

- Because the Consortium brings in all the important research organizations that are involved in fuel cell research and development, there will be ample opportunities for the Consortium members to conduct collaborative work and technology/knowledge transfer for the mutual benefit of the member organizations and, hence, toward the advancement of fuel cell technology. The list of institutions involved in the Consortium is impressive, and given the fact they are all connected to one another, the project is expected to foster many new collaborative activities that otherwise would not have been possible.

- Collaboration with other FC-PAD members is excellent, which is undoubtedly due to the high degree of interactions between the different national laboratories prior to FC-PAD.
- There is good engagement with current partners, including collaborations with universities and industrial partners.
- The presentation does not provide credit on a task-by-task basis to collaborators, so it is very difficult to judge the quality of collaboration. The GDL images appear to be similar to what investigators from Tufts would produce, but no attribution is given. 3M collaboration appears to be just ionomer material inputs. This project sorely needs collaboration with a stack developer to understand what developers have already done and what has not yet been done. There is much good the project could do if it is focused properly.
- Based on the progress shown, it is difficult to know the extent of collaboration with other FC-PAD members and other partners. Modeling is Lawrence Berkeley National Laboratory's (LBNL's) core competency, so it looks like most of the work is done at LBNL.
- FC-PAD is a vehicle for collaboration.

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

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

- FC-PAD is a great Fuel Cell Technologies Office initiative, with a goal to provide technical expertise and harmonize activities of national laboratories with industrial developers. This initiative is relevant to the objectives of the Multi-Year Research, Development, and Demonstration Plan (MYRDDP). The activities are aligned with DOE's goal to address the commercial barriers, such as performance, cost, and durability. This project is in the thrust area "Ionomers, GDLs, Interfaces," whose objective is to focus on fuel cell components, their diagnostics, structural characterization, and modeling for both performance and durability improvements. The objective is aligned with the MYRDDP.
- The overall focus of this project—fuel cell components, their diagnostics, structural characterization, and modeling for both performance and durability improvements—is relevant and a vital key to reach the DOE 2020 targets.
- The impact of these studies will be significant, as this thrust area addresses some important challenges that were not prioritized in the past.
- If successful, this project will make important strides toward addressing issues of cost and durability for fuel cell implementation.
- It is not obvious how this work is focused on addressing the two major technical targets that are not being met: (1) balance-of-plant (BOP) performance with SOA catalyst loadings at high current densities and (2) durability of high-activity alloy catalysts. The principal investigator (PI) could do a better job of communicating how this thrust area is contributing to solving these key issues. Currently, it appears that the work just continues to focus on topics of interest to the PI (ionomer thin film properties and GDLs), but it is not evident how the work may result in significant improvements in the two issues noted above.
- The presentation did not discuss relevance. Furthermore, the presentation did not describe clear goals and objectives. There is some acknowledgement that understanding membrane additive migration and voltage losses associated with thin film ionomers would be good, but outside of this, the objectives of the project are not clearly stated. The FC-PAD effort suffers as a result of a lack of identified customers and deliverables. This particular thrust conforms to this trend.

Question 5: Proposed future work

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

- The proposed future research by the team is aligned with the overall objectives of the Consortium. Investigation of side-chain chemistry and governing structure–property correlations, especially the impact of reinforcement, will help in understanding the impact of side chain chemistry on phase separation and ionomer thin films. The model study to elucidate interactions during solvent evaporation with different solvents will be very helpful in understanding the different stages of phase changes on perfluorosulfonic acid dispersion in transitioning from dispersion state to semi-fluid state to dry polymer state. Understanding of the cell conditioning protocols and their impact on various membrane electrode assembly components will also be helpful in understanding how these components behave during the cell start-up, either at BOP condition or after intermittent shutdown.

- The proposed future work is well aligned with the progress made in the first project period. Developing a thin film structure–property model is of high importance. The initiative to study and develop model interactions and examine scale coupling is relevant.
- Proposed future work is very detailed and essential for the success of this activity.
- FC-PAD allows each thrust to incorporate research findings efficiently. The decision points for including work from collaborators are not completely defined.
- The membrane work is interesting, but it is not likely to have an impact on overcoming the barriers for commercialization of fuel cells. Cerium migration habits are fairly well known. The structure of an SOA perfluorinated sulfonic acid membrane is not something that has a high impact on high current performance or even on durability. The GDL modeling aspect of the project should involve experimental validation, if it is not included already, and it should be done for GDLs with SOA thicknesses. Work related to ink dispersions, ionomer thin films, and membrane interfaces is relevant to overcoming barriers associated with cost, robustness, and lifetime. However, these efforts need to be focused by understanding what specific goals and objectives exist. There also needs to be some understanding of what is most useful in light of the fact that access may be limited with respect to actual manufacturing methods and SOA materials.
- More focus on ionomer interfaces should be added.

Project strengths:

- The project is able to explore some areas that other projects have not, such as membrane interfacial resistance to water transport, ionomer thin film properties, and properties of ink dispersions. These are all areas that are relevant. The project has access to an extraordinary amount of characterization equipment and techniques. Unlike other projects, this project is capable of carrying out a systematic study in which phenomenological boundaries are defined. An example is the investigation of phase separation with ionomer film thickness and equivalent weight.
- The overall proposed research for the Consortium and for individual thrust areas is well-thought-out, thorough, and aligned with DOE goals. The research covers individual goals and necessary activities required to address the challenges related to ionomer, membrane, GDL, and related interfaces used in different commercially available and developmental ionomers, membranes, and GDLs.
- The PI and his team are strengths. A major strength is the challenges this team is addressing; the work will result in very good understanding.
- The focus on fundamentals is a project strength.
- The project features a good team.
- Collaboration is a project strength.

Project weaknesses:

- The project is engaging different commercial entities in the Consortium, but it will be very difficult to manage such activities unless a robust intellectual property/non-disclosure/confidential disclosure agreement is in place. The team should have clear understanding of the intellectual property ownership and legal pitfalls that often come with such a broad coalition of membrane/ionomer/GDL manufacturing and user companies.
- The project has not identified goals and objectives. The project has not interfaced with customers, and it has not defined deliverables. The project does not understand what work it has done overlaps with stack developers and what does not. Collaboration is either fairly light or was not well identified in the presentation.
- To bridge the understanding between interfaces and relations is a challenge. The project would benefit from increased interaction with industrial partners and original equipment manufacturers.

Recommendations for additions/deletions to project scope:

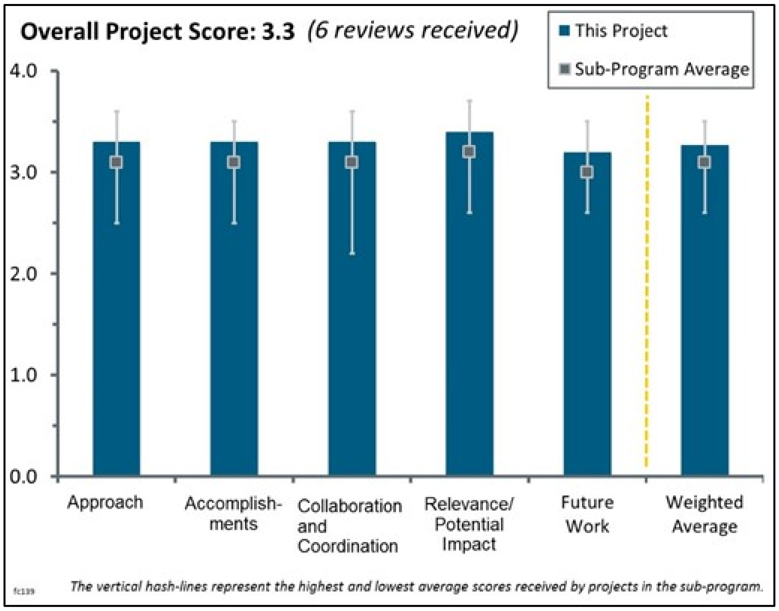
- The project could probably shed the tasks associated with cerium migration, reinforced membrane casting, and GDL modeling. These tasks are less likely to be associated with overcoming barriers to commercialization. The project needs to add considerable levels of collaboration with stack developers in order to identify goals, objectives, and deliverables. The project should maintain emphasis on membrane interfacial resistance and perhaps even consider developing a unit cell hydration model generic enough to assist a wide range of developers, thus allowing developers to figure out how to maintain hydration and stack robustness while limiting the size of the BOP.
- The present investigation of the membrane side chain chemistry and governing structure–property correlations using membrane interfacial resistance measurements is slightly unclear. It is recommended that the project pay careful attention to this.

Project #FC-139: Fuel Cell Consortium for Performance and Durability – Modeling, Evaluation, Characterization

Rangachary Mukundan; Los Alamos 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, three of which comprise this project: modeling and validation, operando evaluation, and component characterization. The project is developing advanced diagnostic, modeling, and characterization techniques to evaluate state-of-the-art (SOA) membrane electrode assemblies (MEAs) and provide insights to improve the durability of the MEA components.



Question 1: Approach to performing the work

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

- The team has correctly approached the operando evaluation and durability to refine accelerated stress tests (ASTs), impurity effects, evaluation of Pt-based MEAs, and development of advanced electrochemical characterization techniques to integrate the analytical results from other thrust groups to conduct modeling on the components.
- FC-PAD is a strong effort to coordinate/bring research focus to address the technical barriers to fuel cell development. The focus of this work covers thrust areas 4 to 6. This effort integrates well with other thrust areas of FC-PAD by providing the performance and durability evaluation of the outputs from thrust areas 1 to 3.
- The approach is strongly focused on meeting U.S. Department of Energy (DOE) targets on development and implementation of characterization techniques and models with the aim to improve performance and durability.
- This thrust area supports the first three areas of FC-PAD with evaluation and characterization. The major efforts are on durability evaluation and developing ASTs to shorten the time.
- There is excellent focus on fundamental understanding in a variety of important topics.
- The development of a combined membrane AST is a direction in which many developers have already proceeded. What is needed instead is a faster mechanical test. A test that isolates mechanical stress is a worst-case scenario for membranes with inadequate swelling properties and, therefore, can be used to define the boundary on how much in-plane swelling is too much. This is needed information for suppliers, and it usually cannot be deconvoluted from a chemical–mechanical combined test. The mechanical test is too long and needs to be shortened. Rather than presuming that the 20× acceleration factor is universal for all materials, it would be interesting to see this approached for an array of Pt particle sizes, ionomer/carbon ratios, carbon types, loadings, and other design parameters. Development of ASTs for PtCo may be of interest, but uncovering failure modes is something that should only be done in conjunction with a stack developer. There are many different types of PtCo, and therefore, PtCo failure modes can vary widely depending on how the cell is operated. The modeling appears to be premised on breaking down mass transport losses into constituents associated with Knudsen diffusion and ionomer thin films, but this kind of task is already being pursued by stack developers. The study of recoverable losses associated with

membrane fragments or SOx was part of a stack developer request. However, the developer was already aware of the association with membrane degradation and with recovery at low potential. The approach to this task should add to what is already known. It is not clear why most of the testing is getting carried out in a serpentine cell or whether a four-channel cell will approximate the same channel flow velocity found in full-size automotive cells or cells for other applications.

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.

- Thorough benchmarking of various ASTs from across the industry is commendable and a great deal of work. Significant progress has been made in terms of AST development and alloy catalyst evaluation and characterization.
- The project made good progress on proposed refined new ASTs, durability of the Pt alloy, and studying and quantifying reversible/recoverable degradation.
- This effort has shown good progress toward evaluating MEAs during cycling. The project has identified degradation mechanisms for evaluation.
- The development of a faster AST for electrocatalyst cycling is good work if the acceleration holds for a variety of cathode catalyst layer parameters. This may assist in shortening development time. The investigators did provide a combined chemical–mechanical test that is shorter than the mechanical test. However, 500 hours is still very long for a combined chemical–mechanical test versus other such tests that exist. The PtCo trends shown with Umicore and IRD catalysts are similar to what has been shown throughout the community. Particularly, it is well known that electrochemical surface area (ECSA) can decrease to a limited extent before performance is affected and that larger particle sizes lend themselves to lower ECSA loss. The presence of Co in the membrane has also been observed. The project needs to go deeper to better understand how industry could do even better to stabilize more active PtCo catalysts. A considerable amount of the modeling context appears to be missing. It is not clear how the model can account for both wet and dry conditions or how well-validated and predictive it is for a wide range of operating conditions. Data from studying recoverable losses appear to mirror what General Motors has already been presenting at the Durability Working Group since December 2014. The project needs to determine how it can proceed deeper to overcome barriers associated with this.
- In terms of percentage of ECSA loss, the old AST seems to be aligned with the Fuel Cell Technical Team drive cycle. The new AST is much more aggressive and does not correlate to the actual fuel cell drive cycle. It is not clear that there is any need for the new AST. It may be faster, but it does not represent the actual degradation pathway that the fuel cell follows during its operation. The study of SOx fragments needs more attention. Typically, SOx fragments are generated when the MEAs are operated under very dry and high-temperature conditions, and the duration of the exposure to such harsh conditions determines the extent of SOx that may form in the MEA. The ex situ result of reduced oxygen reduction reaction (ORR) due to SOx poisoning may look interesting; however, the team members should make sure that in mimicking this condition to develop the AST, they are not overestimating the extent of SOx that may be present in the MEA.

Question 3: Collaboration and coordination with other institutions

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

- Because the Consortium brings in all the important research organizations that are involved in fuel cell research and development, there will be ample opportunities for the Consortium members to conduct collaborative work and technology/knowledge transfer for the mutual benefit of the member organizations and, hence, toward the advancement of fuel cell technology. The list of institutions involved in the Consortium is impressive and, given the fact they are all connected to one another, the project is expected to foster many new collaborative activities that otherwise would not have been possible.

- There is excellent collaboration with both other FC-PAD members and with industry. In addition, international collaboration with the National Physical Laboratory (NPL) appears to be resulting in useful new capability.
- There is strong collaboration among FC-PAD members and external collaborators.
- FC-PAD is a vehicle for collaboration. This effort ties into thrust areas 1 to 3.
- There is clearly a high degree of collaboration.
- Reference electrodes can be very useful in studying fuel cells, but at the moment, it is difficult to see where the reference electrodes from NPL are being implemented for tasks that will help to overcome barriers. Furthermore, the reference electrodes are being implemented in a serpentine cell, which comes with its own questions, especially with regard to how channel flow velocity matches flow velocity in a full-size cell. General Motors, Gore, IRD, and Umicore appear to be materials suppliers. Further depth in these collaborations does appear to be represented in the slides. Collaborations with Ion Power and Tanaka Kikinzoku Kogyo (TKK) are unclear.

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.

- FC-PAD is a great Fuel Cell Technologies Office initiative with a goal to provide technical expertise and harmonize activities of national laboratories with industrial developers. This initiative is relevant to the objectives of the Multi-Year Research, Development, and Demonstration Plan. The activities are aligned with DOE’s goal to address commercial barriers such as performance, cost, and durability. This project is on the thrust area “Modeling, Evaluation, Characterization,” whose objective is to develop and implement characterization techniques and models to improve performance and durability of fuel cells.
- Testing and evaluation will provide the final data to determine quantitatively whether thrust areas 1 to 3 have been successful. Findings from this effort can guide future efforts in thrust areas 1 to 3. Thus, this effort will make important strides toward addressing issues of cost and durability for fuel cell implementation.
- Durability continues to be a major barrier, especially with respect to high-activity alloy catalysts at ultra-low catalyst loadings.
- This is an extremely relevant project and is critical to advancing technology toward the DOE 2020 goals.
- Better, faster, and relevant ASTs can shorten the development time. This thrust area is key in developing those.
- The relevance of the project is premised on measuring durability as well as on developing the ASTs by which durability can be estimated on a component basis. Fuel cell system developers are capable of measuring durability, extending durability with system mitigations, and measuring component durability with either their own ASTs or with more widely adopted ASTs. Therefore, a considerable amount of the project exists to serve the public interest in understanding fuel cell durability. This is not crucial to fuel cell system developers, but it is within the scope of what DOE is trying to accomplish. The development of ASTs is, by itself, not necessary for overcoming barriers. Stack developers have been working with their own ASTs for decades. What the project needs to show is how to provide better ASTs that are shorter and still premised on the same failure modes that exist in a realistic drive cycle.

Question 5: Proposed future work

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

- The proposed future research is aligned with the overall objectives of the Consortium. A complete durability evaluation of PtCo-alloy-catalyst-based MEAs and complete development of a reference electrode set-up is the correct course. However, the evaluation of sulfate infusion’s effect as a function of potential and during durability cycling protocol is questionable. Quantifying the effect of reversible degradation under durability cycling protocol is a logical choice.

- Overall, the future work appears excellent. If possible, it would be good to see the team make more specific recommendations/suggestions on how the community might mitigate certain degradation mechanisms and demonstrate some of these mitigations.
- The proposed future work, as outlined on slide 23, is appropriate and will further progress toward DOE goals.
- This effort has an effective plan for evaluating novel MEAs. It is not clear when inputs from other FC-PAD thrust areas would be evaluated. An “as available” approach may not be effective/appropriate for the success of the FC-PAD effort. The interaction between the FC-PAD thrust areas may need better definition.
- The evaluation of PtCo and PtNi durability appears to be a direct overlap with stack developer work, and it is possibly being done with materials that have not been heat- or acid-treated to a state in which they might be considered to be SOA. In the case of PtNi, if it is from the DOE-funded work, there may be some SOA work proceeding, but the overlap with developer work still exists. This seems to be a benchmarking effort. It is not clear what cell will be segmented with reference electrodes to understand durability effects. If it is a quad-serpentine cell, this may have an entirely different hydration profile from a cell for a full-size stack. Yes, the voltage trends may be there, but hydration and temperature may have very different profiles. Adopting a differential cell for single-cell testing would be a great improvement over what has been proceeding. There is no way that stoichiometry sensitivities that exist for a small single cell will be the same for a cell for a full-size stack.

Project strengths:

- The project should continue to revisit, update, and disseminate AST protocols. Developing insights into loss of mass activity without loss in ECSA is a project strength, as is developing insights into increases in transport losses with loss of Co in PtCo alloys. New local reference electrode capability is being developed. The international collaboration with NPL is productive.
- The overall proposed research for the Consortium and for individual thrust areas is well-thought-out, thorough, and aligned with DOE goals. The research covers individual goals and necessary activities required to address the challenges related to modeling, evaluation, and characterization necessary for evaluating different commercially available and developmental fuel cell MEAs.
- The project has access to considerable characterization and test stand resources. The project personnel in the past have been able to systematically benchmark supplier materials with ASTs. The project has done a better job than other parts of FC-PAD in identifying specific goals and objectives.
- Collaboration and team members, especially the principal investigator, are key strengths of this project.
- Project strengths include efficient teamwork and a well-balanced approach.
- Collaboration is a project strength.

Project weaknesses:

- The project is still using quad-serpentine cells premised on reactant gases delivered in stoichiometric ratios (for most tests) instead of using differential cells. The project has limited access to SOA materials. It would help the project to have a broader understanding of how much variation can exist in catalyst layer design factors as well as in catalyst powders (even while keeping the composition essentially the same) and how these variations can affect durability. The durability of PtCo cannot be fully represented with just two flavors of PtCo. There needs to be greater clarity in how reference electrodes are to be implemented. The quality of collaborations with stack developers can be enhanced.
- Although engaging different commercial entities in the consortia and getting their SOA catalysts for evaluation is a very ambitious initiative, it will be very difficult to manage such activities unless a robust intellectual property/non-disclosure/confidential disclosure agreement is in place. The team should have clear understanding of the intellectual property ownership and legal pitfalls that often come with such a broad coalition of catalyst, MEA, and GDL manufacturing companies.

Recommendations for additions/deletions to project scope:

- There needs to be focus on decreasing the time needed for a mechanical stress test for membranes. A chemical–mechanical combined test cannot serve as a replacement. There needs to be better definition for the modeling work or else it should be removed. In general, the project needs to be able to move beyond what is already understood about failure modes such as those associated with PtCo or recoverable losses due to membrane fragments or SOx. So far, the project is confirming what is known, but it needs to move toward exploring mechanisms or toward working with collaborators to help validate solutions to these problems.

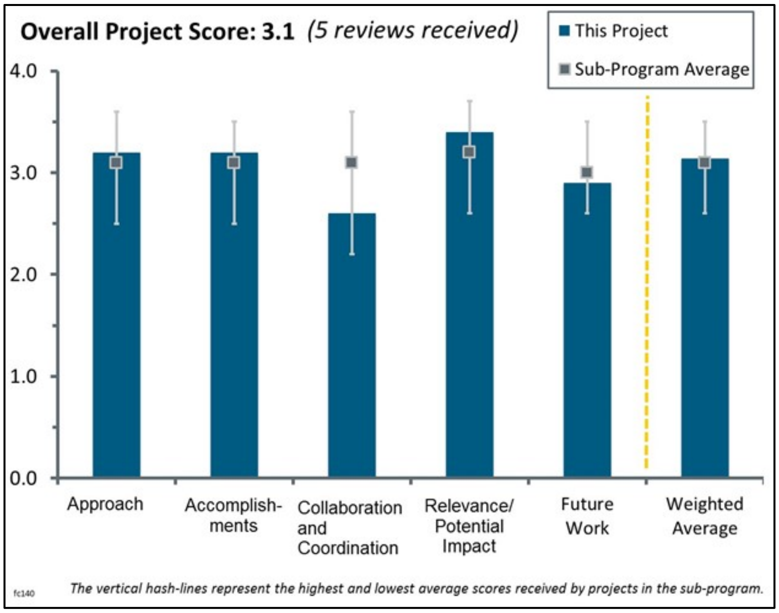
- For newer high-ORR facet-/shape-controlled alloy catalysts, ASTs may need further refinement. The correlation factor from AST cycles to stack durability is very important so it can be explored.
- The project should study the relationship between model durability of MEAs—under both AST and durability cycling protocols—and a real fuel cell system.
- There should be even more engagement with industry and universities (new awards from the latest funding opportunity announcement should help in this regard).

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 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 **3.2** for its approach.

- The project has an excellent approach that builds on outstanding fundamentals to inform the scale-up to complex systems. It would be preferable to have industrial partners to help with scale-up, such as making larger batches of catalysts, which is proceeding but could probably be done faster with companies that specialize in this area.
- The project has an excellent balance of fundamental electrocatalyst study, development of high-activity nanocatalysts, directly relevant analytical capabilities, and strong interaction with electrode optimization experts (Debbie Myers, Argonne National Laboratory [ANL]). The approach of incorporating catalyst scale-up is unique among national laboratories and is a substantial strength.
- The project aptly draws fundamental concepts of catalyst activity and durability into the Program through a coordinated program of catalyst preparation, very detailed characterization, and kinetic activity testing. The project needs to evolve into testing in membrane electrode assemblies (MEAs), reducing its reliance on rotating disk electrode (RDE) testing for activity and performance evaluation.
    - ANL has not paid adequate attention to the limits posed by local oxygen transport at high current density on the practical utility of low-loaded catalysts with very high area-specific kinetic ORR activity but low (<30 m^2/g) Pt specific surface areas. Unless new means of avoiding the local transport resistance can be developed, low-surface-area catalysts will not be cost-effective for applications requiring current densities of ~1 A/cm^2 or higher (which is most applications). ANL therefore needs to give increased attention to maintaining high specific surface areas. Caution should be exercised in thinking that the additional thermodynamic stability of ordered intermetallic compounds versus disordered alloys will necessarily give catalysts with superior durability. The experimental experience with such an approach has given at best mixed results.
- For AuX-PtNi, the concept of stabilizing the Au so that the Au can stabilize Pt is interesting. However, there is much in this concept that would have to go right, so the probability of success is very small. First, AuX cannot dissolve (as of now, it is unknown). Second, Ni still cannot dissolve. In many other PtNi species, this has been a problem, and it has been a problem outside of RDE cycling. Third, a particle has to be made that preserves the layering described for the thin film. For Pt_3Co or PtCo, suppliers have covered this type of particle well.

  - The m-SiO_2 on Pt_3Co is a very interesting structure, although it may suffer from Co dissolution. It is not clear how SiO_2 is eventually removed without affecting Pt_3Co.
  - For Pt nanoframes with better segregation of Pt on the surface, this catalyst will have to overcome what was revealed in the last project. It has to be scalable, and it has to perform well in a cell. Given the activity that has been measured, it is worth trying, but because it is a dealloyed PtNi, the probability of Ni leaching during MEA fabrication and the probability of low high-current-density performance are both high. There are no parts of the approach that discuss what will be done to provide for high performance at high current density. The use of vacuum processes does provide an opportunity to divert from processes that impose numerous acid/heat treatment steps or that introduce impurities.
- There is a real emphasis on fundamental understanding and improvement. The criteria for selection of which technologies and paths are followed are 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.

- Many impressive accomplishments have been made already, including a new experimental technique (RDE + inductively coupled plasma mass spectrometry [ICP/MS]) that is yielding very interesting results and MEA testing results with high performance, which already exceed some DOE targets.
- So far, the major points of progress have been the following: (1) the development of a thin film for AuX-PtNi that has shown stability in glass cell testing, (2) the formation of the three-dimensional novel structure for Pt_3Co using SiO_2, and (3) the enhancement of PtNi nanoframes with Pt segregation. This represents a fair amount of progress for a project that has been active since October 2015. However, none of these developments alone, or even in combination, represents a development that overcomes barriers to commercialization.
  - The scale-up of PtNi nanoparticles is reported to have reached 200 mg. It would be best for the project to move even more aggressively toward batch sizes that are multiple tens of grams. Collaboration with a supplier would be useful. The project needs to move more aggressively toward cell testing, especially for use in durability studies. Too many projects in the Program have been content to report results from RDE only over the first few years of a project.
- One accomplishment is the development of in situ external calibration (EC)-ICP/MS while providing tremendous insight into electrocatalyst durability. Development of a method to stabilize Pt through X-PtNi in a nanoparticle catalyst is very promising. It needs to be demonstrated in an MEA. It appears that a key structural factor leading to variable PtNi nanoframe catalysts has been identified, a critical first step. It is unclear whether the issue has been resolved. There has been excellent progress toward MEA integration of catalysts. MEA hydrogen/air performance is good for the low loading and relatively low electrochemical surface area. MEA mass activity exceeds the DOE target.
- The project's new real-time measurements of Pt dissolution provide critical insights into how to improve durability of Pt-based catalysts against voltage-cycling effects. The correlation of Pt dissolution rates with different surface atomic structures and extents of order should greatly improve the ability to rationally design catalysts with improved durability, and the analysis should be extended to alloy systems.
  - The project has placed the use of subsurface gold in improving the durability of Pt-based catalysts against voltage cycling onto a much firmer basis. The development of "additive X" to prevent Au from segregating to a Pt-based catalyst surface could provide a practical route to more durable catalysts. The identity of X should be communicated to the fuel cell community without undue delay because of patent-filing considerations. Patents filed by national laboratories tend to hinder, rather than promote, incorporation of national laboratory ideas into U.S. industrial development.
  - This project and its predecessors have promised catalyst scale-up at ANL for years without notable results to date. It is time to deliver on this or to give up. Closer ties with industrial firms that manufacture catalysts would likely be a better way to proceed. Modestly larger quantities of catalysts to allow MEA testing to largely replace RDE work are sorely needed, as the relevance of RDE results to real fuel cells is increasingly under question.

- The progress toward the stated goals is good, despite the reviewers' contention that performance must be in relevant systems: MEA, durable supports, and DOE accelerated stress test use.

Question 3: Collaboration and coordination with other institutions

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

- The partners included are good, and the collaboration appears to be good as well, based on the results to date. However, it is unfortunate (but understandable) that the team is limited to national laboratories. Hopefully, this will change soon with the addition of industry and universities to the Fuel Cell Technologies Office's (FCTO's) new consortium models.
- Collaboration with both national laboratories and universities is evident. Inclusion of industrial input is needed.
- Despite the mention of non-disclosure agreements signed with automakers, the majority of the catalyst synthesis and design work appears to be getting done at ANL. The slides speak to Lawrence Berkeley National Laboratory's role in scaling up catalysts, but the actual conceptualization appears to reside at ANL. Greater collaboration in conceiving new catalysts could help the project. Oak Ridge National Laboratory and Los Alamos National Laboratory collaborate, but their roles seem to be to carry out particular tasks for which they are well-suited: microstructural characterization and fuel cell testing, respectively. It does not appear that these partners play a role in strategizing the project approach. Interactions with automakers are represented in somewhat cryptic fashion in the slides. Perhaps even more helpful than automaker collaboration would be collaboration with an industrial catalyst supplier—or even a small company that can carry out vacuum deposition at higher scale—but that appears to be lacking.
- The project needs to improve its ability to get catalyst samples to other laboratories for testing. Selected catalyst types arising from this project need to be synthesized and tested in other laboratories to give full credibility to the results generated solely within ANL. 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 **3.4** for its relevance/potential impact.

- The project has adopted the FCTO Multi-Year Research, Development, and Demonstration Plan catalyst targets as its targets. The g/kW target implies that high-current-density performance has to be acceptable, while mass activity speaks to low-current-density performance. The project is also committed to meeting loading and durability targets. The project relevance is predicated on the relevance of Pt alloys. The one criticism that could be levied is that the Program has not been a stranger to Pt alloys for oxygen reduction. Dealloyed PtNi has been studied in morphologies such as nanoparticles, nanoframes, and nanowires and in nanostructured thin film catalysts. It can perhaps be said that DOE has relied far too heavily on Pt/base metal alloys, to the detriment of other possible materials. This project must show that there is new ground to be broken in the Pt alloy field.
- To meet the ultimate DOE targets, higher-activity catalysts with improved stability are certainly required; however, another major barrier has been the incorporation of these new catalysts (e.g., thin films) into effective high-performance MEAs. Fortunately, the project does include MEA work. However, it is not clear what the team is doing to understand the performance losses in the MEAs (e.g., transport and ohmic) to accelerate the scale-up to this key component.
- This project and its predecessors have been one of the primary sources of ideas for innovation in ORR catalysts for the entire industry. There is more to catalyst performance than kinetic ORR activity—this project needs to start paying attention to the needs for adequate active surface area to avoid excessive losses at high current density due to the apparent local oxygen transport problem. Very high area-specific activities can increase fuel cell efficiencies at low current density, but behavior at high current density is what currently defines the cost of a stack for a wide range of applications.

- The need for better catalysts is the focus of this work; more emphasis on full electrode impact is needed.

Question 5: Proposed future work

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

- The proposed work is excellent; the only recommendations would be to incorporate the following: (1) some scale-up with catalyst suppliers (e.g., industry) and (2) more MEA-level diagnostics to determine what is limiting the performance (this may also require additional collaborators).
- The presentation gave reasonable but overly general suggestions for future work.
- It seems early for this consideration because the continuous process at this stage is so different from what would be used for ton levels. For example, 10 g/car 5 kg per batch per day is 500 cars/day and ~200,000 cars per year. Hence, continuous operation catalyst synthesis is not necessary. The topics highlighted are good, but details are lacking, so commenting is very difficult.
- Most of the material in the presentation pertaining to future work seems to describe how all tasks will continue forward in parallel. New catalyst concepts, new synthesis, characterization, cell testing, and scale-up of synthesis—all of this will just keep going. There is no indication of how go/no-go decisions will be made, or which catalysts hold greater priority toward scale-up and cell testing. The project needs to have some strategy to know which experiments need to proceed first, as opposed to just presuming it is acceptable to proceed in the usual fashion from thin film to small powder batches to glass cell tests to characterization, and so on. Instead, questions should be asked early on. These questions should consider what is most likely to cause failure for AuX-PtNi, the novel structure of Pt_3Co, and what would prevent commercial adoption (e.g., inability to produce at scale). Once these questions are answered, the project should move aggressively and quickly to pursue the most challenging tests rather than wait until the final year of the project.
- The proposed “scaling up” work is not clear.

Project strengths:

- The alloys from which the project is building have been found in the past to have high oxygen reduction activity. The project has access to the very best materials characterization techniques that can be found in the national laboratory system. The project does have the ability to conceptualize novel structures such as the one found for Pt_3Co. The project personnel are some of the best electrochemists in world, especially in fuel cell science. The quality of the measurements is very high.
- The project’s greatest strength lies in its nearly unique ability to correlate surface structure and surface and near-surface composition (all at the atomic level) with ORR activity and durability. Real-time measurement of Pt dissolution is a very powerful new tool that increases still further the value of atomic-scale control of catalyst structure.
- The project strengths are the analysis of technical-like PtNi/C alloy annealing and performance, development of in situ EC-ICP/MS, balancing nano-/meso-scale characterization (ACTEM, XAS, and XRD and some attempt at reproducibility.
- The project strengths are the strong fundamentals with respect to catalyst activity and composition, development of new techniques, and MEA testing.

Project weaknesses:

- Catalyst scale-up is included but appears to be slower than might be the case with more experienced collaborators. Fundamentals with respect to MEA diagnostics are a project weakness.
- ANL needs to realize that high kinetic ORR activity and durability of that kinetic activity are not enough to ensure that a catalyst will have practical utility. One must advance to MEA testing and also probe the catalyst’s utility in MEAs optimized for high-current-density performance in air. ANL needs to increase the strength of its collaborations with other organizations to rise above its current focus on kinetics alone.
- The project has not collaborated with industrial catalyst suppliers, and collaboration outside the national laboratory network is still pending. This is particularly a problem for scaling up catalysts. While alloys have high activity, many of the catalysts being worked on in the project may not be stable in either MEA

preparation or in cell testing. Past projects from the investigators have revealed a tendency to encounter the most challenging aspects of the work in the final year. The project needs to move quickly toward the experiments that might be most revealing or disappointing.

- There is insufficient utilization of in situ EC-ICP/MS for verification. It is not clear what leading technical catalysts that are demonstrated to work in cars do. It is not clear how to improve those using this system. Other project weaknesses are the inclusion of Fe in multi-metallic nanoframe development despite its incomparability with membranes; emphasis on RDE for performance testing; proposed continuous scale-up, which does not make sense; and lack of comparison of performance to leading commercial catalysts (e.g., to PtNi/C, Pt/silica structure).

Recommendations for additions/deletions to project scope:

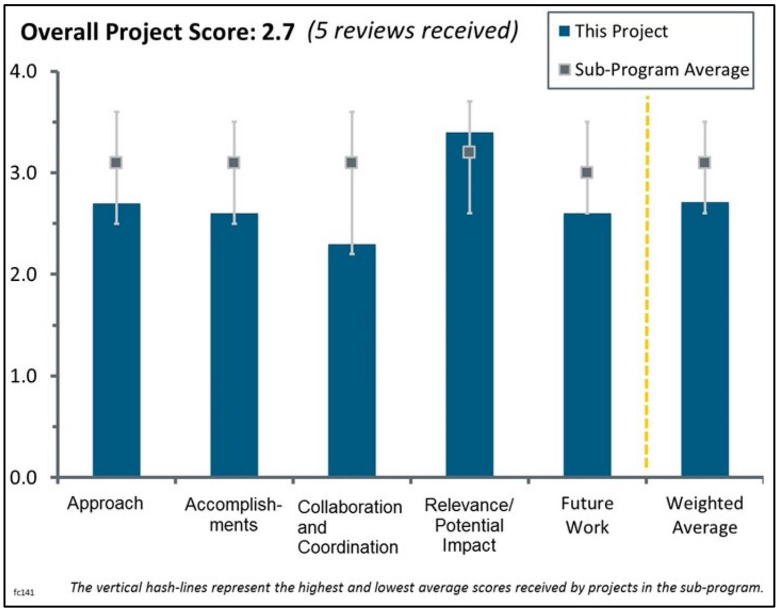
- The project should put a greater emphasis on better understanding and stabilizing a single-catalyst-system approach, rather than pursuing multiple formulations, form factors, and synthesis approaches. Greater emphasis on full electrode testing to confirm stability and durability is also needed. In situ EC-ICP/MS for verification is also recommended. It is unclear what the leading technical catalysts that are demonstrated to work in cars do. It is unclear how to improve those using this system. The project should eliminate Pt/Si whiskers. The very nature of extended whiskers like these is thermodynamically unstable, and they will likely sinter under heating. In addition, integration into an electrode is unclear. It is not clear how one collects current from these efficiently and facilitates ORR. It is not clear how Si is removed. It is not clear how the surface is cleaned off. It is not clear whether cleaning requires heating. If so, it is not clear whether extended structures collapse.
- Collaboration with an industrial catalyst supplier should be added. Faster progress toward scale-up of catalysts, toward cell testing, and toward durability testing in a cell needs to be pursued. Fabrication of PtNi or PtCo nanoparticles that do not appear to advance beyond what has already been done should be eliminated where necessary.
- The project should add major collaborations with organizations experienced in catalyst scale-up and in fabrication of MEAs optimized for performance at all current densities.

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.



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Question 1: Approach to performing the work

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

- The project approach toward development of high-specific-activity catalysts with high Pt utilization, based on Pt monolayer catalysts, is sound. Both activity and durability in rotating disk electrode (RDE) work appear to be routinely assessed. Too little effort is put toward addressing two key issues: use of platinum group metal (PGM) in the cores and high-current-density performance in membrane electrode assemblies (MEAs). Evaluation of durability in MEAs is much more aggressive and is necessary to truly assess the catalyst durability. No such data were presented this year.
- Monolayer catalysts, which can provide good PGM-mass activity along with the high-PGM-mass-specific surface area that is needed to mitigate high-current-density losses due to local oxygen transport effects, are currently the most promising direction for decreasing PGM loadings to DOE targets while maintaining fuel cell performance over the whole operating range. Non-precious cores could provide the above benefits with a wide range of core sizes if durability could be achieved. Precious-metal cores could provide benefits vs. Pt or Pt-alloy catalysts only if the core–shell particle size were kept below the 4–5 nm of reasonably stable Pt-alloy catalysts. Too much of the work still involves cores containing excessive amounts of precious metals. More of the effort should be focused on non-precious cores such as the promising work with NbN. The project should reduce its reliance on RDE testing, which does not always predict behavior in MEAs for novel catalyst systems.
- The non-precious-metal-based cores using niobium–nitride and tungsten–nickel show promise of high activity and stability. The project needs to find collaborators who can successfully translate these improved activities into MEA-level demonstrated activity and high-current-density performance.
- The approach in this project continues to address the needs for reduced-cost and increased-performance catalysts. Questions remain on the utility of replicating an existing approach on various substrates, such as whether there is a rationale to trying Nb, Mo, and Y. The rationale behind the support variance is also unclear; the support variance is a solid and needed addition to the project, but the choice of carbon nanotubes is questionable. Further, the three-dimensional structured supports mentioned are not defined in any way. Lastly, graphene cannot survive carbon corrosion and hence should not be used. The project has some integration with other projects such as developing characterization techniques but does not appear well reintegrated with either MEA project.
- PtY is thought to dissolve, based on prior work. This project should move aggressively to test PtY in a fuel cell.
- In general, Brookhaven National Laboratory (BNL) needs to move much faster toward durability experiments. The project's usual operating mode constitutes surveying a wide range of catalyst samples

before proceeding with cell testing or durability studies (other than those studies pursued in a glass cell, which have little relevance to what happens in a fuel cell). Instead, once a new catalyst is found to be active in a glass cell experiment, the project should move immediately to have inks made (perhaps through collaboration with a supplier or national laboratory), followed by MEA preparation and cell testing. Some evidence of accelerating to cell testing has been shown for nitrided PdNi cores and PdMo cores, but this needs to happen for each catalyst. The use of Au cores (such as with the Ti-decorated Au cores or the AuNi alloy cores) is not likely to lead to a cost benefit versus the use of a PGM core. The nitrided PdNi core and the Pd_3Mo core may also not provide for a cost benefit. There are only two classes of catalyst particles in this project that do not make use of a precious metal in the particle core: Pt monolayers on NbN and Pt/Pd monolayers (and some variations thereof) on a Nb core. Other than niobium or niobium nitride cores, most material approaches risk high cost and low durability.

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

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

- The preliminary work with non-precious NbN cores is very encouraging and should be intensively pursued. The work with small amounts of Pd over Ni-Nb cores may prove useful if the amount of Pd can be held down and if these systems give adequate durability (always a worry when Pd is used). Mo-modified cores rely on strong bonding between Pt and Mo for stability as Mo oxides can be soluble in acid. If these systems give adequate durability, work should proceed to replace most of the Pd in the core with non-precious metals. The $Pt/TiO_2/Au$ system is scientifically fascinating but contains too much Au for practical application unless the total particle size is kept below 4–5 nm. The Pt/AuNi alloy system is also of interest if the particle diameter can be kept down to 4–5 nm. If one could replace Pt in the core of a small particle with Au and maintain stability, one would choose to do so because Au is much more widely available than Pt. The non-aqueous deposition of PtY is a step forward that needs to be followed up with tests of the durability of this system, which would seem to be dubious because of the high reactivity and solubility of Y.
- Mass activity (A/mg of Pt) is exemplary, exceeding the DOE 2020 target by several fold. It is unclear whether progress has been made toward improved mass activity, reduction in core PGM content, or MEA performance since last year (FC-009).
- There are a few catalysts with which mass activity needs to be normalized by total mass of precious metals. Examples in the presentation include PtPdMo, AuPtCo, and PtAuNi. Pt monolayers on PdNi reach about 480 mW/cm^2 at 0.6 V and 22 psig (a realistic high-end operation air inlet pressure) with 0.2 mg/cm^2 precious metals. This is only about 0.42 g/kW (cathode metals only) in spite of a PGM-based mass activity of 600 A/g from glass cell testing. Durability data are needed. Pt/NbN/C holds promise as a catalyst from this project that may decrease precious metal loading and be durable. However, its mass activity from RDE work is 350 A/g, which is decent but could be improved. Similarly, Pt/Pd/Nb is 380 A/g PGM at best on RDE, which also could be improved. Pt/PdMo/C shows approximately 460 mW/cm^2 at 0.6 V and lower pressure for only 0.098 mg/cm^2 precious metals. This appears better in comparison to Pt/PdNi but is still far short of the high-current-density objectives. Again, durability data is needed. The open circuit voltage is very low. Mass activity is only 340 A/g precious metals for Pt/Ti-decorated Au/C.
- Using gold and other PGM cores reduces the overall mass activity on a PGM basis; the state-of-the-art Pt-Co alloy catalysts are achieving over 600 mA/g Pt. Therefore, this project should focus on non-PGM cores and de-emphasize work on using PGM-based cores.
- Despite repeated requests, and published papers indicating instability, stability tests in MEAs of these types of core–shell catalysts are not presented. Without these, it is simply impossible to evaluate the potential of the technology. There is also a significant amount of incomplete analysis in the slides. For example, slide 6 is unclear on how the sample is ball milled. One should not be able to use ball milling to reduce sizes to 50 nm. Either the sample is degrading or the analysis in incorrect. On slide 8, it is not clear why the peak is shifted relative to Pd_3Mo, whether this is segregation of Pd and PtMo or why the Pd_3Mo is asymmetric—perhaps this represents multiple phases. On slide 10, there are striations on one of the particles and none on the other, indicating bulk alloying rather than surface alloying—and negating the core–shell structure. On slide 12 (A), it is unclear why the shapes of the particle in the high-angle annular dark-field imaging are different from the energy dispersive spectroscopy (EDS) maps.

Question 3: Collaboration and coordination with other institutions

This project was rated **2.3** for its collaboration and coordination.

- Good collaborations are in place, but the output/outcome is unclear. This principal investigator has excellent strength in developing new catalysts and new ideas and testing them at RDE level. However, the collaborators do not seem to be engaged in moving this technology forward and scaling up. While some of this technology has been licensed, the licensed catalysts are still very similar to the PtCo alloy catalyst. More work is needed to optimize these at Technology Readiness Level 4.
- The project itself involves little collaboration, but patents from earlier in the project have been licensed and brought into production by an experienced catalyst manufacturer. The project would benefit from more formal collaboration within the project, with catalyst manufacturers and fuel cell developers to modestly scale up catalyst synthesis to allow testing in MEAs. The project should interact with the Fuel Cell Consortium for Performance and Durability (FC-PAD) to do early MEA testing of new catalyst systems. RDE testing of activity, and particularly RDE testing of durability, are of questionable relevance to real fuel cell operation.
- Many collaborations are listed on the slides. However, the presentation does not provide evidence of how collaborations are being used in the past year. Attributions are not given. The slides did not show evidence of where reactive spray deposition had been pursued in the past year. Perhaps this points to collaborations quietly ending. The same might be said for density functional theory (DFT) studies. Technology transfer to N.E. CHEMCAT Corporation is mentioned, but the licensing did take place four years ago. It is not clear whether there has been any follow-up in recent years.
- Collaboration in this project seems limited to Los Alamos National Laboratory.
- It is unclear if any of the work was done outside BNL.

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.

- Pt monolayers on non-precious cores are the most promising pathway to obtain both high kinetic mass activity and good high-current-density performance with low-loaded catalysts. This type of catalyst should therefore get priority-level attention, both within this project and throughout FC-PAD. The project must progress past RDE testing to MEA work if relevance is to be improved and the full impact is to be realized.
- The project has a very high potential to have an impact on the fuel cell industry and can be a game-changer if these catalysts can be mass-produced reliably.
- The project is addressing key commercialization barriers toward cathode catalyst activity, cost, and durability.
- DOE has funded work on Pt monolayer catalysts at BNL for many years. While there is no question about the relevance of developing novel oxygen reduction catalysts for commercializing hydrogen fuel cells, it is fair at this point to begin asking questions about whether the development of Pt monolayer catalysts is helping to accelerate commercialization. It has now been over four years since early 2012 when N.E. CHEMCAT licensed BNL technology. Since then, there has been mostly silence with regard to whether these catalysts have been applied toward commercial programs, despite the initial promise that that would occur. If activity or durability needs to be improved, then that would underline the relevance of continued work. However, if there is something fundamentally flawed with high-volume production of catalysts premised on Pt monolayers, then the relevance of this project is questionable. The use of inexpensive cores and a noted interest to improve performance at high current density are relevant pathways to explore.
- The potential impact of the project is high, but accomplishments and promise are not clear.

Question 5: Proposed future work

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

- Plans for more facile synthesis of Nb-based non-noble cores are good. NbN cores should get more emphasis in the planned future work—attention should be concentrated on non-precious cores even if the RDE activities are not as good as with precious cores. The plans given for improving catalyst response at high current density do not seem to focus properly on the prime advantage of the core–shell—the ability to have high-Pt-mass-specific surface areas to keep local current densities at the Pt surface low thereby reducing local oxygen transport losses. Following this line of reasoning, the core–shell should be able to do the job without messing with ionomer content, derivatized supports, or the like.
- Task 1 is reasonable in goals, but focusing on a single material set is recommended. Task 2 is a welcome route to seeking alternative synthesis routes, but the details seem scattered. Task 3 is provocative but poorly defined. Regarding Task 4, while higher current densities are desired, the approaches suggested are unlikely to succeed.
- Proposed future work sounds good. There is a need for more detailed collaboration with industry and other groups that can integrate this catalyst into MEAs with other best-in-class components.
- The future work describes a future emphasis on refractory metals and hollow cores. It would be preferred for the project to focus on these materials rather than those that include precious metals in particle cores. The approach to reach high current density appears scattered. The use of carbon nanotubes appears novel, but it may be frowned upon in certain organizations because of the expense of handling and safety concerns. Furthermore, there should be some way of facilitating high current density with these catalysts without using carbon nanotubes; otherwise, there is no clear reason to investigate the catalysts as they are. Reducing the Nafion® in the catalyst ink would be a start, but it is not clear how much to reduce it. It is unclear what is providing direction as to how to restructure catalyst layers for high-current-density performance. This project contrasts with other DOE-funded projects in which tasks were included to model catalyst layer structure and figure out how to deposit catalyst inks. More information needs to be shown as to how three-dimensionally deposited layers will solve issues at high current density.
- The project should directly involve industry or a national laboratory partner with demonstrated experience in development of optimized electrodes. Work should have a significant focus on demonstrating viability of the catalyst platform. There should be a focus on fully optimizing Nb-based non-noble metal cores to drive up PGM mass activity to well above DOE targets.

Project strengths:

- Owing to their high Pt-specific surface area, Pt monolayers on non-precious cores are the one clear path to low-loaded MEAs that meet DOE activity targets and performance goals at high current density. The project continues to develop promising new catalyst systems. The project has developed non-aqueous synthesis for the calculated-to-be-good PtY system; the synthesis work should allow the critical durability questions about this system to be answered.
- The project is able to conceive of numerous catalyst species and work at fairly high throughput. The project is able to quickly report RDE activities for catalysts. Some of the ideas entailed in the project include means of removing precious metals from the core of catalyst particles.
- The strong technical team continues to demonstrate novel electrocatalysts with high specific activity and high durability in RDE. Novel approaches toward decreasing overall core PGM content are promising.
- Strengths include catalyst synthesis, generating new ideas, and fundamentally characterizing these new catalysts.
- The project has a strong scientific team.

Project weaknesses:

- Many of the project’s catalyst concepts still displace Pt with other precious metals. Furthermore, reporting of mass activities normalized by precious metal content is not consistent. Fuel cell data for many of the catalysts do not show high power density at potentials near 0.6 V (a rough thermal limit) and at reasonable high-end operation pressure. Furthermore, the plan for advancing performance at high current density has not been shown to be thorough. Rather than leverage collaborations and understanding of porous media or mathematical modeling, the project seems to frequently respond to performance challenges with material novelty. Examples include depositing thin catalyst layers on gas diffusion media or pondering the use of carbon nanotubes. Instead, it would be preferred to see the project find a way to take the catalysts that have already been studied and make them work. The answer to a performance challenge is not to generate an alternative material (usually).

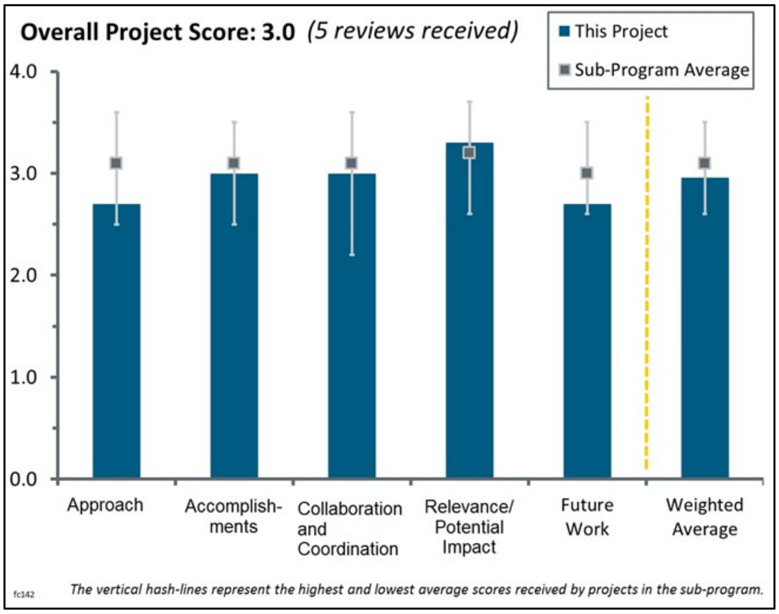
- The project remains overly dependent upon RDE testing of activity and particularly on RDE testing of durability. Most of the core system studies still involve excessive amounts of precious metals other than Pt. This need not be a problem if the total particle size is not larger than the particle size of competing Pt-alloy catalysts, around 4 nm. The currently listed plans to improve high-current-density performance are not the most promising path to utilizing the inherent advantages of core–shell catalysts. The project should go for high-specific-Pt surface areas.
- The materials set is poorly focused. The project has continued focus on “discovering” new materials combinations without appropriate stability testing despite reported stability issues of previous materials. Analysis of the characterization data presented is inaccurate. There is a dearth of more representative analytical methods like small angle XRD, synchrotron XPS for depth/composition analysis, and XAS in structure analysis.
- There is a lack of ways to make these catalysts into meaningful MEAs and translate the same to high-current-density performance.
- There is too little focus on demonstration of performance and durability in MEAs. PGM mass activities remain below expectations for this promising approach.

Recommendations for additions/deletions to project scope:

- The project should try TaN in analogy to NbN with possible durability advantages. The team should rigorously test the (suspect) durability of the PtY system and talk with those who conduct DFT about the results. When the project considers which alloy systems should make useful catalysts, considerations should include stability of dissolved species derived from alloying metals. The project should scale up the preparation of several of the most promising systems (including Pt/NbN) and test activity, durability, and high-current-density performance in MEAs.
- All catalysts performing below 300 A/g precious metals should be removed. The focus should shift toward materials with non-precious-metal or hollow cores. Niobium, niobium nitride, and other refractory cores should be emphasized. The project should seek to minimize Au and Pd as much as possible. Each catalyst should be tested in a fuel cell as quickly as possible to understand durability and high-current-density performance. Much greater emphasis should be placed on how to achieve high-current-density performance—an area in which collaborations would help.
- The focus should be increased on stability (and possibly durability) testing in MEA and on fine-tuning selected compositions that should be stable—such as PtPd.
- The project should collaborate with a team with good knowledge of making MEAs to scale up these catalysts. Work on PGM-based cores should be deleted.
- The project should directly integrate an electrode development and testing partner to enable MEA-level performance and durability evaluation.

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 **2.7** for its approach.

- The project has just begun, but it builds on previous efforts of most of this team. There is good team overlap in specialization, balancing materials synthesis, characterization, and testing. Selection of ALD to attempt the technically challenging goals is a good approach, balancing challenging needs with an industrially viable approach.
- The research team has changed the approach from galvanic displacement to ALD on Ni and Co nanowires to develop a new class of electrocatalysts for oxygen reduction reaction. The team’s previous approach for preparing extended, continuous Pt nanostructure did not show good performance in MEAs, as reported in the 2015 Annual Merit Review.
- The proposed synthesis–characterization strategy is rational and may provide a useful way to overcome many limitations that currently impede the cost-effective commercialization of Pt-based alloy cathode materials. However, there is room for significant improvement. For example, the “nanoflowers” direction should be abandoned because—irrespective of the shape of particles—during operating conditions, the “flowers” will be transformed into a spherical shape with activities that will be dependent on the size of the particles and an optimal segregation profile. The nanowire direction is more promising but also needs to be improved, particularly regarding the stability of low-coordinated Pt and Ni atoms. The ALD method may not be the best one for optimizing the thickness and the composition of the film. The investigator may consider developing a pulsed laser deposition (PLD) method, which is more suitable to “synthesize” well-defined films. Last but not least, the approach focuses on testing rather than understanding, which will not lead to optimization of the PtNi alloy systems.
- The project approach of optimization of Pt overcoated Ni nanowires is generally effective. The approach is lacking in development of improved Ni-leaching mitigation strategies in the MEA. The Ni loading in MEA electrodes is very concerning; it would appear that even if a small fraction leaches, significant fractions of the MEA ion exchange capacity (IEC) will be consumed. More effort toward quantifying and improving is needed.
- The U.S. Department of Energy (DOE) has funded a considerable amount of work on dealloyed PtNi catalysts for oxygen reduction, including PtNi nanoparticles (General Motors Company [GM], Johnson-Matthey Fuel Cells Inc.), PtNi nanostructured thin films (3M), PtNi nanoframes (Argonne National

Laboratory [ANL]), and PtNi nanowires through spontaneous galvanic deposition (the former National Renewable Energy Laboratory [NREL] project). Many of these projects have reached considerable maturity in realizing the barriers associated with Ni-leaching, high-current-density performance, and maintaining performance at larger-scale batches. It is difficult to understand how adding another project investigating a PtNi system adds to what is being done, especially a project premised on ALD, which is widely thought to be a low-throughput means of producing catalyst batches. Data presented so far show samples that have been made with up to 13 wt.% Pt from ALD with oxygen chemistry that have in excess of 600 A/g-Pt mass activity. Furthermore, ALD with hydrogen was said to be able to make nanowires with up to 6 wt.% Pt. However, these samples imply that an extraordinary concentration of Ni will be entering into the process of fabricating MEAs and eventually into fuel cells themselves where the Ni will contribute to aggregating the nanowires in ink (based on findings from FC-106), displacing protons in the membrane, and increasing the hydrophilicity of all porous layers. The researchers understand that Ni needs to be leached, but no data have been shown so far to ensure that activity is preserved after nanowire leaching. The project has recognized that there needs to be an understanding of how to allow high performance at high current density, but so far the results seem to indicate the project is cornered; ionomer levels have no impact on mass transport (except at high relative humidity [RH]), and they have no impact on proton conductivity of the layer. Alternative approaches for trying to devise catalyst layers that will operate at 1 W/cm^2 are apparently not allowed in this project. Essentially, the project is a catalyst powder project in which making layers is almost an afterthought until there is a problem, which there already is. Partners such as GM need to be engaged to systematically begin to address this problem through modeling, an experimental design for ink processing, better diagnostic measurements, or all of the above.

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.

- An impressive number of results have been presented in the report; therefore, the investigators should continue collecting data with the same quickness. However, the quality of the results is more important and will define the success of this project. It will be extremely important for the next review cycle to demonstrate high reproducibility of the results because the preparation of samples with the ALD method will require a careful analysis of “a single nanowire.” For the next review, it would be highly desirable to discuss more about the stability than the activity of nanowires, as the latter is well understood.
- A significant amount of work has been carried out since the start of the project. Pt deposition was carried out on Ni and Co nanowires, and their electrochemical properties have been evaluated. Enormous physical characterization has been performed in order to understand the ALD of ETFECS.
- A good amount of work has been carried out considering the recent start of this project, but results are not promising as of yet. It is clear that modification of the approach is needed.
- The team has continued to make substantial and steady progress toward development of very high-activity electrocatalysts that exceed the DOE target by several fold in rotating disk electrode (RDE). Little apparent progress has been made towards addressing MEA hydrogen/air performance. MEA limiting current densities of <1 A/cm^2 under hydrogen/oxygen, after repeated MEA acid washes, is indicative of substantial (and perhaps continuous) Ni leaching (slide 16).
- Whether or not the project can overcome barriers associated with cost will hinge primarily on the ability to demonstrate high performance at high current density. So far, the project has shown no more than 0.42 W/cm^2 at 0.6 V for 0.16–0.20 mg-Pt/cm^2. At best, this represents 0.38 g/kW gross, which is much higher than the target of 0.125 g/kW net. Granted, this performance level is with samples from spontaneous galvanic displacement, but in principle, the same performance would be expected with ALD. Of greater concern, however, is the lack of options known to improve this performance level. At 100% RH, some mass transport improvement can be found by lowering the amount of ionomer (according to the limiting current measurements), which would likely result in a modest change but not the >100% improvement needed to meet high-power performance with low Pt loading. The project is showing neither the high current performance it needs nor the line-of-sight to obtain high current performance. The amount of Ni in high-activity samples is concerning. High activity has been shown only for PtNi nanowires with 13% Pt or less. Samples with much higher percentages of Pt have been made with acid leaching, but the activities of these samples have not been reported. Higher weight percentages of Pt on nanowires (prior to acid

leaching) can only be generated after greater than 100 cycles on ALD. Some perspective should be reported on the cost implications of this and whether ALD would represent a practical commercial process.

Question 3: Collaboration and coordination with other institutions

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

- The team has an impressive number of collaborators, including an original equipment manufacturer, a catalyst manufacturer for scale-up, and universities.
- This is a team with a good collaborative background, and the geographic proximity makes further coordination likely to continue to be effective.
- It is too early to judge the collaboration; the project has just started.
- The University of Delaware expected to deliver novel Ni nanotemplates, but this still appears to be future work. It is difficult to tell from the presentation, but it would appear that the University of Colorado has been involved in making catalysts through ALD. There does not yet appear to be a contribution from ALD NanoSolutions on the business case analysis, although it would be very interesting to hear more about the business case for ALD. The partnership with GM should be used more to sort out strategy with respect to achieving high-current-density performance. Showing that the project is working with GM to sort out how to achieve the attribute with perhaps the greatest impact on cost would benefit the project significantly.

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 anomalous high performance of extended surfaces continues to be proven, and yet translation to high-surface-area and therefore economic catalysts is needed. This project aims to make high specific activity extended surface area and is attempting to achieve this using ALD rather than the previous focus on galvanic displacement. The goals of this project are high-performance catalysts that are manufacturable, a great need for the Hydrogen and Fuel Cells Program (the Program).
- The project objectives are relevant to DOE’s cost and durability goals. Demonstration of hydrogen/air fuel cell performance and durability would significantly advance the state-of-the-art Pt/C and Pt-alloy/C catalyst.
- There is no doubt that any project that focuses on relationships between activity and stability of cathode materials is of paramount importance for developing a new generation of fuel cells. If the investigators would be able to establish such relationships, the project will contribute significantly to DOE efforts for full implementation of electric cars.
- The project is directly relevant to key Program objectives toward cost and performance.
- New oxygen reduction catalysts can still lead to significant advances in lowering the cost and improving the durability of polymer electrolyte membrane fuel cells. In fact, such catalysts present the greatest opportunity for affecting the cost and durability of the entire system. Extended thin film catalysts are also relevant since their high specific activity represents the scientific opportunity of how active an oxygen reduction catalyst can be, which happens to be much higher than what is conventionally observed with Pt/C or PtCo/C. Furthermore, durability can be higher since particles are large enough to avoid high-surface-area loss through agglomeration. The one question about relevance is whether the project addresses cost by addressing performance at high current density. Technically speaking, the answer is “yes” since the presentation reveals that researchers have already grappled with how to improve performance at high current density. However, if the project does not have the right resources to address this (modeling, ink and layer characterization, processing, diagnostics, etc.), then the entire effort will have been for naught.

Question 5: Proposed future work

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

- The team has specifically proposed to perform catalyst evaluation in fuel cell and durability studies, which are essential to fulfill the project objectives.
- The project in general is strong, but greater emphasis should be placed on more averaging characterization techniques rather than microscopy, which is very "sexy" but highly localized. Future work was not clearly identified, but it can be inferred since this is early in the project. A method for evaluating the stability of the Ni substrates would be very useful (e.g., annealing to determine stability of the flake-like materials [unless they are of the correct crystallographic orientation, they are unlikely to be stable thermally and thus probably also electrochemically]).
- The project should reconsider directing research toward synthesis of "nanoflowers"; it may sound nice, but it could be a huge misdirection. In addition, in the proposed future work, one key direction should be fundamental studies of catalyst stabilities. Testing stability in real fuel cells is usually misleading, providing no quantitative information about the corrosion of Pt and Ni. If the corrosion of Pt and Ni is not understood, it will be impossible to know what type of PtNi alloy should be synthesized.
- The project should significantly direct focus toward understanding Ni leaching in MEA and improving Ni stability in the electrocatalyst. If this cannot be overcome, this promising technology will never be commercially relevant.
- On the future work slide, two of the three major categories shown describe work that is more or less already happening or that does not truly address what threatens the success of the project. Yes, the nanotemplate synthesis with greater shape control could produce more active PtNi nanowires, but it is unclear how they should be integrated into a catalyst layer and how the Ni will be prevented from leaching. Unless the shape control and nanotemplate synthesis routes are being dialed in to address both high current performance and durability, some of this effort might involve considerable guesswork. Similar comments could be made for electrocatalyst synthesis work, although to its credit, this work will contain Ni leaching tasks. Optimization of the electrode structure is where the project needs to go. However, it is unclear how it is known that electrospinning is the answer—that is, how electrospinning will provide an enhancement in oxygen flux or proton transport to enable high current density performance. There should be work devoted to understanding how to make a catalyst layer better before committing to particular methods for doing it. It is unclear why carbon should be added as well as what carbon should be added and how much. There should at least be an experimental design for how to approach this. Isolating voltage losses is good, but it is also unclear whether different types of mass transport losses can be separated out.

Project strengths:

- The project has access to considerable catalyst layer processing equipment available at NREL. The overall concept of the catalyst powder lends itself to high specific activity, which means the "ceiling" for activity and improvement in fuel cell performance is high. The investigators are well-connected to the fuel cell research community and have added an automaker as a partner.
- The project has an excellent team, proven benefit, and a rational approach.
- The team has strong collaboration.
- The project has a novel approach to generate extraordinarily high specific activity and mass activity.
- It is too early to make a judgment about the project strengths. Nevertheless, the project strength might be the existence of many tools that are needed for exploring the feasibility of implementation of extended PtNi thin film catalysts in polymer electrolyte membrane fuel cells.

Project weaknesses:

- The project needs to have better direction with respect to addressing high-current-density performance. It needs to have a strategy for how this weakness is to be addressed. The project might address which variables will be studied and why, how prior knowledge can be used to decrease both mass transport and ohmic losses, and how ink processing should be done. These questions all seem relatively unexplored at this moment (with the exception of the ionomer ratio data). The results of the past NREL project indicate a tendency not to consider fuel cell performance until late in the project. The future work shown indicates that this tendency may carry over into this project. The project is one of many PtNi projects that DOE has funded, which has yielded a common trend: high RDE activity results, followed by poor fuel cell performance, particularly at high current density, which may be related to Ni leaching. The project will have to work hard to avoid this trend and not resemble a duplication of efforts with other DOE-funded projects.

- One weakness is reliance on microscopy rather than more sample-averaged techniques. The “continuous films” produced previously are not continuous, but demonstrated per surface area activity is higher. Without understanding this phenomenon, it is not clear whether this approach is needed. The high variability in deposition due to ALD is worrisome, and it is unclear whether this can be addressed.
- Mitigating measures to inhibit Pt dissolution from ALD ETFECS have not been discussed in the presentation.
- One key weakness is the lack of a clear path toward understanding and minimizing the dissolution of Pt and Ni atoms during fuel cell operation.
- Ni leaching in MEAs is a substantial concern and is not being addressed with enough emphasis.

Recommendations for additions/deletions to project scope:

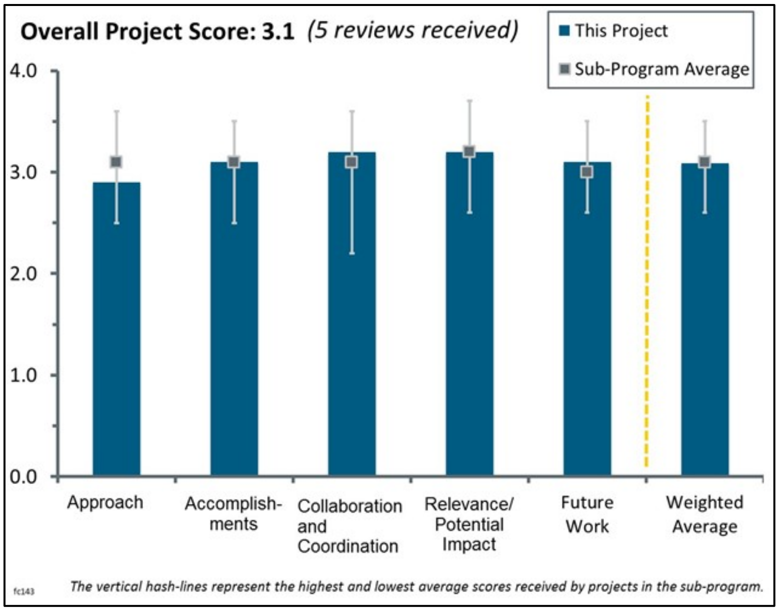
- First, the stability of the hollow Pt-skeleton-like structure (acid-leached 75% Pt sample shown in slide 15) under accelerated stress test conditions is questionable. Second, the team should focus on the methodology for minimizing the wetting problem associated with the ETFECS, which will affect the high current density performance.
- Identifying a method for evaluating the stability of the Ni substrates would be very useful (e.g., annealing to determine stability of the flake-like materials [unless they are of the correct crystallographic orientation, they are unlikely to be stable thermally and thus probably also electrochemically]). The project should shift to more averaging characterization methods (e.g., small angle x-ray diffraction (XRD), x-ray absorption spectroscopy (XAS), synchrotron x-ray photoelectron spectroscopy (XPS)). ALD should be demonstrated on more spherical Ni powders rather than tubes.
- The project should decrease emphasis on making the catalysts and increase emphasis on integrating the powders into catalyst layers. It does not matter that it is early in the project. This kind of task should not be saved until the end; it needs to be planned out now. Collaboration with GM and ALD NanoSolutions needs to be significantly increased with respect to high-current-density performance and the ALD business case respectively. The project needs to work with GM to develop an experimental strategy for assuring high performance at high current density. It should not be enough to say that electrode optimization will happen; questions need to be answered about how this will be conducted from dealloying to ink-making to ink-processing to catalyst layer deposition to cell conditioning. Some indication needs to be given about cost and high-volume throughput that could be expected for ALD—both oxygen and hydrogen chemistries.

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. 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 **2.9** for its approach.

- The approach is good in achieving DOE’s goal for electrocatalysts and supports.
- The approach is good with the focus on NSTF as a platform. Although NSTF has issues for use in some fuel cell applications, as a catalyst platform it is good. One does want to know how it translates to other systems in terms of understanding. The combined experiment and modeling has good potential, although it needs to include explicit solvent techniques to understand how activity will perform under operating conditions. It is unclear whether it is more thermodynamic in nature and it is not clear what mechanisms are being assumed.
- 3M and the NSTF product remain interesting materials for fuel cell applications. Pursuing these materials using a nanoporous and ultrathin film approach seems to be a reasonable mechanism to potentially improve electrochemically available surface area and, thereby, mass activity. Slide 7 shows a high dependence on density functional theory (DFT) to guide/interpret results. It is not clear how valuable/accurate this effort will be in improving the performance or designing higher performing electrodes. The team has already shown the ability to perform high-throughput experimental work in this area and this may be the better route. The inability to share more information about composition/processing inhibits the ability to judge the merits of following different approaches.
- Without any details being given in the presentation on the types of synthesis and/or post-treatment changes used to generate the two modified catalyst types (nanoporous and ultrathin) of this project, it is hard to evaluate the claim that significant improvements require the optimization of a large composition/process space. Previous NSTF projects extensively explored the PtNi composition space and the presentation did not seem to give any data that showed that earlier optimization was no longer valid (other than a lack of composition dependence in the [lower] activity and specific areas of the non-annealed P4A). Increasing the specific surface area of NSTF should be helpful in addressing contamination issues and high-current density performance durability as the ionomer migrates from the membrane to the electrode. However, the area target of 30 m^2/gPt for the nanoporous catalyst is at best marginal and the target of 20 m^2/g for the ultrathin film (UTF) is inadequate for dealing with local oxygen transport. Part of the UTF approach was listed as maximizing the NSTF support surface area. This has always been an obvious approach to addressing robustness as well as activity issues, but past efforts in this area have not seemed productive.

- This project continues to develop NSTF-based catalyst technology and seems to be a continuation of FC-104. This NSTF-based catalyst technology showed significant potential to achieve high catalyst activity. However, the most significant problems are its generic catalyst layer structure, the so-called ionomer-free catalyst layer, and the difficultly achieving the sufficient operational robustness and reproducibility (conditioning). The MEA conditioning is one of the technical questions. FC-104 could not demonstrate a sufficient performance in the short stack at General Motors even though a good performance was shown in the MEA at 3M. The objective of this project is focused on obtaining 200% of the 2020 mass activity target. On the other hand, robustness and reproducibility are not discussed. One of the pre-work activities of this project showed significant improvement of operational robustness of the MEA but the principal investigator (PI) did not disclose how the project achieved it. It seems that the project focuses on the strength of this technology and avoids generic technical problems. Because we see a potential for this unique catalyst technology, it is more meaningful as a pre-competitive research to investigate the catalyst structure in order to gain mechanistic understanding of ionomer-free catalyst layer technology. Some of the approaches that the project discussed, such as high-throughput fabrication, seem to be more like engineering phase approaches than pre-competitive phase approaches.

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.

- Substantial work has been performed and one of the DOE 2020 targets for catalyst durability under accelerated stress test conditions has already been achieved.
- At least one of the advanced electrode structures seems to give improved operation robustness versus traditional NSTF. If this is done without use of a dispersed-catalyst interlayer, it could give significant durability advantages (versus traditional NSTF and interlayer), reestablishing one of NSTF’s prime durability advantages versus dispersed-catalyst electrodes. The addition of “M,” presumably to PtNi, seems to stabilize the specific surface area versus cycling of the nanoporous films, which could improve overall durability significantly. 3M appears to have developed a method that can deposit significantly thinner and more uniform Pt-alloy layers on the NSTF substrate whiskers. This could increase the utility of increasing the area (i.e., length or areal number density) of the substrate whiskers. The combination would be more useful than either development alone. The utilities of the compositionally graded MEAs and the segmented cells seem questionable because processing variables are likely to be more important than composition in the optimization of both nanoporous and UTF catalysts.
- The project was just started, and many technical accomplishments are not expected. Some interesting early work data was presented, such as high performance with hydrogen/air at lowered platinum group metal loading and good retention of performance after the potential cycle tests. Particularly, operational robustness data showed significant improvement over a traditional NSTF electrode. The PI declined to disclose details of this MEA information.
- Accomplishments are limited, but it is a new project. Initial work seems promising, especially the high-throughput work. So far, most new work has focused on performance and not durability, so the feasibility of some of the approaches over time is unknown.
- As the project is still just starting, this category is perhaps more difficult to judge, although the results presented—both for new materials and as background for previous materials—show a lot of samples being screened and some significant improvements in “operational robustness,” allowing advanced electrodes to achieve a broader operating window starting to approach that of Pt/C. Improvements in electrochemical surface area (ECSA) are encouraging, although these have been attributed to “pre-project” work. Performance optimization approaches are difficult to evaluate because arbitrary catalyst variables have limited value. The NSTF work seems more consistent with previous work, and the UTF work seems more novel, but also less advanced. The high-throughput aspects presented should help the project advance.

Question 3: Collaboration and coordination with other institutions

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

- The project has a good team, comprising laboratories, academia, and industry. Everyone seems to play separate roles, so good coordination will be key. It is not clear how many 3M scientists contribute significant time on this project. Because this is an electrocatalyst project, it would be good to provide the NSTF catalysts to others that make MEAs, such as the Fuel Cell Consortium for Performance and Durability and the project from the National Renewable Energy Laboratory.
- The project has a very strong team. It would be nice to have an original equipment manufacturer (OEM) participate so that relevant operating conditions, an area of challenge for NSTF, would be specifically probed.
- The team has very good collaboration with universities and national laboratories.
- Collaborators are addressed in the academic area and they seem to be enough for the characterization of MEAs. Operational robustness evaluations should involve automotive OEMs.
- The kinetic Monte Carlo (kMC) calculations on alloy surface structure predictions at Johns Hopkins University should provide useful comparisons with experimental results. It is not clear what new findings will come out of the DFT calculations at Purdue University. It is too early to know whether the national laboratories' characterization efforts will be significant.

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.

- If successful, the project will meet all or most of the DOE 2020 metrics for electrocatalysts and supports.
- The approach is taking on the critical issues of performance, cost, and durability, and it is only really finding problems at peak power and/or dynamic operation. These are the most significant concerns for automotive applications in which a wide range of operating conditions are to be expected.
- The project objectives are meeting and exceeding DOE 2020 targets, particularly for the project target of catalyst activity (mass activity), which is two times higher than the DOE target. More generically, the NSTF catalyst demonstrated very high specific activity so that the project now needs to significantly reduce the catalyst loading to achieve significant high mass activity. However, the most significant technical barrier of this catalyst technology is operational robustness and reproducibility of the performance. It was considered that the ionomer-free catalyst layer of this catalyst technology could be a problem. There is not enough mechanistic understanding of how this ionomer-free catalyst works. Study in this area is one of the most important work areas. The current DOE research, development, and demonstration (RD&D) target does not address the robustness attribute enough.
- If the new catalysts give operational robustness without dispersed-catalyst interlayers (and if membrane improvements lessen NSTF degradation by membrane fragments), they could enable the full cyclic-durability promise of NSTF to finally be realized in fuel cell applications. There is a significant possibility that the changes generated by this project will provide only incremental improvements that are insufficient to get NSTF into significant fuel cell applications. High-throughput methods are unlikely to contribute significantly to the productivity and impact of this project.
- NSTF in OEM cells is still unproven but the catalyst scaffold could be good to determine. The impact is dependent on MEA tests as well although electrode structure development is outside the scope per the funding opportunity announcement, so it is hard to ascertain impact.

Question 5: Proposed future work

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

- Considering the strengths of various collaborators, the proposed experimental and model studies for the upcoming year can be completed on time.

- Based on project approaches currently addressed, the future work makes sense. DFT models would help characterization work. Again, the most significant technical barrier of this catalyst technology is operational robustness and reproducibility of the performance. It was considered that the ionomer-free catalyst layer of this catalyst technology could be a problem. There is not enough mechanistic understanding of how this ionomer-free catalyst works. Study in this area is one of the most important work areas.
- While quite solid, it is unclear whether performance and durability should be the primary focus (as laid out in the project) or whether "operational robustness," the historic Achilles' heel of NSTF, should have more emphasis. It is also unclear how the kMC and DFT models will lead to improved materials/performance.
- Proposed work seems adequate although a bit nebulous. There is a lot of work to be done and it would be good to know what options they have if certain activities do not occur (e.g., cannot get reliability of techniques). It is not clear how predictive the models will be because they will be based on calibration of data sets. It is unclear whether there is a way to test them.

Project strengths:

- NSTF catalyst technology has a significant potential to achieve high activity and durability. It shows very high specific activity and the project is focusing on the improvement of mass activity. Also, the thin film may have a more bulk-like nature and strong potential to make catalysts durable compared to nanoscale particle-based catalysts.
- Project strengths are the combined experiments, computation, and high throughput. The two main approaches have made seemingly good progress and had good performances in a short time.
- The prime recipient has a track record of continuously working on the NSTF-based catalysts, which is an important factor in understanding the catalyst system and possible commercialization in the near future.
- NSTF is a great platform for high throughput with a tremendous amount of background to build on.
- Two modifications of the basic NSTF catalyst are now available for optimization: (1) porous and (2) thin uniform deposits. One can rationalize reasons why each of these could be helpful, and there are some encouraging initial data.

Project weaknesses:

- There are two project weaknesses: (1) the ultrathin film catalyst layer may not be stable under 0.6–1.0 V potential cycling conditions, and (2) there is no clear proposed work to minimize the water flooding on the thin catalyst layer under high current density operation.
- NSTF has had problems in the area of operational robustness and, while it is good to see that some advances may have been made, it is unclear why/how these occurred and whether they can be further advanced.
- The extent of catalyst improvements achievable versus standard NSTF may be limited by the limited surface area of the NSTF support structure and the thinness of NSTF catalyst layers. The presentation made some mention of increasing the area of the support, but in the absence of detailed plans for this and the limited progress made in past attempts, the ultimate improvements from this project may be incremental at best.
- Stability of gradient structures is unknown. It is unclear as to how rapid the high-temperature route and process is and whether it is truly high-throughput. Also, it is not clear what the focus and evaluation mechanisms are.
- The most significant technical barrier of this catalyst technology is operational robustness and reproducibility of the performance. It was considered that the ionomer-free catalyst layer of this catalyst technology could be a problem. There is not enough mechanistic understanding of how this ionomer-free catalyst works. Study in this area is one of the most important work areas. The project does not cover this.

Recommendations for additions/deletions to project scope:

- The project should make a renewed effort at growing significantly longer NSTF whiskers to increase overall surface area and thickness of the catalyst layer.

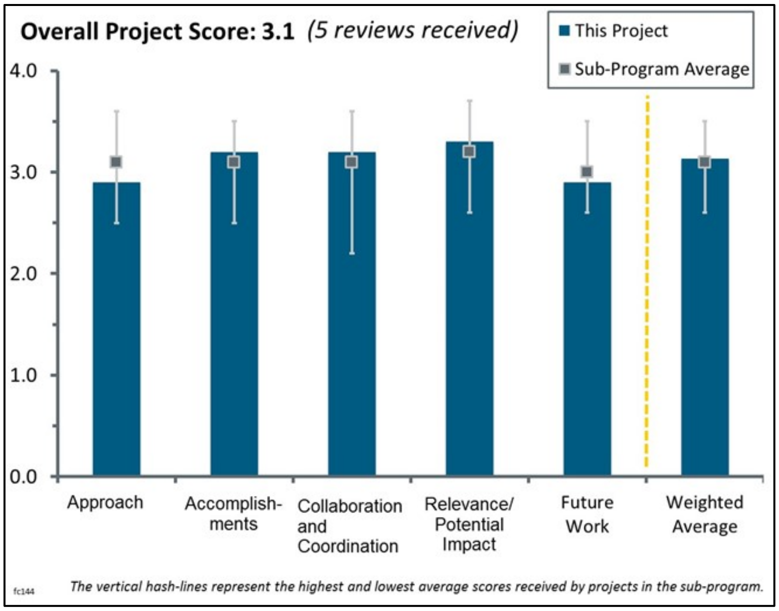
- The project needs to report conditioning procedures in the robustness of these materials. The project should measure transport and surface properties related to operation in cells as well. For dealloying, it would be good to be in contact with the ionomer as well to see whether it continues any leaching, which could explain performance differences between liquid and MEA.
- The project should have less DFT/kMC modeling related to catalyst performance, increase effort in operational robustness, and perhaps add water management/fuel cell performance modeling to help this area instead.
- The project emphasized the strength of this thin film-based catalyst technology to achieve further high-catalyst activity. On the other hand, the project scope does not include the generic problem of this catalyst layer structure—the so-called ionomer-free catalyst layer—and it is difficult to achieve sufficient operational robustness and reproducibility. It is highly recommended that the team revise the project scope and cover the operational robustness and ionomer-free catalyst layer study. Also, DOE RD&D should address the operational robustness in the technical target. The current transient performance target is not enough to address this attribute.

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

Anu Kongkanand; General Motors (GM)

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 heat-rejection and platinum-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 (HSC). The project takes a four-pronged approach: (1) improve oxygen transport with new carbon support, (2) reduce electrolyte–platinum interaction, (3) enhance dispersion and stability of platinum–cobalt particles, and (4) improve understanding and control of leached Co2+.



Question 1: Approach to performing the work

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

- The approach is very sound and is tackling local resistance, although the exact cause is still unknown, so it is a bit of a shotgun approach. It is not certain that the dispersion of the PtCo is the most efficient route, especially compared to the interactions with the Pt and support and ionomer interactions. It would be good to see some more down-select of the different strategies and what happens. It is not clear how Pt findings translate to Pt alloy. There is a good systematic approach for MEA integration but not so much so for the catalyst. Multiscale modeling is a good approach, but how the bridging will occur is unclear. Anchoring of the Pt is a good approach and is interesting.
- One of the approaches for this project—namely, enhancement in dispersion and stability of PtCo particles—has been widely studied and is well documented in the literature. The other approaches, such as improvement in oxygen transport with new carbon support and reduction in electrolyte–platinum interaction, may provide useful information in designing a stable catalyst for the oxygen reduction reaction (ORR).
- There is a problem with the proposed approach. On slide 6, the approach is discussed in light of what the investigators would like to do rather than how they will organize and perform the experiments. Slide 6 summarizes many challenges that polymer electrolyte membrane fuel cell (PEMFC) systems currently present. The strategy or approach is a methodology of how these challenges may be overcome or significantly reduced. That said, the four challenges listed on slide 6 are very important in the quest to move the field of fuel cells forward. On the other hand, in slide 7, the investigators describe what they are going to do rather than how they are going to do it, which is necessary when defining an approach. Furthermore, some questions raised on slide 6 are puzzling. For example, it is not clear what HSC has to do with the kinetics of the ORR. The kinetics of any electrochemical reaction are clearly defined by the electronic properties of the catalysts and should not be dependent on the surface area of either the catalysts or the support. Certainly the number of active sites will control the measured current but the specific activity should be the same. Rigor should be used in defining terms that control activity. Returning to the approach, it would be of paramount importance to discuss what experimental and computational tools will

be used for addressing the many question marks on slide 6. Slide 6 should be followed by slide 20 (selection methods).
- This project is basically taking an empirical approach and characterization with conventional materials. The overall project approach seems to be balanced between material fabrication, modeling, and characterization. It is hard to see what the new development is. Nitrogen-doped anchoring would be one of the new developments but it is a small fraction of the entire project. It is hard to understand why this project is still looking at HSC as a support, and the rationale to choose a conventional PtCo catalyst is also unclear. There is no information about new ionomer materials to be used for this project. Nanoscale diagnostics (visualization techniques) seem to be effective but how it connects to the modeling is still a question.
- This project was awarded and presented as a catalysis project, yet it is nearly exclusively a membrane electrode assembly (MEA) development project. In terms of the approach, half is MEA and half is catalysis; this does not seem appropriate for a catalysis development project. Of the half that is catalysis (New Carbon Support and Enhance Dispersion and Stability of PtCo Particle), even the portion related to the carbon support is described fully as an MEA task. The approach for “Enhance Dispersion and Stability of PtCo Particles” was missing from the presentation; what is intended to be done here is unclear, let alone new.

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.

- For a newly funded project with a start date of April 1, 2016, the amount of information presented under Technical Accomplishment is quite impressive.
- An impressive number of results have been presented in the report; therefore, the investigators should be encouraged to continue with collecting data with the same speed. Understanding the role of Co2+ in oxygen transport is very important. It is somewhat puzzling, however, how it will be possible to resolve this issue simply from testing the fuel cell stack. It would be of principal importance to develop a fundamental program that can provide more reliable data on relationships between concentration of Co2+ and oxygen transport. It was difficult to understand from the presentation what methods will be used to establish dissolution of Co2+ from PtCo during operation and how to separate the loss in activity from Co being leached out from the alloy (a true kinetic effect) versus the effect of Co2+ on oxygen transport. As a consequence, it is very important that effort is directed toward acquiring more reliable data on relationships between the activity and stability of a PtCo alloy and the concomitant effect of Co2+ on oxygen transport.
- The project just started, but some interesting pre-work or early work data was presented. In particular, the pie chart showing the fraction of local oxygen transport resistance in the entire mass transport overpotential is interesting, but whether it is empirical data or conceptual estimation is still a question.
- This project is new, is described as 1.2% complete, and had been ongoing for approximately two weeks before the slides were submitted. It is too early to comment on accomplishments and grade. Past projects involving this principal investigator have gone well.
- This is a new project.

Question 3: Collaboration and coordination with other institutions

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

- The subcontractors for the project include an original equipment manufacturer (OEM), three universities, and a national laboratory. All the individual entities have their proven expertise in the proposed research topic.
- It is a large but structured team. It is unknown how well the interactions— including knowledge and material transfer—will occur throughout the various members. It is not clear how this project overlaps and works with the Fuel Cell Consortium for Performance and Durability because, as presented, it is an MEA project and not a catalyst project. It is not clear whether MEAs will be provided.
- This project is new and subcontracts were not yet in place. As described, there is a good set of collaborators intended for the project but it is yet to be determined how they will interact.

- There is balanced collaboration among academia and industry.
- It is too early to judge the collaboration; the project has just started.

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

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

- The project is critical to the Hydrogen and Fuel Cells Program (the Program) and fully supports DOE research, development, and demonstration (RD&D) objectives. In particular, the project aims at a platinum-group-metal content of <0.125 g/kW, mass activity of 0.44 A/mg, mass activity loss of <40%, and performance at rated power of >1.0 W/cm^2.
- Adding the area-specific current density to the target is relevant to the DOE RD&D and automotive goals.
- There is high relevance with a potentially high impact on the Program.
- The project is addressing a major problem of local resistance. Goals are in line with needed improvements. This is not an electrocatalyst project.

Question 5: Proposed future work

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

- The team has proposed an enormous amount of future work, including carbon support selection, ionomer selection, ionic liquid selection, electrode design selection, and intermetallic alloy development for the first-year work flow.
- Future work is relevant to the project objectives and approaches.
- In general, the proposed future work is reasonably well organized. One direction that needs to be improved is developing methods capable of resolving the issues of stability of Co, as well how to optimize the physicochemical properties of nanoparticles to optimize activity and stability of catalysts.
- Future work seems in line with the goals and down-select. What types of MEA testing will be done, including cell assembly and conditions, is not clear. Although the future work is focused on performance, one must also worry about durability, especially at low loadings. The stability of the ionic liquid during operation is unclear, especially low temperatures and startup. There is no electrocatalyst development.
- The project is new; the entire approach can be considered the future work as there is 99% to go. The project as presented is an MEA project and not a catalyst project. Little to no information was presented in terms of what the project intends to do to develop higher-activity, more-stable catalysts. It appears that this project will primarily select materials already developed and then incorporate them into MEAs to determine the performance. It is unclear how this is appropriate for a catalysis project.

Project strengths:

- The project is tackling a critical problem with divergent approaches. There is a strong team and a systematic approach.
- The goals are clearly defined. This synchronized experimental and modeling effort will guarantee a fast transition from understanding transport limitations and performance of PEMFC.
- The strength is characterization of catalyst layers to improve the performance for both catalyst activity and mass transport at higher current density.
- The approach of minimizing oxygen transport and selecting materials for the highest transport should lead to improvements in MEA performance.
- The team has very strong collaboration.

Project weaknesses:

- One key weakness is the lack of a clear path toward understanding and minimizing the dissolution of Pt and Co during fuel cell operation. Currently, understanding the activity of Pt-based materials is of lesser importance than understanding the stability issues.
- It is hard to see a factor of the new development, such as new materials and diagnostics, in the project. Nitrogen implantation for catalyst anchoring seems to be a part of the new development. The new ionomer material is still unknown.
- How the members of the large team interact with each other is not clear. This is an MEA and not a catalyst project. The project should examine and be concerned about stability and durability.
- The project should comment on the costs of processing with ionic liquids versus cheaper solvents.
- There is no catalyst industry partner to scale up the proposed catalyst(s).

Recommendations for additions/deletions to project scope:

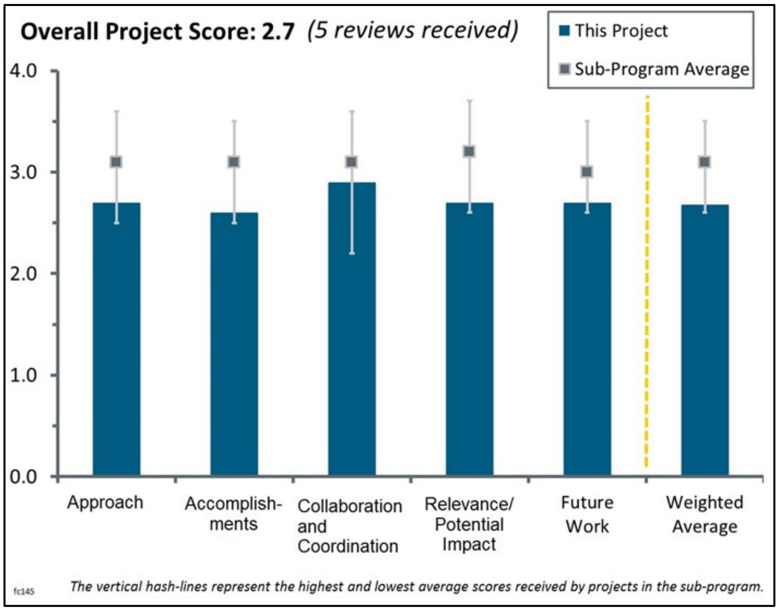
- More factors of new development, e.g., new materials and diagnostics, would improve the quality of the project. So far, it is hard to distinguish from conventional characterization, which is usually pursued by fuel cell OEMs.
- This is supposed to be a catalyst development project and thus catalyst development should be stressed and shown and not just its integration and electrode layer performance.
- If the project is to be a catalysis project, the project should concentrate on being a catalysis development project, not an MEA development project.
- A catalyst manufacturer should be included in the initial stage of the project.
- There are no recommendations at this stage of the project.

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

Vijay Ramani; Illinois Institute of Technology

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 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 platinum, and demonstrate high electrocatalyst performance.



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Question 1: Approach to performing the work

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

- Use of non-carbon supports provides the one real hope for completely materials-based solutions to start–stop and fuel starvation challenges—no carbon can survive these conditions without systems-level mitigations. This project is a rational continuation of the predecessor project that obtained some encouraging results albeit by introducing some potentially expensive and corrodible platinum group metal (PGM) (Ru) into the oxide support. This project attempts to further improve performance while removing all PGMs from within the support. The targeting within the project seems reasonable, and the choice of materials to be tested seems good though not particularly innovative. The leaching of sacrificial silica seems to be an effective way to achieve decent support surface areas. The proposed evaluation techniques seem appropriate. It should be noted that getting an oxide support to work at all in a fuel cell is much more difficult that getting it to work in a rotating disk electrode (RDE). One needs to completely reengineer the electrode layer because of the different hydrophilicity and density of an oxide support vs. carbon and the challenges in maintaining electrical conductivity and avoiding dissolution over the full operating range of the fuel cell. Seeing any fuel cell performance at all with an oxide-supported electrode layer is a major achievement.
- The approach is good, but it is unclear whether this is a catalyst or a support project. Use of modeling to guide materials development is good; however, it is not clear that the density functional theory (DFT) will include solvent to understand surface and leaching under operation. It is also not clear what the Pt particle sizes are and also whether alloys will follow the same trend for interactions.
- The investigators proposed to use DFT calculations to understand how doping may change the electronic structure of TiO_2 and, in turn, its conductivity. Although this is an important step, it is also very important to point out that the deposition of Pt on doped semiconductors will also affect the conductivity of Pt via the very well-known semiconductor–metal interactions (e.g., the Schottky barrier). The investigators also proposed testing the stability rather than using analytical probes such as inductively coupled plasma mass spectrometry to quantify the stability of Pt and the dopant use to increase the TiO_2 conductivity.
- Finding alternatives to corrosion-prone carbon supports is a worthwhile endeavor. Simply improving on the corrosion resistance without maintaining or improving other properties (conductivity, surface area, mass/specific activity) is insufficient. Focusing on Ti, Ta, and Nb is justifiable. Most of the presentation focused on approach; and the value of DFT modeling, which was the focus of two slides, is unclear as is

the value of porous supports (the accessibility of which is uncertain) or scale-up (which is not really relevant at this time—not until high-performance, durable materials are demonstrated).

- The project should clarify lessons learned from previous projects on metal oxide support materials (FC-085). Although some catalysts with metal oxide support developed under the previous project (FC-085) showed impressive durability under accelerated stress tests (ASTs), the catalyst performance was very low. Electrochemical surface area (ECSA) was very low. The basic catalyst performance is one of the most crucial criteria of non-carbon support catalyst material. However, it is not addressed in this project. Doped TiO_2 could be conductive (semi-conductor), and it is still questionable whether it can be adequately conductive as a catalyst support material. The project should clarify the basic idea of how the catalyst performance can be achieved with non-carbon-based support materials. The project should clarify criteria for material to be used as a catalyst support. The project indicated that porous TiO_2 support is an idea and this material may increase the surface area of the support materials. The question is whether the dispersion of Pt catalyst particles can be enough to achieve the catalyst performance, including ECSA.

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

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

- This topic is harder to judge as the project is at its beginning stages. The fact that some data is included is good plus the inclusion of a new support made by sol-gel and the characterization of $Ta_{0.3}Ti_{0.7}O_2$ that can be attributed to this work. Much was made of strong metal–support interactions (SMSI) with either modeling or XPS/XAS (x-ray photoelectron spectroscopy/x-ray absorption spectroscopy) results being suggested to support the statements made. Not enough data were presented (nor was a methodology sufficiently discussed) to make an assessment of the ability to quantify SMSI in a meaningful way or show how it has a meaningful impact on any relevant properties. The BET (Brunauer–Emmett–Teller) surface areas achieved/targeted seem low. All mass activities reported to date are so low as to be unexciting.
- The project was just started, and there was not enough time to show significant accomplishment. Some pre-work and early work were introduced. The project sees potential for Ti-based materials to show good corrosion resistance under the fuel cell operating environment. However, the promising information or data shown is contingent upon adequate electrical conductivity. The project indicated that it was pursuing porous TiO_2 support, which may increase the surface area of the support materials. The question is whether the dispersion of Pt catalyst particles can be enough to achieve catalyst performance, including ECSA.
- Preliminary work has shown some performance of a Ta-doped TiOx-supported electrode in a fuel cell (it is not clear whether this was in oxygen or air). Although that performance is inadequate, this alone is a major achievement. The claims for substantial stabilization of Pt on Ta-TiO_2 via SMSI seem to be stretching the interpretation of the data a bit far. SMSI should help (particularly after strong reduction of the oxide), but how much it should help seems unclear.
- It is too early to judge the accomplishments of the project; the accomplishment slides are a mixture of previous results and “new” data that are very difficult to decouple.
- This is a new project.

Question 3: Collaboration and coordination with other institutions

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

- While limited in the number of participants, the inclusion of two academic institutions with an original equipment manufacturer (OEM) is reasonable for this effort. The best addition to the team would be an industrial entity with experience synthesizing/selling/supplying similar materials to those targeted in this project.
- This is a decent team with an OEM and academia. It would be good to see more interactions with some of the national laboratory consortia. The exact role of the Illinois Institute of Technology (IIT) is not clear as a good deal of materials development is seemingly done by and at the partners.
- The project includes both academic and industry partners; however, the roles of each partner were not well defined. In particular, there is a still question as to who would be responsible for the catalyst performance.

Corrosion-resistant and conductive metal materials are similar to the metallic bipolar plate requirement. TreadStone Technologies worked on doped TiO_2 as the bipolar plate coating under the DOE Small Business Innovation Research Program. TreadStone Technologies can be a candidate partner for the support materials development.

- The project seems highly dependent on Nissan for testing in membrane electrode assemblies (MEAs). As long as such MEA work happens early and often in the project, this is okay. RDE work is close to irrelevant in oxide support work. IIT would be well-advised to develop in-house MEA testing capability, or at least to draw the Fuel Cell Consortium for Performance and Durability's MEA testing capabilities strongly into the project.
- The project was initiated four months ago, so it is very difficult to provide any objective judgment on the collaboration.

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

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

- If oxide supports could be made to work, they could provide a materials solution for startup and fuel starvation. This would simplify fuel cell system operation, would very modestly reduce system cost, and could ease the entry of new fuel cell system developers into the field. It should be noted just how hard it is to engineer new oxide-supported electrode layers with the full performance of carbon-supported electrodes, so the probability of this project's full success should be considered quite low.
- The subject of support is very important, although there is skepticism that it will be possible to replace high-surface-area carbon in the near future. Certainly, such an effort is a logical response to many problems presented by the carbon supports currently used.
- It is not clear how much of a concern carbon corrosion still is as there are system mitigation strategies, although a materials solution would be better. It would be good to know the cost comparison between the new supports and standard Pt/C materials.
- Corrosion concerns are an area that can be improved upon; however, system solutions have been developed that allow today's corrosion-prone materials to meet durability targets (while also achieving performance targets). The poor relative performance of the materials developed in this project (and its predecessor) leave a significant concern that materials with comparable performance can be generated. It is unclear that if such materials are created, they will have a meaningful impact on commercial viability of fuel cell systems.
- Basically, the research focus is shifting from the catalyst itself to the catalyst layer to maintain high kinetics of mass activity *and* enhance the performance at the high current density. In the industry, carbon corrosion has been mitigated by so-called system solutions. The importance of this durable catalyst support is relatively lowered.

Question 5: Proposed future work

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

- Based on the addressed approach, the future work is relevant.
- The testing proposed is reasonable and relevant. The key issues are the performance and durability of materials generated within the project. Based on the data presented and the proposed material sets investigated, it is not clear that performance and durability improvements are possible that would lead to novel supports of high commercial relevance.
- The future plans are reasonable except that essentially all testing should be done in MEAs rather than in RDEs to avoid developing false hopes (although if the support does not work at all in an RDE, it is not worth testing in MEAs). A few more specific ideas on other dopants to use with TiO_2 and on other base oxides to be tried would be helpful in justifying the continuation of this project.
- Future work is a bit broad and so some specificity would be good, especially against metrics. The presentation was vague concerning ways to enhance activity and stability.

- Some elements are there, but a key element that is missing is quantifying the stability of the support and the catalysts.

Project strengths:

- The principal investigator (PI) seems to have a good grasp of the challenges associated with developing effective oxide-supported electrodes and of the methods needed to assess the origins of performance shortfalls. The project plans follow rational if not innovative lines of development.
- The PI has proven in the past that he is able to develop and execute similar projects. The methodology is rather well developed.
- The project has a systematic approach for new materials and supports and for conducting oxides. There is good use of modeling to guide materials development.
- The team is investigating reasonable materials for advanced supports, and the proposed work is scientifically sound.

Project weaknesses:

- The project does not address technical problems of metal support, including the catalyst performance issue experienced by the previous project. Also, this project does not scope how the adequate catalyst performance can be achieved with metal oxide support.
- The proposed approaches are rather obvious and not particularly innovative. It is not clear why they would not already have been followed through to completion under the predecessor project.
- There is no alternative direction if the proposed systems will not work as planned, and there is skepticism that it will work.
- It is unclear that the materials being pursued will ever come close to the performance of current state-of-the-art materials in use today.

Recommendations for additions/deletions to project scope:

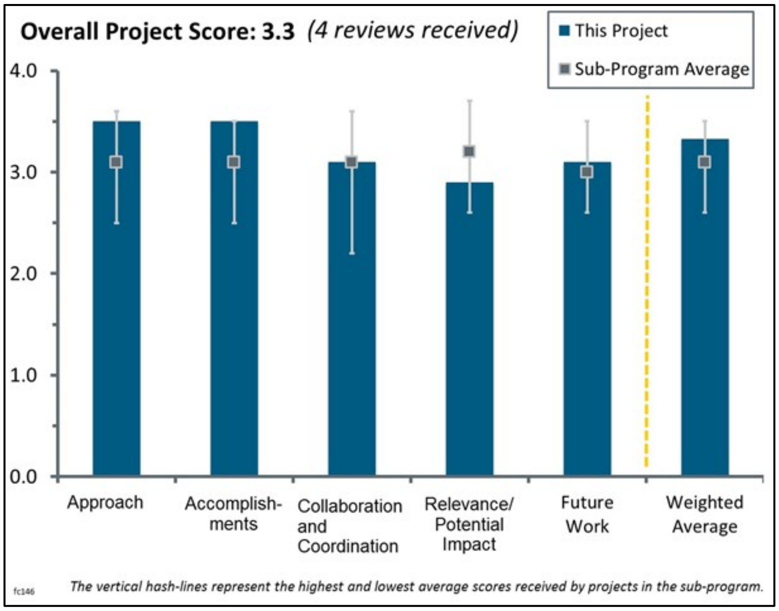
- The project should clarify lessons learned from the previous project that investigated metal oxide support materials (FC-085). Although some catalysts with metal oxide support developed under the previous project (FC-085) showed impressive durability under AST, the catalyst performance was very low. ECSA was very low. The basic catalyst performance is one of the most crucial criteria of non-carbon support catalyst material. However, catalyst performance is not addressed in this project. Doped TiO_2 could be conductive (semi-conductor), and it is still questionable whether it can be adequately conductive as a catalyst support material. The project should clarify the basic idea of how the catalyst performance can be achieved with non-carbon-based support materials. The project should clarify criteria for the material to be used as a catalyst support.
- The project should publish a series of protocols for the evaluation of oxide-supported electrode layers. The protocols should include tests for electronic conductivity after (separate) exposure to electrochemical potentials at both oxidizing and reducing potentials, tests for support dissolution, tests for surface area retention of both the catalyst and the support, and tests of hydrophilicity and changes thereof during electrochemical operation. The project should recommend at least three other oxide/dopant compositions for future investigation. More attention should be given to control of hydrophilicity in oxide supports.
- Key are the interactions between support and ionomer, as well as the surface properties, and so it would be good to add such characterization. It is recommended that the project investigate some high-throughput techniques and approaches to progress the supports faster, which DOE can leverage. The project should focus on and address possible passivation under different operating regimes.
- It would be preferable to see one support family pursued in more detail, with the project trying to define how much improvement in performance and durability might be possible within a single class of supports, as performance to date of these materials falls significantly behind current state-of-the-art materials.

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.


Overall Project Score: 3.3 *(4 reviews received)*

The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Question 1: Approach to performing the work

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

- This team is managed out of Los Alamos National Laboratory, but it includes three groups making alkaline ionomers, which are all being tested in parallel. The team members follow some guiding principles they have developed over the last five years regarding what sorts of chemical functional groups are likely to be stable or not stable in concentrated alkali. In particular, they seek to avoid aryl ethers, which they say will always be unstable in alkali. This approach is sensible and likely to lead to materials with superior properties. Their stability studies are done in such a way that relative stabilities cannot be easily assessed because they simply show that materials are stable under a particular set of conditions. It would be better to compare materials, or to use conditions that eventually lead to degradation, in order to discuss lifetimes and not just the presence or absence of decomposition. On the effect of organocation adsorption on redox reactions, the effect of cations on oxygen reduction reaction (ORR) kinetics is known, but the effect on hydrogen oxygen reduction (HOR) is unclear.
- The approach focuses on the critical barriers of backbone and cation stability in the presence of hydroxide ions. The principal investigator (PI) has a long history with AEM development and has narrowed the approach to a system that has a good chance of success.
- The approach to improve AEM stability by preparing AEMs without ether or electron-withdrawing groups in the polymer backbone in promising. Replacing benzyltrimethylammonium with alkylammonium has been done before, but it is still an effective means of improved stability. It is encouraging that a wide variety of relevant properties were measured and cost was considered in the down-selection, although selection criteria and factor weighting were not defined. The approach for electrode ionomer down-selection is also rational.
- The project is addressing key AEMFC challenges with a multi-faceted approach towards optimization of ionomers for each electrode independently in addition to the AEM.

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

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

- Selected AEM poly(biphenyl alkylene (PBPA1+) has excellent ex situ thermal stability and reasonable conductivity. Swelling is still quite high. The films show reasonable mechanical properties (e.g., 100% elongation). The areal specific resistance (ASR) measured in fuel cell operating mode is also promising. The selected cathode ionomer with side chain shows improved performance, but there is still much improvement needed. The anode binder stability looks good, but the cathode binder is not stable in tetramethyl ammonium OH (TMAOH). Overall, the project has demonstrated excellent progress in a short time period.
- The team has made very good progress, having identified several ionomer systems with high alkaline stability and good ASR in preliminary MEA/single-cell tests. The work on organocation effects on ORR and HOR is good, but it needs a little more fleshing out. For example, the nature of the films on Pt is unclear and could use some clarification regarding thickness, nature of bonding, etc. It would be nice if there was some evidence besides the infrared (IR), which was described in only general terms. It not clear how the IR experiment distinguishes between signals from surface and bulk-solution species. There are several ways to make that distinction, and the PI should clarify how he did it. Regarding other approaches, perhaps some quartz crystal microbalance (QCM) studies would be helpful to determine how much material has deposited on the Pt. Film formation in water may not be indicative of what happens in an MEA. Cell tests with various combinations of membrane and electrode ionomer indicate progress, but a firm understanding of factors affecting cell behavior is still lacking. Perhaps an injection of modeling expertise would help.
- The PBPA+ approach demonstrated very good conductivity and durability. The side-chain poly(phenylene) (PP) ionomer for the cathode yielded significantly improved hydrogen/air performance over the benchmark ionomer. There was good integration of rotating disk electrode studies on ORR poisoning. Understanding the impact of tetramethyl ammonium (TMA) adsorption is key to understanding HOR deactivation.
- While the final goals have not been meet, significant progress has been shown toward meeting the resistance target of less than 0.1 Ω*cm^2 and the performance target of 0.6 V at 0.6 A/cm^2. Good progress has been made towards understanding AEM ionomer stability to hydroxide attack.

Question 3: Collaboration and coordination with other institutions

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

- This highly collaborative project brings together a well-matched team to provide a wide range of materials with high alkaline stability. The PI provides very good support for materials characterization that complements the synthesis expertise of his collaborators at Rensselaer Polytechnic Institute (RPI) and Sandia National Laboratories (SNL). The team members contributing catalysts have not yet played a significant role but presumably will do so as the project progresses.
- The project has a strong team with relevant capabilities. It is unclear which achievements are attributed to which partner organizations.
- The collaborations with RPI and SNL on polymer synthesis are good. It is unclear what role Argonne National Laboratory has had, as only standard Pt catalysts have been tested.
- The plan to include all the partners looks good. It is unclear if the partners have been actively contributing or if their roles will become more important in the future.

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

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

- The project does an excellent job of meeting the Fuel Cell Technologies Office and DOE goals with respect to alkaline approaches to hydrogen fuel cells. It is not yet clear how AEMFC approaches will compete with polymer electrolyte membrane fuel cells (PEMFCs) for hydrogen-based energy conversion; AEMFC approaches are at a much earlier stage, and much is still unknown about the ultimate limits of AEMFCs. Research such as that being done in this project is needed to discover these limits.
- This project is well aligned with the DOE goals and is positioned to advance the understanding and state of the art of AEM membranes. The approach to develop different ionomers for anode and cathode is important for maximizing AEM MEA performance.
- It is unclear what potential impact AEMFCs will have commercially. The apparent need for high-Pt HOR electrode loadings to overcome sluggish alkaline kinetics, coupled with intrinsically lower OH conductivity, ultimately raises significant questions of relevance.
- It is unclear for what applications AEMFCs will be used, and the ultimate targets are not defined. It is highly unlikely that AEMs will ever be competitive with PEMFCs for automotive applications.

Question 5: Proposed future work

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

- The future work seems well aligned with the objectives. More MEA testing and in-cell durability testing to demonstrate that the progress made in out-of-cell testing can be translated to in-cell performance would be appreciated.
- The proposed future work seems reasonable, with the one caveat that the work with fluorinated materials is not well described.
- It is not clear why future work on perfluorinated anion-exchange ionomers is needed. The results on the hydrocarbon ionomers are encouraging, and the National Renewable Energy Laboratory (NREL) is already working on the perfluorinated ionomers. Future work does not include non-platinum-metal-group (PGM) catalysts. Ionomer selection may be sensitive to catalyst type, so binder selection would need to be repeated, especially considering catalyst interaction was a key criterion in the down-selection process. Work to further reduce membrane ASR is recommended to be competitive with PEMs.

Project strengths:

- The relatively wide range of ionomers with apparently good alkaline stability is a project strength.
- The project has a solid approach and excellent synthetic chemistry expertise. The project addresses all key performance and durability properties in the down-selection process.
- The project is addressing key AEMFC challenges with a multi-faceted approach toward optimization of ionomers for each electrode independently, in addition to the AEM.
- The project has a solid approach to address critical issues with AEM fuel cell ionomer development for membranes and electrodes. Significant expertise in the area of AEM development has led to the identification of viable ionomers.

Project weaknesses:

- Targets are not tied to any application. Even if targets are met, the technology will not compete with PEMFCs. There is no in situ durability testing planned.
- The approaches to low-PGM loading or non-PGM were not addressed. This project looks like an ionomer development project, but two of the partners are catalysis experts, and their contributions are not clear at this time.

Recommendations for additions/deletions to project scope:

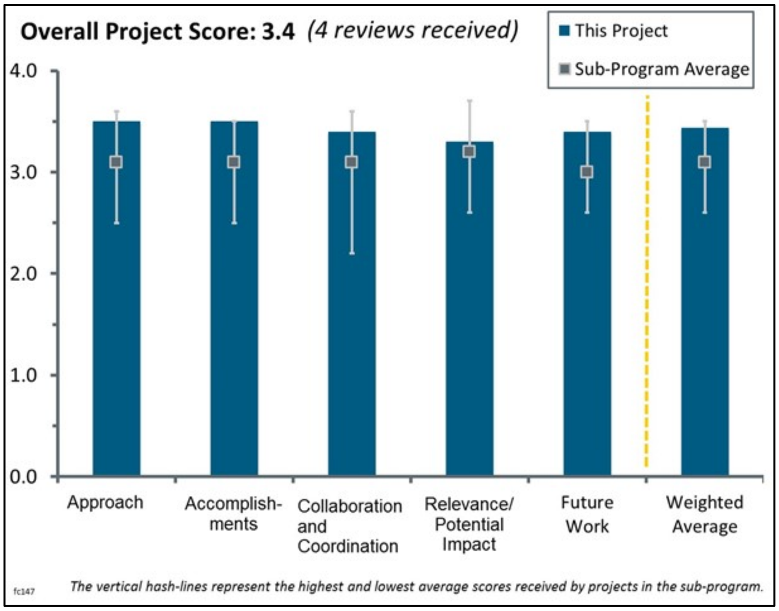
- The MEA/cell building and testing work could possibly benefit from a modeling contribution.
- Gas permeability should be measured rather than estimated from the chemical structure because of uncertainty about the accuracy of the estimate. The project should eliminate the perfluorinated ionomer work or at least ensure that it does not overlap with NREL’s work; the perfluorinated ionomer work is also likely to be more expensive. The project should continue to focus on ASR reduction and work on reducing swelling of membranes. Non-PGM catalysts should be considered in the binder selection process. In situ, non-steady-state durability tests should be run.

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.



The vertical hash-lines represent the highest and lowest average scores received by projects in the sub-program.

Question 1: Approach to performing the work

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

- The principal investigator (PI) continues an approach from a previous project, seeking to modify pre-formed perfluorinated sulfonyl fluoride polymers with diamines with subsequent quaternization to make fluoropolymer anion exchangers. The approach is reasonable, and the work is quite tightly focused on barriers and goals. The inclusion of modeling in the present project is especially welcome because it brings key insights into some of the reasons for different levels of performance in early-stage membrane electrode assemblies (MEAs) and related devices.
- The project addresses the two main barriers for fuel cells: cost and durability. By looking at alkaline membrane fuel cells, which can enable platinum-group-metal (PGM)-free catalysis, the project addresses cost. The project addresses durability of AEMs by utilizing a stable perfluorinated backbone and by investigating methods to add stability to the pendant quaternary ammonium group and sulfonamide linkage. The modeling effort is being used to help guide the electrode and MEA design. The approach of utilizing perfluorinated backbone polymers to enhance water transport is interesting and should provide benefits, as water management is an issue in AMFCs. The perfluorinated backbone approach also complements other U.S. Department of Energy Office of Energy Efficiency and Renewable Energy AEM work that focuses on utilizing aromatic backbone polymers, providing a diverse portfolio.
- The project has a good balance of materials synthesis, characterization, and modeling. Perfluorinated AEMs and AMFCs may provide key ultimate benefits toward high-performing MEAs owing to higher water transport capabilities than hydrocarbon AEMs. Development of AEM models and AEM characterization techniques are immediately relevant to addressing issues.
- The approach is well defined and balanced between synthesis, characterization, and modeling.

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

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

- The PI has made excellent progress. In the past year, the PI has solved several longstanding problems that have plagued this approach, particularly in the area of completeness of conversion for adding amines to sulfonyl fluoride, and subsequently quaternizing the amines to make ammonium salts. As a result, he has in hand multi-gram quantities of ionomer of high quality, which has enabled quick and excellent progress in nearly all other project areas. Conductivity data are quite reasonable in comparison with acid systems. Stability data show problems with the first generation (Gen 1) PF AEM polymer, but the nature of the instability is clear, and the PI is well positioned to make changes that will greatly improve stability; his second-generation (Gen 2) polymer promises to be excellent. Progress on MEA work is very good and reveals a need for deeper understanding of transport losses in alkaline systems, which differ from those in acid systems. The accomplishments from modeling are excellent in such a short time. Diagnostics from CO stripping and hydrogen pumping are also excellent, obviously reflecting the fact that this project is a continuation of prior work with significant investment already in place for both synthesis and diagnostic studies.
- Excellent progress has been made toward development of the Gen 1 PF AEM polymer with good synthetic yield and high conductivity. Durability, however, is very poor. Development of improved durability in the Gen 2 AEM is promising, but conductivity and other properties were not disclosed.
- The project has made very good progress towards the overall target, especially from the MEA performance modeling point of view.
- The project has made good progress developing an alkaline exchange membrane and integrating it into an MEA. The Gen 1 PF AEM polymer performs better at the beginning of life than the Tokuyama membrane. The modeling efforts have identified the importance of anode flooding and water management in the MEA. The project has refined techniques to determine hydrogen oxidation reaction /hydrogen evolution reaction HOR/HER exchange current density.

Question 3: Collaboration and coordination with other institutions

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

- The team has excellent participants well known in the polymer design, characterization, and modeling of fuel cells.
- The team's collaboration is very good, particularly with Lawrence Berkeley National Laboratory for modeling. Collaborations with Oak Ridge National Laboratory were in just one area, and work with Colorado School of Mines was not clear from the presentation, but both are reasonable and probably contributing important information either now or in the near future.
- Collaborations within the team are good. Membranes are being characterized and incorporated into the MEAs. Characterization and modeling feed back into the MEA and membrane preparation. 3M's input has helped utilize the perfluorinated backbone. The project lead is collaborating with other AEM projects as evidenced by the workshop organized by the National Renewable Energy Laboratory and Los Alamos National Laboratory PIs of AEM projects.
- The team consists of recognized leaders in ionomer development and characterization.

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

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

- This is one of the best projects for making progress in alkaline fuel cells. Alkaline systems are at a much earlier stage of development relative to polymer electrolyte membranes (PEMs), particularly with fluorinated ionomers, but work such as that being pursued in this project will rapidly help identify the

similarities and differences between the two systems, which is needed to learn where the correct niche is for alkaline systems.
- The successfully finished project will have a substantial impact on implementation of novel AEMs as a viable alternative to conventional PEM fuel cells (PEMFCs).
- The impact of the AEM membrane work will be dependent on whether an effective PGM-free anode catalyst material can be developed or whether the anode and catalyst loadings can be reduced below those currently observed in PEMFC systems. With Tokuyama advertising their plans to stop supplying their AEM to developers, a membrane material with good performance and high enough stability to allow performance measurements is needed. This project could provide a baseline (or develop a material that could be a baseline with a small company manufacturing and supplying a large benefit to the field) for the AMFC community. A baseline material is needed to help develop catalysts and electrodes, and in this regard, an AEM that can be made at scale would have a large impact by allowing the research community to develop other materials while the membrane is being developed further.
- It is unclear whether AMFCs will ultimately achieve commercial relevance because of poor HOR kinetics and lower conductivity than PEMFCs. Performance under hydrogen/oxygen with high Pt loadings is similar to PEMFCs under hydrogen/air.

Question 5: Proposed future work

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

- The future work is relevant and addresses AEM issues. Variations in conditions for the model should provide useful data to guide the polymer chemistry and MEA design changes. Details about strategies (other than lengthening the side chain and getting rid of the sulfonamide linkage) for increasing stability need to be presented.
- The proposed future work is in good alignment with remaining challenges and barriers as well as with the overall goal of the project.
- The proposed future work is fully appropriate.

Project strengths:

- As far as the reviewer is aware, this is the only current work exploring tetrafluoroethylene/trifluorovinyl ether (TFE/TFVE)-based fluorinated systems for alkaline energy conversion. The project has made excellent progress and is bringing together a team well matched to project needs.
- The project team has good chemistry, and the team members have worked together. 3M's partnership provides a wealth of perfluorinated polymer backbone chemistry.
- A project strength is the understanding of materials design and synthesis. The characterization methods proposed are well established for such types of materials.

Project weaknesses:

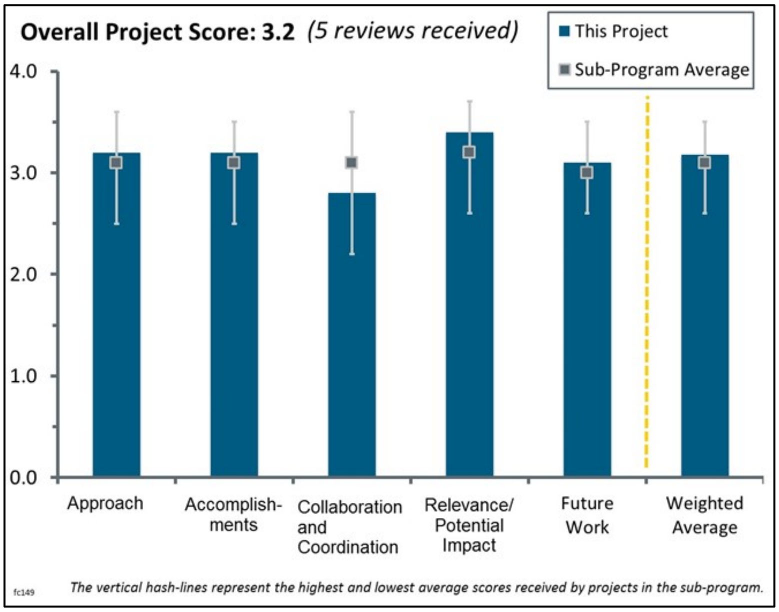
- AEMFC systems in general are not as well developed as PEMFC systems, so the ultimate limits of what can be done are not known. It may be that there are power limits, or durability limits, or other as-yet unidentified limits that will ultimately make AEM systems not competitive. This is a potential weakness of this general area, but it will take projects such as this one to determine whether AEM systems can be competitive.
- A project weakness is the lack of details on MEA fabrication.

Recommendations for additions/deletions to project scope:

- The project should expand the part on achievement of MEA performance using new types of ionomers and membranes. Overall MEA performance depends on many parameters and characteristics of the ionomer, and membranes are only part of optimizing the whole process.

Project #FC-149: Multiscale Modeling of Fuel Cell Membranes

Adam Weber; Lawrence Berkeley National Laboratory



Brief Summary of Project:

Despite broad use of ionomer membranes such as Nafion® in energy research, operando behavior prediction is unavailable. Understanding multi-ion transport in various ion-rich solvents would enable ionomer and system optimization. This project will help optimize and explore design criteria for transport in ion-conducting membranes across length scales in various environments. Researchers will use a novel multiscale modeling methodology to examine and detail controlling interactions for ion and solvent transport.

Question 1: Approach to performing the work

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

- Membrane performance is well defined in the model. It is a unique approach to take a network model to solve the membrane performance with nanoscale phenomena. If this approach can work (i.e., be validated), it can be used to simulate the aged membrane performance with structural changes. Modeling and characterization in this project seems to be duplicative of FC-PAD. The project scope should be distinguished from that of FC-PAD.
- This is a very ambitious project with a relatively small budget. The principal investigator (PI) is addressing, or plans to address, all of the key barriers. The approach of using multiscale models to bridge macro and nanoscale models may be required to model bulk membrane transport properties. To accurately model water uptake and transport, it is likely that long-range polymer motion will need to be included, which will be computationally intensive. It is also unclear how the historical dependence of the microstructure will be addressed. Addressing cation mobility (Ce, Fe, Co, Ni, etc.) should be very valuable, as very little is known about this mobility. The PI may have overreached a bit on the goals of this project, but it is hoped that he is successful.
- This approach is good and is needed. Exploration of design criteria for transport is an objective. The PI could discuss manufacturing constraints with the Fuel Cell Consortium for Performance and Durability (FC-PAD) and with membrane manufacturers before and during the work on developing the design criteria. This discussion is necessary because some design criteria might not be feasible for mass manufacturing.
-
- The multiscale modeling of the membrane seems to be a development of pore-network modeling of electrodes, so the approach seems logical. However, such simulations might be computationally intensive.

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 modeling results seem promising and compare well to the experimental results. It is uncertain whether these results can then be used to predict how a new structure could look.

- Lawrence Berkeley National Laboratory has set up the pore and polymer electrolyte membrane (PEM) networks, has run initial simulations for Nafion, and has been able to obtain tortuosity values by fitting conductivity data. There is still much to do, and they are halfway through the project. The project partners claim they are on track for modeling water flux through a Nafion membrane measured for four different relative humidities and three membrane thicknesses, but it is not clear that they will complete this modeling.
- The model is validated in macroscale membrane performance data (e.g., proton conductivity) and shows good fit. A question is how to measure (or define) the water content of the membrane in the empirical data.
- The conduction network modeling with the pore network is very good.

Question 3: Collaboration and coordination with other institutions

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

- It is good to collaborate with FC-PAD. The modeling and characterization of this project seem to be duplicative of FC-PAD. Membrane/ionomer suppliers should be included.
- There is some interaction with FC-PAD and other developers, but it is unclear whether a team exists for this continued project. It looks like the project needs some collaboration partners.
- Collaboration is limited to discussion only. The project may benefit from stronger collaboration with other FC-PAD members and academia leading in modeling efforts (both in the United States and outside the United States).
- It is unclear how collaborators are contributing to this project. There should be opportunities for the team to work with FC-PAD for both data collection and structural evaluation.
- More interactions with membrane manufacturers are recommended to address feasibility of design criteria for mass manufacturing.

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 structural base membrane performance model is highly useful for membrane development and design of the operational conditions of the fuel cell system. The model is effective for performance and robustness.
- The membrane is one of the important components for fuel cell systems affecting the balance-of-plant system. By defining the transport better, the membrane can be designed more effectively, so the overall system can be more economical.
- If the PI can accomplish what he has set out to do, this project will be of great value to membrane developers seeking to create an ideal membrane microstructure. There will also be great value in being able to model cation transport within a membrane electrode assembly.
- This project has been used to model various aspects of fuel cell performance and electrode interactions. However, the relevance of this year’s work is unclear. Even assuming the project is wildly successful in being able to model the membrane and even recommending a great structure, there is no evident impact on the two biggest issues raised by DOE for fuel cell commercialization.

Question 5: Proposed future work

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

- Most of the future work planned should be very valuable, albeit unlikely to be completed within the timeframe of the project. This valuable future work includes expanding the model to include solvent uptake and transport, making the model dynamic, and expanding the model to include transport of impurities and contaminants such as cations.
- Proposed future work looks ambitious, but it is unclear whether the transient responses can be predicted well. It might help to focus on steady-state response and understanding material interactions. Further, it

might help if this work can develop this model in a highly parallel-processing computing environment (e.g., NVIDIA CUDA®).
- Dynamic modeling is challenging and a very good idea.
- Exploration of design criteria for transport is an objective. The PI could discuss manufacturing constraints with FC-PAD and with membrane manufacturers before and during the work on developing the design criteria. This is necessary because some design criteria might not be feasible for mass manufacturing.

Project strengths:

- The project strengths include using the multiscale modeling approach and the outstanding modeling and microscale structural analysis capabilities of the team at Lawrence Berkeley National Laboratory.
- The project should leverage the partner's knowledge of mechanistic understanding of membrane performance.
- The brilliant PI and his team's capabilities are strengths.

Project weaknesses:

- There is little evidence of collaboration. It is unclear whether the learnings will apply to membranes made from other ionomers. There is not enough time to complete the future work.
- The lack of strong collaboration and the lack of model validation with experimental data are weaknesses.
- There are no experimental interactions with collaborators.
- The project objectives are similar to FC-PAD's and might be duplicative.

Recommendations for additions/deletions to project scope:

- Perfluorosulfonic acid (PFSA) membranes should be included with other side chains (e.g., by 3M, Aquivion) as well as hydrocarbon ionomers. The PI claims that the main benefit of this project is that it can be used to determine an "ideal" structure for optimum conductivity. For this to be true, the model must account for different polymer structures with different chain mobilities. The PI should also address gas (H_2, O_2, N_2) permeance within the model.
- Project objectives are similar to FC-PAD's and may be duplicative. Performance degradation of aged membranes can be analyzed with this project approach (i.e., structure-based membrane performance model).
- If possible, the PI should develop this model in an open-source, highly parallel, multithreaded computing environment.