2019 – Fuel Cell R&D
Summary of Annual Merit Review of the Fuel Cell R&D Subprogram

The Fuel Cell R&D subprogram includes a diverse portfolio of fuel cell technologies to enable low-cost, durable, and high-performance fuel cells for a range of applications. Early-stage research and development (R&D) areas in fiscal year (FY) 2019 include catalysts and electrodes, membranes, fuel cell performance and durability, and assessments. Catalyst and electrode R&D comprises efforts on development and utilization of low-platinum-group-metal (low-PGM) and PGM-free catalysts, with the latter being the focus of work by ElectroCat (the Electrocatalysis Consortium). Membrane R&D includes polymer electrolyte membranes (PEMs), as well as alkaline membranes. Fuel cell performance and durability is the purview of the Fuel Cell Performance and Durability Consortium (FC-PAD). The Fuel Cell R&D subprogram has a portfolio that includes work on medium- and heavy-duty fuel cell applications. The subprogram also includes fuel cell system modeling and analysis, as well as efforts to develop components for unitized reversible fuel cells.

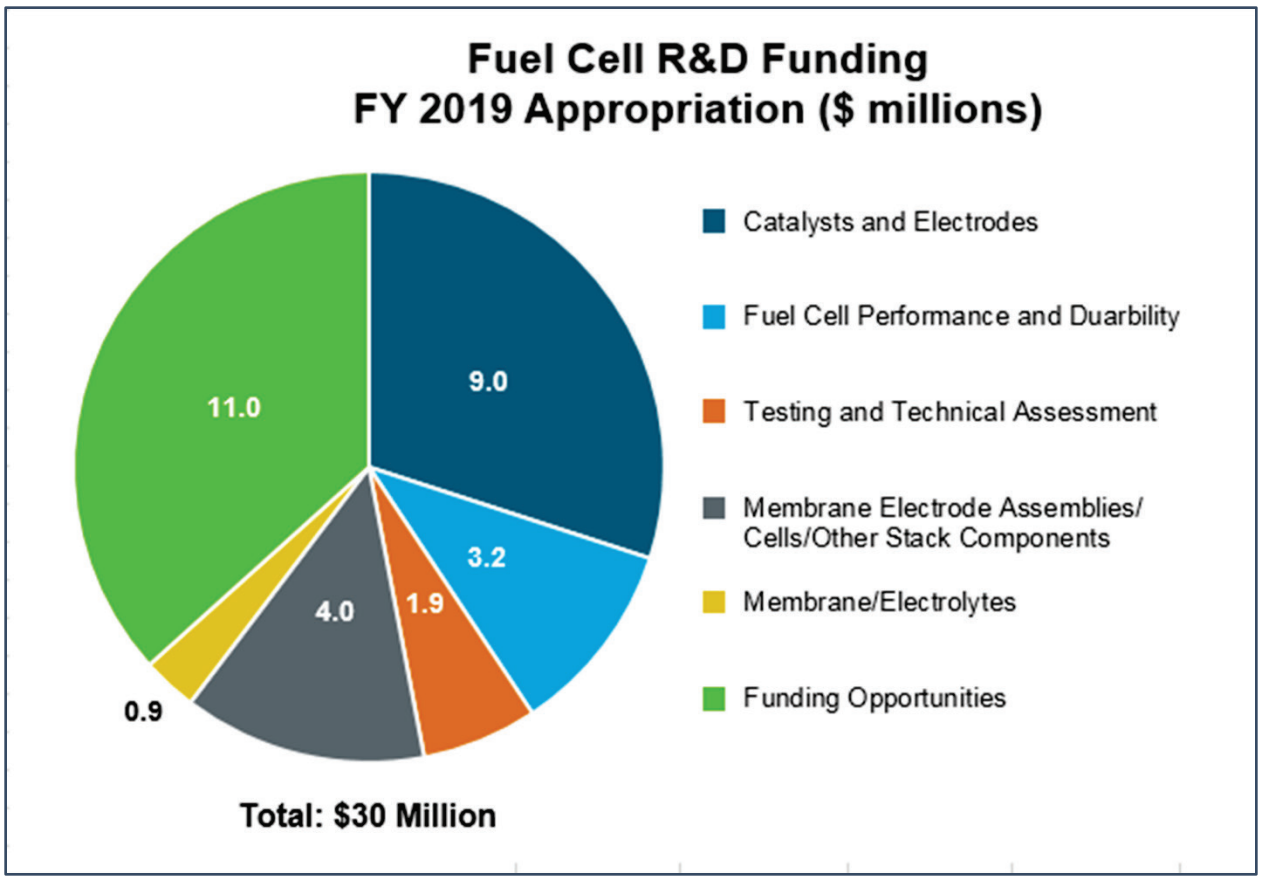
Summary of Fuel Cell R&D Subprogram and Reviewer Comments

The Hydrogen and Fuel Cells Program (the Program) reviewers noted that the Fuel Cell R&D subprogram has a comprehensive, well-structured, and focused project portfolio, with elaborated metrics and milestones, focused on low-technology-readiness-level (low-TRL) research with long-term impacts. The reviewers agreed that the general focus of the various initiatives are in line with the fuel cell industry's long-term needs and that early-stage R&D addresses the primary issues and opportunities. Program reviewers also specifically noted that the consortia approaches with ElectroCat and FC-PAD are well organized to reach the Program's goals and are providing opportunities for more efficient utilization of cross-laboratory capabilities. Furthermore, reviewers agreed that consortia should continue to focus on materials properties work in regard to fuel cell catalysts and fuel cell durability, both of which were repeatedly highlighted. ElectroCat was specifically noted as showing excellent progress in PGM-free catalyst development and performance in MEAs, while working on understanding and mitigating durability issues. Reviewers highlighted the increased emphasis on technology reliability and cycle life as positive. The subprogram was encouraged to continue to support and develop stronger links between the consortia and industry. Reviewers remarked that the consortia- and funding-opportunity-announcement-directed approach to funding will yield highly focused projects. Reviewers stressed the importance of cost reduction in fuel cells, especially in medium- and heavy-duty fuel cell applications. There was agreement that the increased focus on medium- and heavy-duty applications is a strength of the subprogram and will be critically important in advancing adoption in these sectors of industry; the Program was encouraged to continue focusing on these applications. Project reviewers were also impressed with specific project-level highlights and accomplishments, as detailed in the project review reports that follow this introductory summary. Certain individual projects were judged to have unclear or insufficiently defined pathways for hitting their targets and goals within the subprogram and, therefore, to require additional focus.

Forty-eight projects were reviewed, receiving scores ranging from 2.5 to 3.5, with an average score of 3.17. Each of the individual project reports in this section contains a project summary, the project's overall score and average scores for each question, and the project-level reviewer comments.

Fuel Cell R&D Funding

The Fuel Cell R&D subprogram received $30 million in FY 2019. The subprogram focuses on early-stage applied R&D to reduce fuel cell costs and improve performance and durability, as depicted in the figure below. The funding is expected to 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. Future work is expected to focus on meeting performance, cost, and durability targets for fuel cells with continued work through the consortium approach, further reducing PGM content in catalysts, and expanding the knowledge base to advance fuel cell performance and durability. Approximately $11 million was allocated toward funding opportunities that will support H2@Scale initiatives, including advancing reversible fuel cell stack technologies and prototype systems, as well as medium- and heavy-duty fuel cell applications.


Fuel Cell R&D Funding
FY 2019 Appropriation ($ millions)
9.0
3.2
1.9
4.0
0.9
11.0
Catalysts and Electrodes
Fuel Cell Performance and Duarbility
Testing and Technical Assessment
Membrane Electrode Assemblies/ Cells/Other Stack Components
Membrane/Electrolytes
Funding Opportunities
Total: $30 Million

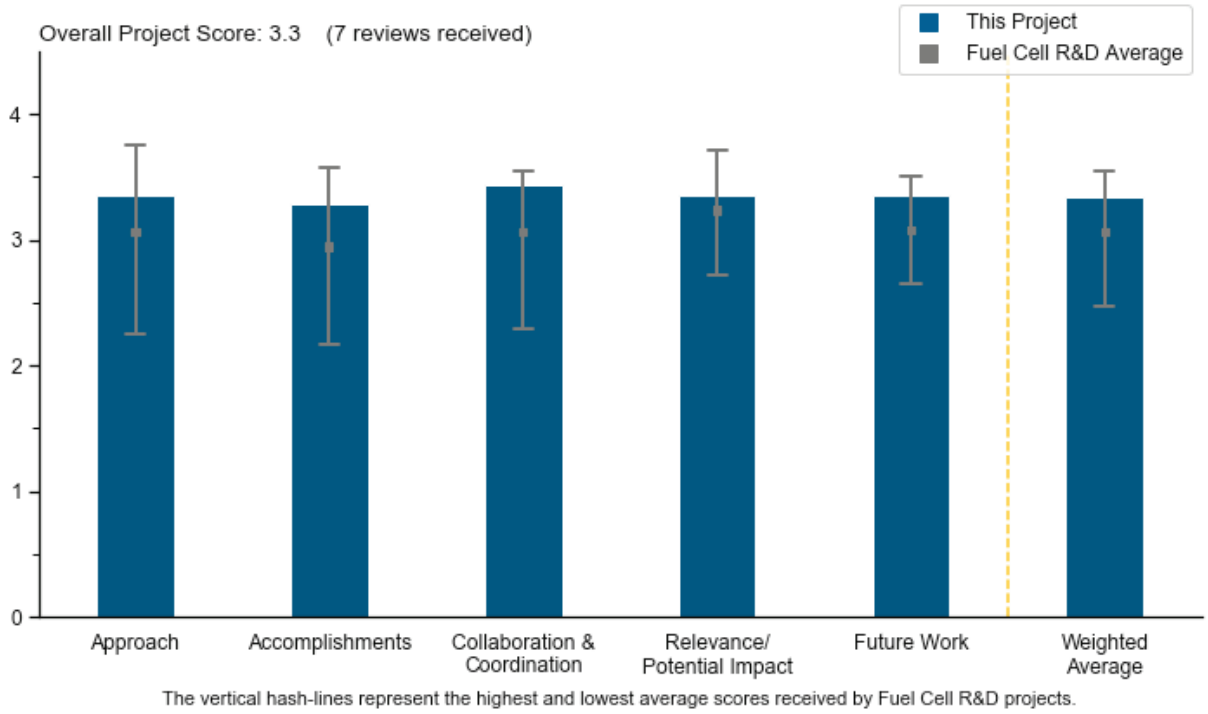
Project #FC-017: Fuel Cell System Modeling and Analysis

Rajesh Ahluwalia, Argonne National Laboratory

Brief Summary of Project

The objective of this project is to develop a validated system model and use it to assess design-point, part-load, and dynamic performance of automotive and stationary fuel cell systems. 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.

Project Scoring



Question 1: Approach to performing the work

This project was rated **3.4** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This work is highly relevant to DOE technical targets and provides a needed stack/system element to help ensure research is moving in the right direction. The approach involves development and refinement of a polymer electrolyte membrane fuel cell (PEMFC) system model, including the latest results from other projects. The approach of investigating various design options, heavy-duty vehicles (HDVs) in particular, and using modeling to predict performance and cost makes sense and is helpful.
- ANL uses a variety of modeling techniques to shed light on important aspects of fuel cell design, including platinum-group-metal (PGM)-free catalysts and electrodes, alloy catalyst performance and durability, and design specifications related to heavy-duty applications. This project has excellent continuity through the years and uses robust approaches to modeling and interpreting fuel cell design choices.
- The approach to modeling is sound and has produced good results year after year. These results are important for understanding systems and can have an impact on moving performance forward, but in an indirect way. The attention to different topics is broad and leverages other efforts, although it is not clear how they are delineated or whether they should be independent projects or nested in other activities such as

the Electrocatalysis Consortium (ElectroCat) and Fuel Cell Consortium for Performance and Durability (FC-PAD), where they support other efforts.
- This work is an important part of the Hydrogen and Fuel Cells Program, and it supports many other activities, such as FC-PAD, ElectroCat, and technoeconomic modeling (e.g., Strategic Analysis, Inc.'s [SA's] work). The principal investigator (PI) does a good job at prioritizing areas of focus.
- The work is quite valuable. It hinges on a wide range of collaborations with complementary projects and interactions with numerous partners and organizations. Given the amount of data on cells in transient and dynamic conditions, a complete system dynamic model would be valuable for shedding some light on barriers related to system thermal management and transient operation.
- The approach used in the project is very good. Evaluating the impact of different materials, designs, and some operations is needed to achieve the overall objectives of the project. Now the whole challenge is to prove the validity of the developed tools.
- While not directly working on technologies that could reduce the barriers, the project's modeling and analysis is effectively integrating the efforts and assessing and/or suggesting promising and relevant pathways to pursue.

Question 2: Accomplishments and progress

This project was rated **3.3** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has many investigations and results. The project progresses well toward the set objectives and the diversity of studied aspects. It also progresses well toward the DOE goals.
- ANL has made significant contributions to understanding important aspects of fuel cell design and the effects of these design choices on performance and durability. The work performed in the last year on PGM-free catalysts and electrodes is very important and useful in helping to guide design of these catalysts and electrodes.
    - That said, the reported results are based on only a single catalyst system (the atomically dispersed Fe-N-C catalyst supplied by Los Alamos National Laboratory). Certain results reported, including the extremely high Tafel slope, seem surprising and may not be accurate.
    - Furthermore, it is likely that related catalysts (for instance, the cyanamide- and polyaniline-based catalysts, or catalysts produced by other ElectroCat partners) may have different properties.
    - Therefore, there is some risk that PGM-free catalyst researchers could go down the wrong path in catalyst and electrode design if they base their design choices on the ANL-reported results.
    - This problem is exacerbated by the fact that there is no experimental validation of the predicted performance enhancements shown in slide 13.
    - Therefore, ANL should be encouraged to perform similar analysis on other PGM-free catalysts.
    - ANL should also try to get experimental validation of the predictions (which would need to be done through external collaboration).
    - The modeling work on kinetic and transport parameters using PtCo data supplied by General Motors is helpful in increasing understanding of the loss mechanism for state-of-the-art catalysts.
    - The analysis of operating conditions and durability related to HDV applications is very helpful in guiding researchers in this increasingly important area.
- The addition of HDVs to the analysis framework is clearly interesting and needed. Quantifying the durability tradeoffs of loading and alloyed versus non-alloyed Pt catalysts will be critical in determining what approaches would be best explored experimentally to validate model findings.
- The results involving PGM-free catalysts are not as compelling; the results highlight how far away the technology is from meeting targets and then also show how activity needs to increase by more than an order of magnitude, with higher loadings and increased active-site densities in engineered electrodes, without presenting any rationale for how this would be accomplished or if it would be possible.
- ANL's accomplishments are relevant, and the team provides excellent data, but it will be interesting to see model validation to give further support to this effort. When presenting the performance and cost targets, a sensitivity analysis of the different parameters should be added to evaluate the most sensitive ones. Investigation of a PGM-free catalyst in the study is relevant, but several types should be used, in particular at least one PGM-free and Co-free catalyst. The project's investigation of HDVs this year is interesting,

and the first results are useful. Nevertheless, close collaboration with FC-PAD seems necessary to use relevant accelerated stress test (AST) and load cycles for durability estimations.
- Many results are presented here. However, the PI should also strive to make stronger conclusions. The results are almost always presented as "preliminary results" or "prone to changes" and really impactful conclusions are never made. For example, it is unclear what decay mechanisms FC-PAD should be focused on, or what HDV hybrid configuration should be focused on.
- ANL has demonstrated progress toward DOE goals. For future work, additional details on the methodology for reaching DOE targets in performance and cost would be helpful, along with details on the mechanisms for improving power density.
- The project continues to meet annual project goals.

Question 3: Collaboration and coordination

This project was rated **3.4** for its engagement with and coordination of project partners and interaction with other entities.

- The PI does a great job of engaging with multiple other project teams and a variety of entities. However, a major improvement would be to increase the likelihood of these other projects' success. An example here would be to model range-extender fuel cell electric vehicles (FCEVs) and show how they can have a significant and positive impact on durability and vehicle efficiency. This may encourage the assessment of range-extender light-duty vehicles (LDVs) (as well as others) by SA, which may result in success in showing reduced cost and/or durability.
- The level of collaboration and coordination, with both industrial and academic relationships, appears appropriate for this type of project. Collaboration with FC-PAD and ElectroCat should be further enhanced, in particular to integrate durability data from single cells and stacks as well as performance from new catalysts and HDV applications.
- The collaboration with other consortia, industries, and working groups seems effective. Collaboration with SA seems relevant, and it may be that additional collaboration with end users could be relevant to obtaining real operation data at stack and system levels.
- The project is well integrated with DOE national laboratories, FC-PAD, and the PIs and projects under the DOE and U.S. DRIVE Partnership portfolio.
- The collaborations from this effort are significant and good, but they are leveraged interactions with other projects, and the ability of this project to specifically request experimental findings to validate models is a minor weakness. If this project is just to validate experimental findings, this lowers the project's impact but improves its collaboration.
- There is overall good collaboration. More experimental collaboration on validation of model predictions, especially for the PGM-free work reported this year, would be helpful.
- The project is based on a strong interaction with other institutions and partners.

Question 4: Relevance/potential impact

This project was rated **3.4** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The developed system model is very relevant, as it is generic and takes different scales into account. Therefore, it is of great interest to DOE, as it supports setting technical targets and redirecting component development. The model also allows for assessing performance and degradation toward DOE objectives, and the collaboration with SA allows for proving relevant data for estimating high-volume manufacturing cost.
- While not working directly on technologies that could reduce the barriers, ANL's modeling and analysis effectively integrate the efforts and assess and suggest promising and relevant pathways to pursue. The durability model, if fully realized, would be a significant addition.
- Examining system designs and modeling the effects of design decisions on performance and cost are helpful to building understanding and informing R&D decisions. The new work on heavy-duty applications is timely and relevant.

- The project is relevant in its role of integrating evolutions of component and sub-component performance and durability as system architectures and subsequent system performance and durability. This project has a strong impact in helping SA better estimate the status of cost. However, the impact might be even higher if the model could have been validated at stack and/or system level, not only at single-cell level.
- The project fits very well within the FCTO Multi-Year Research, Development, and Demonstration Plan.
- The project supports efforts to achieve DOE targets through modeling studies and collaboration. The project itself does not meet or attempt to meet DOE targets.
- To have a higher impact, the P.I. will need to make more solid and impactful conclusions.

Question 5: Proposed future work

This project was rated **3.4** for effective and logical planning.

- It is necessary to (1) examine and evaluate the behavior of the different solvents in the gas diffusion layer and catalyst (carbon and ionomer) environment, (2) explore advanced characterizations, and (3) continue the very interesting work on break-in and conditioning. The proposed future work should be separated into two levels: the first one, which is the "immediate future work" deriving from the conclusions of the actual achievements, and the second level, which is the recalibration of the objectives with regard to the new findings, eventually updating the work directions (taking into account the impact of the current achievements and how performance and durability can be improved).
- The proposed future work includes further support of the SA cost modeling, along with modeling of electrode and catalyst effects for PGM-based and PGM-free catalysts, further HDV modeling, and improved integration of durability into the system analysis. All of this work will be helpful to DOE and to fuel cell researchers.
- Further efforts in medium-duty vehicle (MDV) and HDV markets clearly make sense. Trying to incorporate durability considerations in systems analysis is a challenge, but it is also required. The other proposed future work is a continuation of existing projects and has the ability to leverage other funded efforts.
- The proposed future work is pretty much a continuation of the project mandate of updating technology assessment. Durability model progress and validation can be of significant benefit.
- The future work is well planned. A more detailed modeling for fuel cell systems would be valuable as a future contribution. This should include balance-of-plant (BOP) and thermal management dynamics.
- The proposed future work is good. The project team should add range-extender LDVs.
- The proposed future work is in line with the overall targets.

Project strengths:

- The tool is very powerful in the sense that it is versatile and multiscale (i.e., component, stack, system). It is also being updated with new achievements. The project is collaborating with SA to have up-to-date cost estimation with new material and designs. This year, the investigations of operation modes (approaches to limiting degradation during shutdown and start-up from sub-freeze temperatures as well as idle) are very relevant. The same goes for AST and conditioning (even if the operating conditions for conditioning are not clearly specified and validated).
- The ANL team is highly experienced and skilled in performing analyses of PEMFC components and systems, and ANL works in a well-coordinated manner with the SA team.
- The team has excellent continuity, as they have been working in this space and refining their modeling techniques for many years. Every year, the team expands the envelope of design factors analyzed, making the project increasingly useful.
- The project's strengths include the team's analysis expertise, access to world-class diagnostics, and close association with the DOE and U.S. DRIVE fuel cell community.
- The project's strengths include experience and skill in the area and established methods and approaches, as well as connections to the community and different projects.
- The project can count on a wide network of collaborations, strong capabilities in fuel cell testing, and knowledge in degradation mechanisms.

- The project's strengths include picking good things to analyze and its good engagement with a range of collaborators.

Project weaknesses:

- The project has a very interesting approach to the system modeling; however, the choice of investigations could be discussed, such as investigating operation at high pressure while the trend is to simplify the system, therefore operating at lower pressures. Operating at slightly higher temperatures with new membranes could avoid the investigations around water management.
- ANL should include more sensitivity studies to identify what key improvements can enable substantial improvements in performance and durability. The model should be validated at a stack and/or system level. It is difficult to rely on a model based solely on experimental single-cell testing results.
- The project so far seems to be lacking more insight on the system simulation side, which could be easily validated given the insight available to the authors from their experimental work.
- There is no specific funding for experimental measurements or direct connection to experimental validation within this funded effort.
- More experimental validation of model predictions is needed.
- The impact on the FCTO is less than it could potentially be.

Recommendations for additions/deletions to project scope:

- It is recommended that the project team pursue the work on operating modes such as the impact of different shutdown procedures, conditioning, and idle. The mechanisms behind those operation modes should be investigated. In general, the input and specifications should always come from real operation at system level. This allows for more efficiency in materials investigations. It could be of great interest if the consortium releases recommendations for the most relevant and safest protocols. Investigations of system operation with the BOP components are very relevant. However, investigations should be oriented toward the desired simplified model and its impact on the stack components—for instance, operation with lower pressure and higher temperatures (membrane materials). ANL should also assess the impact of characterizations on durability since they are also part of the real operation. The model should be updated to integrate these operation modes.
- The project team should focus on longer-durability applications in which cost or durability is a major tradeoff, as well as crossing the line to include systems analysis for cost considerations. These applications would best be studied by leveraging SA's efforts but using the models developed here to feed into the cost studies, building on the technoeconomic analyses.
- It would be preferable to see focus on the durability model. Both this and the SA projects are now addressing MDV/HDV fuel cell applications. Both teams would benefit from more clearly defined targets for these systems.
- ANL should investigate a PGM-free and Co-free catalyst in the study.
- The presentation slides do not need to be so busy. The project team should pick just a few key conclusions and show key results that support these major conclusions. The rest should be backup material.
- Additional insights could be provided on the feasibility of the improvements suggested for performance and cost targets.

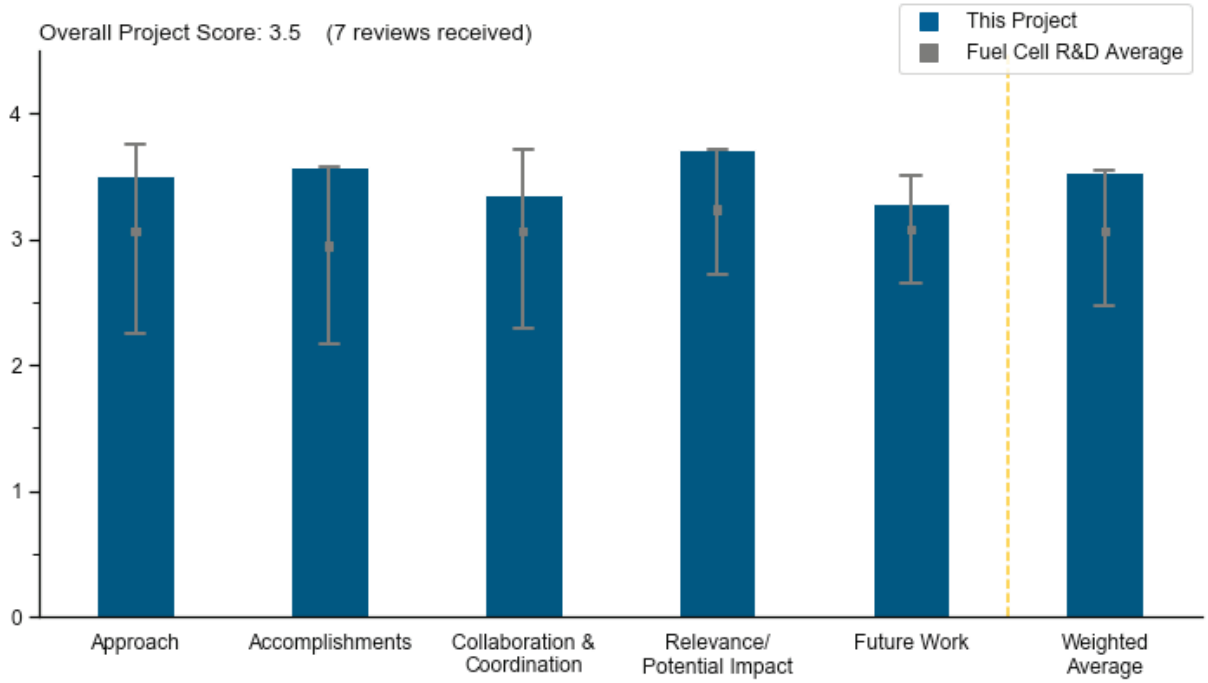
Project #FC-135: FC-PAD: Fuel Cell Consortium for Performance and Durability

Rod Borup, Los Alamos National Laboratory

Brief Summary of Project

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

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **3.5** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- Addressing durability and performance of fuel cells is not a simple task. It needs investigations of different disciplines (thermics, fluidics, material science), with different expertise, at different scales (component, cell, stack, system). It needs also to combine all the generated knowledge in such a way that allows comprehensive interpretations. The approach of gathering the needed capabilities at national laboratories and combining them with different funding opportunity announcements (FOAs) and other collaborations is excellent. The matrix organization between components and cross-cutting thrusts seems very efficient, covering almost all the components of a cell (other stack components need to be addressed as well). The FC-PAD organization succeeded at clearly defining and structuring the sub-objectives.
- The combination of modeling work and expertise, a suite of in situ characterization techniques, and technical expertise has resulted in a more detailed understanding of fuel cell performance and durability. The systematic approach that has been taken, along with the targeted collaborations, has been very successful to date.

- The approach of coordinating the investigation of the performance and durability of fuel cells through a partnership between national laboratories and other organizations in a five-year project is excellent. This project appears to have a very structured approach, combining the strengths and capabilities of the different national laboratories involved.
- The alignment of the national laboratories' efforts in this area is an excellent approach to complement the strengths of each. The result is a generally comprehensive strategy investigating modeling, characterization, cell testing, and durability testing.
- This very strong team is taking a well-crafted approach to improving the science behind component-level integration in PEMFCs. The only area where the work could be clarified from an approach perspective is to be clearer about how samples and information are being efficiently shared among team members.
- The approaches in the different subareas of the membrane electrode assembly (MEA) are appropriate and are addressing the key areas that need to be better understood. Having a coordinated effort among all the parties, in particular the national laboratories, will increase the likelihood of a successful outcome. The way the division of the effort is organized is also excellent. Communication must remain the top priority of such an effort, as it seems to be. The only concern is the groups going off on different, unrelated tangents.
- The approach is sound. Two areas of improvement are recommended for consideration: (1) It should be clearer how the project objectives are set based on input that goes beyond the Fuel Cell Technical Team and encompasses the broader fuel cell international community. For the work to stay relevant, it is paramount that a large number of stakeholders, especially from the industry, can have a say in setting the goals. (2) The project aims at addressing a large number of technical challenges. It seems that this approach may somewhat dilute the resources; further prioritization might be beneficial.

Question 2: Accomplishments and progress

This project was rated **3.6** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The degree of detail and results presented at the review was exceptional. All topic areas showed significant progress in everything from microscopy to electrochemical testing. The modeling should progress nicely as the data become available. The activities are focusing on the right metrics. Regardless of the ultimate outcome, a wealth of knowledge will result that then can (or will) be used to design and engineer the next-generation MEA. The accomplishments are still mostly macroscopic in nature. In time, this should pave the way to more mechanistic, molecular-level investigations.
- To date, progress has been excellent in terms of development of models, detailed characterization of targeted aspects of the MEA, and analysis of the impact of MEA characteristics, including loading, carbon type, ink solvent, etc. The project team has developed useful insight to this point.
- The team is making tangible progress in every area: ionomer and ink processing, catalysts, etc. This is very good work.
- The results presented deliver clear value, especially the work performed to understand the catalyst layer manufacturing properties; this is seen as directly transferable to the industry.
- The objectives of the project are clearly defined and structured, and they are well-aligned with DOE goals. The project is progressing properly toward the realization of these objectives. Certain aspects have been validated, but the accomplishments toward DOE goals cannot be discussed yet. Further validations are still needed.
- The project's accomplishments and progress are effective. The project clearly addresses the DOE targets in terms of performance and durability of MEAs and their components. The impressive results are reflected in 24 publications again this year. However, as there are so many results, they need to be well-structured within a one-hour presentation in order not to be overwhelming. To facilitate the reviewers' work, either fewer acronyms should be used, or a dedicated glossary slide should be integrated.
    - To capitalize on all this knowledge, there should be a dedicated action (on the website) in the consortium to gather all the data resulting from FC-PAD.
    - The platinum-group-metal (PGM)-free catalyst investigated contains cobalt. From a critical raw materials perspective, this means that this issue is not solved. Therefore, FC-PAD should also investigate PGM- and Co-free catalysts.

  - Accelerated stress test (AST) protocols should be developed to investigate component degradation mechanisms. FC-PAD should also investigate degradation mechanisms occurring in stack operation.
  - The ink study has shown interesting results. However, all the experiments referred to electrodes produced by spraying technique. As this technique is not the most-used industrial technique of electrode coating, complementary techniques (slot-die casting, microgravure, screen printing) should also be investigated to quantify the materials and process impacts on durability.
  - FC-PAD investigation and knowledge may also apply to applications other than PEMFCs (e.g., anion exchange membranes [AEMs] or electrolysis).
- The break-in procedure is clearly ineffective to have had such poor performance, but it is difficult to judge what is going on without knowing what the procedure was. The work on conditioning effects is nevertheless useful and important to guide both the ability to get maximum performance out of a given design and to guide factory acceptance testing. It is also important to characterize and understand new material sets as they are introduced. The ink solvent effects are important, and the work is providing useful and interesting results with linkages to expected ionomer behavior. One caution is that ionomer-to-carbon/solvent systems are difficult to generalize, and conclusions should not be applied too broadly. The work is valuable to guide the types of investigations that can be applied to different systems. The characterization of aggregates and agglomerates is very interesting and useful. The study on interactions between carbon pores, water, and Pt locations provides useful work and linkages to the model. For the catalyst AST, which was examined for the loading study, it would be useful to plot the change in the catalyst's specific activity to separate out the change in surface-area kinetic effect from the change in Co-associated kinetics. It is useful to pursue membrane degradation modeling. In reference to Ce migration and its effect on membrane properties and thin films, the results are interesting and deserve further investigation.

Question 3: Collaboration and coordination

This project was rated **3.4** for its engagement with and coordination of project partners and interaction with other entities.

- There is excellent collaboration with other national laboratories. The amount of collaboration with each of the FOA-1412 projects is likely more properly assessed in their presentations.
- The structure of FC-PAD is based on collaborations. This aspect seems to be very well-thought-out and done either in the frame of the FOA (interactions with the DOE-awarded FC-PAD projects led by 3M, General Motors, United Technologies Research Center, and Vanderbilt University), in which regular follow-up conference calls and in-person meetings are set, or out of the FOA frame, including international collaborations. (It is not clear though, how the intellectual property management and information/data exchange are managed in the latter cases.)
- The collaboration between the project's partners is intense and emerges clearly from the presentation. The project team is encouraged to seek more input (or make the input more visible) from the larger community in the prioritization of goals. This work is valuable and must remain relevant to the whole industry.
- The collaboration between the different partners is well structured and well managed and appears very efficient. Enhanced collaboration with the National Renewable Energy Laboratory (NREL) manufacturing team will enable greater understanding of degradation mechanisms, including process impacts on these mechanisms. Collaboration with Argonne National Laboratory and Strategic Analysis, Inc., will be needed to investigate medium-duty (MD) and heavy-duty (HD) vehicle applications.
- The collaboration is excellent based on the coordinated results reported at the review. The project team must ensure that communication among all parties remains the top priority. An area of concern related to collaboration is the dependence on certain suppliers and the subsequent materials selection. Comprehensive studies are being carried out on just a few suppliers' products (gas diffusion layer [GDL], catalyst, carbon source [e.g., Vulcan®], ionomer, etc.). As related to collaboration, the supplier collaborative effort should be expanded. Not all Pt/C catalysts are the same. It is unclear how to improve this activity, especially if the material is proprietary.
- The team has shown good collaboration with partners, both funded through the FOA and unfunded. One of the consortium's goals was to highlight the utility of the national laboratory teams in addressing research

issues and challenges that may be facing industrial and academic researchers. This may be more of a comment for DOE rather than the team associated with this project, but it would be helpful if there were a detailed plan to make these resources available to those outside the national laboratories and the FOA associated with this consortium.
- The rating here might not be fair, but the team did not do a particularly great job in the presentation of describing how the team is integrated. For such a large project, it is easy for all of the members on the team to have their own pet projects, with limited coordination between the individual members. Next time, it would be very helpful for there to be a slide dedicated to this beyond just an organization chart.

Question 4: Relevance/potential impact

This project was rated **3.7** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- Durability and cost are the two main aspects that have been identified by the Fuel Cell Technologies Office (FCTO) Multi-Year Research, Development, and Demonstration Plan (MYRDDP) as key still-missing points for fuel cell commercialization and deployment. In that sense, the work done by FC-PAD is absolutely necessary, not only to assess the advancement toward those two objectives but also to generate innovative breakthrough technologies and materials that are needed to meet MYRDDP objectives. FC-PAD's mission is to generate the necessary knowledge to understand (operation and behavior, components, faults, degradation, interactions, and impacts of operating conditions) and, based on this knowledge, direct the research toward innovative components and elaborate and test them in real operating environments. With that, the mission includes being able to provide clear directions to durability improvement (with cost reduction) to at least reach the MYRDDP objectives.
- The project is totally relevant. The MEA's limitations, performance, and cost factors are clearly impeding, or will impede, the acceptance of the technology by both the producers of the components and fuel cells and the customers. Removing the cost/durability issue will be the primary "win" if the technology can be demonstrated to achieve lifetime, cost, and durability targets.
- Durability, particularly of MEA components, is of critical importance for PEMFC commercial viability. This project details and studies many of the aspects of MEA design that can limit durability. The work completed to date has provided useful insight into many of the mechanisms of degradation and will likely have an impact on future efforts to improve durability.
- The project looked at components under several lenses. It will be important to dedicate efforts to integrate the information and interpret the data at system level. In fact, as stated by the presenter, the belief is that the interfaces between components are the key and must be looked at with a holistic and systematic approach.
- FC-PAD is of very high relevance for the FCTO in achieving MYRDDP targets. Concurrently achieving cost and durability targets is the real key driver.
- The project is providing key insight into all the highest-priority performance and durability mechanisms.
- It would be nearly impossible to find another project more aligned with FCTO targets.

Question 5: Proposed future work

This project was rated **3.3** for effective and logical planning.

- The proposed future work is in coherence with the addressed conclusions. It is in fact necessary to examine and evaluate the behavior of the different solvents in the GDL or catalyst (carbon and ionomer) environment, to explore advanced characterizations, and to continue the very interesting work on break-in procedure and conditioning. The proposed future work should be separated into two levels: the first level, which is the "immediate future work" deriving from the conclusions of the actual achievements, and the second level, which is recalibrating the objectives with regard to the new findings and eventually updating the work directions (taking into account the impact of the current achievements and how to improve performance and durability).
- The proposed work in the near term is appropriate, as the activities reported are not complete. These activities must continue in all areas. From such results, the modeling should evolve into a tool that may dictate changes in the MEA. This is the most valuable outcome. Materials, architecture, conditioning,

particle sizes and distributions, loadings, etc. should all be scrutinized in the results of the current work in order to derive the model. The proposed work supports this. After the current activities have been carried out, the electrode effort should review the hydrophobic/hydrophylic character in the membrane and electrode interface, as well as in the body of the electrode. The microlayer should also be re-evaluated. The following topics should be incorporated in the test plan:
    - Interface chemistry and architecture, especially of the cathode
    - Impact of the fluoride ion released in the first 100 hours and its impact on the electrode properties, in particular the wetting character
    - Impact of various pre-treatments on the carbon used for the catalyst support, GDL, and microlayer
    - Use of carbon other than Vulcan
    - Electrode structural changes and their impact on performance using 3D printing vs. spray vs. doctor blade-like application methods
    - Hydraulic properties of the electrode, microlayer and GDL (possibly using the National Institute of Standards and Technology neutron facility)
    - Energetics of the proton-ionomer dynamics
    - A revisit of the older proton mobility question of McBreen, performed in the 1970s, especially on the cathode
- The project's future work is detailed and well-structured.
- The proposed future work seems to be aligned with industry needs. It is recommended that the project focus the scope even further and consider the interfaces between components as part of the work.
- The proposed future work is mostly straightforward; it would be very helpful if there were more focus on predictive rather than descriptive approaches.
- Regarding requirements for HD applications and research directions, there are a significant range of HD requirements, and not all can tolerate lower power density. The space (or size) of the system can be very important. Cost will also continue to be critical, thereby driving a need for high area utilization of the fuel cell and high current density. Since the durability is critical, the oxygen transport limitations at high current density also become very important later in life as the Pt surface area is reduced. Membrane durability modeling, ionomer film aging, and Ce effects are all areas that should be continued but are not explicitly mentioned in the path forward.
- The move to MD/HD vehicle application is relevant but will induce an adaptation of the ongoing studies, materials, and operating conditions used for the testing. The move has to be well-prepared. Specific AST protocols will have to be defined. Collaboration with the NREL manufacturing team should be foreseen in order to include some manufacturing process aspects (e.g., ink viscosity and coating technique) in the comprehension of degradation mechanisms.

Project strengths:

- The project's strengths include clear objectives, excellent organization for efficient multi-laboratory coordination, different competencies (elaboration, testing and characterization, modeling), means, and different collaborations (e.g., laboratories, universities, and industries). All these points make the project solid and efficient.
- The project's strengths include its highlighting of the critical aspects of MEAs that have directly impact on durability, its detailed experimental and computational analysis of relevant MEA operational processes at a range of length scales, its efficient use of DOE resources, and its adequate integration of research partners.
- The composition of the consortium, the coordination of the project, and the well-defined role of the different partners involved are real strengths. There is a strong focus on publication of the results in a large number of publications to share as much as possible with the international community.
- The multi-laboratory and multi-level approach to studying a broad range of higher-priority degradation and performance mechanisms is a key strength. The outstanding characterization capabilities really drive the in-depth understanding.
- This project has a very strong team and a very clear pathway for development toward FCTO goals.
- This is a well-articulated project with excellent connections between the national laboratories.
- This project is an outstanding coordinated effort by some of the best folks in the country.
- Experimental resources at the laboratories and knowledge of materials and electrochemistry all converged and were coordinated.

Project weaknesses:

- There are no project weaknesses, per se; however, the coordinated effort needs more than one person to manage such a large and important project. The project team needs someone to assist in "seeing the big picture." So much data is being generated that it is necessary to have not just a coordinator of the efforts but multiple people with a higher-level, broader perspective overseeing the technical results. This might already exist.
- It is difficult to find any weaknesses.
- It would be helpful if, in the presentation, the team would dig a little deeper regarding how information moves between partners to realize the strengths of all of the partners. It is clear that good work is being done, but the presentation did not make it clear whether we are seeing the result of several excellent pet projects, or a large volume of information (and possibly materials) is changing hands between the partners. The team is also focusing on a very narrow set of materials, which (although it can provide fundamental insight) can stifle innovation. The modeling work, or at least how it is presented, appears to be very descriptive, and it was not clear what has been done to enable the team to be structurally predictive in the future. The team is not clear about how the information is being used for the predictive synthesis of materials and/or electrode structures.
- FC-PAD is doing tremendous work investigating different components with test/characterization and modeling; this is necessary and should be pursued. However, the real operation vision of the work is missing; in the end, what is needed is an operating system with optimized durability and cost. Now optimizing the components needs to be done in their real operation environment. A global optimization is not the sum of single-component optimizations, especially in a complex, multiphysics and multiscale system like a fuel cell system in which a large number of interactions exist. Optimizing components should be done—but with a stack- and system-level input. The validations should also be done at stack and system level. Even if investigations are done at component level, the problem must be set at stack and system level (including hybridization architectures). For instance, using AST for a component such as a catalyst cannot validate degradation under real operation of the cell (in a stack inside a system) because the degradation mechanisms of each component are not fully independent. There are interactions, and the degradation might be completely different under real operation.
- The presentation's slides lack clarity for some protocols and conditions, such as the break-in procedure. This is a clearly ineffective break-in procedure to have such poor performance, but it is difficult to judge without knowing what the procedure was. In general, although there is a lot of good data, there are a number of places where plots are not sufficiently labeled, conditions are not always given, there are not enough legends, and fonts can be too small to read. The value of the ordered array electrodes is not clear. Modeling to support this work is recommended. The performance with fill carbon is improved but still not good. The value of this work has not been established.
- A weakness of the project is its very large scope, which may lead to some loss of focus in the different activities. Using only sprayed electrodes appears also to be a weakness, as it is not the most representative coating technique for electrodes and leads to the absence of investigation of the manufacturing parameters' impact toward degradation.
- The project needs more industry input, deeper focus on fewer tasks, and a more holistic approach to study interfaces.

Recommendations for additions/deletions to project scope:

- The difficulty of the task is that investigations must be done at different scales, and degradation metrics must be defined at all steps. The researchers should, however, keep in mind that the final durability objective is located at the final object level (stack/system). Connection between durability at material level and stack/system level should always be clearly defined, because the final aim is stack and system durability. The work is great and should be pursued; the problem should be set slightly differently. The specifications should also come from system and stack, and for real operating conditions. Collaboration with higher technology readiness level (TRL) consortia (stack- and system-level partners) is recommended in order to have input for characterization, testing, and real operating conditions. It is the same for validations, which should be done at system level. The model should also be multiscale going from component to system, and fault and degradation propagation between components and scales should be

integrated into the models. The metrics to assess how well the ASTs perform should be clearly defined and located at stack/system level. ASTs should be updated to mimic degradation such as it results from real operation. One way to improve durability is to take advantage of the recovery phenomena (the real operation will certainly include some idle-modes recovery). Therefore, mechanisms behind voltage recovery should be investigated; additionally, how those mechanisms are correlated to aging and operating parameters should be investigated. The influence of different shutting-down protocols (with or without nitrogen) on these performance recovery phenomena is unclear. The parameters defining voltage recovery intensity (soak time, temperature, shutdown procedure) are also unclear. All these aspects are of great interest to improving durability in real operation. Work can be done at low TRL, but the requirement should come from system and stack to be more realistic.

  - Globally, and as a summary, durability in a fuel cell system could be linked to the following: choice of materials (investigated in FCPAD) and architectures; stack assembly, terminal plates (durability, cost, performance), and gaskets (durability under operating condition, leaching pollutants and their effects), as well as stack assembly techniques (pressures); and effect of system operation with real operating cycles, different hybridization schemes). These aspects should be investigated concomitantly; optimizing one single aspect independently of the others is hard to validate in a complex system with a lot of interactions, such as in fuel cell systems. It is recommended that the project team open the consortium to other partners to take into account system consideration, and slightly update the organization of FC-PAD to adjust the problem inputs and validations.

- The next level of research should go into a deeper dive regarding the electrode dynamics. For example, now that there are "visuals" such as the ionomer in the electrode(s), the actual function of that ionomer film or particle, whatever it is, is still a question. This needs to be done on many different levels. For example, it is unclear what the impacts are of voltage, current, or hydraulic effects on the electrode as a function of the distance from the membrane. It is also unclear whether there should be a hybrid approach to an ionomer/ polytetrafluoroethylene (PTFE) electrode based on the results. Answers to these questions might possibly provide additional knowledge for developing a better electrode structure. In addition, with the current resources, it is unclear whether it is of value to understand the energetics of the ionomer–proton transport concept within the membrane (but away from the surface of the membrane). One could ask what the driving force is of an ionomer particle that may or may not be in physical contact with other ionomer particles or films in the electrode—a particle that is microns away from the membrane—to transport a proton. Even if a proton could make it to the far reaches of the electrode, it is unknown how much energy it takes to jump from an ionic-like solution in the pores of the electrode to sulfonic acid sites of the ionomer. It is also unknown what the delta H of solvation is, as well as the transition from a liquid phase to the bound ionic phase. It is unclear whether this impacts performance or whether the ionomer positioned away from the membrane acts only as a wetting site. Lastly, and related to the above, there is a question of how electrode architecture differences affect the key metrics. Perhaps there should be exploratory electrode structure activities.
- The project team should evaluate the effectiveness of AST protocols in predicting component and cell lifetimes in real devices. The team should also use its tools and knowledge to better direct where the technology is going. Of the materials and approaches the team has tested, it is unclear which are very unlikely to meet targets. The team should also develop a robust synthesis and structure a database of material sets. It is unclear whether the project team can leverage what FC-PAD has done or is doing to facilitate more rapid growth in other areas, e.g., AEM fuel cells and electrolysis. Specific to AEM fuel cells, this seems to directly fit into the scope. Granted, three years ago, no one would have seen that AEM fuel cells would get to >3W/cm^2 on H_2/O_2 and 1000+h durability. However, now they have, and there are at least as many fundamental questions as AEM fuel cells. Some of these questions are even more troublesome, with no clear existing pathway for electrode design approaches, ionomer–catalyst interactions, water management, CO_2, etc.
- The project team should initiate close collaboration with the NREL team working on electrode manufacturing in order to include these aspects in the comprehension of induced durability impact. The project should also investigate a PGM- and cobalt-free catalyst.

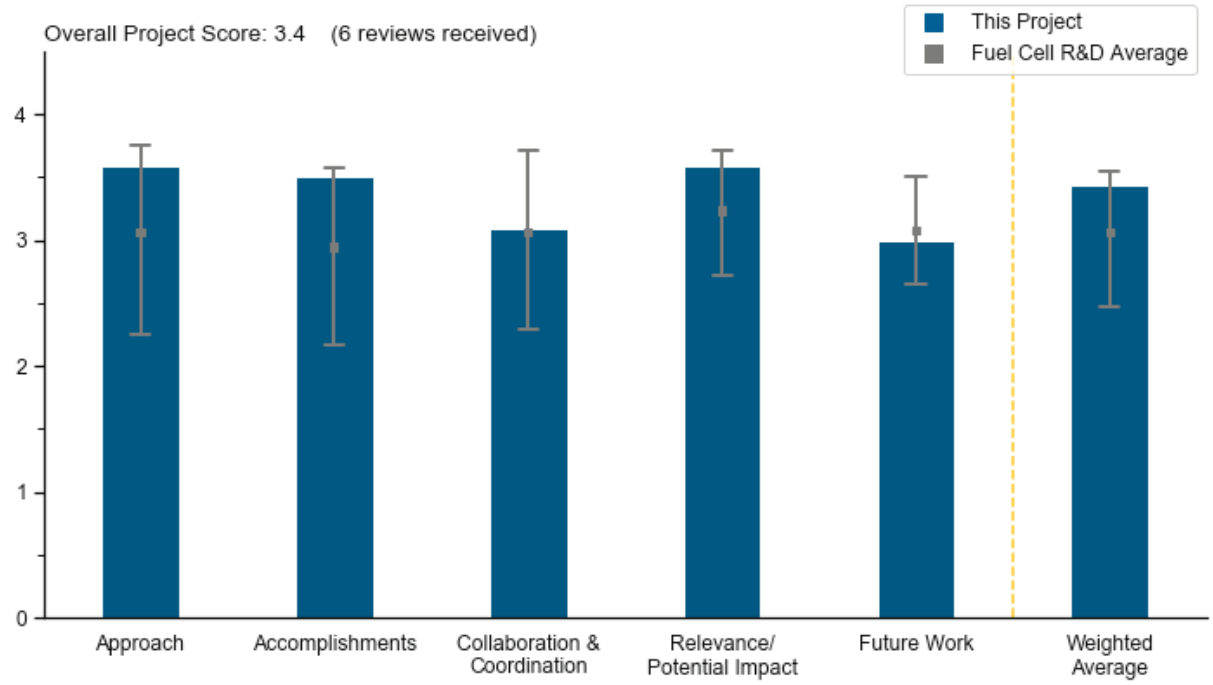
Project #FC-140: Tailored High-Performance Low-Platinum-Group-Metal Alloy Cathode Catalysts

Vojislav Stamenkovic, Argonne National Laboratory

Brief Summary of Project

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

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **3.6** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The catalyst development approach at Argonne National Laboratory (ANL) is unparalleled in terms of the team's abilities to develop and characterize catalysts across the range from fundamental studies of well-defined single crystals to practical scalable thin films and nanoparticles.
- The materials-by-design approach is excellent for improving catalyst performance and durability. Many high-activity Pt alloy catalysts are synthesized in this project, while much effort was focused on understanding Pt or alloy metal dissolution rate.
- The technology based on a materials-by-design approach for the synthesis of low-Pt-alloy nanoscale catalysts is specifically focused on overcoming critical barriers. The approach is feasible and well integrated with other related efforts in the field of designing durable low-Pt architectures.

- The materials-by-design approach taken to designing and characterizing low-platinum-group-metal (low-PGM)-alloy catalysts is comprehensive, including design, synthesis, characterization, scale-up, and fuel cell optimization.
- ANL's approach is novel and has a good chance of "engineering" a catalyst material with better Pt utilization. The approach could be improved if there were more focus on the processes that can make the 5–10 g scale of material, rather than sub-gram-scale batches. The challenge of sub-gram-scale batches is that scale-up is almost always required to make a membrane electrode assembly (MEA) with controllable properties such as loading. The one sample that was made at a 5 g size (a sample of PtNi) showed the best fuel cell performance.
- The approach to project targets is effective, and DOE targets could be met.

Question 2: Accomplishments and progress

This project was rated **3.5** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- It is encouraging to see the scale-up of most of the catalyst in gram levels and very good performance in 50 cm^2 MEAs from these scaled batches. The dissolution dependency study is also very helpful to understand and improve the durability further. ANL exceeded DOE 2020 technical targets for 50 cm^2 MEAs for PtAu, PtNi 5 nm, PtCo 2 nm, PtNi 5 nm, and PtNi/PtCo intermetallic. So far, seven patents have been awarded and six have been filed, which is one of the best outcomes of this project.
- Excellent progress has been made toward stabilization of Pt from dissolution through integration of Au, first at the thin-film rotating disk electrode (RDE) level, and then extended to nanoparticles in MEAs. Additional work could be considered to develop the fundamental understanding of the impact of Au, which may allow integration of other non-PGM materials. Progress toward development of highly active nanostructured PtNi has continued, with very high activities obtained in RDE. However, activity of the PtNi nanoframes in MEAs remains much lower than RDE assessments, and the large decrease in mass activity during conditioning (as seen on slide 18) suggests a very serious durability issue that needs to be resolved. The hydrogen and air performance was quite good for the low-Pt loading used. Another area of note was intermetallic development. Fundamental thin-film RDE studies showed the enhanced stability and activity expected. The transition to nanoparticles yielded good mass activities, but durability (the key expected benefit) was not reported.
- This project has yielded significant insights into structure–function relationships and has yielded several promising classes of catalyst.
- The principal investigator (PI) clearly demonstrated successful progress toward project and DOE goals through the accomplishment of a majority of milestones.
- More fuel cell results are shown this year as compared to last year. Some of the approaches do not appear to pan out in fuel cell performance gains; for example, the higher-mass-activity catalysts do not demonstrate superior performance in fuel cells. It is sometimes difficult to make comparisons, even within the project team's presentation, since at times the researchers run experiments at 250 kPa of air and sometimes at 150 kPa of air.
- Many catalyst systems of PtNi and PtCo have been studied on the stack level for this project. ANL met DOE goals for mass activity and loading for PtNi/high-surface-area-carbon (HSAC) and 2 nm Pt_3Co/C, but did not address the durability goal on the stack level.

Question 3: Collaboration and coordination

This project was rated **3.1** for its engagement with and coordination of project partners and interaction with other entities.

- The project lead collaborated on different technological thrusts with several research groups from national laboratories, academic institutions, and OEMs.
- There is very good collaboration among the team members, which is evident from the results. Now the team is making high-performing catalysts in gram batches, so they are encouraged to share with interested parties, especially with original equipment manufacturers (OEMs) for evaluation. The National Renewable

Energy Laboratory's (NREL's) MEA testing capabilities are well known, but evaluation from outside this team will provide more interest in the industry, considering future technology-to-market (T2M).
- The collaboration on fuel cell performance is limited to one partner, NREL, for fuel cell testing. Again, small batch sizes of catalysts limit the amount of collaboration that can be done since sample size is too small for controllable, repeatable fuel cell testing.
- The laboratory has already developed some high-performance catalysts. Collaboration with the industry sector would be more efficient to transfer technology.
- There is broad collaboration among national laboratories. There could be more indication of interactions with outside researchers and OEMs.
- The project's collaborators consist primarily of other national laboratories. The project would be strengthened by additional collaborations with universities and/or industry.

Question 4: Relevance/potential impact

This project was rated **3.6** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- Though fuel cell electric vehicles are in the commercialization phase, cost still remains the biggest challenge. High-activity, high-durability catalysts are a must for reducing the cost further. This team has developed many high-performing catalysts while also providing insight about durability.
- The project does have the potential to optimize Pt utilization since it is a true engineering approach to catalyst design.
- ANL's achievement is relevant to DOE goals. Research and development of ORR catalysts in this laboratory may have potential impact on high MEA performance for the fuel cell stack.
- The project is directly addressing one of the top key commercialization barriers, the activity and durability of the ORR electrocatalysts.
- There is high potential for impact in terms of new catalysts with high durability and low cost.
- The project is critical to the Program and for the fuel cell community in general.

Question 5: Proposed future work

This project was rated **3.0** for effective and logical planning.

- Taking into account that the project is close to the end, the proposed future work is clear and logically focused on the achievement of final project goals.
- The project has strong future plans in terms of characterization, optimization, and scale-up. Additional effort on theory and structure–function relationships is desirable.
- The proposed future work seems reasonable. It is recommended that the project team add best-candidate-catalyst MEA testing with an institution outside of the project team.
- The proposed future work is appropriate, as it directly aligns with addressing key barriers.
- There are too many catalyst systems that have been investigated so far in the laboratory. The proposed future plan would be more feasible if only the best catalyst and the best scale-up method were investigated. Secondly, RDE-inductively coupled plasma (RDE-ICP) seems to have no clear relevance to the MEA performance.
- The proposed future work sounds like simply more of the same. It lacks the focus to translate to improvements in fuel cell performance. Mass activity is reaching targets, but power density is still far off. More focus should be put on addressing this limitation.

Project strengths:

- PIs at ANL are world leaders in the Pt and Pt-alloy catalyst field. Other team members are supporting ANL's scaling-up efforts, analysis, and MEA testing, which almost completes the circle. The capabilities of each team institute are tremendous for accomplishing the technical challenges.

- The project's strengths include the following aspects: 1) the method for the scale-up is critical for the manufacturer; 2) the use of HSAC for the support benefits of mass transfers at high current density; and 3) the demonstration of the catalysts' performance on 50 cm^2 MEAs.
- This project brings deep fundamental understanding and unique, highly practical analyses to bear against the key barriers of cost and durability.
- The team is very creative in terms of catalyst design, synthesis, characterization, and scale-up.
- The project's strengths include a good engineering approach with a high degree of fundamental studies of the catalyst materials.
- The project is based on a solid scientific hypothesis and solid approaches and is led by a PI with substantial expertise in this field.

Project weaknesses:

- The project's weaknesses include the possibility that (1) the effort to study so many catalyst systems may lead to few gains from each one, (2) there are few durability tests on MEAs with Pt_3Ni/HSAC and 2 nm Pt_3Co/HSAC, and (3) the performance gap between RDE and MEA is still unknown.
- A third-party (outside-project-team) MEA evaluation is missing. This might not be needed from the point of view of the statement of project objectives but is surely important from the T2M perspective.
- The team seems to lack the ability to translate promising catalyst materials to true fuel cell performance potential. A different approach is required to tackle this limitation.
- The project would be strengthened by increased funding to enable increased development scope and rate.
- The project's connections to OEMs for validation are lacking. The theory-backed design could be better demonstrated.

Recommendations for additions/deletions to project scope:

- The NREL team has demonstrated that current commercial catalysts can show mass activities between 0.7 and 1.1 A/mg Pt after the conditioning protocol. Though those high activities of commercial catalyst do not translate to performance, ANL needs to think about how to surpass the MEA performance of the commercial catalysts.
- The PI should consider putting less of an emphasis on increasing the activity of porous PtNi nanoparticle structures and emphasizing stabilization. The PI should consider conducting analyses to understand the impact of catalyst-specific material factors (if any) on the large discrepancy between RDE activity and the MEA.
- Changes to the project scope should be (1) that the project should focus only on one or two of the best catalysts for MEA testing and (2) that RDE-ICP should do more to monitor the loss of Ni or Co.
- An increased emphasis on downstream customers and upstream theory-backed design is recommended.
- There should be more emphasis on scale-up and collaboration with partners with MEA manufacturing experience.

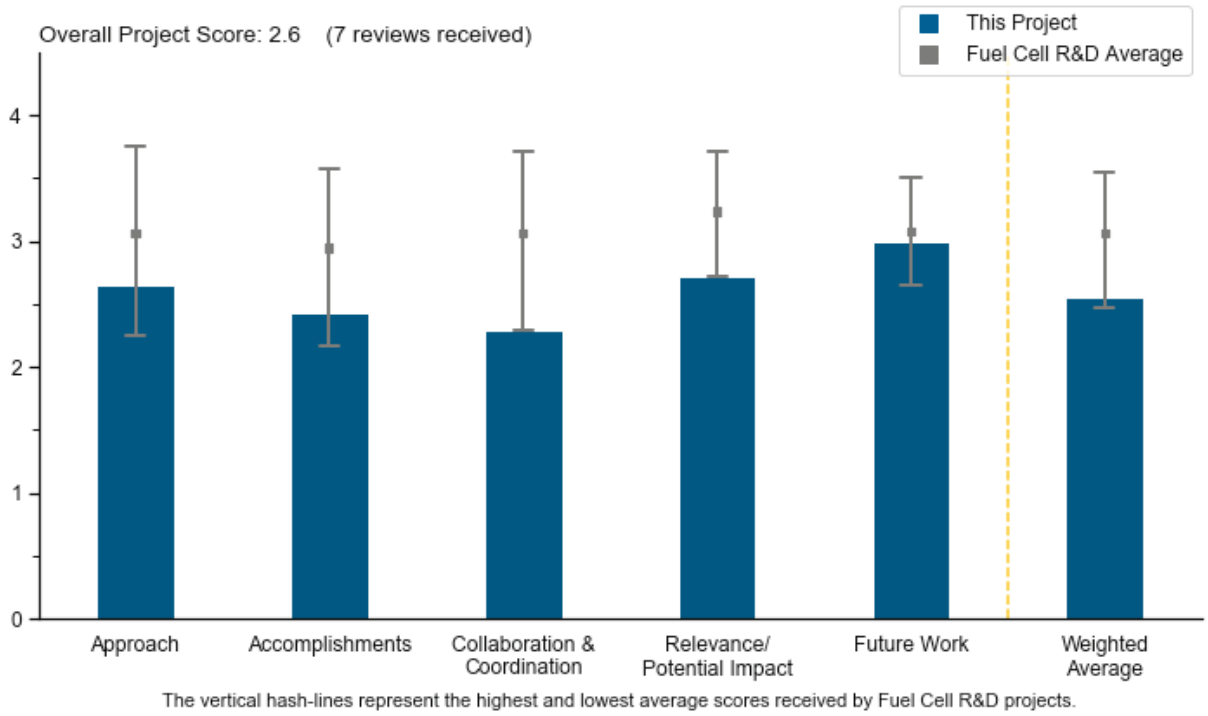
Project #FC-141: Platinum Monolayer Electrocatalysts

Jia Wang, Brookhaven National Laboratory

Brief Summary of Project

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

Project Scoring



Question 1: Approach to performing the work

This project was rated **2.6** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This project seeks to develop Pt-monolayer catalysts that can meet DOE's technical targets for durability, activity, and cost simultaneously. The iterative approach of synthesis, rotating disk electrode (RDE) screening, and characterization (scanning transmission electron microscopy [STEM], x-ray diffraction [XRD], etc.) has led to two catalyst types: $Pt_{skin}(PtNi)_4N)_{core}$ and Pt-NbO_xC. As part of the approach, down-selected catalysts are to be submitted to partners for membrane electrode assembly (MEA) testing, an important activity that was absent this year.
- The approach of this project toward DOE goals is generally effective. The core components for the monolayer Pt atom on nanoparticles are essential, and the modification of support indirectly affects the stability of nanoparticles.
- The project has continued to address the development of novel and promising catalyst concepts (PtNiN and Pt-Nb_xC), the key advancement being the formation of Pt thin skins over Ni-enriched nitride cores, where the Pt skin is stable.
- With respect to material sets being examined and final targets, the approach was presented and appears to align with reasonable goals. However, no outline of the tasks and milestones was presented. It is not clear if

the plan is feasible if this information was not presented. One concern is that the project seems to be focusing on RDE results for optimization. There needs to be MEA testing in parallel to avoid optimizing the catalysts for RDE performance and ending up with a material that does not work well in an MEA environment.

- The proposed project focuses on the barriers: improving the activity, especially the durability, of Pt-based catalysts in polymer electrolyte membrane fuel cells. The two chosen approaches are not fundamentally infeasible; however, one of them ($PtNbO_x$) was given up, and the lack of the MEA data on the other one (PtNiN) makes a fair judgment impossible.
- The project focuses on catalyst development and characterization, with promising RDE results, but with limited effort on MEA translation and demonstration. It would be good to have a Ni dissolution study.
- Brookhaven National Laboratory (BNL) did not show any MEA results from the last year. This is a major weakness in the approach. There is also little to justify the selection of chosen catalytic systems.

Question 2: Accomplishments and progress

This project was rated **2.4** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project focus was on characterizing and optimizing the high-mass-activity PtNiN catalyst while also determining the cause for low MEA activity of the NbO_x-based catalyst. To this end, XRD and analytical STEM were heavily employed. Two key accomplishments involved developing a new intermetallic catalyst and demonstrating that the high activity and electrochemical surface area (ECSA) of the PtNiN catalysts could be retained on a Vulcan support following the catalyst accelerated stress test (AST) in RDE. The latter approach shows great promise for meeting support AST targets. The lack of progress or attention to MEA performance issues was a major omission from this year's progress and accomplishments.
- The PtNiN on Vulcan shows good ECSA and activity stability in RDE catalyst AST. The Ketjen support was explored to control particle size and promote internal Pt for higher beginning-of-life (BOL) mass activity; the activity stability is lessened relative to Vulcan but still meets the project targets after the AST. Unfortunately, this year has not seen an update in terms of MEA-level performance.
- Screening of catalysts was done, and an improvement in catalyst activity was based on RDE performance, which is on track for DOE targets.
- Identification of the no-go on $PtNbO_x$ is still meaningful. The RDE measurements and characterizations on the PtNiN are comprehensive and complete. The RDE performance of PtNiN is good and informative. However, so far there are no MEA data.
- The project has good characterization of an interesting set of materials and presents exceptional RDE mass activity. There is no need to further optimize activity of these materials in RDE. The durability in RDE is not a good measurement of true durability because it is at a lower temperature than an MEA and metal leaching from the catalyst would not have as much impact. Unfortunately, the MEA results clearly trail those of peers in this area for reasons that have not been determined. This should be the focus of all future testing.
- The lack of MEA testing is severely limiting the usefulness of this project. The well-established lack of correlation between RDE and MEA results means that there is no way to know whether any of the catalysts under development hold any promise for fuel cells. The only MEA results mentioned, which are on slide 5, appear to be from the previous year. The use of lattice contraction as measured by XRD to assert that Pt skin formation occurs on PtNiN is questionable. Data from synchrotron studies (extended x-ray absorption fine structure [EXAFS]) or energy dispersive x-ray spectroscopy (EDX) are needed. However, the EDX data on slide 9 do not show any evidence of Pt separation. Therefore, the putative nanoparticle structure does not seem accurate.
- Most DOE targets have been met only on RDE tests. MEA performance should be the final goal.

Question 3: Collaboration and coordination

This project was rated **2.3** for its engagement with and coordination of project partners and interaction with other entities.

- The team is strong and covers necessary aspects of the proposed work. In particular, this proposal involves international collaborations. Although no MEA data have been provided yet, General Motors (GM) is the ideal choice and will deliver reliable data. The presentation did not make the roles of the Korean and Japanese groups clear.
- Since last year, the list of partners and collaborators has dropped significantly, and engagement with existing industry partners appeared minimal. A key deliverable of this project is to provide catalysts showing high activity and durability in RDE to collaborators for verification in MEAs. Without that coordination, this project provides little value toward meeting DOE technical targets.
- Most or all of the results seem to be from the BNL group. Support for MEA testing is desperately needed, but either GM did not provide MEA testing or the results of the testing were not shown.
- Some MEA results from GM were presented, but more MEA iterations are needed to improve MEA performance. It was not clear if any other institution collaborated significantly on this project.
- This year has not seen a major update in MEA performance, which is a role of the project partners. It is unclear what the partners (e.g., GM) have contributed.
- MEA testing could have more collaboration with GM and Toyota.
- The project's collaborations are not strong, and collaboration contributions to the project are not clear at all.

Question 4: Relevance/potential impact

This project was rated **2.7** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The potential impact of this project is high. The high BOL mass activity with the Ketjen support and the highly stable mass activity of the PtNiN catalyst may be a promising pathway to meeting DOE platinum-group-metal (PGM) targets for performance and durability.
- The project's goals for developing stable core–shell catalysts that can meet DOE activity targets are relevant to the DOE Hydrogen and Fuel Cells Program (the Program) objectives.
- The project's accomplishments are relevant to DOE goals. The future work could have the potential to increase the performance of catalyst durability.
- The development of highly active and durable low-PGM catalysts is highly relevant to DOE's mission, especially with the shifting focus to heavy-duty applications. However, to be relevant, the high RDE performance must be transferred into MEA, which has yet to be achieved over the life of this project.
- Based on the excellent RDE activity results, this project shows potential for making progress toward DOE goals for low-PGM-loading catalysts. The likelihood of achieving those goals is low if the project does not focus more on MEA testing.
- During its long history, this project has consistently failed to show good performance in MEAs. Analysis in recent years has demonstrated that RDE testing is barely relevant to fuel cells. Without good MEA results, this project is not very relevant.
- The Program's goals and objectives are based on MEA performance, which is missing from the presentation.

Question 5: Proposed future work

This project was rated **3.0** for effective and logical planning.

- At a high level, the proposed future work appears to be logical. More details on the MEA optimization and who will be performing the work need to be presented to increase confidence that the MEA optimization will be successful. The Nb-nitride materials are being pursued for risk mitigation, but there should be an early decision point for whether to continue exploring Nb-based materials, because so far the results are not promising.

- There may not be much future work since the project is ending. If further work continues, the proposed MEA testing will be the most important part. The proposed future work on corrosion-resistant supports does not seem like a good idea for now since testing the catalyst performance and durability is much more important.
- The primary focus should be on MEA optimization; additional focus on catalyst optimization by RDE or corrosion-resistant supports should be shelved until the factors limiting MEA performance are identified.
- The proposed future work is mostly important here since it proposes to focus on the MEA testing, which is eventually all that matters. Making a nitride catalyst on a corrosion-resistant support is also plausible.
- There is only a short duration left in the project. The proposed MEA testing should be a key focus to identify any major hurdles for future development of these catalysts when implemented in fuel cells.
- MEA validation should be performed, and a dissolution study could be beneficial.
- Validating the catalytic performance in MEAs should be the top priority.

Project strengths:

- The project continues to push the forefront of low-PGM catalyst development, incorporating the advanced characterization tools available in the Office of Basic Energy Sciences user facilities in order to explore underlying mechanisms for high performance and manifest durability in RDE.
- The project has met the DOE targets on RDE for activity and durability. The Ni_4N core is relevant to the stability of the Pt monolayer, and NbO_2 could contribute to the anti-corrosion of carbon.
- The strength of this project is the novel PtNiN catalyst particle structure and resulting mass activity and AST stability.
- Overall, the project is investigating interesting materials, has excellent characterization of the materials, and has excellent RDE results.
- The RDE measurements and the characterizations are comprehensive, complete, and informative. The fundamental science is good.
- The project's strengths include characterization.
- The team has access to good characterization capabilities at BNL, though they could be better utilized.

Project weaknesses:

- The project's weaknesses include the following: 1) the size homogeneity of PtNiN nanoparticles could be improved through the modification of the synthetic method, 2) it is unknown whether there is a Ni leaching problem, and 3) DOE targets have not been met on the MEA level.
- As of yet, no MEA data are available to justify the approach of PtNiN. RDE performance is good only for electrochemical characterizations and screening and is not that relevant to DOE targets.
- The weakness of this project is a lack of parallel MEA testing and apparently weak collaborations.
- The project's weaknesses include weak collaborations and mediocre progress, especially lacking MEA translation of the developed catalysts.
- The key weakness of the project is the lack of integration with MEAs through the project partners.
- The lack of collaborative MEA testing is the primary weakness of this project.
- Lack of any MEA testing severely hurts this project.

Recommendations for additions/deletions to project scope:

- More focus on MEA optimization is needed for the project to be successful. The materials are active enough in RDE; no more RDE activity optimization is required. The Nb-nitride materials are being pursued for risk mitigation, but there should be an early decision point on whether to continue exploring Nb-based materials, because results so far are not promising.
- The project is ending. Based on the lack of progress demonstrated in recent years, there does not seem to be any reason to continue the work. However, if DOE chooses to fund this work again in the future, there should be assurance that a funded partner will be performing extensive MEA testing. The unfunded collaboration with GM does not seem to be providing any MEA results.

- Identifying an invested, engaged partner who will work to overcame challenges of MEA integration should be the highest priority of this project. Los Alamos National Laboratory was listed as a partner last year but was notably absent on this year's slides.
- It is highly recommended that the project run MEA testing on the PtNiN catalysts. Any further characterizations, reasoning, and catalyst optimization should be based on MEA performance. It is reasonable to give up $PtNbO_x$.
- It is recommended that BNL pursue MEA validation and optimization.

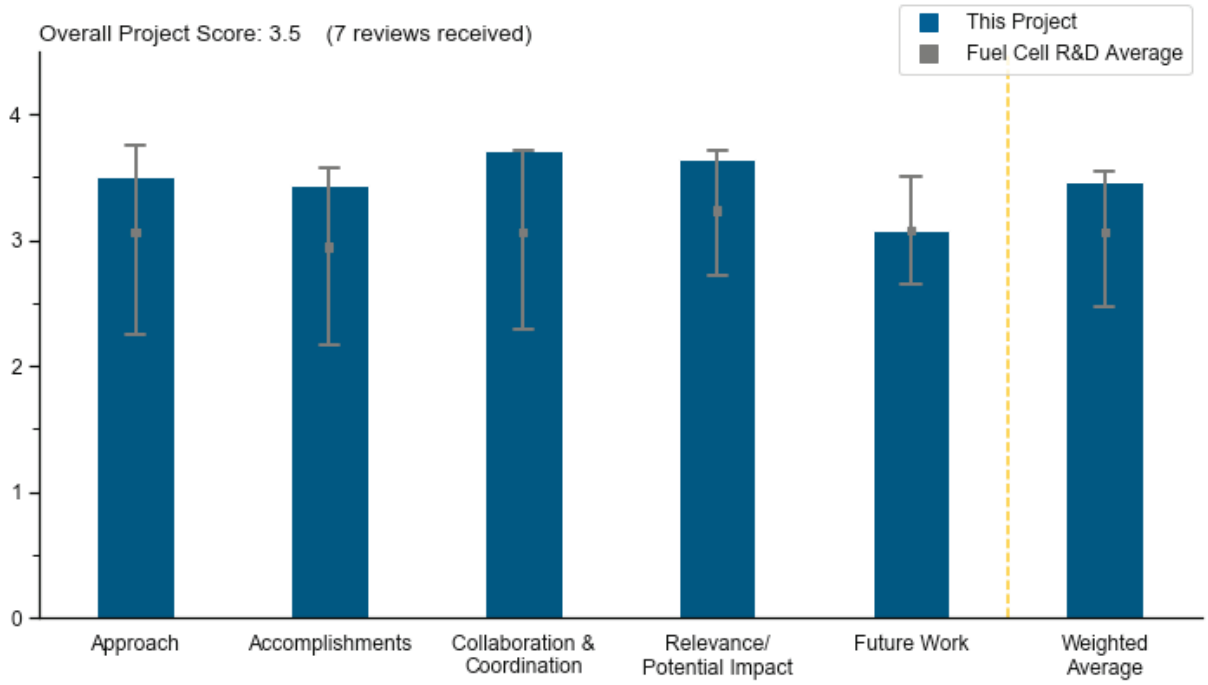
Project #FC-144: Highly Accessible Catalysts for Durable High-Power Performance

Anusorn Kongkanand, General Motors

Brief Summary of Project

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

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **3.5** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The project team utilizes a combined approach, examining catalyst supports and alloyed catalysts and improving transport to the catalyst using ionic liquids (ILs); this is supported by a novel characterization tool for understanding the origins of performance improvements. The approach is technically sound and could lead to key improvements, such as increased oxygen transport, H+ conductivity, and Pt electrochemical surface area (ECSA). The project's findings have the potential to lead to results that could help meet activity and durability membrane electrode assembly (MEA) targets under low-loading conditions. In addition, looking at cation effects, which leach out of the catalyst, or at Ce, which is used as a radical scavenger on the ionomer conductivity, is also important toward achieving the DOE targets.
- The project has an effective approach to understanding opportunities for improved catalyst utilization and performance. The areas that have been selected have provided meaningful results and are promising for

further development; these areas include the accessible carbon supports, the ionomer and IL interface understanding, the ordered intermetallic alloys, and the effects of the cations.
- The approaches adopted are appropriate and comprehensive, covering most important aspects of the topic—including the catalyst, the IL, the ionomer, and the carbon—to improve performance. In addition, modeling has been applied to understand the oxygen and proton transport issues so as to understand the fundamentals and the experimental results obtained.
- The project is making good use of developing different carbon supports to make catalyst particles accessible. The project seems to have moved to ordered intermetallic alloys, which is an approach similar to the Voja and Spendelow projects; maybe a consensus is forming on desiring ordered intermetallics. The IL approach is curious, especially considering that, in the past, Fuel Cell Technical Team (FCTT) members (or General Motors [GM]) have indicated they do not believe that ILs will be stable in electrode layers.
- The project's approach is focused on achieving DOE performance goals at high current density (HDC) by optimizing carbon supports, the electrolyte–Pt interface, the catalyst's activity and durability, and the mitigation degradation effects from transition-metal cations.
- The project takes on a major question about fuel cell behavior at low loadings, and the limitations of low loadings, while giving possible solutions.
- The approach taken by this team to address some of the HCD performance issues and methods used to quantify those losses is commendable. However, it is not clear that the IL that has been used in this project is the best available system for this effort. It is not clear what the fundamental ionic properties are or whether the IL has had any durability issues during the various cycling protocols employed.

Question 2: Accomplishments and progress

This project was rated **3.4** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The intermetallic ordering work is very promising. The IL's effect on stability is quite interesting, and it is good to see that these tests were done. The effects and understanding around them should be studied further. Although the project boosted the utilization of Pt to high levels, it was not able to achieve the rated power target at 150 kPa. The results were nonetheless impressive. However, using a system with such low catalyst loading, below the DOE target, may not provide the best value in terms of lifecycle cost. Focusing on systems with target platinum group metal (PGM) levels is recommended. The accessible Pt-Co stability is a concern, but the trade-off with the higher current density performance still makes this approach worthwhile. Regarding the Co loss effect, the result is relatively as expected. However, the lattice strain mapping is providing some good insight. The catalyst aggregate model and scanning transmission electron microscopy – computed tomography (STEM-CT) work are providing some good insights and are appropriate for better understanding the interactions within the catalyst–ionomer interface. The oxygen diffusivity through the water-filled pores will also feed into this work. The modeling work is a valuable aid in understanding and should be completed.
- The project has made progress toward the goals, has met most of the milestones, and is on track for the others. There is a nice set of variables and parameters investigated in terms of their impact on cell performance as well as investigation of the underlying cause of those changes using novel characterization tools and modeling studies.
- The project team has made very good progress toward achieving DOE targets on PGM total loading and utilization. All targets except the rated power at 150 kPa have been met.
- All the important milestones and go/no-go targets were achieved. Ionomer-related progress is not significant in either practical or fundamental terms.
- The use of ordered PtCo intermetallic catalyst appears to be a significant advantage in terms of durability and lower loss in mass activity (at least with the PtCo/KB). However, the PtCo/KB seems inferior at beginning of life to the PtCo/HSAC-f, and the support corrosion metric has not yet been determined; probably both fail that metric, and thus reliance on system mitigation will still be required. The Pt/Co ratio study and the contrary effects on low current density and HCD are nice contributions. The measurements related to Co dissolution and amount of Co in the ionomer phase raises questions about the effect of the ionomer-phase Co. Further questions that should be explored include whether it reduces the proton conductivity, and thus increases losses, and whether the oxygen resistance is higher for transport through

this Co-contaminated ionomer phase owing to lower water uptake. The measurements are good, but more understanding should be undertaken. The lattice strain mapping should be supplemented with extended X-ray adsorption fine structure (EXAFS) for better understanding of the catalyst surface. This catalyst development project appears to have developed primarily into an MEA development project.
- The project has raised more questions than it has answered, but this is always the case with good research. The finding that the best catalysts are those in pores without direct ionomer contact is something that most would not have guessed, and it will be the basis for much research going forward. Similarly, it is hard to understand how the ILs are limiting catalyst dissolution.
- Sensitivity studies on MEAs addressing durability and HCD performance should be done to understand what the effect of loading on HCD performance is before and after durability accelerated stress tests.

Question 3: Collaboration and coordination

This project was rated **3.7** for its engagement with and coordination of project partners and interaction with other entities.

- The collaboration among partners is evident in the publication list. The accomplishments in characterizing the Co migration and the use of advanced tools, such as STEM-CT and nano-CT to measure diffusivity through water-filled pores, is very effective.
- The team has strong collaborations with both unfunded and funded partners on a variety of techniques and topics. The partners' contributions to the project are clearly defined, and a clear strategy exists for the team to work together to accomplish project goals.
- The project is coordinated very well by GM, with collaboration between Cornell, Carnegie Mellon, Drexel Universities, the National Renewable Energy Laboratory, and 3M.
- This is a very strong team with significant collaboration with a number of groups, including other industries, national laboratories, and academics.
- The collaboration is great. The work from different groups is relevant and well integrated.
- The organization's collaborations appear well managed and valuable.
- This project is truly using some of the best people and facilities in the world doing what they do best. Generally, such a high number of collaborators should be discouraged, but the project team seems to be making it work well. However, it is very difficult to tell how much each group is contributing.

Question 4: Relevance/potential impact

This project was rated **3.6** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The work being performed on this project clearly aligns with the needs expressed in Fuel Cell Technologies Office and DOE research, development, and demonstration goals. The project is addressing the mitigation of cost and performance limitations in low-loaded catalyst layers by investigating a wide parameter set (from carbon types to ionomers and ILs) that has the potential to meet DOE targets.
- The project is well aligned with DOE goals and objectives and has the potential to advance DOE goals by improving fundamental understanding of processes occurring at the fuel cell cathode and integrating this knowledge into real-world polymer electrolyte membrane fuel cells.
- The work is highly relevant, addressing a key technical challenge of HCD performance and durability in an MEA that is impeding progress toward lower catalyst loading.
- The impact of this project will be very high. The project team is not only finding practical improvements but changing the models of how the community thinks these systems work.
- Higher-mass, active, durable catalysts are needed to enable higher levels of vehicle commercialization.
- The project team has made good progress on optimizing the catalyst and demonstrating the benefits of porous carbon for the HCD performance of low-PGM catalysts. However, it remains unclear how the porous carbon helps. The development of ionomers is mild, while this is an important part for the HCD performance.

- The potential impact of this approach for understanding transition-metal migration and durability is significant. However, in this approach, the team should also keep durability in focus and conduct sensitivity studies at higher loading. The impact on MEA cost is understood, but durability is critical.

Question 5: Proposed future work

This project was rated **3.1** for effective and logical planning.

- There should be more follow-up on some of the questions raised by the project's findings. This is especially true for how the ILs are working and will be retained, as well as the mechanism of conductivity within the pores. It would be interesting if the team had a carbon expert and could better tailor the catalysts to a structure that is more optimal with the new understanding.
- The proposed future work appears to be appropriate, covering fuel testing, materials (especially ionomer optimization), and fundamental understanding.
- The project's proposed work is well balanced and combines fundamental and practical aspects of fuel cell development.
- The project team should use target PGM loadings rather than the 0.075 mg/cm^2 loading. The other future work identified is appropriate.
- Future work tasks are well outlined, although it is not clear what type of "new ionomer" development and IL optimization will be carried out.
- For future work, the project team should consider a higher-loading MEA along with a low-loaded MEA for durability. This would help with clearly understanding the impact of this approach. Additionally, implementing a new IL this late in the project would not be an efficient use of funds or time. The team should focus on understanding the impact of the IL system already being used on durability. Exploring "catalyst synthesis paths for intermetallic ordered PtCo with well-controlled size" also seems to be a task starting too late in this project. If this was a new scope in the project from the 2018 Annual Merit Review, then the team should consider dropping this effort.
- It does not make sense to spend significant effort on optimizing IL application when there appears to be no idea of whether these are stable in the electrode matrix and/or durable. The project's response to 2018 reviewer comments regarding IL stability seems ill-considered and rash. Evaluating the IL stability in the matrix seems a critical task in terms of spending more effort related to this, especially when FCTT (specifically GM) has criticized use of IL in the past because it will not be stable. Evaluating the leaching of IL from the electrode matrix is, in part, not as difficult as evaluating effluent water from the fuel cell for the IL, and the task seems a bit straightforward and semi-trivial. Repeated cyclic voltammograms monitoring the capacitance and ECSA may also provide insight as to the stability of the ILs in the matrix.

Project strengths:

- One of the project's strengths is attacking the problem from various angles, which provides an enhanced parameter and material matrix (ILs, porous carbon, ionomer, and annealing effects) that allows for identification of design parameters for improved cell performance. Other strengths include the examination of the cation effects and Co loss studies, a number of characterization studies that clearly contribute to the project, and linkage to foundational work (imaging, strain mapping, etc.).
- The project team is great. The leading organization, GM, is well equipped and extremely experienced in this topic. The expertise of the team's collaborating partners is the right hammer for the nails to be addressed in each strategy. As a result, the team knows what barriers are likely to be overcome by what strategies and has the resources and capabilities to implement those strategies. The project has made steady progress, meeting all the milestones and go/no-go targets at a good pace. Optimizing the catalysts and demonstrating the benefits of porous carbon for HCD performance are important achievements.
- The project combines fundamental and practical aspects of fuel cell development. The project team has implemented state-of-the-art experimental techniques, such as lattice strain mapping, confocal X-ray fluorescence, and advanced modeling, to understand the root cause of insufficient fuel cell performance.
- The project collaborators provided excellent diagnostics, characterization, and modeling approaches to supplement the main studies and increase fundamental understanding. There was also identification and development of concepts and approaches that have proved fruitful.

- There is a strong approach and relevance with great results and implications, as well as strong teaming, with each group working toward its strength.
- The ordered intermetallic PtCo is showing improved durability and mass activity; this is similar to other projects led by Voja and Spendelow.
- The project's strengths lie in the team's expertise in understanding Co migration techniques and correlating that to fuel cell performance curves. Using IL in the MEA, although a novel approach to understanding the Pt–ionomer interface, does not seem to provide value without durability testing. If there is no efficient method for characterizing IL during durability tests, then this effort should be dropped for the rest of the project.

Project weaknesses:

- It appears that the ionomer-related progress lags behind. It has been known that the major limitation of low-PGM catalysts for HCD performance arises largely from the Pt–ionomer interface, such as local oxygen resistance and site-blocking by the ionomer. However, most improvements lie on the catalyst side, probably by improving the catalyst's inherent activity and durability. Although it was demonstrated that porous carbon helps, there is no clear evidence for how this occurs, and the improvement has not been related to the ionomer-related barriers. There are no images in the atomic scale showing the interfaces between the Pt nanoparticles and the porous carbon in the atomic scale. It is appealing to see that the project will make improvements on the ionomer and the Pt–interface parts and will directly address the local oxygen transport resistance associated with deeper understanding of corresponding fundamentals.
- This project seems to be focused only on PtCo catalysts. Perhaps exploring other Pt-M alloy catalysts using a similar approach may have shed more light on the transport mechanisms and impact on HCD performance. The IL seems to serve only as a diagnostic tool for understanding the Pt–ionomer interface. The project does not seem to have addressed the influence of the IL used in these studies, and it is unclear that the results would be different if a different IL had been used.
- The principal investigator has no idea whether the IL is stable and/or durable in these electrodes. It is unclear why the IL benefits do not occur with the HSAC-f support; there is a lack of fundamental understanding on effect. The CO displacement measurements indicating that adsorption of SO^{3-} is lower is not correlated to mass activity measurements. If lower surface adsorption occurs with the ILs, the mass activity should increase, similarly to rotating disk electrode measurements in $HClO_4$ compared with H_2SO_4.
- Something related to one of the project's strengths is that, because of the many interrelated phenomena, it is not clear how to isolate certain effects and identify the dominant factors controlling the performance.
- The project is relying on a PtCo catalyst that has limited potential for improvement.
- As the project developed, the team was not very agile at changing scope or pursuing some of the questions that have been raised but rather has stuck to the script.

Recommendations for additions/deletions to project scope:

- There are no major recommendations, but the scope should be managed carefully to identify the key contributors to the catalyst layer performance and stability. For example, annealing reduces Pt loss, but perhaps it would also affect the CO dissolution. It is also unclear what the major contributor to the PGM utilization is: annealing or carbon support.
- The strain effect in PtCo has been well understood and documented. There is no need to proceed with the fundamental parts and characterizations. It is recommended that atomic-scale imaging be applied to explore the Pt-carbon-ionomer interfaces, which would be best under operando conditions. Efforts made on the catalyst side are excellent; more efforts should be dedicated on the ionomer and interface sides.
- This project needs to evaluate the stability and durability of ILs in their electrodes before more significant work is put into optimizing these electrodes. The lattice strain mapping should be supplemented with EXAFS for better understanding of the catalyst surface.
- The project team should do more work to understand how the IL retards catalyst dissolution. There should be some custom synthesis of carbons to optimize the number of appropriate pores.
- It is recommended that the project drop the scope on new catalyst synthesis paths for PtCo-ordered structures with controlled size and stop all IL work, as there is no method to characterize the IL once it is incorporated in the MEA.

- The part of the project related to introducing a new 3M ionomer does not look very promising.
- Recommendations for additions or deletions are not applicable at this point in the project.

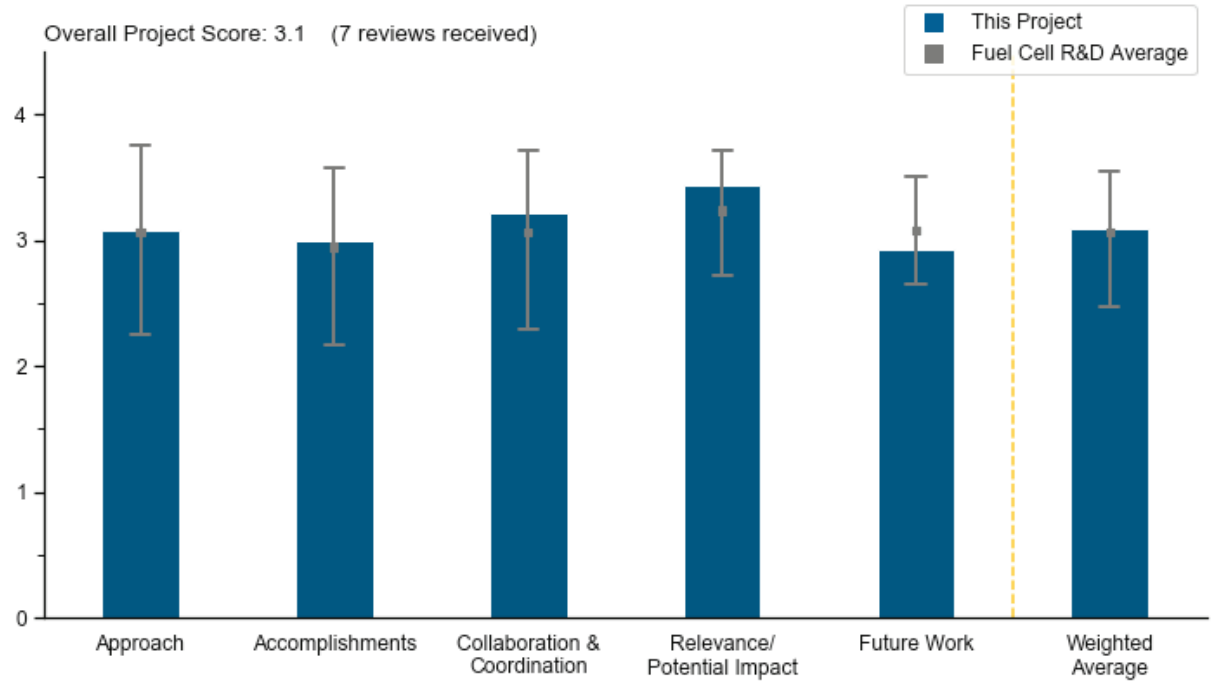
Project #FC-145: Corrosion-Resistant Non-Carbon Electrocatalyst Supports for Proton Exchange Fuel Cells

Vijay Ramani, Washington University

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 (non-PGM) metal oxides as non-carbon alternatives. Along with being corrosion-resistant, the project supports would have high surface area, exhibit strong metal–support interaction with Pt, and demonstrate high electrocatalyst performance.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **3.1** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- Investigating support materials that offer improvements to corrosion resistance over carbon would benefit fuel cell systems. Combining modeling with synthesis and experiment is a viable approach to finding new materials and systems. The choice of doped tin oxide seems very reasonable based on the conductance properties. The choice of and focus on Nb and Sb were not really presented; the doping levels and methods investigated were also not presented in detail.
- It is interesting that this project is looking at such a unique approach by pursuing non-carbon supports. This seems to be a high-risk–high-reward project. The plan to address barriers appears to be working, based on test results.
- The connection to theory seems lacking, and the addition of atomic layer deposition (ALD) seems strangely complicated. Overall, the project has a good synthetic approach, with barriers to overcome and down-selects in the right places.

- The approach includes experimental and theoretical modeling of non-carbon-based catalyst supports (non-PGM metal oxides) to develop high-surface-area supports with good electrical conductivity. Most of the approach discusses TiO_2 and doped TiO_2 supports. Data are related to Sb-doped SnO_2 and Nb-TiOx-aerogel supports.
- This project addresses the need for non-carbon supports to mitigate support corrosion during shut-down/ start-up of fuel-cell-powered vehicles. From the research performed since the last U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program (the Program) Annual Merit Review (AMR), it appears that the project did not focus on achieving its mass activity target. It is not clear from the presentation what Pt-based alloys will be prepared to meet the mass activity target. The presentation does not mention how the project will address the load cycling accelerated stress test (AST) and meet the DOE target of <30 mV loss at 0.8 A/cm^2 after 30,000 cycles.
- Both aerogel and xerogel approaches are good for porous media. The only concern is the scale-up for aerogel, which requires the use of supercritical drying. It is not clear that scale-up for aerogel is realistic. The stability of Sb-doped SnO_2 may also be a concern. It is not clear that Sb-doped SnO_2 is stable in the fuel cell operating condition.
- Alternative supports for carbon can yield benefits in support stability; however, equally important are the catalyst stability and catalyst activity. The approach to the work seems to be lacking in these areas.

Question 2: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has made good progress with respect to improving the performance of non-carbon supports in membrane electrode assembly (MEA) tests. The non-carbon supports appear to work with respect to improving catalyst start–stop durability. In the future, load-cycling data should be discussed to confirm that these catalysts and supports are also effective for the drive-cycle requirements. It is good that the project is presenting cost modeling results; however, the team needs to consider the system mitigation cost, not a stack replacement, as the baseline. It is known publicly that Toyota and General Motors have system mitigation strategies that would not require a stack replacement. Determining the cost of this mitigation may not be easy, but some type of estimate should be derived to determine the real value of the non-carbon support.
- In the past year, there were meaningful improvements in mass activity and performance, one of this project's major limitations in the past. It seems that performance is getting close to that of Pt/C while offering significant potential durability benefits and that one of the big advances was improving the catalysis by modifying the synthesis pathway to include Pt in aerogel formation. This was not discussed in much detail in terms of why and what was learned and what this could mean for additional advances. The catalyst cost analysis showed clearly that antimony-doped tin oxide (ATO) was not a critical cost concern compared to Pt, but the system cost analysis was not particularly compelling, as most systems will not tolerate stack replacements. This perhaps changes when considering heavy-duty applications in which these materials may offer even more durability advantages.
- The observed durability is excellent and the performance good, despite the apparently lower kinetic performance due to Pt deposition issues. It is likely the move to alloys will help with this.
- Rotating disk electrode results show better mass activity for Pt/aerogel-NbTiOx. However, the MEA performance is well below that of Pt/C. Even worse, the durability of the Pt/aerogel is substantially worse. Comments were made that this is due to loss of conductivity, which is likely going to happen under other conditions as well. Thus, these materials do not seem overly promising for long-term durability. It seems like work can end on these materials.
    - The in-cell mass activities of Pt/Pt-aerogel-ATO do not appear comparable to Pt/C (slide 15, beginning of polarization curve). While they show better performance at very high current densities, those performances are not comparable to state-of-the-art (SOA) Pt/C electrodes (they get <0.4 V at 2.0 A/cm^2, whereas the SOA is ~>0.6 V at 2.0 A/cm^2).
    - The carbon corrosion results (slide 15) do show that these materials have improved durability in comparison to Pt/C. However, the degradation over 1,000 and 5,000 cycles is substantial. The real competition is not Pt/C with shut-down/start-up cycles but Pt/C with mitigation strategies incorporated into the vehicle system to avoid high potentials. While a material solution is preferred, it needs to be competitive with the overall system strategy; this is not.

  - The cost modeling assumptions are simply inaccurate. Original equipment manufacturers (OEMs) are not going to make systems in which the catalyst has substantial degradation; OEMs are putting in system mitigation strategies to limit the shut-down/start-up voltage excursions. Thus, current Pt/C MEAs do survive 5,000 cycles; the issue is the additional system complexity, which equates to some cost and probably some loss of efficiency—but nowhere near that associated with replacing the stack. Thus, the cost modeling assumptions are simply incorrect, and it is an erroneous conclusion to state that the Pt/ATO is cheaper than Pt/C. The Toyota Mirai is working very well with Pt/C.
- The support stability for aerogel–niobium-doped titanium oxide (NTO) is questionable; mass activity loss is about 50% after only 500 cycles (slide 11). Aerogel–NTO is not as stable as carbon-based support. It is not clear why the project still focuses on the aerogel–NTO support. Beginning of life of Pt/aerogel–ATO polarization curves is much lower than that of Pt/C within the 0.9–0.6 V range. It would be good to know the cause. The H+ conduction seems to be an issue. The principal investigator (PI) may consider improving the H+ conduction within the catalyst layer. Mass activity loss is quite significant: 50% for Pt/aerogel–NTO. The loading is high: 0.2 $mgPt/cm^2$. Backpressure is high: 100 kPa.
- Incremental improvements toward addressing mass transport limitations were presented, but catalyst activity is still significantly behind. The cost analysis did not seem to consider the industrial engineering controls to avoid high voltage spikes. If engineering controls are considered, it is not clear whether the cost analysis with the expensive support will be an attractive alternative.
- Not much progress has been made since the 2018 AMR. The technical target/current status table on slide 5 is identical to the one presented in the 2018 AMR.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- This project appears to have effective and strong collaborations.
- The teamwork is very impressive.
- Collaboration levels seem mixed. Nissan Technical Center North America (NTCNA) is strong. The University of California, Irvine, seemed to have a good start at the University of New Mexico but was delayed by the PI move; what remains is unclear. The Fuel Cell Consortium for Performance and Durability (FC-PAD) interactions are not clear; it seems that some stability testing would be helpful.
- The team has broad relevant skills appropriate for the project. What seems to be missing is an industrial participant that would potentially be able to supply the developed materials at commercially relevant scales.
- Collaborations seem appropriate, although additional collaborations might prove useful, especially in the area of mass activity. The project could use help preparing Pt-alloy catalysts.
- Interactions with NTCNA in terms of testing and MEA optimization should be beneficial.
- This project involves two universities and an OEM. Interaction with FC-PAD is suggested.

Question 4: Relevance/potential impact

This project was rated **3.4** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- Higher-mass active catalysts with stable supports to eliminate degradation mechanisms such as corrosion and particle coarsening would be a tremendous benefit to fuel cell commercialization.
- This is an interesting project that is investigating unique non-carbon supports. The technology may be unlikely to be adopted for automotive MEAs in the near future because it is relatively immature; however, these supports could prove useful in electrolyzers or other types of fuel cells that require harsh operating conditions and in which carbon corrosion is a bigger concern.
- The potential of this approach and the achievements greatly advance Program goals.
- Replacing standard catalyst material supports with high-performance and more durable replacements would have a significant impact. Initial durability testing using ASTs has shown promise. Performance is not yet

to a fully competitive level, and whether ASTs developed for current systems are relevant for these new systems is not clear.
- Using metal oxides as a catalyst is a good approach for long-term durability of polymer electrolyte membrane fuel cells (PEMFCs). However, the cost of metal oxides may prevent their application in PEMFCs, despite the benefit of the support stability.
- If successful, the new supported catalysts will benefit fuel cell development. However, as fuel cell manufacturers continue to advance their technologies, the new low-PGM electrodes using this class of supports might not be relevant.
- The project focuses on corrosion-resistant metal oxide supports. Metal oxide supports have been suggested as catalyst supports in the literature. However, to date, none of the proposed metal oxide supports meet the DOE target for catalyst durability and support stability.

Question 5: Proposed future work

This project was rated **2.9** for effective and logical planning.

- The future work all seems appropriate; the project should concentrate on catalyst development related to mass activity and durability. Cost analysis should not be a priority for this project; however, if any is done, realistic assumptions related to how OEMs are actually operating cars should be incorporated (i.e., new catalysts are a trade-off with system mitigation strategies). Future work should include use of higher-activity alloys, such as Pt-Ni and Pt-Co. Future work should also include understanding the support electrical conductivity degradation and whether that reduction in electrical conductivity will occur at steady-state operating conditions.
- The Pt alloy investigations make sense to increase mass activity. Varying the ALD conditions to try to further improve Pt distribution seems reasonable, as this seems to have had a strong impact on this year's performance improvements. Gaps include a lack of detail in future work plans and a discussion that includes mechanistic understanding of the processes affecting properties.
- The goals of improving mass activity and electrodes make sense. Additional support approaches from the University of California, Irvine, also make sense. However, density functional theory (DFT) at this stage of the project makes no sense, especially as the effectiveness and connection between the early DFT and results is tenuous. Pt ALD seems like an overly complicated approach to Pt deposition and is poorly suited to the high loadings of Pt needed.
- Preparing Pt-alloy can lead to improved mass activity and meeting the project milestone of achieving 0.3 A/mgPGM. However, the PI did not specify the type of Pt-alloy catalysts that the project will prepare in the third year.
- The future work is logical and appropriate. Examination of 0.95–0.6 V cycling should be presented to enable an understanding of whether Pt adhesion to the support is an issue for these materials.
- The proposed future work is appropriate.
- Pt alloy development should have started earlier in the project. It is doubtful the project will be able to accomplish good activity performance in such a short time before the final deliverable. There is no mention of catalyst durability testing. While the support stability seems reasonable, the Pt stability on the metal oxide supports might be affected.

Project strengths:

- This project is attacking catalyst durability through a unique approach relative to other projects. The team has made significant progress at improving the performance of the non-carbon support catalysts in MEAs and has obtained encouraging initial results with respect to durability.
- The metal oxides are a good approach for catalyst support stability. The aerogel and xerogel approaches are effective for obtaining high surface area, which is critical for support. This project is a good exploration of non-carbon catalyst supports with stability superior to carbon.
- The project has strong new results for a more corrosion-resistant support system. Performance is approaching that of Pt/C, with improved durability.
- Catalyst stability is excellent, even with low mass activity. There is a close connection between Washington University and NTCNA.

- Collaboration with NTCNA adds strength to the project, as Nissan has provided significant contribution to the research. The PI is well experienced in PEMFC research.
- The strength of this project is the development of materials that should not be susceptible to corrosion during normal operation or shut-down/start-up (e.g., carbon corrosion).
- The team is developing more stable supports, as compared to Vulcan® XC carbon.

Project weaknesses:

- The cost analysis is overly flawed with assumptions. Pt/C is never going to be operated straight away like this. The catalysts, to date, are not competitive with SOA Pt/C, let alone PtCo/C. The Pt/C polarization curves (e.g., slide 15, iR-corrected) are not representative of what can be achieved with Pt/C; the fact that those are iR-corrected but the Pt/C slope is rather large shows there is a problem with that baseline. The supports have not adequately improved catalyst durability to remove vehicle durability mitigation strategies; thus, they are still in a developmental phase. The durability needs to be improved rather dramatically, it appears, in both electrical conductivity and Pt activity stability.
- Catalyst AST testing (0.6–0.95 V, 3,000 cycles) should be included to assess the usefulness of the materials under realistic conditions. The cost model needs to have a more realistic comparison to system mitigation strategies, not stack replacement. Performance at low current density needs to be addressed.
- The project does not focus on catalyst durability under load cycling between 0.6 V and 0.95 V. The metal-oxide-supported Pt and Pt-alloy catalysts tend to show poor activity after a few hundred or few thousand cycles owing to Ostwald ripening and agglomeration processes.
- The catalyst development needs more fundamental aspects. More characterization of the Pt/aerogel–ATO/NTO catalyst is encouraged to build the property–structure–performance relationship to better guide MEA development.
- Mass activity of catalysts is poor, possibly stemming from poor Pt deposition. The proposed remediation via ALD is dubious. The utility of DFT and implementation of DFT are unclear, even at project closing. FC-PAD is underutilized (for stability, DFT, and analysis of Pt quality issues).
- The catalyst mass activity seems to have reached a maximum with these supports. With fuel cell engineering controls, the need for this work is not clear since the catalyst's corrosion-resistant attributes do not justify the sacrifice in catalyst activity.
- The project lacks details and mechanistic discussion or understanding regarding critical processes related to performance and durability.

Recommendations for additions/deletions to project scope:

- More mechanistic studies are needed to quantify degradation rates and mechanisms. The project should conduct additional in situ testing (Sb, Sn leaching and effluent analysis), ex situ testing (conductivity studies as a function of thermal or electrochemical oxidation), and post-mortem analysis (Pt size, migration/coalescence).
- The project should emphasize the direct mass activity comparison with SOA PtCo/C catalysts and evaluate the catalyst in terms of durability, in terms of both Pt mass activity and corrosion. Appropriate metrics for durability are not comparisons with Pt/C but whether a catalyst with improved durability can replace Pt/C with the various mitigation strategies present in operating systems.
- The project should remove DFT and focus on improved synthesis of supports and Pt-alloys (that will likely stabilize on oxides). Also, there should be more interaction with FC-PAD for stability and support degradation analysis.
- In cost modeling, the team needs to consider the system mitigation cost, not a stack replacement, as the baseline. Load-cycling data should be reported to confirm that these catalysts and supports are effective for the drive-cycle requirements.
- Understanding the interaction of metal oxide support and catalyst will be valuable for support and catalyst stability.
- The project should perform load cycling ASTs for at least one of the promising catalysts.
- Catalyst ASTs are needed, in addition to the support ASTs.

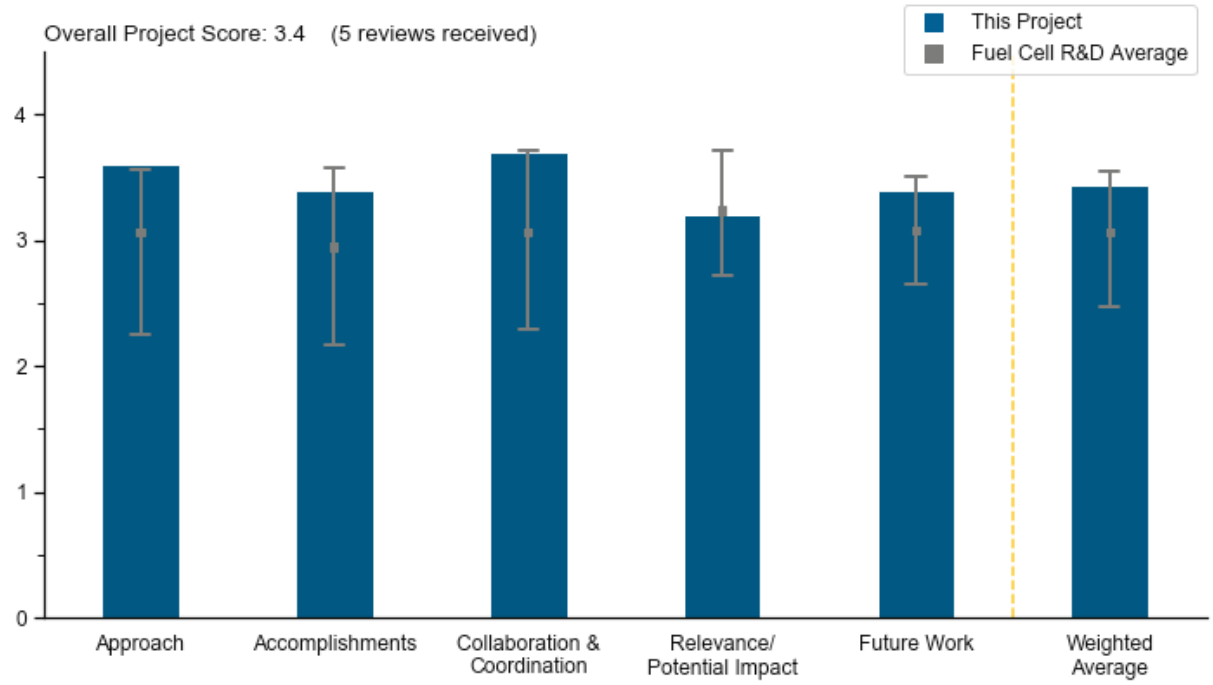
Project #FC-146: Advanced Materials for Fully Integrated Membrane Electrode Assemblies in Anion Exchange Membrane Fuel Cells

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.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **3.6** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This project has done a very good job of systematically defining and solving new challenges in the AEMFC field, some of which (such as phenyl adsorption on Pt, cation co-adsorption, etc.) have not been tackled or discussed, essentially, by any other groups. This project has been timely in many aspects and has been one of the truly successful projects in the field globally.
- The project addresses key barriers of durability, performance, and cost of alkaline membranes and AEMFCs. Key barriers of stability of the backbone polymers and cationic groups have been identified and are being addressed. Key issues concerning interactions of the ionomer and ionomer degradation products with the catalysts have been identified and are being addressed.
- The approach addresses critical limitations of AEMFCs. An initial focus on membrane stability, followed by a shift to addressing cell performance and durability, addresses critical barriers; the project is well integrated with other AEM efforts.

- The project's approach is a logical progression from membrane production and testing, to AEMFC performance evaluation, to AEMFC durability testing.
- The main idea behind bringing in AEM materials was to create fuel cells with inexpensive electrodes, which means no platinum group metals (PGMs). A significant portion of this project's attention seems to be on the interaction of the ionomer and Pt. Maybe, if a non-PGM catalyst were employed, no phenyl/phenol-Pt or cation-Pt interactions would be important. If phenyls create problems, the polyphenylene polymer appears to be the worst choice for AEMs. Also, it would be good to pick a "champion" ionomer and go through all the tests with that polymer. Showing good durability data with an AEM of conductivity of 10–20 mS/cm at 80°C (which is practically useless material from the perspective of fuel cell power output) is not fair and will not lead to any practical conclusion.

Question 2: Accomplishments and progress

This project was rated **3.4** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has developed a new route to complete quaternization in ionomers; now conductivity loss due to interaction between catalyst and ionomer is addressed, and the project has a very stable membrane. The project team identified performance limitation caused by cation co-adsorption and designed an ionomer to minimize adsorption. The team is finishing 5000-hour durability and observing better performance at high current density, thanks to a lower potential for oxygen reduction reaction. The project is on track to meet final project milestones and complete deliverables. This is the final year of the project, which is an extension of the original three-year project. The project is 98% complete toward an end date of September 30, 2019, and nearly all money has been spent.
- Significant progress has been made, especially in the area of determining degradation modes and some of the causes for poor AEM or MEA performance. The project has worked to determine that the reason for the conductivity loss and degradation of Diels Alder PolyPhenylene (DAPP) membranes is cross-linking due to unsubstituted Br reacting with hydroxide, followed by dehydration and ether formation in the Hexane-6-TriMethylAmmonium (HTMA)-DAPP membranes. This has led to pre-cross-linked material with higher durability for AEM materials. The results indicating phenyl oxidation to phenol and poisoning by the phenol group are important for future durable membrane design and high performance in AEMFCs. Work from this project has been instrumental to increasing understanding of AEM durability issues and catalyst alkaline–ionomer interactions. As seen on slide 6, showing alkaline stability results using a logarithmic scale for the conductivity is misleading, masking degradation and making it look like materials are more stable than they are. The initial decrease in conductivity (over the first 1000 hours) for the HTMA-DAPP shown in this slide is substantial and could be as high as 50%, but it is difficult to determine from the logarithmic plot. Because of the choice of a logarithmic scale for the Y axis, it is difficult to see if there is continued degradation after the first 1000 hours. The neutron reflectometry results are interesting, but it is not clear how the simple molecule analogs relate to the real-world situation. In membranes, the cations are tethered to a polymer backbone, and the tether and polymer backbone will have a large influence on how densely the cationic groups can pack near the Pt surface.
- Good progress has been made in regard to improving performance and the durability mechanism. However, the loss in performance after 500 hours is a concern, and the target is unlikely to be met with remaining project funds.
- The only target that has yet to be met is durability. Otherwise, very good progress has been made.
- Practically all project milestones have been met on time.

Question 3: Collaboration and coordination

This project was rated **3.7** for its engagement with and coordination of project partners and interaction with other entities.

- The project partners appear to be collaborating well with each other. The project lead has been collaborating with others in the AEM community, including other national laboratory, academic, and industrial collaborators, and has been a leader in the AEMFC community.

- The collaboration seems strong, with team partners contributing. There are also a number of other non-funded partners with some level of interaction.
- It appears that this team has worked very well together and that team members have all made significant impacts on the project.
- Many collaborators, materials, and much knowledge is flowing in both directions.
- Strong inter-laboratory cooperation is evident.

Question 4: Relevance/potential impact

This project was rated **3.2** for 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 has been one of the most successful AEMFC projects out there. The team has pushed the performance and improved the stability of ionomeric components ex situ. Even though they have not yet met the durability target, a couple of years ago, 500 hours of AEMFC durability with 12% degradation would have been unheard of. Even now, it is very good, and only a few other groups are known to have had the ability to accomplish it.
- AEMFCs are an appropriate low-technology-readiness-level area for DOE to investigate. They offer the potential to operate with reduced- or zero-PGM loadings, potentially leading to reduced costs and a large potential impact. This project addresses key AEMFC issues of durability and MEA performance. The applicability of AEMFCs to automotive applications is unclear, as there are issues with carbonate formation and extra system complexity needed to scrub CO_2 from air.
- The commercialization of AEM materials with Xergy, Inc., and Aldrich is impressive and should be impactful to the community.
- Relevant progress has been made in developing AEMFCs with higher performance. There is a continuing need, however, to develop more durable cells that can produce high levels of performance with non-PGM catalysts.
- The reviewed research can be classified as basic. Except for the ionomers developed at Rensselaer Polytechnic Institute, everything will be of moderate practical importance toward achieving the general goal of bringing AEMFCs to the market.

Question 5: Proposed future work

This project was rated **3.4** for effective and logical planning.

- The proposed future work addresses critical issues, including improved durability, non-PGM catalysts, and carbonation.
- The one remaining task cuts right to the heart of AEMFCs now: durability.
- The project is nearing completion. The proposed work is appropriate for the time remaining in the project.
- The proposed future work is appropriate to finish the project by September 20, 2019.
- Investigating the degradation of pathways due to the presence of Pt is a waste of time. It would be better to try some non-PGM catalysts. Also, fuel cell performance tests with O_2 oxidant should be done with air, or at least with synthetic air (with no CO_2), instead.

Project strengths:

- The project addresses the stability of AEM membranes and the performance and durability of AEMFCs. These objectives are critical barriers. Good progress was made in particular with improving cell performance and meeting the majority of milestones.
- The project has developed stable AEMs, identified causes of performance degradation, and developed mitigations for those causes. New ionomers have been commercialized and will be available through Aldrich. The project has numerous collaborations.
- The project team has done exceptional work determining degradation mechanisms.
- This is a very strong team, and they have made very good progress.
- The project is strong, but not too strong.

Project weaknesses:

- The membrane durability remains a concern, and targets will likely not be met. Future work can potentially address other AEMFC limitations, including non-PGM catalysts and carbonation. It is also unclear how or to what degree steady-state operation projects to lifetime durability.
- The project has no real weaknesses, though it may tend to oversell the technology slightly (for example, using a logarithmic plot when showing degradation).
- The team does not show compelling evidence that the real cause for in-cell degradation is the cathode ionomer oxidation.
- There is too much focus on the effects of Pt. Non-PGM electrode-based AEMFCs are what is needed.
- The project's weaknesses include the delays to the project schedule. Ongoing questions remain about the long-term stability of AEMFCs.

Recommendations for additions/deletions to project scope:

- The team might benefit from reaching out to another group outside of this project with a more cathode-stable ionomer–electrode formulation, in order to reach the durability target. Also, it is unclear or unstated whether the cells are operated with wet KOH at the beginning of life or whether the KOH is completely removed before operation. If the former, it could be that it is the KOH mobility outside of the cell area that is decreasing performance with time. Finally, the team could also try to run the durability experiments with air (both with and without CO_2). This can often help with durability by either (1) making the reacting environment less oxidative (lowering oxygen concentration), or (2) the presence of a small amount of CO_2 at 80°C leaving a few carbonated groups, which are less nucleophilic than OH-. One of these approaches might also improve stability enough to reach the target. Also, there is disagreement with the previous year's reviewers about CO_2. That would be a distracting task from the primary goals and skills of the team.
- The project is nearing completion. No additional scope is suggested. This is an area that would be appropriate to continue, but perhaps with more of an emphasis on electrolyzer conditions.
- The project team should focus on the best-performing ionomer type only and test it thoroughly.
- The project is nearly complete, and changes to the project scope are not recommended.

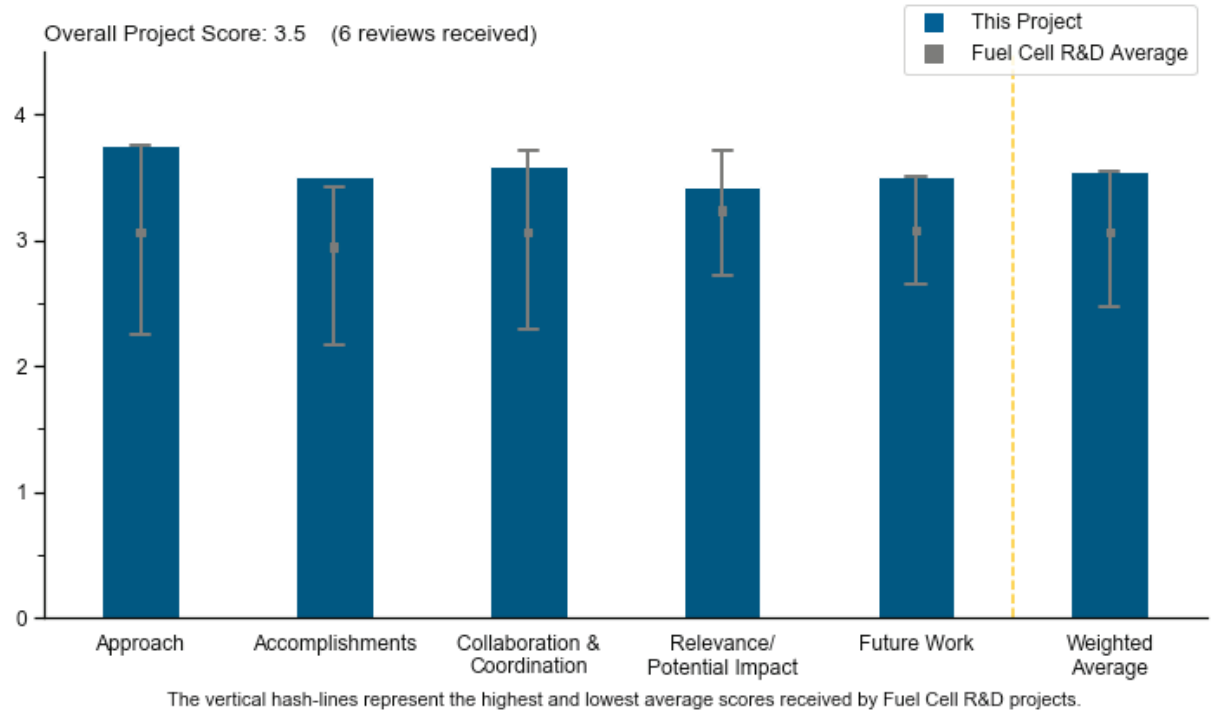
Project #FC-147: Advanced Ionomers and Membrane Electrode Assemblies for Alkaline Membrane Fuel Cells

Bryan Pivovar, National Renewable Energy Laboratory

Brief Summary of Project

Anion exchange membrane fuel cells (AEMFCs) 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 AEMFCs, and apply models and diagnostics to AEMFCs to determine and minimize losses (water-management-, electrocatalysis-, and carbonate-related). Researchers will synthesize, characterize, and optimize AEMs and fuel cells for performance and durability.

Project Scoring



Question 1: Approach to performing the work

This project was rated **3.8** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The project identifies durability, cost, and performance as key barriers for AEMFCs. The strategy to use perfluorinated materials to address water management is notable, as water management is a key issue affecting performance in AEMFCs. This project is also addressing the impacts of CO_2 and carbonate formation, another key barrier often neglected in AEMFC studies. The approach of using perfluorinated materials may be detrimental to cost but should be beneficial for water management.
- The team is clearly extremely experienced and competent enough to carry out this work; this is evident in the approach. This is an extremely challenging project, given inherent issues with AEMs and their application to fuel cells. The team has done an excellent job sequencing through different synthesis paths for the polymer and developing different configurations for membrane electrode assemblies (MEAs). The data are encouraging.

- It may be that, just like with polymer electrolyte membrane (PEM) systems, the path to practical AEMFCs will lead through perfluorinated backbone ionomers. The project team does the syntheses, optimizations, and modeling very well.
- The project has a systematic approach from membrane to MEA and fuel cell testing. The project team has developed a combined experimental and modeling approach with key collaborations and partnerships.
- The project is well designed; the critical barriers and technological risks are understood, and the mitigation strategies have been proposed.
- The project team has made great progress.

Question 2: Accomplishments and progress

This project was rated **3.5** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The AEMFC performance and durability demonstrated by the project's principal investigator (PI) are in the top 3% of the world's best achievements in the field. A majority of milestones have been successfully completed.
- The demonstrated performance, methodology, and identification of problems and avenues for improvements were excellent.
- There has been excellent progress in MEA developments and investigating different catalyst approaches (gas diffusion electrode vs. catalyst coated membrane). The team has done good work looking at water management in AEMFCs, particularly modeling work and experimental validation with segmented cells. The status of the Gen-3-type membranes (tethered with all C linkage) is unclear, specifically whether a go/no-go decision was made for this path or if this option is still being pursued. Gen 2+ work is important since data has shown the RN(Me)3+ cation degrades, but progress in Gen 2+ seems slow, and not much progress is apparent since last year. International Electrotechnical Commission conformity is a bit low, and durability data for longer durations is needed. It would be nice to know what cation is being utilized in the Gen 2+ work.
- The project is 85% completed toward project goals and ends on September 30, 2019. The Gen 2+ membrane shows material degradation in KOH testing, which could complicate conductivity measurements. There are interesting modeling results from Lawrence Berkeley National Laboratory (LBNL) that point to water management as key to stable performance. There is more work to be done on experimental validation of the CO_2 modeling results.
- All the past milestones were met. The fact that the team attempts to employ CO_2-free air in the fuel cell experiments is a plus. Now, let the project move toward non-platinum-group-metal (non-PGM) electrodes, as promised to the public years ago. Modeling CO_2 effects and water management indicates that the team is attacking truly important barriers.
- To the extent that the project laid out key performance metrics and achieved them, the work is excellent. The team achieved 600 hours of continuous operation and good current densities. However, there is still a gap in requirements for commercial systems.

Question 3: Collaboration and coordination

This project was rated **3.6** for its engagement with and coordination of project partners and interaction with other entities.

- The in-project collaborations appear to be working well. Collaborations with the AEMFC community are apparent, especially through the electrode work. National Renewable Energy Laboratory (NREL) has been supplying its Gen 2 membrane to multiple institutions.
- The collaboration between NREL, LBNL, Colorado School of Mines, and 3M was mutually beneficial and accelerated the project's progress.
- It is evident that the team worked well together. Each team member appears to have done its part, and NREL has coordinated activity well.
- There is excellent collaboration between three national laboratories, a university, and an industrial partner.
- There has been good collaboration with all partners and other projects.

- There is an effective use of collaborators' strengths.

Question 4: Relevance/potential impact

This project was rated **3.4** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- AEMs offer a route to lower the cost of fuel cell systems (an anticipated reduction in precious metal catalyst content and costs). This is of vital strategic importance in DOE's overall strategy to support a transition to hydrogen fuel cells in order to meet global targets for CO_2 reduction and eliminate dependence on fossil fuels. The project is clearly relevant and has high potential impact. There are, however, multiple routes to achieving this goal. DOE should explore many alternatives simultaneously. It would be important to understand why this chemistry is a good candidate to be in that down-selected group. The perfluorinated backbone is a big plus. It would be good to know if there are other considerations.
- AEMFCs are an appropriate low-technology-readiness-level area for DOE to investigate. They offer the potential to operate with reduced- or zero-PGM loadings, leading to reduced costs and a large potential impact in some applications. The applicability of AEMFCs in transportation applications is questionable, as there are added complexities of dealing with carbonate formation. Some analysis of the necessary cost and size of an automotive system to remove CO_2 from the air stream may be beneficial for guiding decisions on directions for AEMFC work. This project addresses key AEMFC issues of durability and MEA performance.
- Durable ionomer and the important studies of CO_2 and hydration are surely well aligned with the Hydrogen and Fuel Cells Program and DOE research, development, and demonstration (RD&D) objectives. This project has the potential to advance progress toward DOE RD&D goals.
- AEMFC development is critically important for advancing inexpensive fuel cells with completely PGM-free anodes and cathodes. The project under review fully supports the goals and objectives in this research area.
- The performance of AEMs continues to be of concern, but at least now NREL has demonstrated good performance is possible.
- The project team has developed new insight into mechanisms of performance degradation in AEMFCs.

Question 5: Proposed future work

This project was rated **3.5** for effective and logical planning.

- The project's future work involves continuing the development of better polymer structures based on learnings from the first two rounds of synthesis, integrating modeling analysis into component development, and further development of electrode assemblies. This is consistent with the overall project approach.
- The proposed future work identified continued integration of modeling efforts with cell testing as a key next step in developing this class of AEMFCs. The proposed work elucidating performance losses in the MEA will be beneficial.
- The team has a clear understanding of how to address remaining challenges and barriers through the proposed future work.
- Based on the figure summarizing the effects of AEM improvements versus electrode improvements, it can be concluded that the focus should be on the ink and electrode optimization.
- The project is nearing completion. The proposed work is appropriate for the time remaining in the project.
- While the modeling and the weaknesses of ionomers is well described, the path to improvement is not clear.

Project strengths:

- The project's strengths include its combined theoretical and experimental approach, its good use of collaborator strengths, and its identification of weaknesses in Gen 2 material and engineering of a solution in Gen 2+.

- The project's focus on perfluorinated membranes provides unique properties that other AEMFC projects lack, including the benefits of perfluorinated systems in water management.
- The project's strengths include its excellent analysis and expansion on understanding of AEM-based fuel cell performance and electrodes. The project has a strong team and collaborations.
- The PI demonstrated a significant breakthrough in AEMFC technology by successful development of the team's approach, passing critical milestones and go/no-go design points.
- The team understands the challenges with AEMs and has performed excellent work with 3M to develop these materials and test them in fuel cells. The team is very experienced and accomplished.
- This is a very strong project; it has a great team and great results.

Project weaknesses:

- AEM development is inherently challenging. From the data, it looks like a membrane reinforcement strategy would have been a plus, to integrate into the component development. It would have been beneficial to have seen some data with alternate (non-platinum) catalysts. Ultimately, testing should be standardized on CO_2-free air, versus hydrogen or oxygen. It is hard to compare the data with data seen outside this project in the commercial sphere. But these are minor issues; the fundamental project effort has been excellent.
- The project needs to do more experimental validation of predictions related to water management and CO_2. Membrane materials still demonstrate mechanical failure in KOH.
- AEM stability is still very low. AEM performance lags far behind that of PEMs, while PEMs improve. It is not certain that AEMs really make sense in PGM fuel cell applications.
- The project has made no effort to test non-PGM electrodes.

Recommendations for additions/deletions to project scope:

- From the data, it looks like a membrane reinforcement strategy would have been a plus, to integrate into the component development. It would have been beneficial to have seen some data with alternate (non-platinum) catalysts. Ultimately, testing should be standardized on CO_2-free air.
- A shift to electrolysis may make sense since the economic case may be better. Now that performance with Pt is proven, it is also recommended that the project incorporate PGM-free, at least on the cathode, to reduce Pt loading.
- The project should compare the performance of Gen 2 PF AEMFCs with PGM and non-PGM electrodes, using synthetic air (with no CO_2) as the oxidant.
- The project is nearing completion, and there are no recommendations for added scope. It may be beneficial to focus future AEM work on electrolyzer applications.

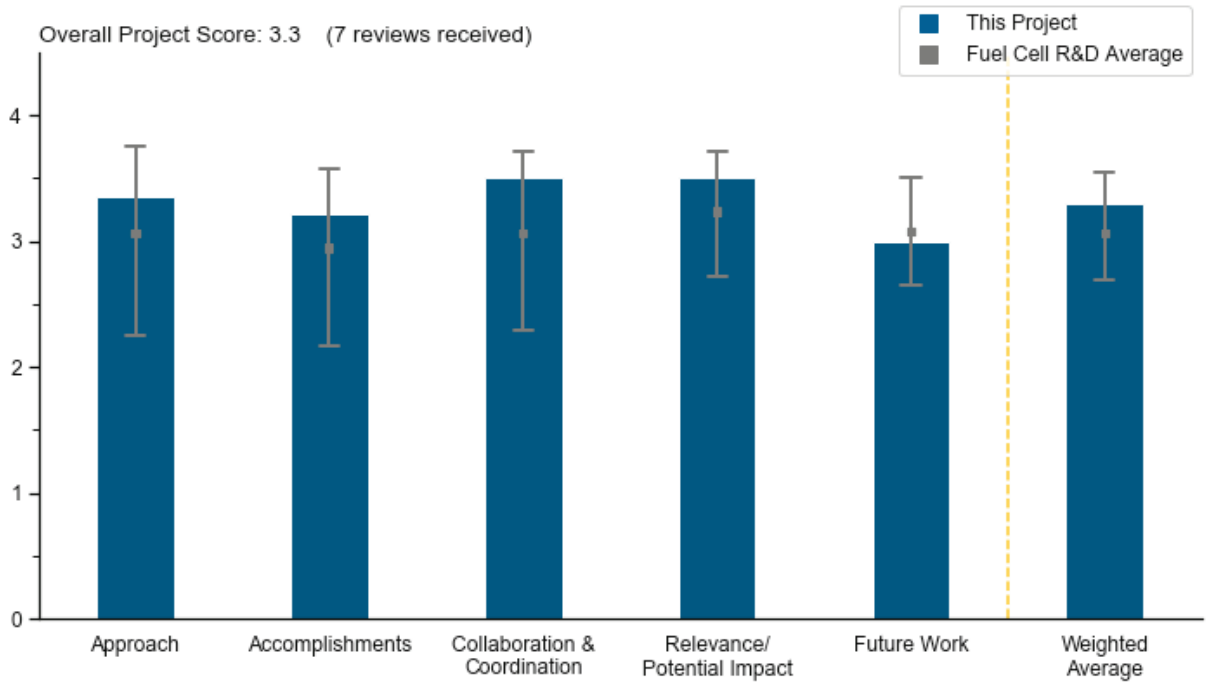
Project #FC-155: Novel Ionomers and Electrode Structures for Improved Polymer Electrolyte Membrane Fuel Cell Electrode Performance at Low-Platinum-Group-Metal Loadings

Andrew Haug, 3M

Brief Summary of Project

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

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **3.4** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The project scope addresses the cost, performance, durability, and robustness barriers as outlined in the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan (MYRDDP). Specifically, this project addresses the localized mass transport limitation of oxygen on the cathode side of the fuel cell. The project addresses this through the understanding and characterization of oxygen transport in ionomers and by designing new ionomers, integrating dispersed NSTF (dNSTF) into catalyst layers, and evaluating the benefit with respect to improving oxygen transport, while also maintaining performance and durability. The ionomer approach aims to improve ionomer conductivity and transport, allowing for a reduction in ionomer content, which could result in an overall reduction in local resistance. The project approach is in direct alignment with the MYRDDP and the goal of reducing mass transport limitation.
- It is good that 3M is taking a hard look at different ionomers. The researchers seem to have an open mind, and they have found that proton conduction, rather than oxygen transport resistance, is the limit. They use

their well-known NSTF catalyst for the work. The research deficit of the proposal is that the chemistry of the ionomers is not disclosed (IMIDE#1, etc.), so it is hard for the community to get a better understanding of the science behind the results. Overall, the project is making good progress toward U.S. Department of Energy goals.

- This approach is very insightful, especially coupled with the work General Motors is doing, as both are answering the oxygen transport problems at low loading from very different perspectives. The one thing that cannot be judged is whether this approach is cost-effective. The only mention of cost is when it is listed as one of the barriers. One of the great things about NSTF it that it was a roll-good item that could be directly transferred to the membrane. Now, there are multiple layers, higher loadings, and additional processes, all of which will significantly increase costs. It is also not clear how this helps solve the flooding problems generally seen with NSTF, but the catalyst seems to operate fine at high relative humidity.
- The approach is excellent; however, there is a concern about how the technology being developed here will be successfully utilized by the fuel cell community, since 3M recently announced that it is not going to make membrane electrode assemblies (MEAs). Since only 3M makes the NSTF catalyst, it is not clear how MEAs with NSTF are going to be produced. It should be clarified whether 3M will sell the NSTF catalyst as a product so that this technology might be usable.
- The approach addresses many of the elements of creating a catalyst layer, including the catalyst itself (dNSTF), oxygen transport through ionomer, and proton transport through ionomer. However, not much is mentioned in the slides about the carbon content of the layer or how water management might be addressed. The use of modeling eventually points to the higher coverage of whiskers with ionomer, but it is not clear whether this has led to better results in testing at higher current density. The approach emphasizes the improvement of kinetic parameters, such as mass activity and ECSA, to meet performance goals. The approach leaves considerable uncertainty as to how high-power targets are to be met, outside of the modifications to the ionomer.
- Electrode ionomer is an important area of work. However, the dNSTF was not well planned. It would have been better if the project had done some preliminary work prior to the start of the project. The risk and reward are quite low considering the relatively low mass activity, low electrochemical surface area (ECSA), and additional processing costs. It is unclear how productive it is for Michigan Technological University (MTU) to pursue modeling to investigate the root cause of the dNSTF problem, considering its non-uniform structure and performance perspective.
- Attempting to improve ionomer performance through increasing permeability and conductivity is worthwhile, as is investigating dNSTF in MEAs. The synergies of the two approaches are not clear.

Question 2: Accomplishments and progress

This project was rated **3.2** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has made excellent progress toward the project goal. The researchers have met all of their milestones and targets. All tests are done in accordance with procedures outlined in the MYRDDP. However, not all of the targets are met with one membrane–ionomer–catalyst system, and some testing or targets are done at 70°C or 80°C, so it would be nice to see consolidation in the future work. On the ionomer side, imide-based ionomers have shown through this project that oxygen and H+ transport can be improved. Multi-acid-side-chain (MASC)-based ionomers do not appear to be characterized or reported here. The Fuel Cell Consortium for Performance and Durability (FC-PAD) characterization indicates the reasoning behind that, namely the local structure of the polymers on Pt, orientation, and swelling. This should be extended to other imides and MASC ionomers. On the catalyst side, dispersing NSTF is a logical extension of the approach, and it circumvents issues with NSTF while resulting in a new catalyst layer structure enabled by dNSTF.
- There are impressive results with respect to both the development and demonstration of new ionomers in catalyst layers and new catalyst layers with dNSTF. It is good to see that dNSTF may be able to overcome some of the major issues with conventional NSTF (e.g., those listed on slide 3 of FC-143: operational robustness, contaminant sensitivity, and break-in conditioning). For many years, some have been recommending this approach; 3M's response used to be that dNSTF would not be cost-competitive. The team should clarify how or whether this concern has been addressed.

- The team has made remarkable progress, having reached almost every DOE 2020 target, in some cases by a wide margin. At the time, these were very aggressive targets. Similarly, the team has completed nearly all of the project objectives while providing new insights into transport and degradation mechanisms.
- The project has made very good progress toward DOE goals. As of February 2019, the team has achieved the DOE goals and seems to be on track to meet all of the targets.
- The project has had some encouraging ex situ results on ionomer development. However, it is late in the project, and the benefit has not been realized in fuel cell performance. There are some interesting fundamental analyses by partners and FC-PAD. It is not clear how the project will implement the learnings, as it is late in the project. Data quality is a concern; it is unclear whether there was any measurement replication. The sample description is lacking. It is not clear what data belong to which MEA configuration. The targets were not met with the same or a similar MEA configuration. It is not clear whether there were any positive outcomes or learnings from dNSTF. If not, this should stop. The MEA diagnostics did not translate to fuel cell performance.
- The project has had some positive results surrounding the ability to go to lower ionomer-to-carbon (I/C) ratios, to reduce the transport resistance at high current density, and to achieve reasonable performance and durability metrics with dNSTF. The project still has a number of challenges, including voltage loss at 0.8 A/cm^2 and questions regarding conductivity within the electrodes that need to be further elucidated. It was difficult to relate the results to tasks, as the task topics showed up only in the summary at the end of the talk. Clearer comparisons of single variable changes that are focused on just ionomer changes or just catalyst changes would have been beneficial in assessing the impact of different factors on performance and durability.
- The mass activities are still below project targets. It is not clear if the gram-per-kilowatt measurements are bounded by $Q/\Delta T$. Good progress has been shown on addressing robustness. Some imide ionomers may still be a problem for electrode conditioning. The researchers were able to understand that high-current performance may be limited by proton transport. They were able to realize transport gains with more carbon and less ionomer. Support-corrosion accelerated stress tests (ASTs) show that the losses are within target. However, higher current density losses following 0.6–0.95 V cycling point to a need to address agglomeration and slow proton transport.

Question 3: Collaboration and coordination

This project was rated **3.5** for its engagement with and coordination of project partners and interaction with other entities.

- 3M has done an excellent job leveraging its university and DOE collaborators. Combined with 3M's excellent test methodology (and a good gas diffusion medium), the results are very compelling.
- Nearly all of the collaborators made contributions, even though 3M certainly did most of the work. Argonne National Laboratory processed the electrode, while Lawrence Berkeley National Laboratory provided direction on which ionomers would lay flat on platinum. Tufts University (Tufts) evaluated electrode tortuosity and looked at the coverage of whiskers with CO-stripping measurements. MTU provided a model to support increasing ionomer coverage. The National Renewable Energy Laboratory measured mass transport resistances, while Los Alamos National Laboratory ran fuel cell tests that showed performance was limited by proton transport. The one collaborator whose role was difficult to find in this year's slides was Oak Ridge National Laboratory (ORNL). The presumption is that ORNL is called upon whenever images are needed. MTU's work supported what was found experimentally, but the polarization in the validation plot seems to reflect a shape different from the polarization that was measured.
- There are many project partners, including FC-PAD, MTU, and Tufts. It was great to see progress made by sub-partners this year, as highlighted by the project slides. Important contributions were made to project scope and the prime's capability to meet the targets. Regarding slide 10, it is not clear which ionomers Tufts analyzed by alternating current (AC)/direct current (DC) or what the conclusion is. The team should clarify with which perfluoro imide acid (PFIA) the I/C ratio was measured and why MTU performed water uptake at such high temperatures. More information should have been included here.
- The prime, 3M, appears to be getting many useful results from the project partners and FC-PAD.
- The team, including the FC-PAD members, has appropriate skills.

- The project lists nearly 50 individual contributors and 8 organizations; some of the capabilities seem to overlap, and from an organizational standpoint, it might be better and easier to stick to a tighter team. Having said that, there are strong contributions from all of the major players, though 3M and FC-PAD seem to be doing the lion's share.
- 3M does all of the development work. The project's partners and FC-PAD contribute only to fundamental analyses, and it is not clear whether or how much any of the learning was feedback to the materials development work.

Question 4: Relevance/potential impact

This project was rated **3.5** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The project goes directly to the heart of one of the biggest challenges facing automotive PEMFCs: the limit of low-loaded catalysts with air. The project has provided much insight, especially considering transport in these systems. It also offers real strategies with demonstrated improvements. Not much more can be asked from a project.
- The work being performed on this project clearly aligns with the needs expressed in the MYRDDP. The project approach to attack oxygen transport limitations using both ionomer and catalyst layer changes has the potential to meet the DOE targets in terms of performance and durability at low loading. Importantly, the project focuses on both ionomer and novel catalyst structures to understand the issues and provide mitigation strategies.
- The project supports many of the targets familiar to lowering cost and enhancing durability for proton exchange membrane fuel cells with low amounts of platinum. The project uses a catalyst that can be produced at relatively high commercial volume, although 3M's commitment to producing fuel cell electrodes at high volume is questionable. The impact of ionomers on conditioning will affect the utility of the material, which will also affect relevance.
- The team is working on the key issues to enabling MEAs with ultra-low platinum-group-metal (PGM) loadings. However, it is not clear how some of the learnings might be used by the fuel cell community if only 3M has the NSTF catalyst and 3M is not selling the NSTF catalyst or MEAs.
- Based on the reported results and performance, the ionomer development efforts may have more relevance. They have shown the ability to go to lower I/C ratios and decrease transport resistance, and they have the potential to be applied to a variety of different catalyst systems. The dNSTF approach is clearly interesting, but it needs further development and may not be able to achieve state-of-the-art performance.
- This project supports the DOE's goals well. The only problem is that, ironically, the project has no corporate transition partner, as 3M has minimized its fuel cell business. Perhaps 3M will sell one of these wonderful new ionomers and advertise how to make good catalyst-coated membranes with them.
- Electrode ionomer development is important, but dNSTF appears to be an unproductive choice. Many unnecessary efforts were put into topics with low return on investment (ROI).

Question 5: Proposed future work

This project was rated **3.0** for effective and logical planning.

- The project's Gantt chart, milestones, and future work plans are clearly presented and show the pathway to ultimately achieving project targets, as well as new understanding about this important issue.
- The future plan is good with respect to completing the technical milestones. However, it is not clear how this technology is going to be successfully handed off to others for use in future MEA product development.
- Most of the "key items" have to do with processing, toward which the researchers do not provide much insight. The project is coming to an end, and one of the focus items is to reach the voltage durability target, which is the only one they have not yet met; this is a good focus.
- The future work contains many important areas (e.g., proton conductivity and improving the ionomer coverage of the whiskers), but it is balanced more toward enhancing performance rather than durability. With the losses at higher current density, more of the future work should be directed toward durability. The

project end date is September 30, 2019, so it may be wise to cut new material developments from the future work.
- The project ends in September, so the researchers are just wrapping up. It would have been helpful to hear about a transition plan and about how well the ionomers work with non-NSTF catalysts.
- It is quite late in the project. Effort should be put into realizing benefits in fuel cell performance. Electrode ionomer development is important, but dNSTF appears to be an unproductive choice. Many unnecessary efforts were put toward low-ROI topics. Some interesting fundamental analyses were done by partners and FC-PAD. It is not clear how the project will implement the learnings, as it is late in the project.
- The proposed future work is reasonable, but it does not provide prioritization and was not discussed during the presentation. The slide included is a "laundry list" of relevant activities that does not provide clarity of focus or direction on most critical issues.

Project strengths:

- The project's greatest strength is the uniqueness of the NSTF approach. These catalysts are different and behave differently from any other catalyst systems that others are studying and provide a very different perspective. Any model that one comes up with has to fit both this approach and more traditional Pt/C approaches. The unique NSTF, coupled with a strong team, has led to the ability to meet or exceed almost all DOE milestones while keeping to the project goals.
- The project is using a catalyst that can be made at a high manufacturing volume using scalable and repeatable vacuum processes. The primary investigator is knowledgeable in manipulating catalyst, catalyst-layer, and ionomer parameters to create higher performance. The project has made good use of all collaborations in the national laboratory network.
- The project has a good approach. NSTF catalysts are a clearly differentiated catalyst architecture, and therefore it is great to see that 3M is (finally) taking a different approach to potentially addressing the major issues with NSTF catalysts. The new ionomers are great new materials for the fuel cell community.
- The project's strengths include its strong collaborations and its utilization of both sub-partners' and FC-PAD's capabilities and expertise. The project has an interesting and promising approach for both ionomers and catalysts/catalyst layers to achieve targets.
- The project's strengths include its advanced ionomer development, in which 3M is a world leader, and the NSTF catalysts that offer a unique platform of interest for advanced fuel cells.
- The project has very good and very credible results. The members of the large team are all contributing appropriately.
- The project's strengths include its ability to develop perflouro polymers.

Project weaknesses:

- The project should make more use of FC-PAD's capabilities to understand the role of ionomer structure on properties, the impact of ionomer in catalyst layer structure on performance, and durability at low loading.
- There are limited synergies between ionomer development and dNSTF approaches. The presentation included significant data, but it was difficult to distill differences between the results.
- Only 3M makes the catalyst, and it would be difficult to imagine it being made by other suppliers. Most stack manufacturers will likely continue to rely on dispersed powders unless something very convincing emerges from this project. There are still some remaining issues with conditioning that appear to relate to ionomer choice. This could be a significant barrier to implementation. The project still needs to show compelling power density and durability. Without these, the odds of implementation in a commercial fuel cell system could be very low.
- Cost analysis is not in the project or not reported. This is troublesome because it is difficult for an outside observer to tell if what the team is proposing is practical. Along those lines, adding Ir, the rarest of PGM catalysts, as a solution does not seem like a very viable pathway, but maybe it is so low an amount that it does not matter.
- The useful ionomers are not disclosed, and it is not clear that anyone could ever get them. It is also not clear whether the ionomers work with catalysts that are not NSTF.

- The project's collaborations are weak. It is unclear if having partners and FC-PAD was helpful or simply a distraction to 3M.
- The path forward, with respect to technology handoff, is not clear.

Recommendations for additions/deletions to project scope:

- 3M should provide the new ionomers to others so that other groups can see whether higher performance can be obtained using other conventional catalysts. This could greatly increase the interest in these new ionomer options. 3M should clearly communicate what the path forward is with the NSTF catalyst. NSTF seems to have great promise for electrolyzer applications. However, if 3M is not going to sell MEAs or NSTF, the team should consider how electrolyzer developers will utilize NSTF, as well as how fuel cell developers will potentially use NSTF catalysts.
- It is recommended that the team work on a transition plan to commercialize the ionomer and advertise the results. If 3M does not want to make the ionomers, perhaps 3M can license them to another company.
- The project is in its last six months, so there are no additions to suggest. Removing new material developments would be wise in the late stages of the project. The project should remove most of the activities that are not related to lowering conditioning time and enhancing power density and durability after 0.6–0.95 V cycling.
- There should be more tie-in of materials that meet component-level targets into AST and performance testing—for example, the high-oxygen-permeability ionomers in MEA testing. There should be more characterization of the catalyst layer produced from dNSTF regarding what it looks like and how it is unique or different from conventional catalyst layers.
- Clearer comparisons should be made between single variable changes, and the impacts on performance and durability for each of the systems should be studied.
- The project is wrapping up and is rightfully focusing on increasing durability.
- It is not clear if there were any positive outcomes or learnings gained from dNSTF. If not, the work should stop.

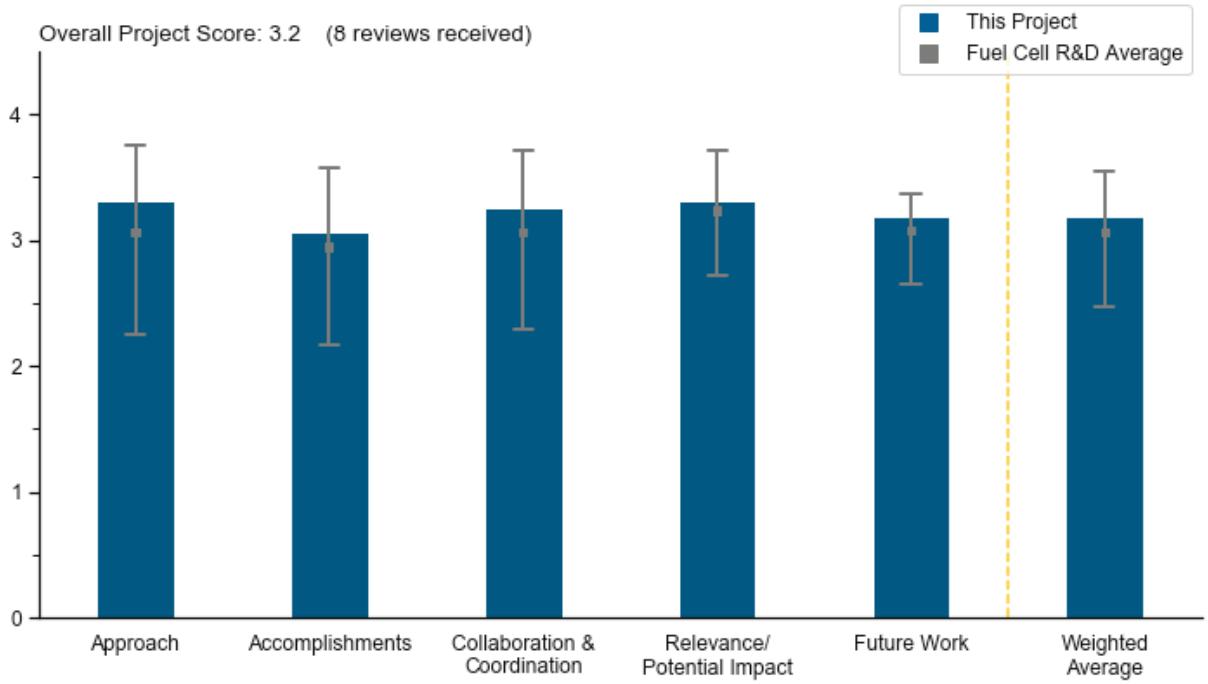
Project #FC-156: Durable High-Power Membrane Electrode Assemblies with Low Platinum Loading

Swami Kumaraguru, General Motors

Brief Summary of Project

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

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **3.3** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The proposed approach is clearly described and based on a multi-step, multi-factor improvement design matrix. The milestones and go/no-go decision points are adequately selected and allow for the estimation of progress being made toward Office of Efficiency and Renewable Energy final targets from the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan.
- The project strives to identify benign operating conditions that will enable enhanced MEA durability through the use of in situ and ex situ testing, analysis, and modeling. The overall project approach appears sufficient to meet the project objectives.
- The project has performed very comprehensive study under realistic operating conditions. Various voltage cyclings are performed to develop fundamental models of mechanical stresses for combined degradation. Tasks are very well distributed among the team members. The team is starting with SOA MEA, which eliminates many variables in the study and provides more confidence in the results.

- The approach is detailed and well suited to identifying the detrimental conditions to avoid in MEA operation. The use of two-factor analysis of the load cycling experiments can provide some very useful insight and should be pursued further.
- The project has a sound approach; particularly appreciated was the use of "design of experiments" to accelerate the testing campaign.
- The approach addresses the two major classes of failures: those caused by voltage cycling and those caused by either low or cycled hydration. The project includes variations in voltage cycles and deeply analyzes aspects of membrane degradation that are a tier below the operating condition stressors, such as cerium migration and hydrogen crossover. Interactions of voltage cycling and hydration with temperature are well addressed. One question that could be raised with the approach is whether all cerium-containing additives behave alike. It is unclear whether the results shown here would be repeated for any cerium additive package. Given that many milestones from budget period 2 still remain incomplete, there does arise a question of whether the project scope is too large. The voltage cycling runs could be completed, but it is uncertain that the membrane degradation modeling will be completed. There is also a model based on Pt and Co dissolution that needs to be completed. The Pt dissolution model does not have an equation for conversion of PtO to Pt2+.
- The voltage cycling of significant run variations may end up providing valuable results, but at this point in the project, not all data appear to make sense. This is an extensive and methodical way to generate the test data, but improved understanding on repeatability and error-checking (or at least insight into this) would be appreciated in order to ensure that the data will be meaningful when the testing is done. As mentioned last year, the use of only one material set does raise concerns that relationships may change. No indication of the material set was provided. This should be provided in at least general terms. It is recommended that the team run tests on air and hydrogen to more closely mimic actual operation, as there will be changes in the catalyst layer's local conditions and some effect on mechanisms. These are not expected to be significant enough to invalidate the approach, but this will need to be checked.
- The project's strengths revolve around the SOA materials, performance, and durability of the systems being investigated. The approach itself seems strongest in budget period 1, with decreasing clarity and impact in subsequent budget periods. The meaning of the task numbers given on accomplishment slides is not clear, and those tasks do not connect to budget period task numbers. All the areas of interest at least have scientific relevance, but most are not directly tied to overcoming barriers and will instead help provide fundamental understanding.

Question 2: Accomplishments and progress

This project was rated **3.1** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Some important and practical results have come from this project: the realization that mechanical stresses accelerate chemical degradation (even at the cell outlet), the understanding that convection is the primary transport mechanism for cerium cations, and the understanding that cerium content affects the uniformity (or lack thereof) of membrane-thinning over the cell. The trends of electrochemical surface area (ECSA) loss and Co migration with higher relative humidity (RH) cycling were probably understood in the community prior to the work done by this project. Conclusions are still needed from the voltage-cycling experimental design. The project is still having difficulty understanding operating condition effects on gas crossover since the crossover rate appears to be independent of initial cell shorting.
- The go/no-go result of the effect of RH is not a new result. Previous DOE projects have demonstrated the strong effect of RH on cathode degradation. The project has interesting results on Co distribution in the MEA cross-section. In regard to the membrane highly accelerated stress test (HAST) work, the combination of accelerated testing with the diffusive crossover maps and Ce X-ray fluorescence (XRF) maps is very interesting and is providing useful insights. The follow-up work on Ce transport modeling and understanding mechanical stress impacts on chemical degradation is also valuable. For the membrane thickness effect on chemical degradation, the effect of the open circuit voltage (OCV) value on the membrane degradation rate should be considered. The OCV value will vary with membrane thickness. Further understanding of the local shorting effects is valuable.

- Very good progress has been made so far in studying various H_2–N_2 cycling to understand the effect of operating conditions. The effect of RH was studied extensively, with detailed analysis of losses. Significant progress has been made on model development and HAST testing.
- The project's progress has been strong in meeting DOE goals. Performance and durability have been demonstrated largely at the SOA level and at the level of DOE targets.
- The competition of main milestones and go/no-go points is on excellent track. The improvements made in the activity and durability of novel MEAs are significant.
- The team has made good progress on assessing the impacts of different MEA operating conditions on performance degradation. The demonstration of the impact of RH on Co dissolution, alloy catalyst performance, and ECSA loss is a very useful result.
- After two years, it appears the project may be behind from the perspective of milestones and total funds spent, assuming annual budget periods and go/no-go points. The membrane and Ce-additive-specific work appears to have made very good progress and is on track, but the H_2–N_2 and hydrogen–air voltage cycling appears behind, and the review slides on the model appeared primarily to provide the framework, with relatively few results and conclusions.
- The team is on track to deliver project goals.

Question 3: Collaboration and coordination

This project was rated **3.3** for its engagement with and coordination of project partners and interaction with other entities.

- Collaboration with the Fuel Cell Consortium for Performance and Durability (FC-PAD) is well established, although this is a General Motors (GM) project for the most part. The National Renewable Energy Laboratory is engaged on cell testing, and Argonne National Laboratory is included on degradation modeling. The collaboration with Giner, Inc., appears to have yielded results that speak to the primary mechanism for cerium transport (convective). The collaboration with the University of Texas at Austin (UT Austin) has provided the results showing greater cobalt movement to the membrane with higher RH cycles.
- This project significantly benefited from close collaboration with FC-PAD, which allowed the team to demonstrate steady progress toward final project goals.
- The work distribution across partner organizations appears reasonably coordinated and appropriate for each organization's capabilities.
- The structure with FC-PAD and subcontractors leads to good collaboration.
- The team has made good use of outside resources.
- Although collaboration with project partners was mentioned, the presentation did not highlight the specific role played by each of the partners. The team is encouraged to prioritize the information so that the partners' work becomes more evident.
- The collaboration group is limited, but GM's breadth makes this less necessary. There are clear roles for the sub-tier partners but limited capability for bringing in outside strength beyond FC-PAD participation.
- The contribution of the UT Austin is not very clear. GM has done most of the work. The contribution of Giner, Inc., is evident from the results. FC-PAD's collaboration also seems less substantial.

Question 4: Relevance/potential impact

This project was rated **3.3** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- Operational durability is of critical importance for polymer electrolyte membrane fuel cell integration. Activity and durability, especially for catalytic materials, are often inversely proportional. Improving the durability of MEA performance through the optimization of operational conditions has a higher chance of immediate success than an approach focused on the development of new materials.
- There is no question the project is highly relevant. At this stage in fuel cell development, material sets exist that can enable a degree of commercialization, as evidenced by vehicles that are actually on the road. However, much of the scientific phenomena that would enhance system mitigations, and therefore system durability, still remains unexplored. This requires a low-technology-readiness-level (low-TRL) kind of

effort to enhance high-TRL applications. This project attempts to do exactly that by probing the phenomena that got "left behind" as developers were racing to produce vehicles. The only question regarding relevance might be with respect to how well the project can expect to complete its goals before time expires. There is still much work to do, and the scope is very large. Although the project does focus on one particular material set, the material set is adequately representative of what the community is using.

- The combination of high power density and good durability with low Pt loading is clearly one of the main challenges that are left to be solved in reducing fuel cell cost and enabling the industry to grow. This work addresses the challenge directly.
- The results have been promising and strong when considering cell performance and durability. Other efforts, such as modeling studies and Ce migration, are relevant but do not provide the same value to the project as MEA optimization.
- The project team is aiming to achieve DOE's performance and durability targets. With GM being the principal investigator (PI), most of the tests are performed under realistic operating conditions and, most importantly, using SOA MEAs.
- The project is critically important for the DOE mission toward development of inexpensive and robust MEAs with low-platinum-group-metal content.
- The project is directly addressing MEA durability, which is one of the most significant barriers for the widespread deployment of fuel cell technology. The overall approach should provide significant benefit to the field. However, one concern is that the SOA MEA components are not disclosed, and as such, the results generated will be only indirectly useful. Without knowledge of the components, other research groups wishing to extend the results generated in this project will likely need to generate their own datasets with their own materials.
- The work is highly relevant. There is no doubt that much of the work will be of high value. However, the overall potential impact is difficult to assess at this point in the project.

Question 5: Proposed future work

This project was rated **3.2** for effective and logical planning.

- The proposed future work is in line with what is necessary to successfully complete the work. The focus should be on measuring the true intrinsic dissolution rates of Pt and Co at realistic conditions, such as the presence of Nafion™ and the effect of RH. The project team may want to look at different carbons, as they could affect metal dissolution and ECSA loss.
- The project still has an overwhelming amount to accomplish. The voltage cycling experiments need to be mapped, the dissolution rates of many species need to be measured, and a model needs to be made for membrane degradation, among many other things. The future work certainly acknowledges all of this. The future work mentions a durability improvement in the accelerated stress test. It would seem enough for this project to clarify the relationships between stressors and resulting material failures. Durability improvements would seem more the domain of actual developers in vehicle products, especially now that the technology status is based on vehicles.
- The planning of future work is logical and based on previously reported progress, including several risk-mitigation scenarios.
- The proposed future work is logical and aligned with the project plan.
- The scope of the work is appropriate but does little to suggest significant additional advances beyond those already demonstrated within the project; it is not certain these are even included in scope. The focus of the future work is on developing models and improving fundamental understanding, but these seem to be topics of a more academic focus rather than ones that advance the current SOA.
- The proposed future work seems reasonable for achieving the project goals.
- The future work seems to center around a new simulation model. The scope of the model is not entirely clear. It is recommended that the project team make the model validation plan and strategy very clear for the next meeting.
- The project team should provide more explicit data on the test station variability, as well as which DOE runs will be repeated and how many times.

Project strengths:

- GM's vast experience with MEAs and their degradation is the biggest strength of this project. The team is well established, too. Understanding combined degradation in order to help develop fundamental models that can predict the degradation is a really good approach. The team is looking into various H_2–N_2 cyclings instead of just using the established durability protocols.
- The work is relevant and is applied to high-priority degradation mechanisms. The HAST membrane work is very valuable. Although not shown in the presentation, the understanding of the oxygen transport mechanisms as the electrodes degrade likely will be applied and of value. The dataset is being generated on an MEA that meets DOE Pt-loading targets.
- The project is led by a vehicle manufacturer keenly aware of the relationships between operating stressors and material failures. This manufacturer understands that mitigations can be generated in system operation to prevent material failures. Not all developments have to come from improving materials. The project is well connected to the national laboratory network for analysis and modeling. The project has appropriately divided failure modes into two categories: those that arise from voltage cycling and those related to membrane failures.
- The PI demonstrated deep understanding of degradation processes in catalysts and catalyst layers, derived from a quite complicated design of experiment. The obtained knowledge will facilitate the completion of milestones and the final project goal.
- The project's strengths include detailed analysis of intrinsic mechanisms of instability in MEA performance, two-factor analysis of MEA operating conditions, and effort centered on finding the true dissolution rates at relevant conditions.
- The key project strength is in the project approach: utilizing in situ and ex situ testing, advanced characterization, and modeling.
- The project's strengths include the materials used, as well as the performance and durability demonstrated.
- The experimental approach is sound.

Project weaknesses:

- The project workload is extreme, which has resulted in a heroic list of tasks in the future work section. The project sometimes finds relationships that were already understood in prior work (e.g., ECSA losses increase following cycling at higher RH). The final product of the project may prove difficult for other developers to use in designing system mitigations.
- The project's weaknesses include its design-of-experiment data-generation format, which makes it difficult to assess emerging trends. The project is also specific to only one material set, and no information is provided on the material set being used. The team indicates that these are being provided with no restrictions on analysis, so more information could be provided in the DOE Hydrogen and Fuel Cells Program Annual Merit Review (AMR) material.
- It is recommended that the next AMR presentation be more focused on covering collaborations and high-level project takeaways.
- The primary project weakness is that it is using proprietary materials that will not be disclosed, limiting the direct usefulness to the field.
- It does not seem like the future work offers much opportunity for further advancing the performance and durability of fuel cells.
- The collaboration and contribution of other team members and FC-PAD are not very clear.

Recommendations for additions/deletions to project scope:

- Once the project has made sufficient progress in establishing the feasibility of the overall project approach, the team should consider assessing other catalysts and MEAs comprising disclosable material sets; this should be done earlier in the project rather than at the end of the project, as stated on slide 27.
- The project team should provide information on the material set used, test some variations on the material set once the degradation model has been developed, and include air–H_2 testing.

- The project team should look at different carbons—for example, high surface-area carbon versus Vulcan®—as porosity can affect particle sintering and dissolution.
- No additions are proposed. Any work associated with improving durability should be eliminated. This project should be designed for understanding the relationship of operating stressors and material failures. If repeated tests to observe crossover trends with local shorting do not show trends, then this part of the project should be eliminated.
- Much of the modeling work and fundamental studies seem of limited value. It is not clear what else could be done to further advance performance and durability, but approaches along these lines would provide more value.

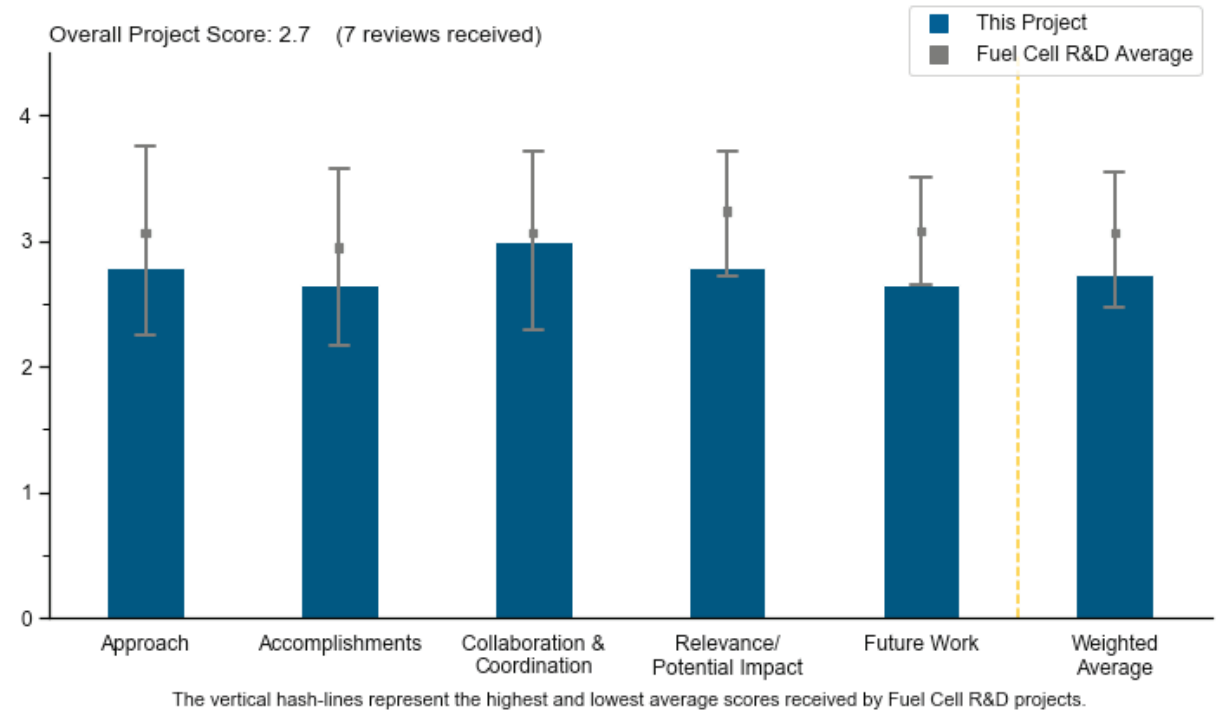
Project #FC-157: High-Performance Polymer Electrolyte Fuel Cell Electrode Structures

Mike Perry, United Technologies Research Center

Brief Summary of Project

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

Project Scoring



Question 1: Approach to performing the work

This project was rated **2.8** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The project scope is not clearly aligned with the barriers in the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan (MYRDDP); however, the project essentially addresses performance, cost, and durability, as inferred from other slides. Specifically, this project addresses the localized mass transport limitation of oxygen on the cathode side of the fuel cell, using modeling to inform catalyst layer development. The project approach is in direct alignment with MYRDDP targets of reducing mass transport limitation in low-catalyst-loaded electrodes and under low-humidity operation. This is accomplished through the understanding of transport phenomena. The approach of developing a detailed, geometric, microstructural model of the catalyst layer is a novel pathway and, compared to other projects, the focus on understanding is key. The project is validating models with real-world MEAs.

- The project's approach is to develop detailed geometric models of cathode catalyst layer (CCL) microstructures. The team will project and close the performance gap by using the model to design and build optimum electrode structures.
- As a model for addressing gas-phase transport losses, the project does well in determining which models consistently agree with experimental results observed for changes in platinum loading, ionomer-to-carbon ratio, and platinum mass fraction. The project then goes on to apply the appropriate models. Much of what the project has studied can be applied from an in-depth understanding of recent literature. However, it is not entirely obvious how the model is addressing condensed phases. There is some mention of how effectiveness factors and Thiele moduli change with water filling, but how water filling arises is not shown, and neither is how hydration of the catalyst layer and membrane is determined. The nanocolumnar catalyst is interesting, but there are many details missing, e.g., sputtering costs, reproducibility, batch size projections, and other information.
- This project uses a good mix of computational model development and validation with outside MEA data and their own. The model results have been used to make MEAs with lower transport resistances. It appears that this was brought up by reviewers last year, but it is hard to see the relevance or utility of the thin-film catalysts. The project would be more suited if it focused on carbon-supported, nanoparticle-based catalysts.
- The recently published hierarchal model helps with understanding the transport phenomena in the catalyst layer. The model advises that smaller agglomerates can improve performance. However, it is unclear how the team plans to control the agglomerate size.
- This project attempts to create a somewhat universal model to explain fuel cell behavior. The 2018 paper by Darling is an excellent paper, but it has some limitations, mainly due to its use of 21 variables. While some of the variables are easy to define—such as temperature, pressure, etc.—some are harder to define, such as ionomer-film thickness. The model seems to go in with the assumption that oxygen resistance is the most important factor, and then the model proceeds to prove its theory. While the model accounts for the density of primary particles and porosity, it does not account for pore size distribution. There have been some papers out showing that the pore size distribution of the catalyst layer is very important. Eikerling also emphasized porosity in his model. The authors do not try to match their model to a broad number of catalyst-coated membranes (CCMs). Another complexity is the gas diffusion media (GDM), which will clearly change the results. They use a United Technologies Corporation (UTC) gas management device, but no information is given. Without some more validation, it is hard to know whether the model is really useful beyond the researchers' laboratory.
- This project was not set up for success. The project promised to achieve DOE targets by first understanding electrode transport loss, then validating it, and then fixing it. However, the approaches for understanding the phenomenon are essentially the same as what others have done in the last nine years. Unsurprisingly, no particular learning was made. No specific plan for model validation was laid out. Therefore, this project will likely be a repeat of what others have already done. The project lead does not have access to the resources or expertise needed to prepare electrodes or MEAs to validate the model or to pursue the development of materials to achieve DOE targets.

Question 2: Accomplishments and progress

This project was rated **2.6** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project was delayed, and a no-cost extension is being implemented. In the meantime, the project has made progress toward the project goal. The team has met most of the project milestones and targets and is on track for others. The team should include dates with milestones, as its unclear how the no-cost extension shifts out the quarters. For example, it is unclear whether Quarter 6 (Q6) is delayed or on track, or when the go/no-go is. All tests are done in accordance with procedures outlined in the MYRDDP. There is a clearly outlined path for meeting the Q8 target.
- The team has made good progress in model development at different length scales. However, this has not yet translated into MEAs that meet the target metrics.
- The team made progress toward the project goal, but it is not clear if they asked the right question in the first place. They went into the study saying that oxygen transport resistance was the key parameter, and

they then proved it. A later talk by 3M showed that proton resistance in the ionomer has a big impact. Pore size distribution cannot be ignored.

- The recently published hierarchal model may oversimplify the diffusion in the catalyst layer. The model does consider the porosity and tortuosity to some extent (by dividing the bulk diffusivity by 4, it seems), but a catalyst layer with a similar porosity could have very different tortuosity and diffusivity. Maybe the incorporation with Lawrence Berkeley National Laboratory's (LBNL's) full-scale MEA model can better handle the mass transfer in the electrode.
- The model has been incorporated into the LBNL two-dimensional (2D) MEA simulation, but the modeling results shown depict radically low limiting currents that vary with particle size, which is hardly the reality with realistic fuel cells. For small cells with gas-phase transport, the model appears to predict performance losses well, accounting for three levels of oxygen resistance (nanoscale, agglomerate, and catalyst layer) as well as ohmic losses. The nanocolumnar catalyst still has low mass activity, which continues to raise questions as to whether this catalyst is a worthwhile part of the project. Substantial cell resistance needs to be removed, and a PtCo catalyst needs to be used to meet the Q8 targets.
- The project has not generated new experimental data to validate and test the critical parts of the model. The modeling approach has been established; its impact on the development of an optimum electrode structure is not clear. The statement that agglomerates smaller than 150 µm are needed is not particularly innovative. The suggested improvements (e.g., Pt-alloy catalyst, General Motors [GM] cell, thinner membrane, 950 equivalent weight) have no relevance to the optimized structures being generated through model predictions. GM and others have already reported results using these MEA features. The Q6 milestone (1 W/cm^2) has not yet been reached.
- The project is delayed. No new progress to the field has been made.

Question 3: Collaboration and coordination

This project was rated **3.0** for its engagement with and coordination of project partners and interaction with other entities.

- The United Technologies Research Center (UTRC) team already has extensive collaborations with Ion Power, Inc. (Ion Power), the University of Arkansas at Little Rock (UALR), and several national laboratories, and the team is planning to work with Vanderbilt University (VU) and Los Alamos National Laboratory (LANL) on alternative CCL architecture.
- The team has made good use of partners and resources, including the Fuel Cell Consortium for Performance and Durability's (FC-PAD's) resources.
- The team expanded its collaboration with other members of FC-PAD.
- The project's sub-collaborations and FC-PAD collaborations are clearly defined, and a clear strategy exists for the team to work together to accomplish project goals. The project is leaning on the strengths of its FC-PAD partners. The UALR structures are interesting, but they appear more like a side project to obtain a different structure for the model as opposed to a direct effort in utilizing those structures in catalyst layers.
- The project's collaborations outside of FC-PAD include Ion Power, to deliver custom sub-MEA or MEA components, and UALR, to develop the nanocolumnar catalysts. Both have contributed to results, although the results need to be improved substantially to meet future targets. The model has been delivered to FC-PAD, particularly to the LBNL's 2D MEA model. Oak Ridge National Laboratory has assisted with transmission electron microscopy (TEM) characterization of the UALR catalysts. The project's collaborations could have been considerably enhanced by an automotive presence to guide the project toward the true modeling issues in the development of fuel cell systems.
- There is good collaboration with UALR and national laboratories. The contribution from LBNL's modeling work was not clearly presented.
- The project relies heavily on Ion Power to provide "innovative" electrodes for model validation. No progress has been made.

Question 4: Relevance/potential impact

This project was rated **2.8** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the MYRDDP.

- Detailed transport models at different length scales will have a significant impact on MEA architecture development and identify areas for potential improvement.
- The work being performed on this project clearly aligns with the needs expressed in the MYRDDP. The unique approach to developing and testing models to overcome oxygen transport limitations is relevant and has potentially very high impact in the field. What is less clear is the benefit of UALR's approach and its impacts.
- Most of the project aspects align with the DOE Hydrogen and Fuel Cells Program and DOE research, development, and demonstration objectives. It is unclear whether the durability test is a part of the project's tasks.
- Both LANL and UTRC have tested UALR's thin-film-catalyst-based MEAs, but the power density has not reached the 1 W/cm^2 target. The hierarchal model (with many assumptions) has been validated with two old datasets available in literature. On the basis of progress to date, the future contributions to novel catalyst and electrode development may be limited.
- Although understanding performance loss in low-Pt electrodes is crucial, model validation is difficult, and modeling alone cannot fix the problem.
- The project may not be able to deliver any true assistance to developers. Nearly all fuel cell developers have already found some way to treat oxygen transport resistances that is reasonably functional. The project does not take great lengths to address the biggest hurdles to development, which all have something to do with how hydration is managed in a cell. The balance between drying and wetting will affect how systems are designed. As the emphasis turns more toward medium-duty (MD) and heavy-duty (HD) trucks, a project focused on new catalysts and gas-phase transport may not be relevant for higher-platinum-loading systems that need high durability.
- It is not clear how this model could be of general use to help improve CCMs. If there is a belief of high oxygen resistance, the model does not explain how to fix it, nor does it explain the idea structure for low oxygen resistance. Agglomerate size is recognized as an important parameter, but it is not clear how this should be changed.

Question 5: Proposed future work

This project was rated **2.6** for effective and logical planning.

- Adapting and applying the developed model to others working on alternative CCL architectures adds to the impact of this project. It would be helpful to provide more details as to how the team plans to use modeling insight to make MEAs that meet DOE targets.
- Considering the state of the project, the future work needs to be more specific. There are remaining questions, including who will provide materials for model validation, what the metrics are, and what new approach in modeling is to be attempted. All of the noted topics have been heavily studied by others over the years.
- The future work reflects where this project needs to go. It would be nice to see more clearly the timelines and the revised dates based on the no-cost extension.
- Hopefully, the team can go into next year's work with an open mind and not just attribute results to oxygen resistance.
- The project will continue to improve the model based on nanocolumnar catalysts and gas-phase transport, and attempt to achieve a better match between model and experiment. It is questionable whether this will result in a real impact on developers. The work done on alternative CCL structures may be difficult to achieve in the time remaining for the project. It may be good to remove the thin-film catalysts from the project since these catalysts may still have problems with mass activity, batch size, cost of production, etc.
- The proposed plan will overcome some research barriers. It is unclear what improvement is expected on the models. Using nano computed tomography (nano-CT) at Argonne National Laboratory may not be able to provide too much help for this project because of the limited spatial resolution of the nano-CT (~25 nm). The approach mentioned in Padgett et al. seems more feasible ("Connecting Fuel Cell Catalyst

Nanostructure and Accessibility Using Quantitative Cryo-STEM Tomography," *Journal of The Electrochemical Society* 165, no. 3 [2018]: F173-180. doi:10.1149/2.0541803jes). The alternative CCL architectures (collaborating with LANL and VU) are not clearly presented.
- The proposed directions lack specificity. There are no clear-cut approaches or recommendations for developing a deeper understanding of the electrode structure or multiphase, multiscale transport processes.

Project strengths:

- The project participants are knowledgeable regarding all previous fuel cell models that address oxygen transport resistances. The models being used to address oxygen transport resistances are consistent with experimental results. FC-PAD and Ion Power are able to deliver results such as TEM, MEA components, and higher-level models.
- The project's strengths include the multi-length scale model that has been validated with multiple data sources and the application of the model to a range of CCL architectures.
- The project's strengths include its three journal publications. With FC-PAD support, the team is able to develop models, characterize microstructures, and measure cell performance.
- The project's focus on modeling first is unique and has high potential impact to help design electrodes and structures.
- The project has strong collaborations with industry, universities, and national laboratories. The multiscale models provide guidance on catalyst design and optimization.
- The project's attempt to build a physical model of a CCM is a strength.

Project weaknesses:

- The model seems to have a predetermined answer: oxygen transport resistance is most important. The project would do better to really push on the results with advanced GDM and ionomers and to try to look at the effects of pore size distribution.
- The project may not be in tune with the existing needs of the fuel cell development community. Some parts of the project feel like a catalyst development led by mathematical modelers. This is probably not the ideal means by which new catalysts are developed. The project has yet to demonstrate target performances at the MEA or catalyst level.
- There have been no novel contributions from the model to illustrate pathways for developing high-performance polymer electrolyte fuel cell electrode structures.
- The UALR structures are similar to nanostructured thin-film catalysts and do not add significant value here.
- The plan is not clear or specific. There is no momentum in the project.
- It is unclear how the thin-film catalysts developed here will have any sort of impact.

Recommendations for additions/deletions to project scope:

- The authors should check the literature again and read the work of Eikerling with an open mind. They might also look at results from state-of-the-art CCMs from companies such as W.L. Gore. The team should refer to "Manufacturing of Low Cost, Durable Membrane Electrode Assemblies Engineered for Rapid Conditioning" (report number: DOE-GORE-18052). The team should also look at the Freudenberg GDM and the effects of relative humidity to help elucidate the effects of water management.
- The project should delete the catalyst development and add a focus on hydration and condensed phases. If there was some way to begin addressing durability, this would be better for the community. This appears to be outside of the scope of this project, but the goals associated with MD and HD trucks (e.g., 25,000-hour operation) appear to make durability a more pressing need than understanding oxygen resistances at low platinum loadings.
- The project should pursue a pathway to integrating the UALR structures into MEAs and consider the potential to match or contribute to models.
- UTRC should consider another round of brainstorming ideas.
- The project should stop the work on the thin-film catalysts.
- It is recommended that the project be discontinued.

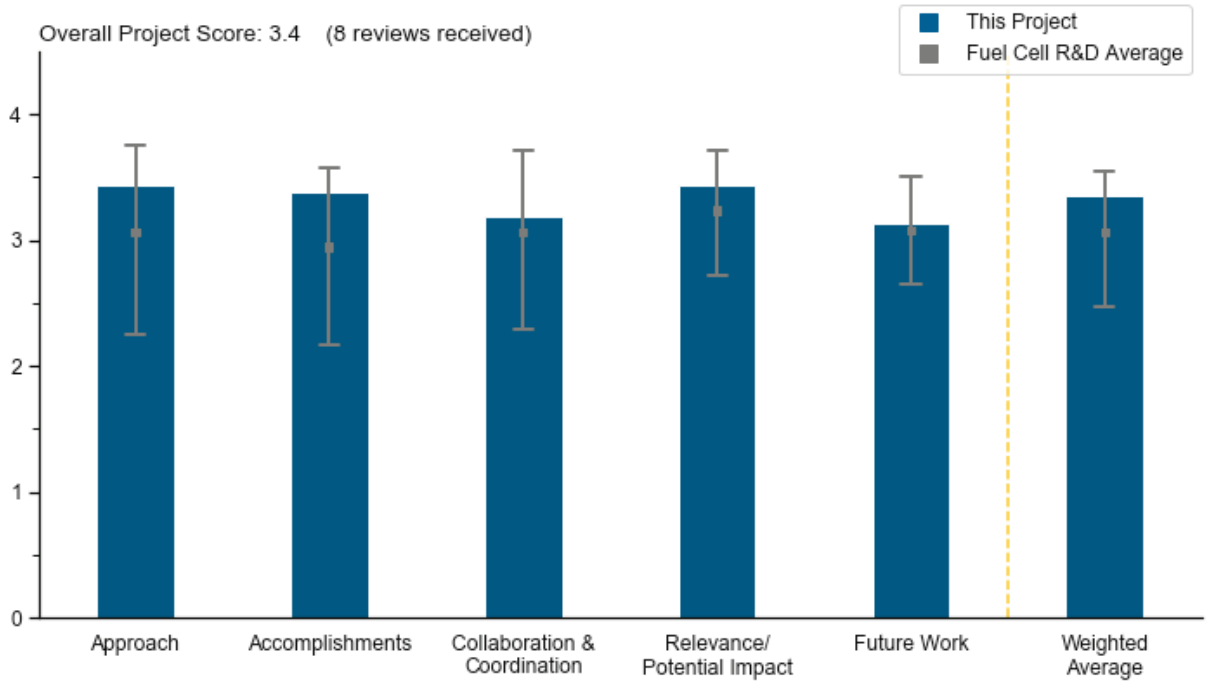
Project #FC-158: Fuel Cell Membrane Electrode Assemblies with Ultralow-Platinum Nanofiber Electrodes

Peter Pintauro, Vanderbilt University

Brief Summary of Project

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

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **3.4** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The objectives and barriers are well identified. The key issues of improving performance with low catalyst loading and at low relative humidity (RH) are being explicitly addressed. The proposed electrode mats will be easy to integrate with existing polymer electrolyte membrane (PEM) systems.
- This was a very nice presentation, and the project is on track to make some innovative catalyst-coated membranes (CCMs). The fibers seem to have high performance. While the fabrication approach is very good, the test conditions are questionable. One major problem with the work is the very high flow rates that are used in the test conditions for the CCMs. The project uses 80°C, 200 kPa (abs), 125 sccm H_2, and 500 sccm air for 5 cm^2 cells, while 4000 sccm H_2 and 8000 sccm air are used with all MEAs for cells with an active area of 10 cm^2. It is hard to understand the results under these very unconventional flow rates. The authors should use standard stoic rates of 1, maybe to a maximum of 2. The flow rate should vary with the

current. The Sigracet-29-BC gas diffusion layers are also obsolete. Running at 100% RH is also questionable.

- The project scope is not clearly aligned with the barriers in the Fuel Cell Technologies Office (FCTO) Multi-Year Research, Development, and Demonstration Plan (MYRDDP); however, the project essentially addresses performance, cost, and durability, as inferred from other slides. Specifically, this project addresses the localized mass transport limitation of oxygen on the cathode side of the fuel cell through the use of electrospun, fiber-based catalyst layer structures. The project approach is in direct alignment with the MYRDDP targets of reducing mass transport limitation in low catalyst-loaded electrodes and under low-humidity operation. This is accomplished through the understanding of structure–function relationships in the electrospun electrodes. The project team is also developing functionalized fibers for the electrospinning process that would help produce catalyst layer structures to improve mass transport. This should be compared to conventional electrodes made with the same polymers.
- This is a unique approach to generating catalyst electrodes. It is very flexible but also has the potential to be very insightful because of the different structures it creates. Economically, it is very hard to determine whether this is a valid approach. Peter Pintauro has spoken toward this in the past, stating that the approach will be valid if these can be made at low cost, but it bears repeating, especially if there is more evidence or if the team's thinking has matured. This is especially relevant as the larger-scale electrodes looked and behaved differently.
- This is a PEM fuel cell technology project looking at MEA fabrication using an electrospinning approach. The project has a large interactive component, and the contributions of various laboratories, showing interactions and the utilization of Fuel Cell Consortium for Performance and Durability (FC-PAD) laboratories. The electrospun electrodes are expected to improve performance at low RH conditions.
- The project's approach is novel and has impressive results in terms of power density.
- The project can still benefit from further systematic variation of compositions and process parameters, combined with FC-PAD's diagnostic and characterization tools, to correlate effects and enhance understanding of the underlying physics of the processes.
- The technical approach of the project is highly relevant to the FCTO mission. However, the focus seems to be on achieving only high activity and not high durability. It is not clear how the team plans to address durability requirements.

Question 2: Accomplishments and progress

This project was rated **3.4** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The authors have made excellent progress since last year. They seem to be on track to have a high-performance CCM. It is difficult to determine the true performance because of the unusual test conditions. The imaging of water was a really interesting experiment.
- The project's accomplishments are excellent, with good demonstration of power density and improvements in durability.
- The project has made progress toward the project goal. The team has met most of the project milestones and targets and is on track for others. The drive-cycle durability target was a miss, and the team should outline the path to resolve this. The milestone table makes it difficult to interpret when milestones are due; the past milestones over the course of the year leading up to the DOE Hydrogen and Fuel Cells Program (the Program) Annual Merit Review (AMR) are unclear. All tests are done in accordance with procedures outlined in the MYRDDP, although not all targets are met with one membrane, ionomer, or catalyst system. The Generation 2 (Gen 2) fibers using electrospinning show promise to achieve targets. The project is missing a clear understanding of the effects of the structure. There seem to be benefits such as in Co retention and low-humidity operation.
- There are outstanding results on the MEA performance side. The high performance at low RH shows that the approach works. The new materials are maintaining performance between high and low RH at high current densities. High performance is accompanied by good durability. The performance-versus-humidity data are missing comparison to typical baseline material at benchmark conditions. The measurements were shown at 80°C, 40% and 100% RH, and 200 kPa. Performance data are typically benchmarked to 80°C, 100% RH, and 150 kPa.

- There is good repeated performance and results on the performance, especially with the Co retention, though it is unclear why these electrodes should be better in this regard; this is certainly an area for further study. The project team should compare their results to the benchmarks more often or show the DOE targets in the presentation charts, which would make judging much easier. There are moderate results on the scaling portion, but undoubtedly, when the project team tunes the parameters, this will be achieved. The cost target and progress would have been good to see as well.
- There are interesting results at very low RH (40%), combined with neutron radiography results, which suggests water retention. The project still needs further development and progress to be state of the art at nominal RH.
- The targets relating to power output and durability have been met.
- The ionomer-to-carbon (I/C) ratios of the Gen 1 and Gen 2 MEAs are based on energy-dispersive X-ray spectroscopy (EDX) analysis, which has limitations on depth of penetration. The I/C ratios that were targeted for making MEAs are also not very clear.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- The project team has made good use of FC-PAD, using many of the different characterization tools at the consortium's disposal. The team has Lawrence Berkeley National Laboratory down for "possible modeling of the electrodes." This should be engaged, as there is not a good understanding of why these work better. It is nice that the team has an industrial spinner to make sure that the process is scalable.
- The project is starting to see good results from collaborating with FC-PAD (e.g., imaging results from Oak Ridge National Laboratory [ORNL] and neutron radiographic water-mapping at the National Institute of Standards and Technology). The Nissan Technical Center of North America (NTCNA) baselines are improved but are still not clearly state of the art. Collaboration should be exploited to enhance the characterization of nanofiber electrode results. It is unclear whether the eSpin Technologies, Inc. (eSpin) work is complementary or mostly duplicative; the project could use more clarity on the respective roles.
- The neutron imaging work is excellent use of the FC-PAD laboratories. This project can really benefit from seeing the MEA water profiles since operating at low RH seems to be one of the primary advantages of this technology.
- There are very good collaborations, although the researchers would benefit from some more mainstream testing methods.
- There is clear exchange and collaboration between laboratories. There is apparent separation of tasks and expertise, with exchange of information between laboratories also apparent.
- The project lists good project partners with a scale-up partner, an industrial original equipment manufacturer partner, and FC-PAD laboratory partners; however, it is not clear what, if any, of the results were made with the partners' input or contribution. For example, the Nissan data shown on slide 10: it is unclear if that was with MEAs made at Nissan or simply tested at Nissan. It is also unclear why Nissan is testing with fixed flow polarization curves at a strange, high stoic. Many of the polarization curves are with a fixed flow that is excessively high, based on the required flow for proper stoichiometry. This can affect low-RH polarization curves dramatically.
- The NTCNA's participation seems to be very limited on the project with 75% of the work completed; it is not clear what the Center's contribution is, and there is a question of integration of the work. Further FC-PAD tasks are described under "Collaboration," but there is limited evidence for water distribution other than neutron imaging and scanning transmission electron microscopy (STEM) analysis of Co; perhaps this was done at ORNL, but it is unattributed on slides 7, 10, and 16. While mass and water transport in these novel structures seems to be critical, it is not clear when that work is to be performed and integrated, and this is not included in the future work slide.
- The work scope of eSpin is not clear. It is unclear whether the company is involved in any optimization of the nanofiber ink formulation for the manufacturing or is involved only in optimizing the manufacturing process parameters for the catalyst slurry provided by Vanderbilt University (VU).

Question 4: Relevance/potential impact

This project was rated **3.4** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- MEAs with electrospun mats are showing remarkable performance improvements at low RH with low catalyst loadings. This is very relevant to DOE goals. Reducing catalyst costs in PEMs is a major objective, and this project directly addresses that.
- The project topics are right on target with the technology advancement and the Program goals. The project would result in decreased catalyst layer loading while upholding performance—and even improving it at low-RH operating conditions.
- High-performance, stable, low-platinum-group-metal-loaded, robust electrodes are key to programmatic success. Further progress is still needed in the high-power-density performance of the nanofiber electrodes, but it shows promise in stability and low-RH performance. High performance at low RH is also an enabler, given that target power density is tied to the Q/ΔT metric (i.e., driving higher temperature, which results in reduced RH). The project team is encouraged to further identify the reasons for this somewhat surprising low-RH performance that has been exhibited.
- The work being performed on this project clearly aligns with needs expressed in the MYRDDP; the project's approach to attack oxygen transport limitations using both ionomer and catalyst layer changes has the potential to meet DOE targets, in terms of performance and durability at low loading. While the practicality of the electrospinning process on a mass-market basis and MEA fabrication is not discussed, it should be evaluated.
- Based on the excellent performance results, the impact is substantial.
- Because this is a unique way to make MEAs that is showing great promise, understanding the design and manufacturing parameters is critical for large-scale adoption and for understanding the cost implications. The responses to the comments from AMR 2018 are not sufficient. As additional data with more slurry properties are available and new binders are investigated, it should become easier to use design-of-experiment concepts to systematically understand the parameters and to help optimize the process variables.
- Though the project's performance has been impressive to date, it is not clear what tweaks the team has left for further improvements in light of DOE goals. It would be interesting to see the project use the better 3M ionomers or the better catalyst that General Motors and 3M have been using in conjunction with these structures. The project will always have some impact just because it is different; this system will need to be better understood and modeled in order to understand why the team is seeing performance gains.
- If the authors' work can be scaled, etc., it may become an important approach. It is not clear how it would scale to a roll-to-roll process.

Question 5: Proposed future work

This project was rated **3.1** for effective and logical planning.

- The proposed future work covers the logical continuation of the project and addresses the questions that have arisen during the project. One could not have predicted that Gen 2 MEAs at either electrode would give high power at low RH, but it is definitely a question worth investigating.
- The project shows the benefit of the fiber format for the electrode; some imaging was done to show the water profile in the cell. More modeling or analysis work should be done to determine the mechanism for a fiber format of the catalyst layer showing improvement. This mechanistic understanding will likely lead the research team to even more improvements.
- There is no dispute with the proposed future work, aside from the need to normalize test conditions with DOE standard methods.
- Overall, this is a very sound project plan moving toward the completion of the work.
- It would be beneficial to see more systematic parameter and process variation, combined with FC-PAD diagnostics, to further identify the mechanistic impacts of nanofibers on performance, RH dependence, voltage stability, etc. Voltage loss breakdowns should also be performed to elucidate the low-RH

performance and measure potential mass transfer effects; for example, the high I/C ratio of the "core-shell" morphology may negatively impact local oxygen resistance.

- The team is right to want to try new ionomers of lower equivalent weight and new catalysts. The team should have done more work to fundamentally understand why these structures are better; modeling with Lawrence Berkeley National Laboratory would help in this case. There is no mention of future scalability studies or economic analysis, which should be done or, if it is not an issue, at least that should be reiterated; however, this seems like more steps than what the best processing methods are using today, especially if the project team is casting in the salt form.
- The project's future work mainly focuses on reaching performance targets, but there is little focus on integrating fundamental understanding and the use of FC-PAD capabilities to understand what is happening in these structures and help accelerate the development. It does not seem to make sense that the project team should continue (or include) conventional catalyst layer development with the functionalized ionomers for electrospinning.
- The only recommendation is for the project team to focus on one catalyst for mat electrodes made at eSpin until the optimization variables are understood.

Project strengths:

- The team is very experienced with electrospinning ion-conductive polymers. Electrospinning enables the production of nanoscale architectures with several features that are inherently beneficial in fuel cell electrodes (e.g., porosity, connectivity, etc.). Engaging with an industrial partner early in the project will help convince others of the potential impact of the results on a large scale.
- The project's strengths are in the principal investigator's (PI's) slurry development and in the industry partner for manufacturing mat electrodes. More focus on ionomers and design for manufacturing would help industrialize this method of making MEAs.
- Electrospinning produces novel catalyst layer structures that seem to have the potential to make an impact toward project targets.
- The unique structures being generated with tunable parameters should make for an excellent tool for understanding performance. This is a good use of the FC-PAD team.
- The PI has a strong background in nanofiber technology. FC-PAD's characterization capability is a key attribute that has been utilized to greater extent in the second year.
- The project has excellent performance results on a novel approach to catalyst layer design.
- The project has an innovative approach to making CCMs and seems to be getting high-performance results.
- The project has a sound approach and a good team and working strategy.

Project weaknesses:

- There are not many weaknesses. The project shows good progress overall.
- The project teams have the capability to fully characterize the performance (e.g., R-O_2 local, mass transport, electrochemical-surface-area loss, specific activity), but they have not yet used that capability sufficiently.
- The transfer of the electrospinning process from VU to eSpin appears to be a problem since the eSpin mats lack the durability of the VU mats. This is probably an issue with scale-up, but it begs the question of whether the mat-spinning process can be achieved on a useful scale.
- More could be obtained from the project with better project management and collaboration with sub-partners and FC-PAD. The conventional electrodes with electrospun polymers should be deemphasized after having initially demonstrated no benefit.
- The project's teaming could be improved. Polarization curves should be run at constant stoic rather than constant flow.
- The project's unusual test conditions (i.e., the very high reactant flow rates) make the data hard to understand in comparison with standard CCMs.
- There is a lack of economic analysis. The project's approach will take more steps than have been contemplated today. Though the baseline materials being used are relevant, it would be good to see more variation.

Recommendations for additions/deletions to project scope:

- The researchers should consider talking to a manufacturer about how to use the VU process in a practical system. This will prevent the project from falling into the "valley of death." The national laboratories are not a transition sponsor. Perhaps the small company American Fuel Cell (AFC) might be willing to talk to the team (although AFC might have been bought out by Plug Power).
- A comparison of mats made with Pt catalysts on different carbon supports could shed light on the MEA performance. It would be beneficial to see some additional focus on the durability of the mat electrodes.
- The project team should add more characterization and modeling to understand the performance differences between these systems and cast systems. The expected cost in comparison to existing methods should be shown.
- There are questions on the role of eSpin. It would also be good to see more voltage-loss breakdown analysis. More systematic parametric variation is recommended to elucidate the mechanisms and benefits of nanofiber electrodes.
- The project team should make some effort in modeling and understanding the mechanisms of improvements in catalyst layer performance based on the fiber format.
- The conventional electrodes with electrospun polymers should be deemphasized.

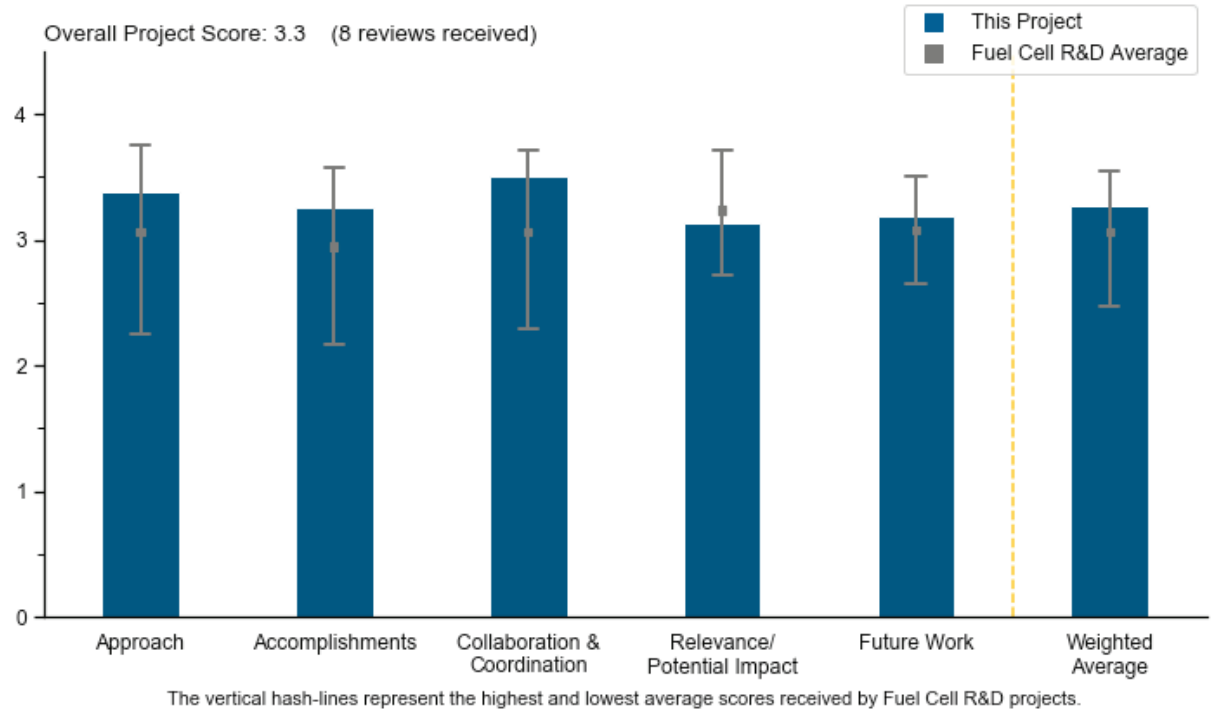
Project #FC-160: ElectroCat (Electrocatalysis Consortium)

Deborah Myers, Argonne National Laboratory, and Piotr Zelenay, Los Alamos National Laboratory

Brief Summary of Project

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

Project Scoring



Question 1: Approach to performing the work

This project was rated **3.4** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The project covers large, sprawling work, with multiple parties and activities. Nevertheless, it is surprisingly focused on the two key points of PGM-free catalysts: activity enhancement and durability extension. This is the type of effort needed to truly understand this class of catalysts and is the most appropriate for the advanced resources and personnel of the national laboratories. There is a nice combination of theory, analysis, and experimental and synthetic work.
- The project aims to develop advanced PGM-free catalysts and electrodes aimed at enabling significant cost reductions for fuel cell electric vehicles. To date, the project approach appears to focus primarily on increasing performance via catalyst activity and electrode transport; this work is excellent. While achieving performance is ultimately necessary, much more focus needs to be placed on durability in the immediate term.

- The project's approach is to develop and implement PGM-free catalysts and electrodes by streamlining access to unique synthesis and characterization tools across national laboratories, developing missing strategic capabilities, and curating a public database of information. This project has very strong leaders from four national laboratories, each with proven capabilities and expertise in some of the key technologies that are critically important for the success of this project. At this stage of the project, additional expertise is required on the catalyst layer and on degradation.
- The science content of this project is world-leading. The researchers are clearly very receptive to feedback from the community, which keeps them in touch with reality.
- The overall project approach appears strong. Progress is clearly being made on improving catalytic activity. One suggestion is that additional modeling of the electrode could be used. The electrodes are thick, and there may be some polarization associated with ionic conduction, triple-phase boundaries, etc., not just intrinsic catalyst activity. If this might be the case, some dedicated experiments and modeling could help clarify the issues, especially when looking at getting to the ultimate performance targets (which may require even thicker electrodes). Another suggestion is that durability under practical operating conditions has clear potential to be a showstopper for this class of catalysts. While performance is improving, durability is paramount to ultimate success. Additional work on durability, and articulating whether this class of catalysts may ever have sufficient durability, is encouraged. Scavengers can help (although they reduce activity), but it appears this effect is more or less short-lived.
- The project approach is well thought out and well structured and is designed to systematically answer the key questions in the development of PGM-free catalysts. The combination of the high throughput, characterization, automated learning, and performance evaluation work is an effective approach to developing relationships and advancing the performance and understanding of catalysts and catalyst layers. More modeling work would be useful to guide the work on electrode structure. Atomic and mesoscale models are being used to guide the catalytic behavior and active-site understanding. The durability work has increased with some good progress. However, some of the suppositions require further confirmation. It appears that the team is depending mostly on electrospun electrodes to provide improved mass transport and high-current-density performance. However, no indication was provided on feasibility or the rationale behind those expectations.
- The approach of working on material discovery and tool development at the same time is really good. There are many unknowns for PGM-free catalysts, including new tools needed to improve performance or even new material discovery by understanding the current materials.
- Much investment has been put into this one material for some time. It is clear that it is of no practical use without magnitudes of improvement in stability. The focus should be only two areas: determining the degradation mechanism and whether it can be prevented. Any other area is a waste of funding.

Question 2: Accomplishments and progress

This project was rated **3.3** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The accomplishments and progress are excellent and truly a testament to the capabilities of the national laboratory facilities and researchers in characterization and materials development. Of particular relevance were the identification of a correlation between the atomically dispersed (AD)Fe and the oxygen reduction reaction (ORR) activity (as seen on slide 14), the high-throughput materials fabrication and utilization of machine learning (slides 23–25), the accelerated stress test (AST)-induced loss breakdown (slide 28), and perhaps most importantly, a roadmap to achieving the required performance (slide 36). To date, the progress made toward addressing durability appears to be modest, consisting primarily of assessments of decay rate versus operating conditions (e.g., potential, cycling under air versus N_2) and the impact of a radical scavenger, which are important initial steps. It appears that the durability of the catalysts assessed is in the order of tens of hours.
- The work is structured via well-defined milestones that are on track or have been exceeded. Overall, there has been excellent progress on improved fuel cell performance over the past 10 years. Over the report period, improved performance was achieved both in the kinetic region, via catalyst understanding and synthesis approaches, and in the mass transport region, via electrode structure. The reported performance of 36 mA/cm^2 is highest, not the average. The goal was still exceeded, but the project should be quoting the

average, with error bars. The work on electrode structure is still relatively limited, although some interesting results are being achieved. The ionomer layer studies are interesting, showing better performance with decreasing hydrophilicity (high equivalent weight, lower ionomer percentage) nearer to the microporous layer is expected to result in improvements in water management and increased utilization. The use of the high-throughput approach should provide useful insights. There still does not appear to be much use of model correlations or a model-driven approach in this area, although the future work does mention that the project will be using models. There was some reasonable progress on understanding durability issues and mechanisms, the mitigation of shelf-life issues, and the use of cerium to address peroxide degradation mechanisms. Cycling in air versus nitrogen highlights the importance of using a range of realistic conditions to explore degradation mechanisms and impacts. The cerium work can be further explored to confirm mechanisms involved. There was a nice analysis on the improvements needed to achieve power density targets, with a path laid out to achieve them. However, the feasibility of achieving these steps is not explicitly stated. They will certainly be very challenging. The high-throughput approach should provide valuable trends that can then be examined and understood with model approaches. However, error bars should be shown on the plots. The progress on the data hub is not shown; the milestone status indicates this work is in progress.

- Significant progress has been made in tool development. Nuclear resonance vibrational spectroscopy (NRVS) with NO as the surface probe for detecting surface Fe sites is very helpful in conjunction with Mossbauer analysis for active sites. The project team has also put efforts into understanding the effect of carbon source and dopant on Fe in order to gain some insights; however, so far this project is lacking in material discovery. The same polyaniline (PANI)-type catalyst has been used to show good performance in meeting milestones. An improved version of PANI is adequate for proceeding, but it limits other material discovery. Hopefully, the team can show a few more catalyst candidates that meet both performance and durability targets. The layered electrode structure results are promising, and with some more optimization and lower catalyst loading, perhaps MEA performance can be further improved.
- The hypothesis for the most likely failure mechanism for the loss of catalyst activity is well supported (i.e., the formation of the hydroperoxyl radical). The improvement of the highest levels of activity was only mediated by the need for an additional 12x improvement in order to make this class more feasible for implementation. Additional focus on the interactions implicit between electrode structure design and ionomer may help bring about greater activity.
- The project team has made good progress in a challenging area.
- This project team has shown and continues to show progress in each of the four key areas of development: catalyst development, electrode and MEA development, high-throughput fabrication, and modeling. However, the rate of progress in the areas of catalyst-layer structural optimization and performance stability has decreased compared to previous report periods, putting the project's goals at risk. The area of Fe migration remains a fundamental concern. Fe that reaches the membrane is known to severely limit the membrane's durability because of the Fenton reaction. Since this project introduces Fe into the catalyst, it must define a solution path. The authors also need to resolve the cause and mitigation of the new results found during AST. The fact is that there are large losses in the kinetic region when cycling in air, but the Fe-reversible redox couple remains relatively unchanged. This seems to suggest a counterintuitive, possibly new, decay mechanism.
- The performance milestone was achieved. Catalyst durability is clearly being looked at.
- Most of the project's effort was devoted to milking out the most initial activity from a very unstable material. This will have no practical use. More work should be done on understanding the degradation mechanism and whether it can be prevented.

Question 3: Collaboration and coordination

This project was rated **3.5** for its engagement with and coordination of project partners and interaction with other entities.

- The work of the national laboratories in the consortium appears to be well coordinated. The other funding opportunity announcement partners should also provide effective overall collaboration, with a variety of techniques and approaches used to improve the PGM-free results.

- Collaboration among the four national laboratory team players is really good and evident from the results. The collaboration with many partners outside of ElectroCat is also very encouraging.
- It was particularly pleasing to see a group of researchers starting with very fundamental catalyst discovery transition all the way to fuel cell testing.
- Collaboration across all of the national laboratories appears well coordinated, extensive, and effective.
- Interactions, both "within" and "outside" the consortium, are very good, extensive, and strong. It is unclear how the fruits of this effort will transition to commercial adoption later, and whether feedback from commercial entities has been codified enough to know what is an acceptable commercial or lifetime target (independent of DOE targets). In other words, the project team needs to define a minimum viable product (MVP), with industry input. The product should be defined for first-entry markets and not necessarily transport.
- There is good collaboration, although it is quite limited to selected project members. There is also good use of the national laboratories' advanced and extensive capabilities.
- It appears coordination is being done well.

Question 4: Relevance/potential impact

This project was rated **3.1** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The success with PGM-free catalysts supports DOE's goal of acceptable cost for high-volume automotive applications of hydrogen fuel cells. Baked into this goal is lifetime durability, which also addresses relevance and impact. As a methodology that aids the fuel cell community, the inclusion of machine learning and high-throughput catalyst development has additional relevance but may not be as goal-oriented as the key performance indicators.
- This work can definitely have an impact on DOE's long-term goal of fuel cell commercialization with cost reduction, but it is still far from where it needs to be. DOE has spent millions of dollars on PGM-based catalysts, so one hopes the Department will continue to support PGM-free catalyst development as well—for a long enough time to give this team an opportunity to meet the ultimate targets.
- Durability and cost are the primary challenges to fuel cell commercialization and must be met concurrently. The PGM catalyst is projected to be the most expensive single contribution to the cost of the fuel cell stack. Eliminating PGMs while maintaining the current level of specific power, power density, peak energy efficiency, and durability will significantly accelerate the deployment of fuel cell systems.
- Much progress has been achieved in PGM-free catalysts; however, the eventual applicability to high-current-density and high-power-density durable applications is still much in doubt. With the improvements obtained, it is likely that these catalysts will find areas of application, thus increasing the relevance of the work.
- The project is clearly pursuing the reduction of PGM loading for ORR, which is directly on the pathway to achieving the polymer electrolyte membrane fuel cell (PEMFC) cost goal. In addition to doing good science, the project appears focused on the performance objectives.
- While excellent progress is being made toward developing improved activity catalysts and electrodes with improved performance, these materials have a low probability of meeting any relevance toward reducing fuel cell system costs unless the durability is substantially improved (not by tens but by thousands of hours). It is unclear whether the non-PGM catalysts based on atomically dispersed metals can be sufficiently stable because of demetallation and active-site degradation. Additionally, it is also unclear, and perhaps unlikely, that these materials will be able to meet support durability; it is not known whether start/stop degradation will occur.
- Eliminating PGMs does not necessarily mean lower cost. Because the cost of all the other components combined is large, the cathode catalyst with the highest activity, power, and durability will give the most cost-effective fuel cell stack. The current material is Fe, which is not compatible with PEMFCs, as it has very low stability or usable activity.
- From the presentations and discussion after, it is difficult to see what cost benefit these catalysts could provide. This is not a failing of the project, as the team is still developing the catalyst, so solid cost numbers are not expected. With the additional processing and increased thickness of the electrode, it is not likely that this electrode system would be significantly lower in cost than a conventional electrode. This is

opinion only, and those who are more familiar with the project will likely be able to provide a more considered comment. It would be beneficial to include a rough estimate of what savings could be achieved for an electrode if the project is successful.

Question 5: Proposed future work

This project was rated **3.2** for effective and logical planning.

- Durability is the number one consideration on the list, which is a good sign, given the importance of this metric to the overall effort for developing PGM-free catalysts.
- The project team has a clear plan for improving performance and has already demonstrated good progress.
- There is a comprehensive summary of proposed future work focus on slide 36. This slide details the relative expected impact for improvements in the power train of the MEA (e.g., activity, mass transport, resistance, etc.). The project should address lifetime with inhibitors.
- The proposed future work is as expected; durability needs some serious consideration while the project team continues improving the performance. The use of developed tools should lead to new material discovery to meet the ultimate targets. Some focus on data hub and machine learning is encouraged, but that should not distract from the main focus of PGM-free catalyst development. Based on these preliminary results, layered electrode structure would need more attention in future work.
- Achieving the power density improvements will be very challenging. The team is depending on electrospinning for mass transport and some ionomer approaches. However, achieving 12x activity is undoubtedly the most challenging and may not be achievable. There was little mention about other approaches to electrode design, although the appendix shows some reasonable approaches around using the combinatorial cell. The multiscale modeling efforts mentioned should be pursued to guide the electrode development.
- The project's future work aims at improving the performance and durability of the PGM-free catalysts, directly in line with the key needs. However, it is unclear what fraction of effort will be devoted toward durability, the largest barrier to date and where most development is needed. Only one of the four bullets on slide 38 addresses durability, and only a few of the quarterly progress measures and milestones on slides 5–8 address durability.
- Short-term durability has identified some potentially significant issues. Decreases in catalyst activity of 50% and 70% suggest that fundamental changes to the basic catalyst may be necessary to produce a stable catalyst. If this catalyst cannot be stabilized, alternative processes need to be identified. The potential risk with Fe is high, and so a solution path is needed.
- The project should focus on degradation.

Project strengths:

- The project's strengths include the outstanding teamwork among many members and organizations; the excellent state-of-the-art analytical methods being used to support or eliminate hypotheses; and the various talents and expertise being harnessed for real improvement of state-of-the-art PGM-free catalyst activity, the understanding of limitations for active site density, and potential failure mechanisms.
- The project has a very capable team to accomplish its tasks. The principal investigators (PIs) are leading these efforts from the front. New tools have been developed that can assist in material discovery in coming years. The collaboration with partners outside ElectroCat is also great.
- The project is extraordinarily effective at utilizing the excellent national laboratory capabilities (including facilities and expertise) to rapidly improve the activity and performance of PGM-free catalysts and electrodes.
- There are very systematic, comprehensive, and aligned efforts toward improving catalyst activity, durability, and stability. Extensive characterization and catalyst-level modeling has been incorporated, and the project has a strong team.
- This project group is very experienced, as shown by their previous work. The PIs and their partners are capable of successfully managing this project.
- The technical knowledge and the quality of the science in this project is world-leading.

- There is a good balance of focus on performance targets and scientific investigation. Improvements in performance have been shown.

Project weaknesses:

- The same catalyst, or version of that catalyst (PANI), has been used to meet the targets. The project needs to look into new and different catalysts as well. The same MEA is not used to show the ORR activity and mass transport in air targets. A higher-loading MEA (~6 mg/cm^2) was used to meet the ORR target and lower loading for other MEA performances. It is expected that all targets should be met with the same MEA, or at least with similar loadings. Finally, the durability is very poor and needs to be improved without sacrificing performance.
- Durability concerns have increased during this report period. Alternatives that are more stable need to be defined. The fundamental mechanism responsibility for the losses in the kinetic region need to be identified. Alternative catalyst-layer structures need to be defined and evaluated, and the stability of the hydrophobicity of this layer needs to be a primary parameter. Developing a solution path to verify that Fe will not leach out of the catalyst layer over time, at all operating conditions, is required.
- The project needs a definition of an MVP that is at an intermediate performance between automotive goals and other, more near-term applications. The project team should also start the transition to air tests. It is acknowledged that air data was posted in the presentation, but now that catalyst activity is sufficient to actually consume enough oxygen to start mass transport limitations, the development of thick three-dimensional electrodes poses significant challenges that mean that earlier efforts are needed.
- This is not a weakness of the project per se, because it looks like there is not currently a metric that is not being met, but overall, more metrics and an emphasis on durability, even at this stage of performance, seem important. Going for performance first and then thinking about stability should be avoided. Performance and stability need to be pursued in parallel. However, it is understood that this is still early for developing PGM-free catalysts, and the team does appear to recognize this.
- This is perhaps not a weakness of this project, but in general, there is an over-emphasis on capital cost and performance within all DOE projects. This is not a bad approach at this stage, as cheaper fuel cells would be great, but a greater emphasis on lifetime expectancy would be appreciated. If PEMFCs cost roughly what they do now but had an 80,000 hour+ lifetime, that would open up additional markets for hydrogen as an energy carrier and would be highly disruptive. This project team should apply themselves to a long-life, reasonably low-performance fuel cell for stationary power. Perhaps this is not possible in this project and may require additional membrane experts, but it would be a good use of the approach and expertise used in this project.
- Approaches to electrode structure could be better developed.
- The project's focus should be on degradation.
- The project is not placing enough emphasis on improving durability.

Recommendations for additions/deletions to project scope:

- The project should strongly consider de-emphasizing activity and performance development and dramatically increasing focus on developing durable materials. It is clear that the activity and performance may reach commercially acceptable levels, but the durability demonstrated (at tens of hours) is very far from Pt-based materials. It is also unclear what the cost savings of a PGM-free MEA will be, relative to a PGM-based MEA. As Pt contents continue to decrease with state-of-the-art PGM-containing MEAs, the need for PGM-free may be decreased. Current reported results by General Motors and Los Alamos National Laboratory (Spendelow) show feasibility of <0.10 g/kW (albeit also without meeting requisite durability). This translates to 9 g of Pt, or about $300 at current Pt prices. The other costs associated with the catalyst and electrode are likely to be similar for PGM-free. Analysis should be conducted for where the "break-even" point is for PGM-free compared with PGM, e.g., what level of performance and durability are required to match the cost of current commercial or laboratory technology.
- The project team should prioritize degradation work. Eliminating other non-value-adding work can save much funding.
- The project needs to focus more on durability and, perhaps, some more on electrode modeling, especially if catalyst loadings rise and electrodes get thicker.

- There needs to be more focus on material discovery and improving durability.
- A recommended addition would be electrode structure modeling.

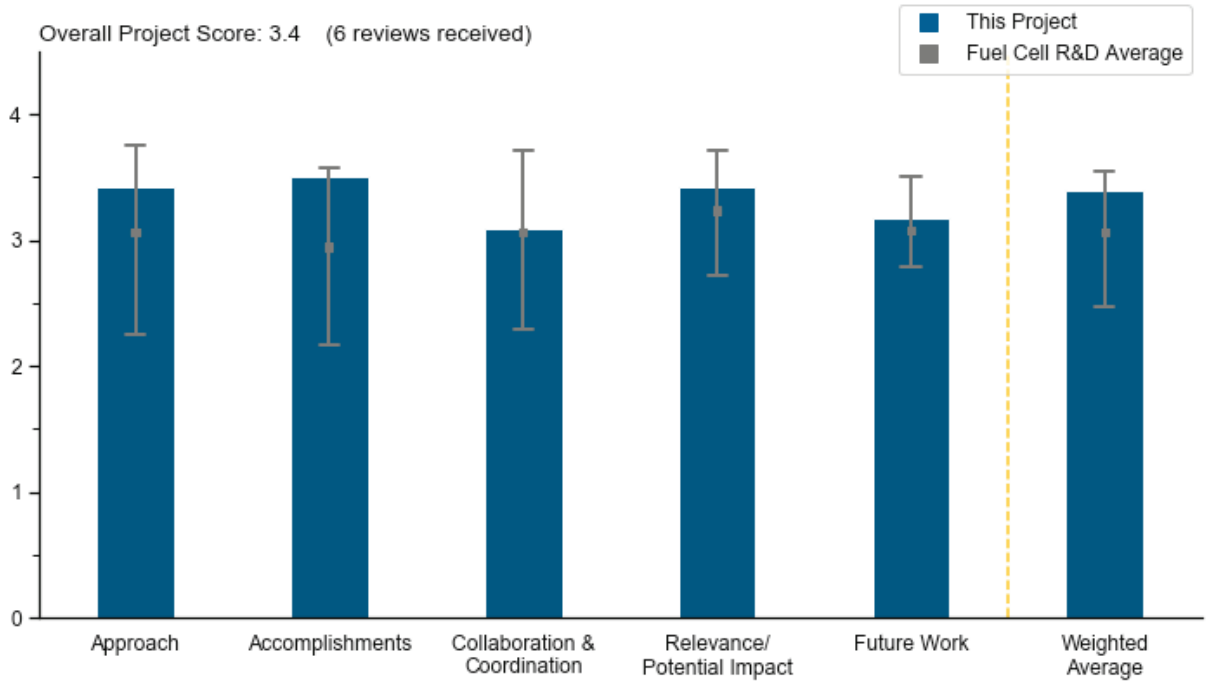
Project #FC-161: Advanced Electrocatalysts through Crystallographic Enhancement

Jacob Spendelow, Los Alamos National Laboratory

Brief Summary of Project

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

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **3.4** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The project approach is excellent, pursuing the development of highly active and stable intermetallic catalysts on supports with improved durability. The project appears to use modeling, materials fabrication approaches, and materials characterization effectively to drive the technology forward.
- The project put forth several techniques to achieve the ordered intermetallic catalysts and appears to have down-selected to the scalable LANL synthesis. The project combines synthesis, theory, and electrode integration for a comprehensive effort. There is also an interesting avenue of research in using the University of Buffalo's (UB's) platinum-group-metal (PGM)-free-type materials as a support.
- Stabilizing PtM-alloy catalysts is a good approach. However, one reviewer asked why the team limited itself to transition elements for the M_1M_2Pt system. It should also be made clear why the team chose Vulcan carbon for the PtM synthesis, and if there is a plan to explore high-surface-area carbon with a similar approach. The team should also elucidate why they use N-doped carbon supports and state whether

other types of carbon will be used. These are fundamental questions that the national laboratory teams should address.
- The project approach is good, with different synthesis methods guided and supported by modeling, testing, and characterization and well-defined plans for scale-up and MEA validation. It is not clear whether there is a thermodynamic basis for the use of intermetallics. It is also unclear whether there are any phase diagrams used in the design of ternaries.
- The approach targets many of the barriers in catalyst development. The approach is very similar to currently funded and previously funded projects.
- The approach being pursued on the project is feasible and logical.

Question 2: Accomplishments and progress

This project was rated **3.5** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Impressive accomplishments have been achieved. It is promising to see that the ordered structure remains after the catalyst accelerated stress test (AST) in MEAs. The $L1_0$-PtCo catalyst is also showing significantly reduced loss of Co content. Most of the targets have been met with this catalyst. The work with the hydrogel supports is very promising and shows very good activity and stability. Optimization of the electrode integration is needed and may be completed by the structure of these supports.
- The team has made exceptional progress in developing highly active and durable intermetallic catalysts. One catalyst has met several DOE targets, including mass activity and electrocatalytic (cyclic) durability. The hydrogel supports look promising for support durability with Pt catalyst.
- The team has achieved excellent accomplishments toward highly active PtM alloys. More work needs to be done in durability and in transferring this performance into an MEA and obtaining the same high activity, but this is a good step in that direction.
- The project appears positioned to achieve the DOE 2020 targets for catalyst performance in a single material, which is an outstanding accomplishment.
- Progress has been made with the new PtCo catalysts, some of which meet the DOE performance targets. The reduced Co leaching is promising.
- The presentation slides mentioned that particle size remains mostly unchanged after 30,000 cycles. However, a larger image that includes various particles is necessary to see if there is any Pt-Co agglomeration due to particle sintering and Ostwald ripening processes under 0.6–0.95 V cycling conditions. The conclusion should at least be supported by particle size distribution or X-ray diffraction fit. It is not clear if the Mn hydrogel is a new support or is derived from the polyaniline as N-doped carbon support with Mn. In any case, "Mn hydrogel support" can be oxidized to higher valency during 1.0–1.5 V potential cycling or during normal operating conditions. Leached Mn can affect the membrane and act as a Fenton-like reagent.

Question 3: Collaboration and coordination

This project was rated **3.1** for its engagement with and coordination of project partners and interaction with other entities.

- The project has good collaboration; the partners' contribution to the project is clearly demonstrated.
- The collaboration between LANL and IRD Fuel Cells and the collaboration between LANL and UB are both clear and effective. One area for improvement would be explaining exactly how the results from the University of Pennsylvania (Penn) and Brown University (Brown) fed into the materials prepared by LANL. It was also not clear if the best catalysts were prepared at LANL, Brown, or Penn or if the results at Brown and Penn even fed into the work at LANL.
- The project has very good collaboration; however, the contribution from partners is not clear in the presentation. There are three entities preparing catalysts using various techniques, but the presentation is not clear as to which technique is considered successful. The project ends in September 2019, and there may not be sufficient time for MEA scale-up production and catalyst or MEA validation by EWII Fuel Cells (EWII). EWII's involvement in years 1 and 2 is unclear from the presentation.

- There are multiple catalyst development partners in this team. It is not clear what the focus was for each of those partners or how they contributed to the overall project. Future MEA fabrication and scale-up methods need to be better elucidated.
- While the roles of each organization were well described on slide 21, it is unclear how much of the catalyst development work was done at LANL versus Brown or Penn.
- It is unclear what the current roles of Brown, Penn, and EWII are in the project.

Question 4: Relevance/potential impact

This project was rated **3.4** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The project's scope and goals align with the DOE Hydrogen and Fuel Cells Program's technical objectives, and the team has the potential to help make progress toward research, development, and demonstration targets for performance in low-loaded catalyst layers.
- This approach shows much promise for developing highly ordered PtM alloys with good activity, and the approach has the potential to meet DOE cost targets.
- The team is directly addressing key barriers for commercialization, catalyst activity, and durability.
- The potential impact is very high for achieving PGM-loading and durability goals.
- The approach is very similar to currently funded and previously funded projects, and therefore a similar outcome is expected.
- Based on the MEA results, this project appears to be achieving DOE's intended goals for this funding opportunity announcement. One slight criticism is that LANL's method was stated to be scalable, but the details of the processing were not provided. Traditionally, preparing these alloys in small nanoparticles is challenging, so it is not clear what LANL is doing differently if the team is not using a protective coating, as stated in the question-and-answer session. If the method used is not scalable, the technology would obviously not have the intended impact. The final months of the project will examine scale-up, so it is likely that that question will be answered soon.

Question 5: Proposed future work

This project was rated **3.2** for effective and logical planning.

- The proposed tasks are in line with the current accomplishments and overall project goals.
- The future work appears to be logical and is likely to be successful.
- The proposed work looks very good. The scale-up of the down-selected $L1_0$-CoPt catalyst seems appropriate. Work needs to be done to select the most appropriate support. Perhaps the principal investigator might consider a more conventional high-surface support instead of Vulcan, if he has not already. The hydrogel support requires significant optimization and scale-up.
- The proposed future work is logical, based on the project's current status and the remaining gaps to the project's targets. Some additional effort should be placed on support durability with the advanced catalysts on hydrogel supports.
- The proposed work is a logical continuation of the presented effort. AST and accelerated durability test protocols are normally recommended in 25–50 cm^2 MEAs. Without knowing the catalyst preparation process used, it might be difficult to obtain gram quantities with good quality control for the final deliverable.
- The proposed work is very generic and does not elaborate on how the team plans to increase the high-current performance of N-doped supports. This would be needed to understand the source of the lower performance at high current density first. To guide them further in their optimization work, the researchers should plan to study ionomer–transition-metal interactions within MEAs in detail.

Project strengths:

- The team has been exceptionally effective at developing highly active catalysts with improved durability and demonstrating a good balance of fundamental and practical materials development, modeling, and characterization.
- The MEA results and progress on this project made Jacob Spendelow's 2019 Hydrogen and Fuel Cells Program Awards Fuel Cell R&D award well deserved.
- The team has strong partners in catalyst synthesis and support development. The role of the industry partner for the MEA scale-up is not clear.
- The strength of this project is the high activity and the stable catalyst that maintains its intermetallic structure through the catalyst AST.
- The catalyst–support combination study is interesting. The team has made progress toward MEA validation and demonstration of scale-up.
- The team has made very good progress on addressing most barriers.

Project weaknesses:

- The project has few weaknesses. One is perhaps the electrode integration, but this seems to be a focus of the future work.
- Scalability could be a concern, depending on the catalyst synthesis procedure that is down-selected.
- More understanding on certain findings would be good. For example, it is not clear what is preventing Co from leaching, whether there is an effect of the non-PGM particle size, or how the hydrogel support helps with performance.
- Along with maintaining mass activity, the team also needs to address the maintenance of specific surface area. Where shown, the specific surface area losses were significant. Meeting durability in MEAs will also require having stable surface area to avoid performance loss due to the increased local oxygen transport losses that are anticipated to occur.
- The project is focused on only transition metals to provide ordered structure, even for M_1M_2Pt alloys. It is suggested that the team explore the possibility of other metals to provide stability to the ordered PtM structure. The focus on support development should be minimized until significant progress is made in the understanding of this catalyst's durability, primarily regarding the catalyst–support interactions.
- The scalability of the process is unclear because the exact synthesis approach for the best materials was not revealed.

Recommendations for additions/deletions to project scope:

- This team seems to be addressing some critical aspects of alloy catalysts. It would be good to see additional scope devoted to understanding the role of other metals (other than transition metals) in enhancing the durability of M_1M_2Pt catalysts. The team seems to be missing a partner who can help in understanding durability. The interaction of this team with the Fuel Cell Consortium for Performance and Durability (FC-PAD) is not clear, and once the project researchers have reproducible MEAs from their industry partner, they should consider teaming with FC-PAD to understand durability.
- The project team should consider assessing the intermetallic catalysts for Pt and Co dissolution kinetics in coupled rotating disk electrode testing/inductively coupled plasma mass spectrometry (ICP-MS) testing at Argonne National Laboratory (Vojislav Stamenkovic) to determine whether Pt dissolution is improved relative to Pt random alloys.
- It would be good to see comparisons to commercial PtCo catalysts.

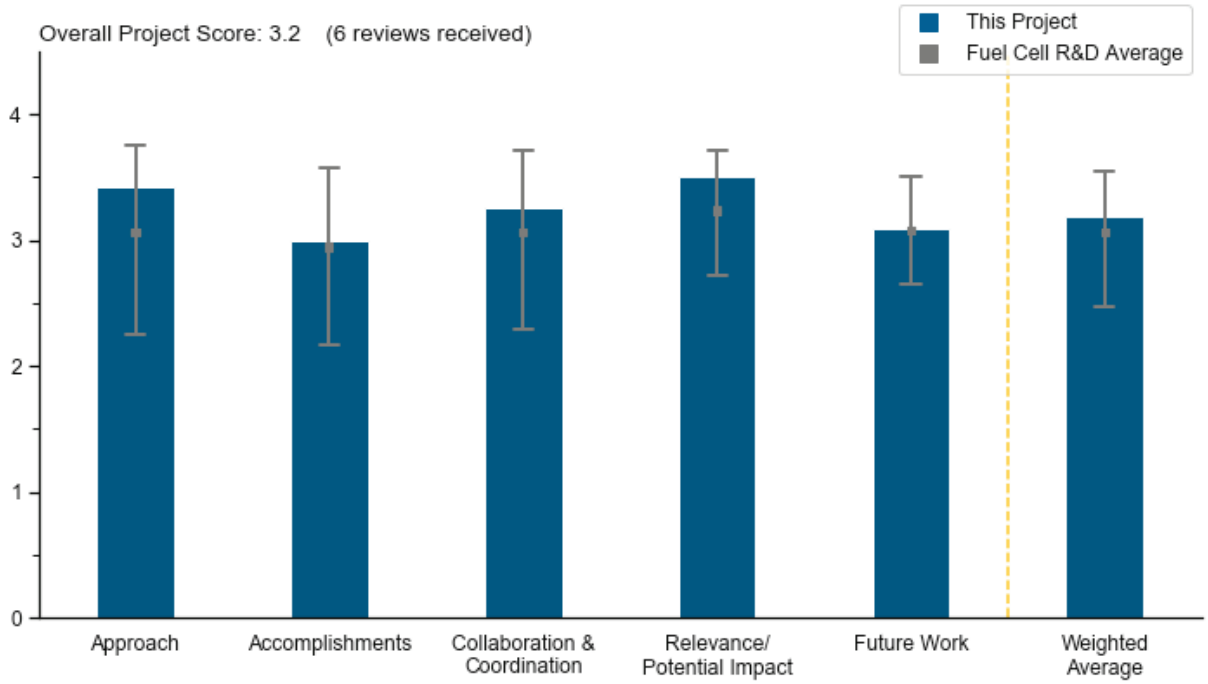
Project #FC-162: Vapor Deposition Process for Engineering of Dispersed Polymer Electrolyte Membrane Fuel Cell Oxygen Reduction Reaction Pt/NbOx/C Catalysts

Jim Waldecker, Ford Motor Company

Brief Summary of Project

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

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **3.4** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The approach to develop and validate a new Pt-based Pt/NbO_x/C cathode catalyst by developing and optimizing a vacuum powder coating PVD is valuable and novel. It does deserve attention and support since such a methodology has not been thoroughly examined. The project is supposed to address all relevant steps in the development of new materials, such as electrochemical evaluations of the catalyst for high durability, power density, and mass activity. In addition, the effort is aimed at developing the manufacturing process by demonstrating scalability and cost-effectiveness.
- Improving the durability of carbon-supported platinum group metals (PGMs) is critically important for the fuel cell industry. The proposed approach of achieving it through the synergistic interaction of platinum and carbon with NbO_x compounds is feasible and integrated with other activities in the field.
- This continues to be a more original project on low-PGM oxygen reduction reaction (ORR) electrocatalysis than most of the run-of-the-mill efforts that have for decades focused (often successfully) on Pt alloys, core–shell structures, etc. The same applies to the selection of vapor deposition as a method of catalyst

synthesis. However, by being more original than most, this project is also more risky. Given the existing fundamental science challenges, the project has made a move in the right direction by revising the scope to have greater emphasis on materials characterization and durability, including the nature of interactions in PVD Pt/NbO_x/C catalyst and durability comparisons between Pt/NbO_x/C and Pt/C reference catalysts.
- The project approach is logical and directed at barriers of cost, durability, and performance. The project has appropriately adjusted its approach to include higher NbO_x content, which should be more likely to demonstrate the effects of metal–support interactions and potential for improved durability.
- Utilizing niobium oxide to stabilize the Pt/C catalyst is a novel approach. The addition of NbO_x has a significant effect on the support stability and catalyst durability.
- The approach has improved from last year in terms of including more direct measurements of the Pt–Nb interactions, thus including some science in the project. One of the important aspects that can come out of this project is understanding the degree to which Pt and metal oxide interactions occur and how much they can change and/or stabilize the active catalyst particles. It is unclear what the advantage is of having catalysts made by both Oak Ridge National Laboratory (ORNL) and Exothermics, Inc. (Exothermics). It is not clear, with the catalysts made to date, why large-scale batches of catalyst are needed in this project.

Question 2: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has made tremendous progress since the 2018 Annual Merit Review. The principal investigator (PI) explained the interaction of NbO_x with Pt catalyst and its role in stabilizing the catalyst using X-ray photoelectron spectroscopy (XPS) and X-ray absorption spectroscopy (XAS) techniques.
- The PI presented excellent progress toward project objectives. Milestones that were not critical for the DOE mission were re-evaluated, while critical milestones were completed.
- The accomplishments and progress are clearly presented and show promise that the approach of this effort would lead toward novel catalysts with improved durability. Nevertheless, while the losses in mass activities for various samples are satisfactory, voltage losses for the majority of samples are still substantial. The system of choice demonstrates promise for the future work by performance that was measured in 5 cm^2 single-cell tests with the Exothermics 180920 catalyst. These results show that the sample meets the second go/no-go mass activity target, with the beginning-of-life (BOL) mass activity of 389 A/gPt and end-of-test (EOT) mass activity of 290 A/gPt. Considering that the focus of budget period 2 was on improving durability with increased NbO_x content, the project demonstrated progress, even though other technical targets need to be addressed as well.
- The project has met the year 2 milestone for determining Pt–NbO_x interactions. The project has met the milestone of <40% loss in mass activity and <100 mV loss at 0.8 A/cm^2 in the electrocatalyst durability cycle. The project is behind on reaching the performance milestone of >500 mV at conditions meeting the heat rejection target with a loading of <0.125 mgPt/cm^2 and P 150 kPa or less. The project is behind schedule for meeting durability in support corrosion tests. The reasons for the low maximum power performance are not yet understood. Additional diagnostics should be performed, including loss breakdown to determine pressure-dependent and independent mass transfer resistances, high-frequency resistance, and H+ transport resistances.
- The XAS and electronic interactions are informative for this project and other projects. It appears that there is a lack of Pt–Nb interactions; however, there is a suggested interaction between Pt and O from NbO_x. This data set is important for understanding why there is not a direct Pt–Nb interaction and why the Nb XAS signal does not change with Nb content. It is unclear why the BOL mass activity of 389 A/gPt was reduced to 290 at EOT. It could be particle growth or loss of bonding with the NbO_x. These are important questions that should be answered going forward. There does not seem to be a good correlation between the electrochemical active area (ECA) and mass activity loss. Not many data were shown to understand whether this is due to conditioning or an unrelated effect. The project should show more than ECAs and polarization curve performance. Data such as particle size distribution and change in distribution with mass activity should be shown. An explanation of the cause behind the mass activity loss is needed. The ECA on slide 15 does not seem correct (58.68 reduced to 5.43, meaning a 90% loss). There is either a ~10% ECA loss (if it should be 54.3 m2/g) or a ~90% ECA loss (if it should be 5.43); neither correlate well to the mass

activity loss of 25%. It is curious that the project PI does not bother to do any quick rotating disk electrode (RDE) cycling measurements to evaluate the catalyst durability, when for so long the PI insisted that RDE measurements needed to be done for everything.
- An increased emphasis on fundamental science, while commendable, has not provided clear answers to questions about the ORR mechanism on $Pt/NbO_x/C$ catalysts. That being said, the XAS studies at Northeastern University (NU) have afforded some interesting insights but, as is typical for such studies, few firm conclusions. A more complete picture of interactions between the three components in this complex catalyst system (including carbon support) would be desirable, helping with the design of future $Pt/NbO_x/C$ catalysts with higher activity (including the high current range) and durability (over a wide range of NbO_x content).

Question 3: Collaboration and coordination

This project was rated **3.3** for its engagement with and coordination of project partners and interaction with other entities.

- This project has an outstanding collaboration involving three industries, two universities, and a federally funded research and development center. The contribution from each partnering entity can be seen throughout the presentation.
- The collaboration is well executed, and the partners are well coordinated.
- This is a very strong team with complementary capabilities. Problems with EWii Fuel Cells, LLC (EWii) testing (i.e., electrode conditioning) indicate possible miscommunication between partner organizations. Collaboration with external partners who have interest in materials of the type under development in this project and the synthesis approach being used could help overcome at least some challenges.
- Collaborations appear to be working, for the most part. There appear to be substantial differences in observed performance between the partners. The reasons for the differences need to be determined, and comparable performance needs to be measured for the same materials by the different partners. Interactions with collaborators outside of the project (the Fuel Cell Consortium for Performance and Durability [FC-PAD], for example) are not apparent.
- The team collaborated between partners and with members of FC-PAD.
- The NU measurements appear to be the science behind this project. It is unclear what the advantage is of having catalysts made by both ORNL and by Exothermics. It is not clear why, with the catalysts made to date, large-scale batches of catalyst are needed in this project. EWii seems to be present to make scaled-up membrane electrode assemblies (MEAs), but the catalyst does not seem to have progressed enough to date.

Question 4: Relevance/potential impact

This project was rated **3.5** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The project focuses on high-performing and more durable catalysts, as well as a reproducible and scalable manufacturing process. For that reason, the relevance and impact to DOE goals could be significant if the project is successful.
- The project aligns well with the DOE Hydrogen and Fuel Cells Program (the Program) and DOE research, development, and demonstration (RD&D) objectives; the project has the potential to advance progress toward DOE RD&D goals and objectives.
- The project focuses on developing a $Pt/NbO_x/C$ catalyst for high durability, high power density, and mass activity to meet the DOE 2020 technical targets for electrocatalysts and catalyst supports.
- The work is relevant and could have impacts on cost, durability, and performance. The work could provide an alternative catalyst preparation route that is amenable to mass production. The metal–support interactions could be expected to lead to improved durability.
- More durable, higher-mass active catalysts are important for the commercialization of fuel cells. This project correctly addresses a major cost and durability issue with fuel cells.

- The project is very relevant to the Program's objectives and, by using a different approach, promises improvements in performance of nanodispersed Pt catalysts, especially in terms of durability. However, the tendency for Pt agglomeration needs to be overcome.

Question 5: Proposed future work

This project was rated **3.1** for effective and logical planning.

- The proposed future work focuses well on the remaining challenges and is aligned with the remaining 40% of the project timeframe.
- The proposed future work will lead to further understanding of this new class of catalyst material.
- The proposed future work is well aligned with project goals and DOE technical targets.
- The proposed work is logical and well justified. Ultimately, by the end of the project, it would be beneficial to have some kind of a judgment call on the viability of this class of ORR catalysts. Existing discrepancies between the performance of small-scale samples from ORNL and large-scale samples from Exothermics should be understood and eliminated.
- The proposed future work is logical and directed toward meeting the upcoming go/no-go decision point and determining loss breakdowns. Additional work to try to correlate properties with microstructure would be beneficial (for example, how H+ losses or pressure-independent mass transport resistances correlate with NbO_x or Pt content or connectivity).
- The continued XAS studies are good; they could be expanded more. The cost estimates will probably not be very relevant until a promising catalyst is produced. The durability measurements are important, as improved durability is a major objective.

Project strengths:

- The project has demonstrated the Pt/NbO_x catalysts without added resistance from the PVD catalysts. The surface analysis (i.e., XAS) from this project is important for understanding the particle interactions and what value these types of interactions will have in terms of stabilizing and/or changing the electronic state of Pt.
- Substantial progress has been made in materials composition reproducibility. There was a significant increase in batch size. The project is completing critical milestones on MEA performance and durability.
- The project has an original approach, well-executed collaborations, promising performance, and durable and novel materials.
- The project has demonstrated good activity for Pt catalysts (as opposed to Pt alloy), which should show good durability.
- The originality of the approach, promising improvements to catalyst durability, is the biggest strength of this project.
- The project has shown excellent collaborations and tremendous research progress.

Project weaknesses:

- The project has looked at only a few ionomer-to-carbon ratios and what are relatively low NbO_x contents. The current electrodes that were made suggest that the project has a problem with high currents. The catalysts do not appear to be showing better durability to catalyst cycling, which is a major (or the major) objective of this project: more durable catalysts. If the project is not showing better durability, then the project needs to show higher mass activity, but it is not even looking at active particles like PtCo, so it will not compete with those types of catalysts.
- The PVD process appears to be less controllable than was predicted at the outset of the project. Particle size distribution and other parameters do not appear to be as easy to manage as was originally thought. Based on the RDE results, MEA performance has been poorer than expected.
- MEA evaluations in 50 cm^2 are needed, as are improved power performance and optimization of the catalyst composition.
- The shift of the project scope toward fundamentals has not yet provided the level of understanding needed to design better $Pt/NbO_x/C$ catalysts in the future.

Recommendations for additions/deletions to project scope:

- It is unclear what value the cost analysis adds if the catalyst cannot show improved durability and/or mass activity. The project should concentrate on showing the improvement in performance before spending time figuring out how much more expensive these catalysts will be compared to traditional catalysts. The science from the XAS measurements should be expanded to other interactions, if possible, so that the community can know whether the proposed metal oxides have potential. Metal oxide supports have been proposed for a long time, with little real promise to date; the XAS can help evaluate this potential. The project should measure catalyst properties such as particle size distribution and understand the causes behind the loss in mass activity, which could be particle growth or loss of bonding with the NbO_x. These are important questions that should be answered going forward.
- The project should focus on better understanding the higher mass activity obtained by the Pt/NbOx/C catalyst compared to state-of-the-art Pt/C catalysts.
- The still-high voltage loss at 0.8 A/cm^2 and the large scatter of the durability test data are of concern and should be addressed.
- Evaluations should be done in 50 cm^2 MEAs.
- The project should facilitate MEA fabrication and testing activity.
- There are no recommendations for additions or deletions.

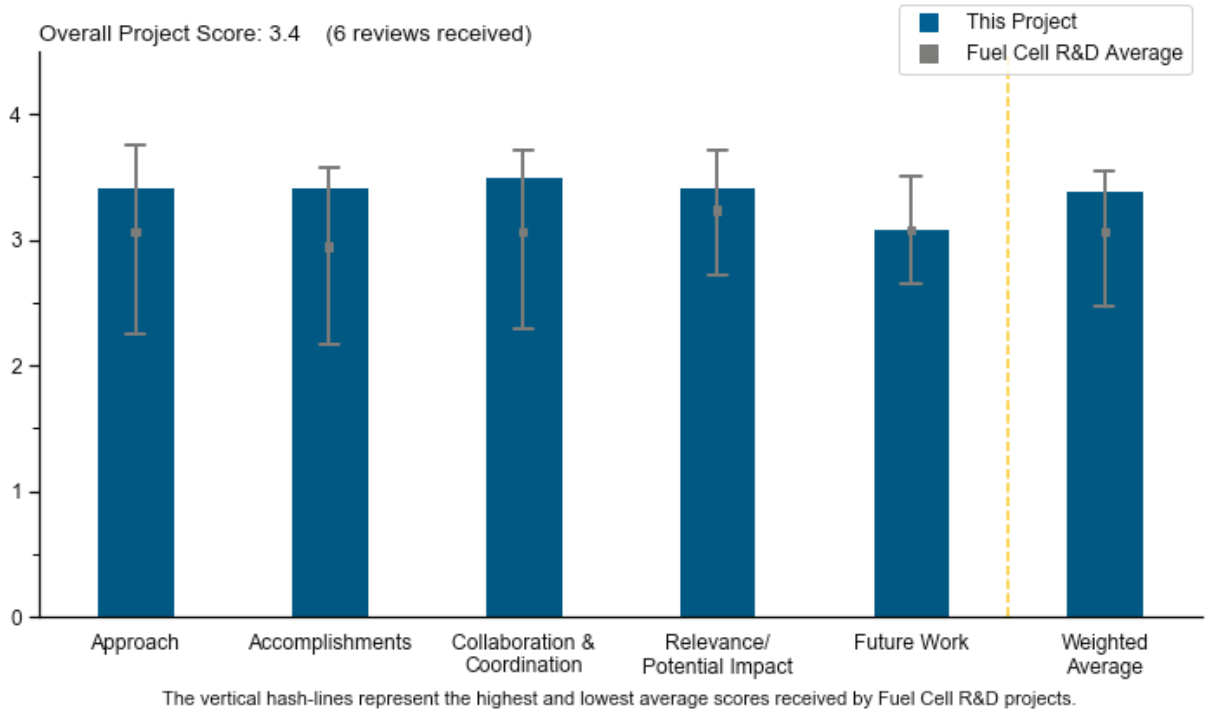
Project #FC-163: Fuel Cell Systems Analysis

Brian James, Strategic Analysis, Inc.

Brief Summary of Project

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

Project Scoring



Question 1: Approach to performing the work

This project was rated **3.4** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The project has a very good approach to assessing current and projected costs (in 2020 and 2025) of automotive, bus, and truck fuel cell systems at high manufacturing rates, using the design for manufacture and assembly (DMFA) analysis combined with the questionnaire sent out to system developers. This year, the choice is to put aside the light-duty vehicles (LDVs) in order to concentrate the efforts on medium-duty vehicles (MDVs) and heavy-duty vehicle (HDV) trucks, which is an excellent strategic choice to accompany the imminent development of fuel cell trucks. The report on cost will be released in September 2019. The project also identifies low-cost pathways and presents very interesting side studies quantifying the impact of durability on cost and completing the ways of improving durability.
- The approach taken by Strategic Analysis, Inc. (SA) is almost always appropriate and good. SA performs an essential service in quantifying costs for the community that would otherwise not be available. The pivot toward HDVs is a great addition. Some of the choices employed need further analysis, but they are off to a strong start. The areas beyond HDVs are of slightly less interest and are less impactful. A project like this has a hard time directly affecting DOE targets.

- The approach is valid and helps in overcoming the listed barriers. Insight on the operational costs (e.g., fuel, maintenance, consumables, etc.) and stack replacement costs (given the knowledge on the degradation performance) would be valuable. An interesting result would be the dollar-per-mile rate compared to another conventional solution with an internal combustion engine or a different powertrain.
- The DFMA method for cost estimation is appropriate. The continuously close interaction with component suppliers and system developers is key to fairly accurate cost estimations. Updating the LDV model biannually now while focusing on MDVs and HDVs makes sense.
- While not directly working on technologies that could reduce barriers, the project's cost analysis plays a necessary role in assessing the DOE Hydrogen and Fuel Cells Program's (the Program's) progress toward overall goals.
- The approach to the work is comprehensive in looking beyond component cost reduction and also including recycling and manufacturing techniques.

Question 2: Accomplishments and progress

This project was rated **3.4** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Great accomplishments have been globally presented again this year. This progress will help DOE achieve its goals by tracking how technology changes increase or decrease cost. However, switching from metallic to flexible graphite bipolar plates (BPPs) remains questionable. Ballard Power Systems, Inc.'s (Ballard's) arguments have been well presented, but it would have been interesting to have the feedback from other industry partners, in particular from Nikola Motor Company (Nikola). The already demonstrated lifetime in buses and the reduced cost projections have been underlined. However, the preliminary cost results indicate that BPPs are not considered as part of the major cost difference between LDVs and MDVs/HDVs. However, using flexible graphite BPPs will lead to decreased mass and volume stack densities. Quantified impacts may be compared with corresponding DOE targets. Even if the Toyota Mirai is currently using Ti materials for BPPs, Ti should not be used as a reference material in the quantified cost impact of durability measures. All the other original equipment manufacturers (OEMs) are developing coated stainless steel. The integration of two-dimensional manufacturing is very relevant and may be regularly updated in the future. Analysis of end-of-life vehicle recycling and disposal cost is of high interest.
- The work done on HDVs is a critical emerging need. To date, the work is built around the light-duty framework and a focus on capital cost, which is insufficient for the heavy-duty market, where total cost of ownership (TCO) is more important. The team recognizes this and has done a good job of putting the first study in place so that it can be built upon and refined. The flexible BPP work was somewhat surprising, as it was cheaper and more durable without any shown performance losses. The question that immediately comes to mind is why this approach would not displace metals in light-duty application. The end-of-life recycling and disposal cost was interesting, but perhaps less impactful.
- The project has defined objectives on MDV and HDV trucks. Achievements are aligned with the project objectives with reporting on cost. Results are due in September 2019. This choice is also of immediate interest to DOE on fuel cell truck development. Therefore, the accomplishments and progress of the project are excellent in the direction of DOE objectives as well.
- The accomplishments demonstrate success in moving the focus and analyses from LDV to MDV/HDV technologies and markets. This has further aligned the project with DOE objectives. Ballard has been identified as the sole supplier that can achieve the required performance and cost targets. This could create a future situation that drives higher cost owing to market rather than technical factors. Alternatives should be identified.
- The project shows advancement with respect to 2018 milestones and, most importantly, achievements that indicate the feasibility of DOE targets.
- The project continues to stay current with the inclusion of MDV/HDV cost analysis and cost assessment of developing technologies.

Question 3: Collaboration and coordination

This project was rated **3.5** for its engagement with and coordination of project partners and interaction with other entities.

- The project has a close collaboration with the National Renewable Energy Laboratory and Argonne National Laboratory (ANL) for acquiring knowledge, data, and review of the assumptions and results. Other collaborations with Ballard, 3M, and Precors Technologies have been set. Therefore, the project has a very good approach for seeking information and feedback by setting up relevant collaborations, which also allows the team to be up to date on new developments to integrate into the cost model. However, this year, it could be relevant to collaborate with HDV truck developers (Nikola, for instance), as the effort is concentrated there. These partners could bring very interesting guidance on objectives and priorities and give a relevant review of the assumptions, etc. In addition, these kind of collaborations will help with the choice of system and hybridization architectures. Specifications should come first from the system and then from the components.
- SA is doing an excellent job of collaborating, mainly with the ANL systems analysis project but also by expanding with many component and system developers, suppliers, and end users. This interaction gives confidence in the results. However, neither material nor system configuration selection should rely on only one partner.
- The project has a few funded partners to fill in specific gaps but does a great job of leveraging the community, particularly since the team is investigating areas that are often highly proprietary. Outside of having direct OEM insight, it is likely the best option.
- Strong collaboration is demonstrated with the project partners, and the project has utilized other industry expertise to inform the approach. While the presentation refers to a list of ~30 industry participants, this list is not included in the backup material.
- Close interaction with the ANL systems analysis project is key and effective. The project has an extensive list of collaborations as required for project effectiveness.
- There is very good collaboration and networking within the project.

Question 4: Relevance/potential impact

This project was rated **3.4** for 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 is very relevant to supporting the Program and to advancing progress toward its goals and the FCTO Multi-Year Research, Development, and Demonstration Plan (MYRDDP). The project provides up-to-date cost assessment by integrating very recent technology achievements and is very reactive and flexible to DOE interests, thereby allowing DOE to have very efficient follow-up on costs. The focus on MDVs/HDVs this year is an instance of this flexibility. The project is also helping progress toward MYRDDP objectives by assessing ways to reduce cost to achieve DOE objectives. The project also provides guidance on where future focus (and funding) should be directed. The project has very interesting side studies, such as its study of the impact of recycling on cost and its investigation of end-of-life recycling. The project should also open a study on the second life of a fuel cell as a means of lowering cost and expanding durability. The business model could have interesting results. The project will make great gains in efficiency if it associates with OEMs that can provide a final system vision as the input while keeping the excellent current collaborations.
- Both the relevance and potential impact of this project are strengthened by the addition of MDV and HDV topics and the studies beyond the baseline that analyze multiple cost drivers and pathways. The potential impact of the project is strong with the inclusion of direct input and feedback from the MDV and HDV developers. The shift of focus from LDVs and buses will also allow the project to inform future DOE programs and objectives in the near term.
- This project provides a powerful tool for forecasting the costs for fuel cell systems and allows for systematic updates based on achievements of other projects or of industry interviews. This really contributes to identifying fuel cell system cost drivers to help DOE to refine and update its targets.

- Quantifying cost is critical to demonstrating that DOE targets have been met. In particular, the HDV analysis will be critical in helping establish targets once the TCO issues are better understood.
- The project plays a necessary role to DOE in providing a status update on the key cost metric. Thus it is of more use to the Program and managers than to having direct impact on the technology. However, it has indirect value in identifying promising technology enablers (or, just as important, areas and proposals of limited potential benefit).
- The project is critical to understand the feasibility of DOE targets and guiding future funding opportunity announcements.

Question 5: Proposed future work

This project was rated **3.1** for effective and logical planning.

- Incorporating durability into cost modeling is a very good choice, as are sensitivity analyses for HDVs. There should be a stronger focus on the TCO for HDVs, rather than the laundry list of lower-priority issues listed in the proposed work section (e.g., fluorine recovery, perfluorosulfonic acid synthesis, platinum-group-metal [PGM]-free synthesis).
- The proposed future work represents a comprehensive and ambitious set of analyses. This list should be prioritized with a plan to execute in order of priority, to better address the disclaimer that "any proposed future work is subject to change based on funding levels." It will be informative to further understand if LDV manufacturing can be leveraged for MDVs/HDVs.
- The proposed future work is in line with project objectives. The identified remaining challenges and barriers have been well identified. However, not all of them are considered in the proposed future work, some of which in particular can be considered as crucial for MDV/HDV development.
- The project has played an important role in assessing the potential cost impact benefit of DOE fuel cell projects, but often only when the projects have reached a more mature stage. It would be worthwhile to provide earlier preliminary analysis to help guide and assess the potential benefits of projects' approaches and proposed work.
- The proposed future work is aligned with the current achievements. It lacks ambition toward longer-term goals. Some questions about different investigations exist, for instance, as to whether PGM-free catalysts are realistic for MDV and HDV trucks. Concerning the application, it is not clear where the priorities are in terms of materials or what the sound compromise is between material price and durability.
- Future work should include a more detailed analysis of operating costs. Also, an assessment of the second-life potential of fuel cell stacks could be another interesting contribution of the project.

Project strengths:

- The project has an excellent approach to providing current and future cost projections, integrating the latest technology achievements. The project is very active in setting up relevant collaborations, either to feed and update the model or to have guidance and review on the assumptions and results, which will increase the robustness of the results.
- There is excellent experience in DFMA and cost analysis, with excellent collaboration with the ANL systems modeling work. The project has employed a stable calculation methodology over the years and continuously incorporates feedback from industry and works closely with industry.
- The project demonstrates strong collaboration with broad expertise. This allows the project to conduct studies with impacts beyond the baseline cost; for example, the addition of recycling and disposal cost analysis is very valuable for the developers as both a cost and environmental consideration.
- There is a strong basis for this work. The project has a great approach as well as transparency on its approach and data. The results fill a significant gap that would otherwise be a mystery to the community.
- The team's strengths include its extensive fuel cell cost analysis history, expertise in cost assessment, and list of collaborations.
- The project benefits substantially from the contribution of its industrial partners. The cost projects are extremely valuable.

Project weaknesses:

- In the past, a weakness would have been "access to state-of-the-art materials," but that has been effectively addressed with the Fuel Cell Consortium for Performance and Durability partner projects and strengthened collaboration with the ANL systems analysis project.
- The cost analysis is partially relying on the results of performance and durability modeling from the ANL project. As these results are based on single-cell testing with no validation of the model at stack and system levels, this appears to be the main project weakness.
- Given the collaborations with industrial partners, it would be valuable to understand when the reported production volumes will be met. Moreover, longer-term perspectives (after 2025) might be explored. The expected projection uncertainties should be included in the charts.
- The project should diversify the stack and system suppliers to have less bias on the results. Although there is work done on systems, the project lacks a system vision and guidance. It needs also to update priorities to application needs.
- The proposed future work is ambitious and should be expressly prioritized according to how it best aligns with DOE objectives.
- The project is perhaps taking on too many different topics rather than maintaining a sharper focus.

Recommendations for additions/deletions to project scope:

- The questionnaire is a very good point. It should go further toward actual collaboration with developers. With trucks, for instance, there is a need to update the vision and constraints for truck applications. It is not very relevant for trucks to have Pt-free loading, and it may be that the compromise is more on durability. The powertrain architecture should be extended to the range extender, as this could define the developers' choice to upgrade to fuel cell MDVs and HDVs. If the scenario of a second life for fuel cells is integrated with different applications, an impact on lowering cost and extending durability will be seen. In that case, the durability will be assessed on the whole life cycle of the product. The project team should (as much as possible) diversify stack and system suppliers, including internationals, to have less bias on the cost projections. It could be relevant to assess the impact of operation modes such as start-up and shutdown in terms of "penalty" on durability and cost, as this is part of the real operation of a system. Similarly, as part of real operation, recovery phenomena should be also included (in terms of a "bonus" for durability and cost). The same goes to the break-in and characterizations, which are part of the real operation. Again, collaboration with end users for input on that point could lead to very efficient assumptions.
- MDV/HDV development is currently of high interest, and the project should therefore intensify its investigation by considering the following:
    - Investigating the TCO and not only the fuel cell system cost means taking into account capital expenses and operating expenses, as a huge amount of hydrogen is consumed and system efficiency appears more critical than for LDVs.
    - Potential synergies between the LDV and MDV/HDV components and production plants should be investigated, as should how far, in terms of cost projections, MDV/HDV systems could benefit from LDV developments.
    - There should be a sensitivity analysis on the level of hybridization between the fuel cell and battery on the MDV/HDV TCO.
- Ideally, the focus would be sharper on HDVs in the short term. There should be a stronger focus on TCO for HDVs (meaning the tradeoffs in cost, performance, operating strategy, and durability, perhaps getting more input from the ANL HDV modeling efforts). At this time, it is not clear what the most cost-effective way is to run these systems that have very different tradeoffs from LDVs, and this team has the ability to help make an impact in this area.
- It would be interesting to understand whether the cost of Pt is expected to substantially influence the cost of the system in the mid-future. Given the expected rise in production volume, it would be good to know how platinum costs will change.
- There should be more focus on early cost assessment of other DOE project proposals to better assist in approach directions.
- The project team should explain why the identified system solutions are not currently in models and what additional barriers need to be overcome.

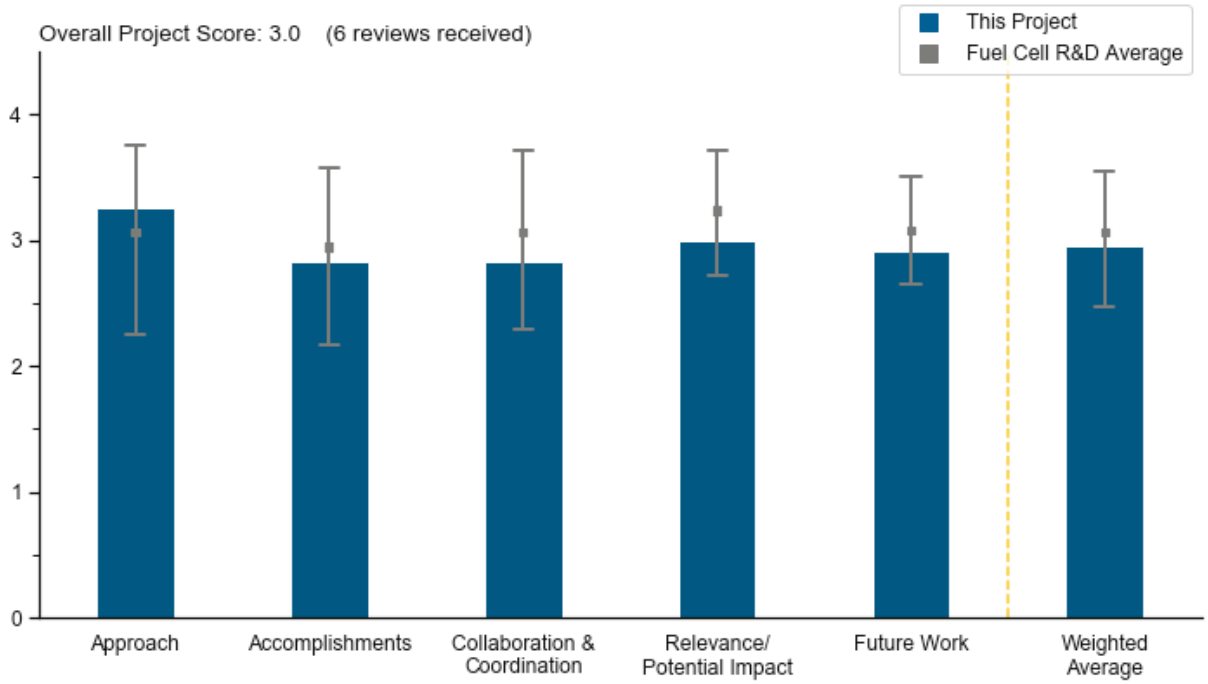
Project #FC-170: ElectroCat: Durable Manganese-Based Platinum-Group-Metal-Free Catalysts for Polymer Electrolyte Membrane Fuel Cells

Hui Xu, Giner, Inc.

Brief Summary of Project:

The project objective is to develop a Mn-based platinum-group-metal-free (PGM-free) catalyst and membrane electrode assembly (MEA) as a replacement for current PGM catalysts. The developed catalyst and MEA will have lower cost/cost volatility, improved corrosion performance, improved de-metalation performance, and reduced membrane degradation compared to the baseline. The developed catalyst and MEA will be tested on a development fuel cell stack.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **3.3** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This project is sharply focused on overcoming critical barriers in the development of PGM-free catalysts for polymer electrolyte membrane fuel cells (PEMFCs). The team combines experimental measurements and theoretical computations to search for better catalysts.
- This project is focused on developing durable, Mn-based, PGM-free catalysts by implementing chemical synthesis in aqueous environments. This would substantially lower the cost of production, with a proposed increase in performance.
- The approach covers necessary development areas such as the study and optimization of the catalyst synthesis and corresponding density functional theory (DFT) modeling, as well as optimization of the electrode.
- The project effectively balances new materials development, characterization, and modeling to advance toward the project's objectives.

- The team appears to be focused on the milestones and pursuing the right approaches (experimental and computational) to further understanding and improve the performance of project materials and MEAs.
- The approach seemed promising, but the execution seems less so. The synthesis and electrode seem to be progressing, but the DFT and diagnostics do not. Also, the assumption that these catalysts do not hurt the membrane has not been proven.

Question 2: Accomplishments and progress

This project was rated **2.8** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Significant progress has been made in material synthesis, as demonstrated by the polyhedron structure for the Mn-N-C catalysts developed in aqueous solution. Moreover, the particle size can be controlled by tuning the concentration of metal salts; particles did not agglomerate, hence preserving morphology after high-temperature treatment. These parameters allow insight into the structure–function correlations of Mn-based PGM-free catalysts. It was found that large particles tend to form optimal electrode pore structure. Another important result, obtained by X-ray diffraction (XRD), shows that the Mn does not affect the crystalline structure of zeolitic imidazolate framework-8 (ZIF-8), and Mn-oxide was not detected. Catalytic performance in RDE testing indicated that half-wave potential reached 0.82 V. After electrochemical evaluations, atomically dispersed and N-coordinated Mn sites, along with porous and partially graphitic carbon, are preserved in the Mn-N-C catalysts, indicating promising durability. A higher degree of graphitization relative to Fe-N-C catalysts may explain the enhanced durability. In terms of MEA performance, there is significant improvement from last year; however, this is still substantially lower when compared to the state-of-the-art (SOA) Fe-based catalysts. All of these achievements are well aligned toward DOE technical targets.
- The team has demonstrated excellent progress toward project objectives through clear and measurable performance indicators (such as activities of catalysts and fuel cell performance). The team has made important contributions to overcoming some critical barriers.
- The project has made good progress toward improving the performance (four-fold) and activity (five-fold). The catalyst's durability in rotating disk electrode (RDE) testing is excellent, but it has not been demonstrated in the MEA.
- The apparent progress in electrode performance is substantial, and there are some catalyst synthesis trends to back it up. However, the quality and consistency of the results are scattered. "Breakthroughs" abound without explanation, and unsupported conclusions have been drawn. Some examples include the N-content by Raman spectroscopy that was claimed but is not an accepted trend.
- It looks like the team did achieve the year 1 goal for activity in an MEA at 0.9 V (10 mA/cm^2), although just barely. The figures showing the impact of things like particle size, ionomers, catalyst synthesis route, etc. are helpful for seeing the range of approaches that the team is pursuing to make improvements. It would be helpful to see a clear statement about the conditions for which it seems it will be possible to hit the targets (i.e., what particle size, what active site density, etc.). The presentation does not appear to have much information on degradation. It appears degradation was not a milestone yet, so this is reasonable, but it would be helpful to see baseline data on durability and degradation.
- The performance gap with the SOA Fe-based catalyst is still significant, as the stability is not superior. The team presented progress of 4 mV less of voltage loss in RDE testing when compared to 2018; this is not convincing because of the lack of batch statistical data.

Question 3: Collaboration and coordination

This project was rated **2.8** for its engagement with and coordination of project partners and interaction with other entities.

- The collaborations between team members are well coordinated, and productive outcomes have been demonstrated.
- The team is well coordinated, and the roles of the participants are clearly defined.

- The collaboration with Giner, Inc. (Giner), SUNY State University of New York at Buffalo (UB), and ElectroCat is good. The project is involved with the University of Pittsburgh, but the actual interaction is unclear; the collaboration with General Motors is also unclear.
- The interactions among the groups in the project appear good. It is somewhat hard to tell if interactions with the wider DOE laboratories are taking place or whether that is important for this team to succeed.
- The team of collaborators is well established; however, it is unclear how the data at Giner and UB compared with each other, especially in tasks where there is duplication of test effort, such as in Task 3.
- While the roles of each collaborator are well described on slide 26 and are logical, it is unclear what specific contributions each organization made.

Question 4: Relevance/potential impact

This project was rated **3.0** for 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 aligns well with the objectives of the DOE Hydrogen and Fuel Cells Program and DOE research, development, and demonstration (RD&D). If successfully conducted, the project will have the potential to advance progress significantly toward DOE RD&D goals and objectives by dramatically reducing the cost of catalysts for PEMFCs. The development of low-cost catalysts is vital to the broader commercialization of PEMFCs.
- If successful, this project may have significant impact on DOE's PGM-free strategy.
- PGM-free catalysts are a major goal, and the best so far are Fe-based, so Mn is a better approach.
- This team is clearly addressing the goal of developing PGM-free oxygen reduction reaction catalysts and electrodes.
- While the project is making good progress toward advancing activity and performance, durability has only been minimally addressed. If the durability of the Mn-based PGM-free catalyst is similar to many other PGM-free catalysts (i.e., low in MEA testing), it is unclear how relevant this class of materials will be toward addressing key commercialization barriers.
- The potential impact of this project is still uncertain owing to the lack of progress in the catalyst's performance and stability.

Question 5: Proposed future work

This project was rated **2.9** for effective and logical planning.

- The project is continuing to advance the catalyst activity and appears to be rightly emphasizing durability.
- The priorities in the proposed future work appear to be good. It is good that studying durability, improving performance, and working on MEAs are all priorities.
- The proposed future work is well aligned with project goals and DOE technical targets.
- The proposed future work focuses on overcoming the remaining technical barriers. The tasks are well thought out and scientifically sound.
- The project should increase its focus on understanding and improving the stability of the proposed formulations, specifically in quantifying the dissolved and oxidized content, in addition to achieving performance targets.
- The general approach makes sense, but the details are lacking. One main issue is that the connection between size and electrode structure is contradictory; on slide 24, the claims around ionomer penetration and coverage do not make sense for a small particle.

Project strengths:

- The project's strengths include its well-defined systems and systematic approach. There is a high level of control of physicochemical parameters that is leading to improved understanding of catalyst performance and is providing guidance for the future work. This is a well-executed team effort with clearly defined achievements.

- The key concept of the project is well conceived, excellent progress has been made so far, and the future work is focusing on the remaining challenges. The results have the potential to reduce the cost of catalysts dramatically for PEMFCs.
- The project's strengths include its alternative approach to the Fe-based catalysts. The team did achieve the year 1 performance goal, and it looks like there is a reasonable balance between performance improvement and scientific work.
- The new approach to non-PGM catalysts appears to be effective. The project has made excellent progress in one year.
- The project has a complex approach to understanding the impact of synthesis conditions on performance.
- The project is showing promising catalyst and electrode improvement.

Project weaknesses:

- It is not clear whether the project is on track to hit its year 2 performance goal. No MEA durability has been shown yet, although it looks like this is not yet a milestone. The team should consider whether the DFT modeling is proving helpful for the synthesis and durability efforts.
- The project has presented many misreported, inconsistent, and "optimistic" conclusions. It also has unclear differentiation from other projects, except for the Mn approach. The DFT contribution is also lacking.
- The claim of improved stability is not supported by any sufficient datasets. There is a lack of statistical data from the catalyst batches and no inductively coupled plasma data on the main leaching elements.
- The project's weaknesses include the poor performance demonstrated in MEA testing. It is hard to envision that any of the PGM-free systems would be able to meet expectations in real systems.
- As of yet, the project has not placed sufficient emphasis on addressing durability.

Recommendations for additions/deletions to project scope:

- The project team should include statistical data on multiple batches and quantify the formulation's stability to acid wash. The team should also address transport losses with high content of an ionomer more specifically related to the MEA structure.
- The project team should add membrane stability testing to confirm that Mn-N-C catalysts do not degrade the electrode. The researchers should also use consistent testing to ensure that their conclusions make sense.
- Extensive durability studies are needed.

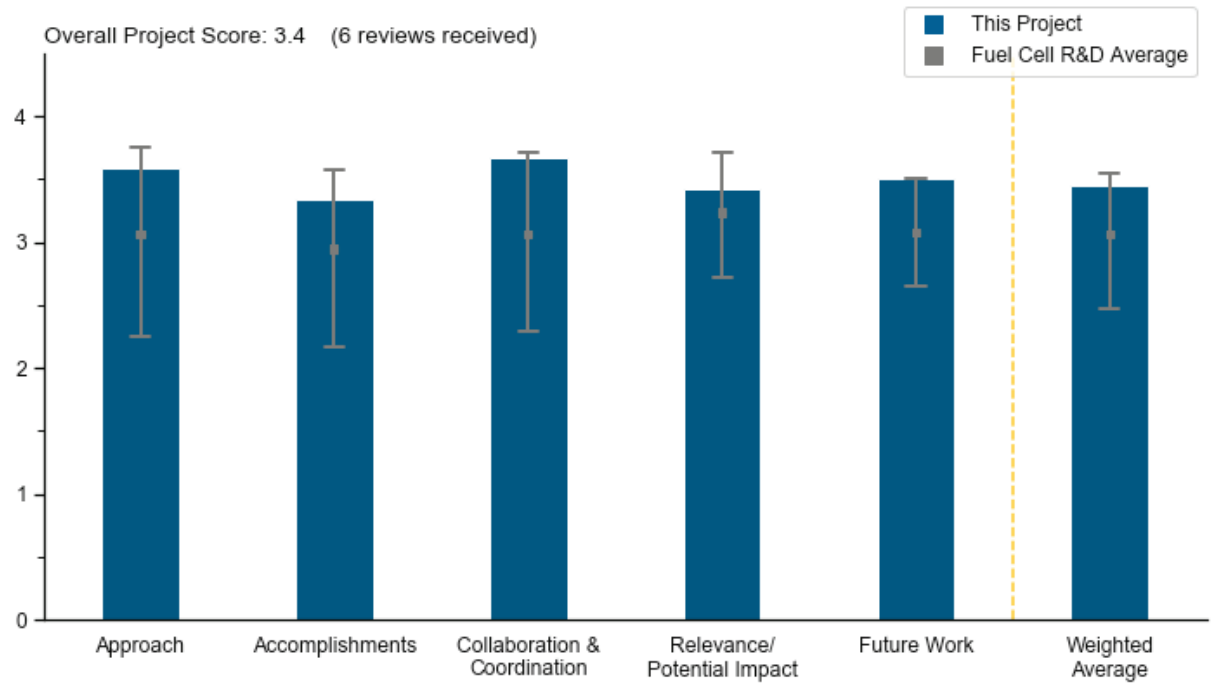
Project #FC-171: ElectroCat: Advanced Platinum-Group-Metal-Free Cathode Engineering for High Power Density and Durability

Shawn Litster, Carnegie Mellon University

Brief Summary of Project

This project is developing platinum-group-metal-free (PGM-free) oxygen reduction reaction catalysts for polymer electrolyte membrane fuel cell cathodes. The team is undertaking a thorough approach that combines advanced, atomically dispersed metal–organic-framework (MOF)-derived Fe-N-C catalysts, PGM-free specific cathode architectures, and advanced ionomers. This project seeks to (1) enable high-power density and improve durability with new cathode structures designed specifically for PGM-free catalysts; (2) increase PGM-free catalyst activity and stability through novel synthesis approaches, including using a simplified, low-cost method; (3) mitigate PGM-free cathode flooding for fast oxygen transport across thick electrodes; and (4) integrate advanced ionomers into the electrode structure for optimal performance and durability.

Project Scoring



Question 1: Approach to performing the work

This project was rated **3.6** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This project effectively combines catalyst synthesis, membrane electrode assembly (MEA) testing, electrode engineering, advanced microstructural characterization, and modeling to demonstrate improvements in PGM-free MEA performance, along with improved understanding of transport and reaction phenomena and structure–function relationships.
- Overall, the approach appears excellent. The use of modeling and numerous experimental techniques is sharply focused on meeting the project goals. The approaches appear appropriate and well organized.
- The project's approach is impressive.
- The project is well organized and covers all the major elements of the catalyst and electrode in a direct, highly focused scientific approach. It would be beneficial to see the group focus on not only the

macroscopic view of the electrode but also a more physical, chemical, and analytical approach for the catalysts. It is suggested that the team look at Mössbauer spectroscopy as a tool for understanding the Fe electronic structure and the impact on electrocatalysis. It would also be good to see the group re-investigate the older, well-known, non-ionomer-based (polytetrafluoroethylene [PTFE]) electrode layer compositions (or hybrid versions) rather than just the current conventional approach.

- The model-based design approach is sound, and it has produced excellent results. It would be good to see less emphasis placed on catalyst activity and more placed on understanding material issues and transport limitations in the electrode.
- The project's approach is to use a state-of-the-art (SOA) catalyst, demonstrate performance, and then scale up the catalyst fabrication process and develop and demonstrate catalyst layers and MEAs that meet performance. An SOA PGM-free catalyst was received and used to demonstrate performance. Good progress has been made at the fundamental level as measured using a rotating disk electrode (RDE), but MEA performance is poor, and stability at the MEA level is very poor. The data suggest issues with both the cathode catalyst layer and the catalyst-layer gas diffusion layer (GDL)–GDL interface. The project needs to optimize these components and their interaction.

Question 2: Accomplishments and progress

This project was rated **3.3** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Compared to the milestones, it appears this project is ahead of schedule. It also looks like some interesting analysis is being done on the durability question, which may lead to strategies to improve durability (i.e., changing carbon phases). There is excellent use of modeling and various types of electrochemical measurements, as well as additional characterization techniques.
- The project is making outstanding progress. Significant accomplishments were made in the past year in improving catalyst and electrode performance. The project is breaking new ground in high MEA performance and is meeting and exceeding its goals. Durability is still a major concern, especially since a large part of the performance improvement appears to be due to the use of higher operating temperature, which could accelerate degradation. The project should be reporting on all MEA performance metrics using the same MEA construction. It appears that the researchers are using different membrane thicknesses and catalyst loadings to optimize MEA performance separately for the high-current and low-current targets; this practice may be common in the field, but it should be discouraged since it makes results less relevant to the real world.
- The results have generated significant progress and therefore have contributed to furthering the advancement toward achieving the goals. The key now is to better understand the implications of the results and refine the model, as well as the experimental plan. As progress is made, the initial challenges change. The efforts required to address the challenges are also in need of refinement. It would be helpful to see how such challenges change over the lifetime of the project. The team could maybe use a spider chart to track progress and include the primary metrics, such as conductivity, catalyst activity, degradation, durability, performance, and even some form of a water management metric.
- The team has made good progress toward its milestones. Moving forward, it is recommended that increased focus be put on the high-current-density, transport-limited regime and less focus be put on catalyst activity.
- This project has met the initial half wave potential goal of 0.87 V versus reversible hydrogen electrode as measured by RDE, but the power produced is well behind that shown by other developers. Roughly 40% of the project has been completed, but only about 25% of the budget was used. This suggests that additional resources are needed to accelerate development to improve the cathode catalyst layer and GDL. High high-frequency resistance (HFR), very high accumulated water-induced mass transport losses, and 30% performance loss in less than 40 hours will each require significant effort to resolve. The authors report reversible and irreversible losses. However, their data show the decay rate after recovering the recoverable losses is much higher than the earlier decay rates, so the reversible losses are not really reversible.
- The electrode development is impressive, as is the catalyst work. It is unclear how much progress is occurring in catalysts, with the overlap with other projects.

Question 3: Collaboration and coordination

This project was rated **3.7** for its engagement with and coordination of project partners and interaction with other entities.

- Fantastic collaboration appears to be the key to this project's success. Outstanding catalysts developed by the University at Buffalo group have contributed greatly to the good results.
- There is good collaboration apparent with all partners, including the Electrocatalysis Consortium (ElectroCat) and others.
- Key personnel need to reach out to teams that are further along in the development of this type of catalyst, such as ElectroCat, Energy Materials Network (EMN), and consortium members, including national laboratories. The proposal clearly and completely defines the roles and contributions of each team member, and the final team, facilities, and equipment required to complete this project are fully in place, ready and available. This project has full commitment from senior management and the corporate officers of its partners.
- The team has strong collaborations among university, industry, and national laboratory partners.
- The project appears well organized, and the team members appear integrated.
- It is not clear how fuel cell initiative changes at 3M will impact the company's involvement in the project. It is not clear that Giner, Inc., is the right partner for synthesis scale-up. This should involve a chemical company.

Question 4: Relevance/potential impact

This project was rated **3.4** for 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 is probably the most promising PGM-free project, with exception of the Fe effect on membranes.
- PGM-free catalysts are critically important for commercializing fuel cells. Fe-doped MOFs have shown significant progress, and additional performance and stability improvements are required. Focusing on reducing HFR, water management, and other mass transport issues, as well as reducing decay, will provide significant benefits for this technology.
- The project is quite relevant, as a successful outcome of all the non-precious-metal development efforts, including this one, will likely make the technology more attractive cost-wise, as well as reduce dependence on platinum sourcing from outside the United States. A successful project will also help redefine the cost and performance mode for all fuel cell applications and provide guidance. It is not clear yet that non-PGM fuel cells will be significantly less costly, because if the current–voltage behavior is lower than that of platinum-based systems, additional membrane surface area may be needed. In other words, membrane costs may increase, as may additional internal cell hardware, to make up for the lower performance of the non-PGM catalysts.
- The project has strong potential impact, as transport limitations are a significant loss for high-loading PGM-free catalysts at high power densities. The focus should continue to be on the nature of these losses.
- The project is clearly focused on DOE goals. Limited durability at this point does raise the question of whether these materials are serious contenders to meet the durability targets, but this project will help answer that question.
- Within the context of PGM-free research, this project is highly relevant and is already having a large impact on the field. However, the relevance of PGM-free catalysts to the industry is still questionable. With the development of extremely low-PGM-loaded MEAs in recent years, PGM cost is no longer the major obstacle that it once was. Furthermore, the durability of the catalysts developed in this project is still far too low to be relevant in transportation or most other applications.

Question 5: Proposed future work

This project was rated **3.5** for effective and logical planning.

- The proposed work is consistent with the development plan and should provide additional insight.
- The future work is solid and well planned. The increased focus on durability is a good idea.
- The project's proposed future work is excellent.
- The goals seem informed by current results and project targets.
- In their future work, the researchers need to reach out to teams that are further along in the development of this type of catalyst, such as ElectroCat, EMN, and consortium members, including national laboratories. The approach and plans should be provided.
- Model-driven design should be more emphasized, with model and theory results leading to improved treatment and understanding of performance and stability. At this point, the description of future efforts is general and could be more precise.

Project strengths:

- The project plan is addressing the right metrics, and the team is doing a good job at focusing on using relevant external testing methods. The team is very good and has a handle on looking at the key variables so as to further the development of the model and PGM-free catalysts.
- This project group is very experienced in working on the scale-up of laboratory processes and in MEA development and fabrication.
- The team has strong background in catalyst synthesis, characterization, and scale-up.
- Overall, the project looks well organized and is making progress toward hitting the technical targets.
- The project's greatest strength is its excellent collaboration with a strong team.
- The project has great methodology and execution.

Project weaknesses:

- It would not be called a weakness, but it would be good to see the team expand beyond the current conventional wisdom regarding electrode composition and structure. The project needs a chemical company at some point to address the synthesis and scale-up of the catalyst.
- A reduced role of modeling is detected in the near future, which reduces the potential for impact.
- The project has a reliance on Fe-based materials and an unclear catalyst focus.
- This project needs to add resources to accelerate the rate of development.

Recommendations for additions/deletions to project scope:

- The following are suggestions for the project. (1) The project should include the evaluation of the Fe-based catalysts using Mössbauer spectroscopy. The impact is on gaining an understanding of the differences in the catalyst. (2) The project should develop a hybrid hydrophobic–hydrophilic electrode composition by determining whether adding some hydrophobic character will address performance. The impact is on water management behavior in the electrode. Alternatively, the team should concentrate the ionomer at the membrane–catalyst interface. (3) The researchers should look at the PTFE content of the microlayer as well. They should even consider removing it and looking at a higher-PTFE GDL character.
- The project should maintain a model-based design approach by continuing modeling activities. The team should compare performance accomplishments to model predictions.
- It may make sense to include other catalysts from other projects.
- There should be more focus on durability.

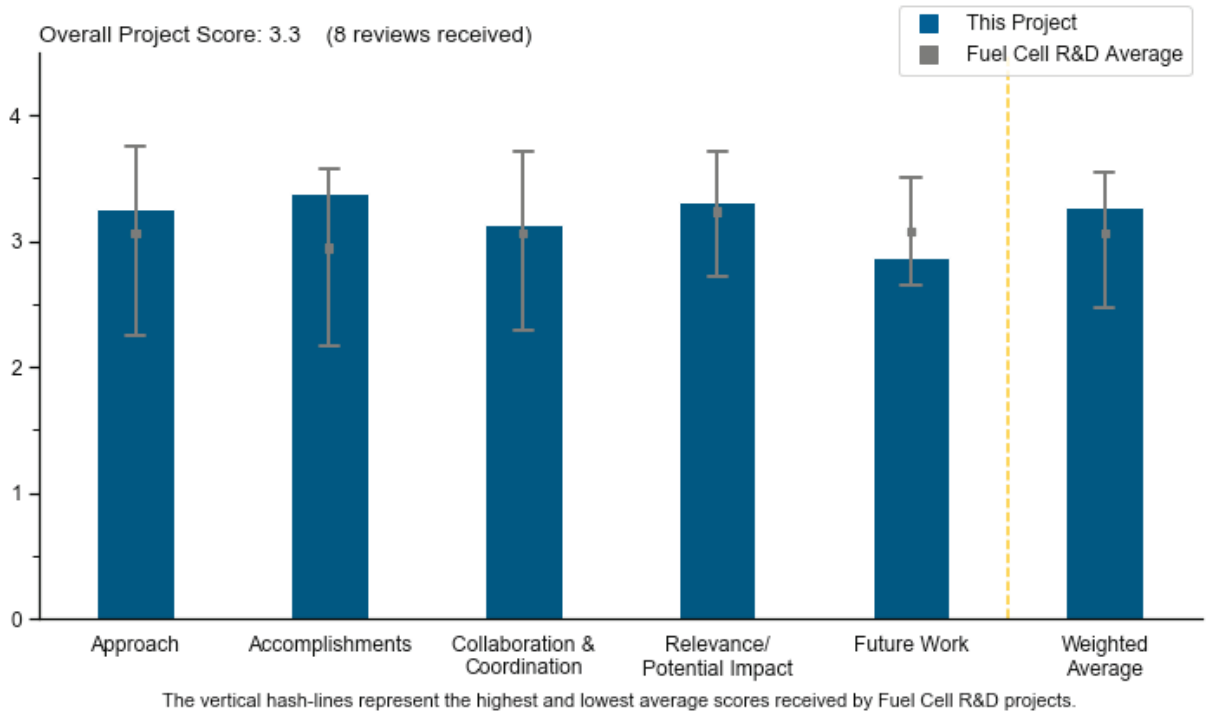
Project #FC-172: ElectroCat: Highly Active and Durable Platinum-Group-Metal-Free Oxygen Reduction Reaction Electrocatalysts through the Synergy of Active Sites

Yuyan Shao, Pacific Northwest National Laboratory

Brief Summary of Project

The project objective is to improve the activity and durability of platinum-group-metal-free (PGM-free) oxygen reduction reaction (ORR) catalysts through dual active sites for enhanced oxygen reduction and hydrogen peroxide (H_2O_2) decomposition. Materials and synthesis innovations include (1) dual active sites for ORR and H_2O_2 and (2) thermal shock activation for high activity through increased active site density. The developed catalysts will lower cost, reduce H_2O_2 formation by 50%, maintain the activity level, and double the durability compared to baseline platinum catalysts.

Project Scoring



Question 1: Approach to performing the work

This project was rated **3.3** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- Use of the dual active sites is a unique and effective approach that can address activity and stability simultaneously.
- The radical scavenger approach is good, and the results are promising. The team needs to further improve membrane electrode assembly (MEA) performance.
- The goal of the project is very clear: increasing the durability of PGM-free catalysts in polymer electrolyte membrane fuel cells (PEMFCs) by reducing the H_2O_2 yield. Although it is inconclusive whether H_2O_2 is the major reason for the poor durability of PGM-free catalysts, recent studies have suggested so. The approach to address this issue is also very clear: creating dual sites with FeNC and MOx that reduce H_2O_2 via the 2X2 mechanism. The results are promising, showing that mixing FeNC with certain metal oxides does increase stability and reduce H_2O_2 yield in rotating disk electrodes (RDEs) and increase the durability

of PEMFCs. Overall, this approach appears to work. What is missing in the logic chain is direct evidence showing that the improved durability in PEMFCs is indeed related to the reduced H_2O_2 yield identified in RDEs. There is also no comparison between the MEAs with or without oxides subject to long-term PEMFC operation. The thermal shock synthesis was not sufficiently justified.

- Peroxide formation likely contributes to the degradation of PGM-free catalysts, and this project seeks to reduce the effects of peroxide formation through the addition of ceria. This project appears to be more focused on reducing degradation in comparison with other work being done on PGM-free catalysts, which is needed. However, it is not clear that ceria decomposes peroxide byproducts significantly faster than the peroxides destroy the active sites. It is strongly recommended that the project team design experiments or perform modeling to determine the extent to which this approach will work. Approaches that prevent peroxide formation may be required to meet the durability requirements for commercial adoption.
- The approach overall appears good. Various types of synthesis are taking place and being translated into MEA performance testing. While the goals are being met, it is a little hard to understand the strategy behind the work for the coming year other than trying out a variety of new synthesis approaches. It would be helpful to see quantitative gaps, and how the proposed approach would be able to meet those gaps. It is not clear that the approach to improving durability will work. The work shows improved durability, but it is still far from the ultimate targets, and simply trying new scavenger compositions is unlikely to address the challenges. It may make sense to try to approach the durability in a more fundamental way to help create ideas that would get closer to achieving the ultimate targets.
- The overall approach is good, and the team seems to be on the right track to address durability issues arising from peroxide radicals. However, the effect of the peroxide radical on catalyst degradation (i.e., CO_2 emissions) and membrane degradation in an MEA is not well elucidated. The team should focus on understanding these effects via a clear design of experiment and the support of good analytical tests. The effects of loading and the tolerance to peroxide radicals due to catalyst loading are also not very clear. Slide 7 shows a good comparison between FeNC and FeNC–NCeOx system durability, but it is difficult to compare the effect of loading, as it was not provided for the FeNC system.
- The project's approach is to use rational design to develop durable, highly active, PGM-free ORR electrocatalysts. The project is also developing dual active sites for ORR and H_2O_2 : MNx and radical scavenger. The thermal shock process has been tested and shown to be ineffective for the removal of Zn. NTaTiOx has shown promise. Modifications are needed to address these processes. Priority should be given to identifying and verifying a workable process.
- Other reviewers may disagree, but the focus on catalytic activity is misplaced. The focus should be on building dual-catalyst functionality and evaluating the effect of this technique on the performance, stability, and chemical environment of the cell. To that end, it is time to drop the thermal shock approach, as it has not contributed significantly to these goals. Also, as a national laboratory is conducting the project, the nature of the "direct loading" approach should be fully revealed.

Question 2: Accomplishments and progress

This project was rated **3.4** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has made significant progress and achievements. The activity already met the final target, and the performance met year 2 milestones. In addition, the durability improved dramatically. The validation of the method of mixing FeNC with metal oxides suggests that peroxide is likely the major cause for the poor durability of FeNC.
- Mass activity reached 35 mA/cm^2 at 0.9 V (H_2/O_2), for which the DOE target is 44 mA/cm^2, and achieved 138 mA/cm^2 at 0.8 V (H_2/air). Both of these are the best in the field.
- The project has made significant progress since the last DOE Hydrogen and Fuel Cells Program (the Program) Annual Merit Review, with more promising fuel cell data. The current density of 30 mA/cm^2 in an H_2/O_2 cell represents a result similar to that of the state-of-the-art. However, at low potential, the polarization current density is still low.
- Relative to the milestones, it appears that progress is excellent. The bigger challenge is around durability, which has not really shown up in the milestones to date.

- The team has made excellent progress in achieving reduced CO_2 emissions in the presence of the scavenger. There has been more limited progress in thermal shock synthesis.
- The project is achieving targets with respect to activity and is showing improvements in durability. Relative to the other ElectroCat projects, these are good results. There is some question of whether the dual-site catalyst is really more stable or only appears more stable because it starts at lower performance (the degradation graph was "normalized"). It would be helpful to show the non-normalized data.
- The project's progress toward overall goals is promising. However, some of the fundamentals are not covered sufficiently. For example, slide 15 does not show the differences between the various thermal shock samples. It is not clear what parameter was changed between samples 1 and 4 that led to either improved or poor RDE performance. Significant improvements that result from any process variables should be accompanied by sound scientific explanations, which are lacking in the presentation slides. No qualitative explanation has been offered to explain the effect of thermal shock.
- This project is underachieving and underspending. Additional resources need to be applied to development tasks. RDE performance on Pt is low, and using that Pt performance gives a claimed delta for Pt performance that is artificially high. There has been significant progress at the RDE level, but there is a long way to go, and the iR-free performance at the MEA level is low. In addition, both the high-frequency resistance and decay are very high. The fundamental causes for these losses are not understood.

Question 3: Collaboration and coordination

This project was rated **3.1** for its engagement with and coordination of project partners and interaction with other entities.

- The team is strong and appropriate to what was proposed for implementation. The collaborations with national laboratories are great. The whole collaboration nicely covers all the necessary aspects, including synthesis, characterizations, modeling, RDE, and MEA evaluations, and these aspects were well integrated. The thermal shock method is new and works in one case. The detection of CO_2 is a good method. X-ray absorption spectroscopy (XAS) is also very informative in detecting the Fe species. However, there is no direct evidence, either microscopic or spectroscopic, for the close proximity of the FeNC site and the metal oxide sites. High-resolution transmission electron microscopy (HRTEM) and XAS may do it.
- This is a good, complementary team. For the next presentation, it is suggested that the project team specify each institute's efforts and achievements.
- The project has good collaboration among multi-institutions.
- The project's coordination appears adequate.
- The presentation could have made clearer what the contributions of Washington University and Ballard Power Systems, Inc. (Ballard) were to the presented results. There is a good integration of ElectroCat resources.
- Pacific Northwest National Laboratory and Washington University appear to be collaborating well. The motivation for the thermal shock approach was not clear, and those results seem to be a separate side project. The motivation for pursuing this approach needs to be described.
- The project's key personnel are qualified, as demonstrated by the completion of previous projects and by their publications on work in this and related fields. The team, facilities, and equipment required to complete this project are not fully applied. This project has the full commitment of senior management and corporate officers of partners.
- The collaboration between organizations is not very clear. It does not seem like some of the expertise that lays within this project team has been efficiently utilized. For example, no explanation has been offered for the improvements observed in hydrogen–air performance with cycling. Moreover, the principal investigator (PI) suggested that the team had not performed the polarization curve tests until equilibrium was reached. This clearly suggests that the expertise of collaborators such as Ballard has not been fully utilized to understand this phenomenon. It seems like these tests were not performed at Ballard.

Question 4: Relevance/potential impact

This project was rated **3.3** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- If this project were successful, there would be potentially very high impact on durability for both PGM and non-PGM catalysts. Detailed understanding is key, however, and the perspective of original equipment manufacturers, who have undoubtedly considered such approaches, will be essential.
- The project shows progress toward improving the activity and durability of PGM-free catalysts. The potential for a breakthrough result that completely solves the degradation issues with these materials seems unlikely, although the approach may contribute to the solution.
- PGM-free catalysts are critical for PEMFCs because the cost and dependence on foreign precious metal can be resolved by a PGM-free catalyst.
- The team is focused on Fuel Cell Technologies Office goals.
- The work has advanced the development of PGM-free catalysts, but the performance needs significant further development.
- Reducing PGM loading is critically important to the successful commercialization of PEMFCs. The potential for this conceptual approach to be successful has not been demonstrated.
- It is not clear if this project is targeting new highly active catalysts or targeting a subset of non-PGM catalysts and working to improve their durability. With many of the slides lacking fundamental explanations for observed improvements or decreases in performance, it is difficult to judge potential impact.

Question 5: Proposed future work

This project was rated **2.9** for effective and logical planning.

- This project's most important contribution to the Program's goals and objectives would be to provide a clear way to improve the durability of PGM-free FeNC catalysts, which would implicate the original causes of the poor durability of FeNC catalysts. The poor durability of FeNC is basically the most important issue and the toughest barrier impeding the commercialization of FeNC catalysts. Mixing oxides mildly reduced the ORR activity of FeNC, but the reason behind this is not given. A counterbalance between activity and durability may be important for the advancement of PGM-free catalysts.
- The catalyst performance has made significant improvement. However, the more stable catalyst has lower MEA or fuel cell performance. Therefore, a new idea on how to improve both stability and performance needs to be addressed.
- The proposed future work should seek to build understanding of these catalytic systems. During optimization, the team should consider both the fractional and absolute loading of the scavenger, as well as the influence of "direct loading" on electrocatalytic activity. Optimization should also include the energy and coulombic efficiency of oxygen reduction since scavenging may represent a loss of electrons. Thermal shock should be de-emphasized since it does not appear promising.
- It is suggested that the project team focus on the MEA ink formulation, which will provide a better understanding of the factors determining performance. The team is encouraged to work on MEA structure characterization to build the property–structure–performance relationship needed to guide MEA development.
- The project's future work needs to focus more on verification of the process that produces a catalyst with the required performance and durability. In addition, MEA scale-up and catalyst layer structural issues are formidable and will require significant additional development attention.
- Better dispersion of ceria is a logical approach, but the team does not appear to have an understanding as to whether this approach will be enough. Modeling and analysis of the degradation mechanism would be a beneficial addition to this project. As the team seems to have strong characterization capabilities, using those to understand degradation mechanisms would help in this field.
- The future work, as outlined on slide 24, is not clear or precise. For example, the optimization parameters that would be targeted to improve performance or durability are not clearly justified with supporting evidence in the content of the slides. It is recommended that the project team refocus on their specific areas

of strength, which seem to be in catalyst synthesis. It would be beneficial to see more catalyst systems investigated using a similar approach to the one taken so far (i.e., creating dual active sites to mitigate radicals).
- The future work appears rather exploratory and is not based on clear hypotheses generated from a fundamental understanding or a quantitative gap analysis. It should be determined whether pursuing scavengers containing Ta will introduce cost challenges and what the criteria will be for selecting scavengers. Overall, the proposed work seems lacking in detail.

Project strengths:

- The overall strength of this project lies in the great performance of the catalysts in terms of both activity and durability, as well as in the validity of the proposed strategy. The reported activity of FeNC is comparable to the state-of-the-art, while durability was dramatically improved, a big deal in the PGM-free field. The proposed strategy is very straightforward and targeted: mixing FeNC and metal oxides. This makes the logic flow and the big picture of this project very clear. Characterizations, including the CO_2 detection and XAS, are very relevant, informative, and insightful.
- This project group is experienced in working in this field, and the project has chosen partners that have been proven capable of successfully completing this type of research.
- The project's strengths include its unique approach of using dual active sites and the complementary team on catalyst synthesis, MEA, and characterization.
- Relative to other work on PGM-free catalysts, this project appears to be focused more on reducing degradation, which is needed more than increasing performance.
- The team has made strong progress with promising results and good impact on durability and decomposition.
- The project has a good idea and has made good progress.
- The project is hitting the performance milestones.
- The strengths of this team are seen in their catalyst synthesis approaches. Although the team has very good partners in Washington University and Ballard, those partners do not seem to have been utilized well for electrode or MEA design and testing. For this project to be successful, the PI would need to engage those partners effectively.

Project weaknesses:

- The primary project weakness seems to be that the overall objectives and targets are not well defined. The team has a good industry partner that can provide guidance and direction to target in order to improve the technology readiness level of this technology.
- To meet development goals, this project needs to apply resources to increase the rate of progress. The processes and procedures needed to verify catalyst layer performance and stability over time at all operating conditions are expected to overwhelm the current level of resources.
- The project needs a more fundamental understanding of the mechanism of dual active sites. It is critical to understand how much H_2O_2 the scavenger can handle, namely, what the scavenger's capacity to stabilize the catalyst might be, as well as whether it would be consumed in the course of fuel cell operation.
- Direct evidence for the adjacent dual sites is missing. It is unclear what kinds of oxides are good choices and can effectively reduce H_2O_2. While the dual-site system works, its potential has not yet been fulfilled. While thermal shocking works for one case, the oxide NTaTiOx is rather unusual, which projects cost concerns.
- The project could use a deeper focus on hypotheses that drive fundamental understanding to make improvements. It is not clear that the durability is on track, although it does not appear to be in the milestones yet.
- More careful performance evaluation is needed. ElectroCat should do this.
- It is not clear whether ceria decomposes peroxide byproducts significantly faster than the peroxides destroy the active sites. The potential for a breakthrough result that completely solves the degradation issues with these materials seems unlikely if only a dual-site approach is pursued. The motivation for the thermal shock approach was not clear, and those results seem to be a separate side project.

- This is not the most transparent project, as some of the new technology is hidden. Relatively little scientific understanding can be gleaned without full disclosure.

Recommendations for additions/deletions to project scope:

- The project team should design and perform experiments or modeling to determine the relative rates of peroxide decomposition on ceria versus carbon corrosion. The team's wide array of characterization techniques should be used to better understand the degradation mechanisms. The team should also show the non-normalized degradation data to prove that the approach is working.
- The team should fulfill the potential of the dual-site system by systematically varying the type of metal oxides, the molar ratio of FeNC and oxide, the catalyst loading, etc. The team needs to understand why some oxides work and some do not, and the project should directly image the adjacent FeNC and oxide sites.
- The team is encouraged to look into the capacity of the scavenger, namely, the mechanism of the scavenger that deals with H_2O_2 and how long it can last in the course of fuel cell operation.
- The project scope may need to be redefined so that the team focuses on its areas of strength, which seem to be catalyst synthesis.
- The thermal shock approach should be rolled back since its ability to contribute to the project is questionable. At this point, catalyst development should focus solely on the effect of the scavenger on performance and durability.

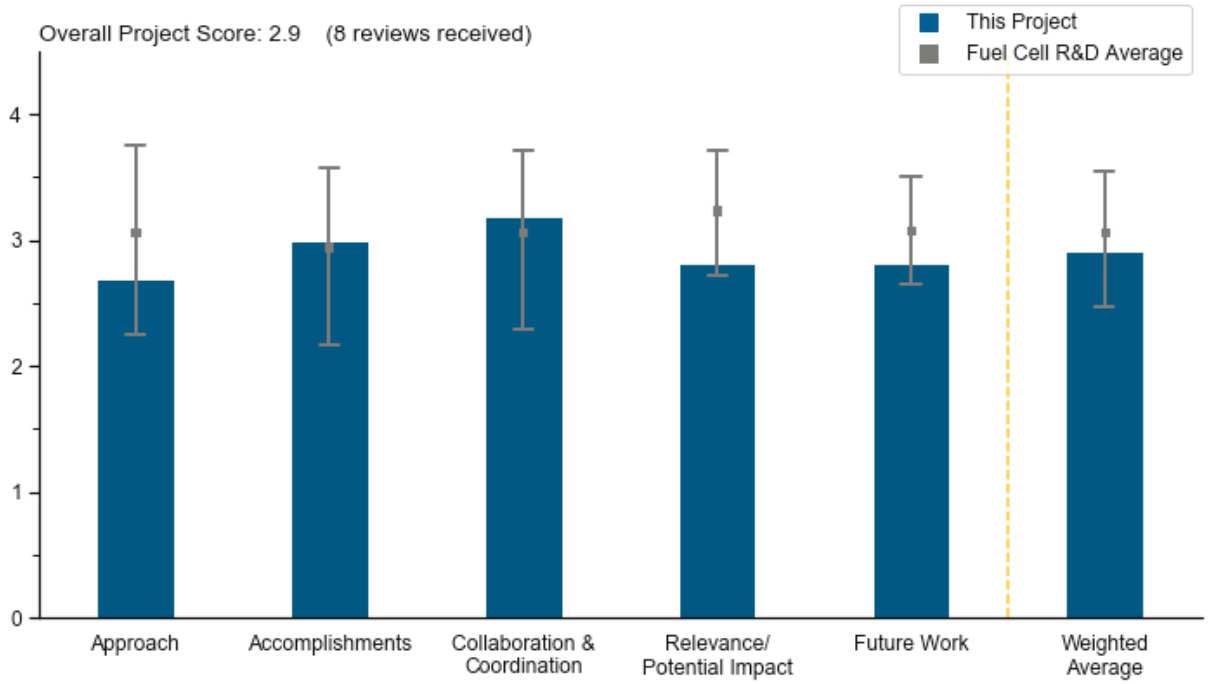
Project #FC-173: ElectroCat: Platinum-Group-Metal-Free Engineered Framework Nanostructure Catalysts

Prabhu Ganesan, Greenway Energy, LLC

Brief Summary of Project

The project objective is to develop durable, highly active electrocatalysts for the oxygen reduction reaction (ORR) through a unique, bottom-up, rational design to enable a better understanding of the platinum-group-metal-free (PGM-free) active sites and improve activity. Fiscal year (FY) 2019 objectives include (1) catalyst development based on high-surface-area polymers, (2) membrane electrode assembly (MEA) optimization and fuel cell testing, and (3) achievement of 25 mA/cm^2 in a H_2–O_2 fuel cell test at 80°C and 100 kPa.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **2.7** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The plan to improve catalyst activity was presented and is feasible. The team also appears to have an understanding of the primary issue with low MEA performance. The area that is most lacking from the approach is the identification of durability barriers and a strategy for addressing durability. The team's strong modeling could be applied toward helping understand degradation mechanisms. Even if the team cannot match "world-record" PGM-free performance, providing insight into degradation mechanisms would be beneficial. The team is encouraged to focus on non-Fe catalysts to improve durability.
- The project's approach is to develop durable, highly active, PGM-free oxygen reduction electrocatalysts using rational design. The project is structured to focus on hydrogen and oxygen performance using fully humidified reactants. The performance goals for hydrogen and oxygen were met, but the decay is very high, with 60% loss in four cycles. The development progress and spending are on schedule, and milestones have been met for the hydrogen–oxygen effort. The fact that the development efforts have been

almost exclusively on hydrogen–oxygen tests to date is a significant concern. The ultimate requirement is a hydrogen–air MEA.

- The polyporphyrin approach represents a good direction. The synthesis support from Northwestern University (NU) is also very good. The catalyst performance progress is slow.
- There is an adequate combination of synthesis, half-cell testing, MEA testing, and computational analysis. It is hard to see how this will improve upon already existing PGM-free catalyst materials.
- The objective of this experiment and modeling project is to develop durable, highly active, PGM-free oxygen reduction catalysts for hydrogen and air fuel cells using a rational design approach. The project began on September 1, 2017, and is scheduled to end on December 31, 2020, so it is about 50% completed. The principal investigator (PI) has developed catalysts based on high-surface-area polymers that are pyrolyzed. Experimental results were shown of poly-phenylporphyrin and a zeolitic-imidazolate-framework-based (ZIF-based) Fe/MN catalyst with unusually large particles (200–300 nm) and low metal loading (0.08–0.35 at%). On the other hand, the surface area of this catalyst after pyrolysis was very high, at 1263 m^2/g. The project examined the composition and morphology of these Fe-based catalysts using electron energy-loss spectroscopy (EELS) and x-ray absorption near edge structure (XANES). There was also a modeling component to the project. The team demonstrated active site and reaction pathway modeling for Fe-porphyrin and FeN_4 in graphene, but it was not clear how the modeling work is giving the PI new insights for improved catalyst performance. There was also some fuel cell testing of poly-phenylporphyrin. While the FY 2018 target for fuel cell activity was met, catalyst stability and durability remain issues. High degradation rates were observed at a voltage hold of 0.4 V, with less degradation during a potential hold at 0.7 V. It is not clear how or whether the durability will be improved. Modifications were made to the polyporphyrin synthesis to improve fuel cell performance, but details were not given at the Hydrogen and Fuel Cells Program (the Program) Annual Merit Review (AMR). It appears that part of the low-fuel-cell-power problem is due to a poorly designed cathode with the PI's PGM-free catalyst. It is recommended that the PI devote more time to proper fuel cell MEA preparation.
- It was difficult to see what actual approach is being used for what problem in FY 2019. On slide 4, four red dotted boxes were used to highlight (1) synthesized porphyrinic polymers, (2) added heteroatom and peripheral functionalities, (3) a proposed model for active sites, and (4) the optimized MEA for optimized catalysts. It is unclear whether all of these are for increasing surface area, for increasing the density of active sites or activity per site, for improving durability, or for identifying the key issues. Apparently, the team is trying to answer many challenging questions. However, the approach seems not clearly described.
- Some project targets are set, and progress is being made, for example, with the 900 mV iR-free oxygen performance. However, some other significant targets have had 0% progress, with the project 40% complete. For example, no progress has been made with the more realistic power density target set for air operation at 800 mV cell voltage, with a current density target of >150 mA/cm^2 on air; not even one chart of air performance was shown. The last point is about durability: no target has even been set for the project.
- Much investment has been put into this one material for some time. It is clear that it is of no practical use without magnitudes of improvement in stability. The focus should have been put in only two areas: determining the degradation mechanism and whether it can be prevented. Any other area is a waste of funding.

Question 2: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The team has made progress toward meeting the 2020 performance target for MEAs operating on oxygen.
- In FY 2019, the project team met the MEA activity target of 30 mA/cm^2 at 900 mV iR-free cell voltage. The rotating disk electrode test result was improved from 2.03 to 2.78 mA/cm^2 at 800 mV, exceeding the 2019 target of 2.0 mA/cm^2. This is significant progress. However, the performance enhancement factor was not clearly identified, and durability needs to be improved.
- While the catalyst requires additional development efforts for operating on hydrogen–oxygen, the additional known issues that involve the cathode catalyst layer suggest that early testing on hydrogen–air should be a major part of development, even at this early stage. Mass transport and even some portions of kinetic losses are known to be significantly more severe when operating on air. An early understanding of

the fundamental mechanisms associated with operating on air should be used to guide the catalyst development. The ultimate requirement for the catalyst wettability may impose requirements on the catalyst fabrication process.

- The PI has made good progress regarding the activity of his PGM-free catalysts. Durability tests were performed this past year, as suggested by a 2018 AMR reviewer, but the results were not good. There was a significant drop in current density during a 5 hour 0.4 V hold, with less current loss over a long period of time when the potential was fixed at 0.7 V. This means that the durability issue was quantified via direct fuel cell performance data, but it has not yet been corrected.
- The team is making progress toward improving activity and appears to be meeting the project targets in that regard. However, highly active catalysts that rapidly degrade are not useful for any applications of interest to DOE. More progress and focus are needed to address degradation.
- Good progress has been made for the current density target at 0.9 V. However, the team is strongly encouraged to focus on developing the precursor conversion to reap the benefit of the precursor.
- The project's activity targets have been met. The durability of the material at potentials where H_2-O_2 is evolved is limiting.
- The project has made good progress from this point last year. It would be good to see more results on durability.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- The final team, facilities, and equipment required to complete this project are in place, ready, and available. The team's personnel are qualified, as demonstrated by the successful completion of previous projects and publications. This project has full commitment from senior management and corporate officers of partners. Ample facilities are available to support and complete this work.
- The collaboration and partnership with Savannah River National Laboratory (SRNL) and ElectroCat appear quite strong. It is not clear whether Ballard Power Systems (Ballard) has contributed significantly to the project. NU provided characterization. Overall, the project appears to be a collaborative effort.
- The results were obtained from collaborative efforts. The lack of contribution from Ballard (who is a no-cost partner) is understandable at this stage. Studies using modeling may aim more at the materials on trend to guide the search for suitable heteroatoms.
- A wide variety of partners is on the project, from small businesses (including the lead) to national laboratories, universities, and a leader in polymer electrolyte membrane fuel cells (PEMFCs) (Ballard). However, no data were contributed from Ballard, whose role is the "evaluation of promising electrocatalysts."
- This project has very good multilaboratory collaboration. The presentation showed that the project has leveraged the works from many laboratories, including ElectroCat. However, it is not clear whether the lead institution has made a major impact.
- This is a focused and capable team.
- The collaboration is adequate.
- The collaboration and coordination of the various team members were somewhat weak and not clearly described in the AMR slides. The modeling work and results seem separate from the catalyst development efforts. The modeling results indicate a specific improvement in ORR kinetics (e.g., a new reaction pathway for FeN_4 on graphene). This will be important only if the PI can see and/or monitor the reaction pathway. Otherwise, he will not know whether such a new pathway exists. Similarly, it is unclear how the PI will modify the catalyst synthesis to create additional open Fe axial sites, as suggested on slide 12. The specific accomplishments of the NU researchers were not clearly identified; slide 6 indicated that they prepared a poly-azoporphyrin catalyst, but the properties and activity of this catalyst were not discussed. It is not clear how California State University, Northridge, has contributed to the project or whether there are plans for Ballard to perform experiments in year 3. Al Anderson and Edward Holby seem to be doing modeling work, but it is unclear whether these two researchers are working together or separately.

Question 4: Relevance/potential impact

This project was rated **2.8** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- There is a need to develop high-activity, PGM-free oxygen reduction catalysts for hydrogen–air fuel cells.
- Reducing PGM loading is critically important to the successful commercialization of PEMFCs.
- The project objective is relevant to the long-term goal of eliminating the use of PGMs for a large impact on cost reduction. It is still unclear that significant advances can be made toward this goal, as the durability results add more challenges. Ultralow-PGM catalysts, with non-PGM active sites on support, may be more promising.
- The project shows progress toward improving the activity of PGM-free catalysts. The approach for addressing degradation issues was not presented. To be useful for any DOE-relevant applications, orders of magnitude of improvement are needed in the durability of PGM-free catalysts. The team's modeling and approach for preparing catalysts could be directed toward exploring degradation mechanisms and generating ideas for addressing durability, but it is unclear whether the project will attempt this.
- DOE has a subset of targets and goals for PGM-free catalysts, and this project has hit one of those targets; however, it still appears to be far off from the others. To make a meaningful contribution to the overall goal of the Program, much progress still needs to happen.
- Non-PGM catalysts have significant promise for future fuel cell integration. However, it is hard to see what this project will add to the already existing knowledge and materials for non-PGM catalysts.
- Eliminating PGMs does not necessarily mean lower cost. Because the cost of all other components combined is large, the cathode catalyst with the highest activity, power, and durability will make for the most cost-effective fuel cell stack. The current material is Fe, which is not compatible with PEMFCs. There is very low stability and usable activity.
- The PGM-free catalyst performance from this project needs to catch up quickly with the results from other projects.

Question 5: Proposed future work

This project was rated **2.8** for effective and logical planning.

- The proposed future work appears to be adequate to meet the specified targets.
- It is not clear what the lead organization will contribute in regard to the future work. Polyporphyrin still represents a promising direction. The team should give more focus to MEA performance optimization at both high-V and high-I regions.
- To be fair, the presentation ran long, and the future plans were not presented in detail. The plan for improving MEA performance was logical. A plan should be added for addressing durability that combines modeling and the many parameters that the team can control with catalyst synthesis. Even if the team cannot solve degradation, presenting a better understanding of degradation would move this field forward.
- For future work, the PI proposes to explore new catalysts (i.e., different porphyrin links and the use of different transition metal centers). It is not clear how this work will improve catalyst durability, which is the number one issue with the PI's catalysts right now. Early on, during year 3 of this project, the PI should down-select one or two catalysts for MEA optimization and fuel cell testing, with durability as the focus. The modeling work should be de-emphasized unless the PI can show how it is directing or steering research work.
- The proposed future work appears to be more of the same. A durability target appears to be missing in the work. The approach should have a durability go/no-go decision point; if the material were fundamentally flawed with respect to durability, then future work on the material would be questionable.
- Future work on this project needs to devote significantly more effort to fabricating and testing MEAs on hydrogen–air.
- It would be good to see more work on durability.
- The proposed future work is mainly a continuation of multiple tasks. It is unclear how the electrochemically active surface areas were determined. Studies using modeling could screen metals as possible active centers.

Project strengths:

- The team is making progress toward improving activity and appears to be meeting the project targets in that regard. The collaboration and partnership with SRNL and ElectroCat appear quite strong. The modeling results were insightful.
- The PI has made highly active catalysts. The end-of-project catalyst activity target (30 mA/cm^2 at 900 $mV_{IR\text{-}free}$) has been met. This is the primary strength of the project.
- This project group is capable of conducting the research and development on PGM-free catalysts. Scaling up and verifying the stability of hydrogen–air is likely to present significant challenges.
- The project team has met performance targets and demonstrated the need to address catalyst durability at high current densities.
- The project addresses a challenging subject. The team is well qualified to carry out the proposed tasks.
- The novel PGM-free material is making progress on oxygen power density.
- There is very good collaboration in this project.
- This is a focused team.

Project weaknesses:

- The durability of the PI's catalyst is poor. The project team did not present a path forward for rectifying the durability problem. The PI discussed only "modification" of the catalyst, with no details. Cathodes with the PI's catalyst in a fuel cell MEA performed poorly. It is not clear how the results of the modeling work will be translated into new catalyst design. For example, it is not clear how new catalyst designs in year 3 will target open Fe axial sites, as per slide 12.
- The direct link between the computational work and the synthesis and experimental work is not very clear. Material durability is a critical weakness that could be difficult to fix.
- Testing MEAs on hydrogen–air should be given priority.
- There did not appear to be sufficient focus on understanding and addressing catalyst degradation.
- More work should be put toward the weaknesses of durability and air power density.
- The project has not led with strongly innovative ideas so far, which may take time and more experience.
- There has been slow progress.

Recommendations for additions/deletions to project scope:

- The team's strong modeling and the flexibility of the catalyst synthesis approach should be applied more to help understand degradation mechanisms. The team should consider non-Fe metals, such as Co or Mn, to avoid membrane degradation. Even if the catalysts are atomically dispersed, the Fe is likely released through degradation.
- It is recommended that the PI devote more time to (1) optimizing electrode and MEA fabrication, (2) testing the catalyst in a fuel cell under different temperature, relative humidity, and gas flow conditions, and (3) improving the durability of catalysts in fuel cells. The project team needs to collaborate better and coordinate tasks more effectively. There should be better utilization of talents in year 3, and the PI should better explain the contributions of each team member in next year's AMR presentation.
- It would be beneficial to see more durability work.
- The project needs a go/no-go decision point for durability.
- The team should propose strategies for improving durability.
- The cause of activity decay should be studied.

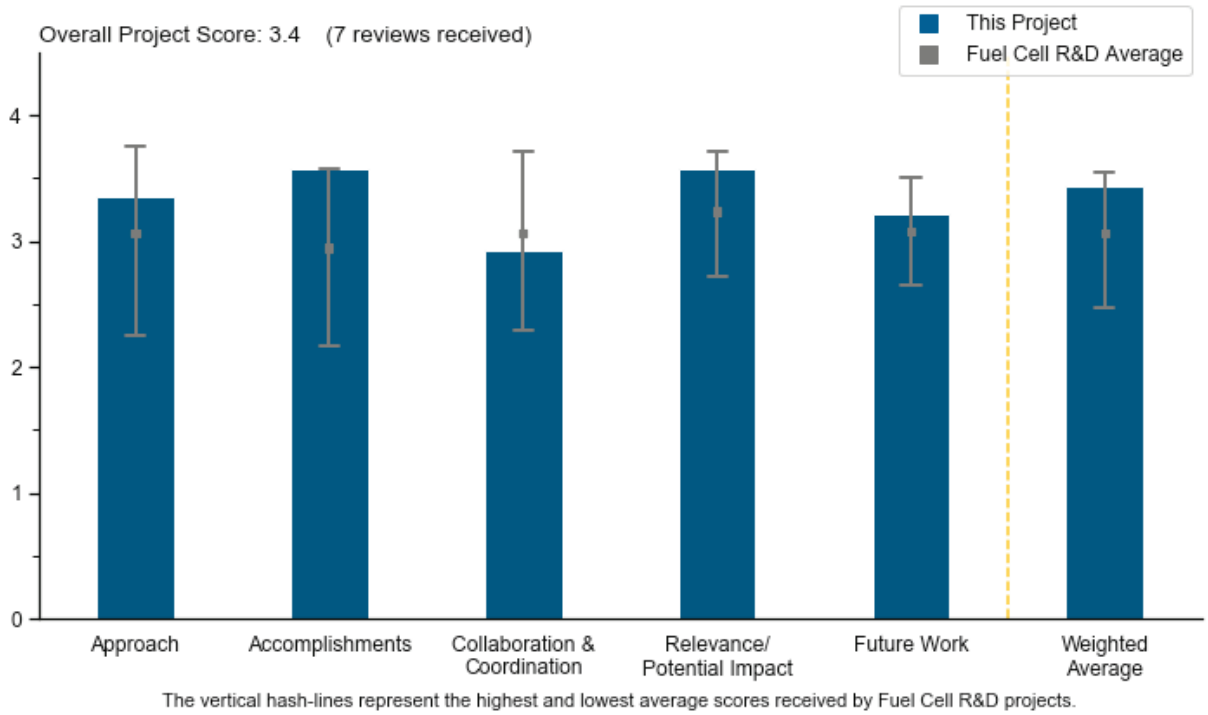
Project #FC-174: Highly Efficient and Durable Cathode Catalyst with Ultralow Platinum Loading through Synergetic Platinum-/Platinum-Group-Metal-Free Catalytic Interaction

Di-Jia Liu, Argonne National Laboratory

Brief Summary of Project

The project objective is to develop ultralow-platinum@platinum-group-metal-free (PGM-free) nanofiber cathode catalysts that achieve all U.S. Department of Energy (DOE) fuel cell catalyst/membrane electrode assembly (MEA) performance metrics, particularly in the high-current/power-density region. The approach reduces platinum usage through synergistic interaction between ultralow-platinum and PGM-free sites by improving catalyst activity and durability/transport by using a porous nanofibrous network catalyst support instead of a conventional carbon support.

Project Scoring



Question 1: Approach to performing the work

This project was rated **3.4** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The project approach is focused on development of an ultralow-Pt-loading cathode catalyst that can achieve DOE targets at high current densities. This unique approach is very well thought out, with well-defined direction to move forward.
- Targeting the durability and mass/charge transport barrier through utilizing a porous nanofibrous network allows for high specific surface area. Using metal–organic-framework (MOF)-based synthesis that allows for use of industrial processes provides flexibility and scalability to the material.
- This project demonstrates a promising approach combining a PGM-free catalyst support produced by electrospinning and ultra-low loadings of Pt-Co catalyst.

- This low-Pt approach with the synergetic PGM-free site is a very effective way to meet all the DOE targets on an MEA level.
- The idea of using PGM-free supports to create an ultralow-Pt electrode is worthwhile, especially in the context of an ordered support such as a MOF, which would be expected to distribute Pt uniformly. The use of electrospinning to generate a more hierarchical electrode is interesting, but it would also be good to see evidence of why mass and charge transfers were expected to be enhanced. Enhancements for surface area and corrosion resistance are more obvious. Since PGM-free supports are known to generate hydrogen peroxide, it would have been good to have seen how this could be addressed in the initial approach to the project. It should have been understood at the beginning that this would need to be mitigated.
- The approach is somewhat duplicative of multiple approaches: high-surface-area materials, in situ Pt-Co alloy formation, etc. The project approach is very much similar to what Popov has done. Further, actual details on the approach are unclear and were not clarified during the question-and-answer (Q&A) session. However, project results are promising.
- This work seems diffuse with multiple non-overlapping goals. A focus on the synergistic catalyst effects is recommended.

Question 2: Accomplishments and progress

This project was rated **3.6** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- To date, overall accomplishments have been excellent. In-depth material analysis and testing clearly helps evolve an understanding of what the material is doing and how it behaves under various conditions. This is essential to advancing and progressing to a more mature and finished product that can be transitioned into functional fuel cell stack hardware.
- The project results are unbelievably good. For example, polarization curves on slide 17 are nearly perfectly straight in air, which is something even original equipment manufacturers (OEMs) cannot achieve.
- The results on a single selected MEA met all the DOE targets. The project accomplishments are excellent.
- The approach has yielded exceptionally high mass activity with low loading through a synergistic effect between the support and catalyst. The power density is still lower than desired. Given the very low loading, there could be a modest increase to reach power density targets while still having a very low MEA PGM content.
- The team has made good progress toward milestones and has generated interesting insight into catalyst mechanisms.
- The project has clearly shown the ability to achieve extremely high mass activities, and the polarizations are beginning to achieve 1000 mW/cm^2 under the heat rejection limit (0.675 V). Durability to 30 K electrocatalyst cycles has also been shown for both nanofiber and non-nanofiber electrodes. However, the nanofiber electrodes are having difficulty meeting high power density. The Q&A session revealed that there may still be some robustness issues related to peroxide generation at high temperature and low relative humidity. Electrodes have not yet been tested at low temperatures. No support corrosion accelerated stress test (AST) data were shown this year.
- The project met all 2025 DOE targets in terms of catalyst and MEA performance.

Question 3: Collaboration and coordination

This project was rated **2.9** for its engagement with and coordination of project partners and interaction with other entities.

- Collaboration on this level of work is acceptable and seems highly coordinated. Additional collaboration partners who are interested in moving the technology to the next level would be ideal.
- There seems to be a good collaboration with Purdue University on modeling the interactions between the catalyst and support. It is important that the project partner with an OEM for optimization of the MEA for power density.
- The team has incorporated collaboration with university partners, particularly in modeling.
- There is a well-coordinated effort between experimental and modeling teams.

- It appears that Purdue University helped on the modeling that indicated that hydrogen peroxide should theoretically be reduced by nearby Pt, but very little evidence exists of other outside collaborations. The information presented does not clearly show where there are collaborations with the Fuel Cell Consortium for Performance and Durability (FC-PAD), nor what the impact was.
- Validation of performance from the industry sector would be wonderful.
- Project collaboration is not clear.

Question 4: Relevance/potential impact

This project was rated **3.6** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The project offers a novel, very promising, and scalable approach for making an ultralow-Pt/PGM-free catalyst that already meets all 2025 DOE goals in terms of performance. The project has potential to advance polymer electrolyte membrane fuel cell technology.
- This project is quite relevant to the DOE strategy of using low-Pt or PGM-free catalysts. The demonstration from the results shows the potential impact on getting a high-performance MEA with low Pt loading.
- The project work performed, and results shown, indicate a clear and impactful potential contribution to the industry. It could be quite a huge step if the material can be manufactured in a more cost-effective manner than traditional fuel cell stacks while maintaining performance.
- The development of a high-activity, durable catalyst is central to the mission of the DOE Hydrogen and Fuel Cells Program. This project features catalysts that are able to achieve high activity and durability. Support corrosion metrics and status appear to be missing from the target table. Robustness metrics are missing from the target table.
- The project has potential for significant impact. The proposed approach requires a technical assessment to evaluate cost. However, this would be beyond the scope of the presently funded work.
- This project can potentially have high impact if the proposed mechanism of synergy can be proven.
- The targets of performance and costs are highlighted.

Question 5: Proposed future work

This project was rated **3.2** for effective and logical planning.

- Testing the material at high temperature is a highly relevant and important next step to improving overall fuel cell performance. Each step in the proposed plan is logical and will provide useful information.
- The future plan is well balanced between catalyst development, integration into MEAs with different types of membranes, and MEA independent validation.
- The future work slide consists of six bullet points, two of which are mystifying: the use of high-temperature membranes and thin membranes. This seems to be a tangent, considering that not all the work with the catalyst is complete. The project scope should focus on the catalyst; if the membrane directly affects the catalyst, then membrane work could be justified, but that does not appear to be the case. Unlike a company such as 3M or Plug Power, Argonne National Laboratory has not and should not be in the business of developing MEAs. Rebalancing the platinum from anode to cathode is a fair future task, as well as further optimizing the catalyst layers. Addressing robustness to peroxide formation in dry conditions and observing performance at low temperature and wet conditions should probably be emphasized more in the future work than it is.
- The future work plan can be improved by focus on mechanistic understanding and reduced consideration of MEA issues.
- There are too many avenues of future work proposed. Key efforts would be the optimization of cathode loading for power density and PGM content as well as the National Renewable Energy Laboratory testing.
- The future work should focus on validating the performance in H_2–air MEAs.
- Degradation product studies are needed to confirm stability.

Project strengths:

- The project team demonstrates excellent material and catalyst analysis. Multiple different approaches were analyzed and compared against a state-of-the-art material. There is a clear and logical proposed path forward.
- The project proposes a unique approach to make catalyst and support in a one-pot synthesis. Flexibility of the synthesis allows one to control support porosity and also catalyst activity and durability at the same time. Modeling is used as a tool for understanding synergetic interactions between the catalyst and support.
- The team has made an excellent start on potentially exciting scientific contributions. The following are project strengths: (1) the project met the DOE targets on the H_2–air fuel cell stack, (2), the project discovered synergetic effects on the Pt_3Co and CoN_4 PGM-free site, and (3) the laboratory met DOE targets with ultralow Pt loading using a MOF frame.
- The project has demonstrated a catalyst capable of extremely high mass activity. The project has shown durability with respect to the electrocatalyst AST within a fuel cell. The project began with a novel and ordered concept: the integration of platinum into a PGM-free catalyst that has its own activity, as well as a set structure.
- The project strength is the combination of Pt and PGM-free catalyst supports to achieve high mass activity with reasonable power density.
- The project team demonstrates good performance.

Project weaknesses:

- The project still needs to address robustness to a wide array of automotive conditions, including dry conditions and wet. The project sorely lacks collaboration, especially with industrial partners. Generation of hydrogen peroxide under certain conditions could become a showstopper under realistic automotive conditions.
- Project claims are overstated. The project approach combined several approaches that have limitations but does not address them. Benchmarking is suspect.
- The following are project weaknesses: (1) cobalt nanoparticle dissolution could degrade the Nafion™ film, (2) the understanding of this synergistic effect must be clearer, and (3) H_2–air high current performance is still poor.
- The weakness of the proposed synthetic approach is the limited ability to control the amount of cobalt nanoparticles that leach and degrade fuel cell performance.
- A current weakness is the below-target power density.
- The work plan for this project is too diffuse and should be more focused.
- An interested transition party would round out the project as complete.

Recommendations for additions/deletions to project scope:

- Future work associated with novel membranes should be deleted. Industrial partnerships should be added to understand robustness needs and issues that arise under realistic mobile application operation. Material should be added to slides related to the batch size and scale of manufacturing achieved to date.
- Electrode stability by FC-PAD is needed to confirm the issues others reported with this approach (membrane degradation, Co and Pt leaching, and migration).
- The proposed mechanism should involve copious generation of desorbed peroxide. Rotating ring disk measurements are strongly encouraged.
- Additions to project scope would be validating the low Pt loading and the high performance through collaboration with automotive OEMs.
- The project should evaluate the effect of cobalt leaching from cobalt nanoparticles on fuel cell performance.

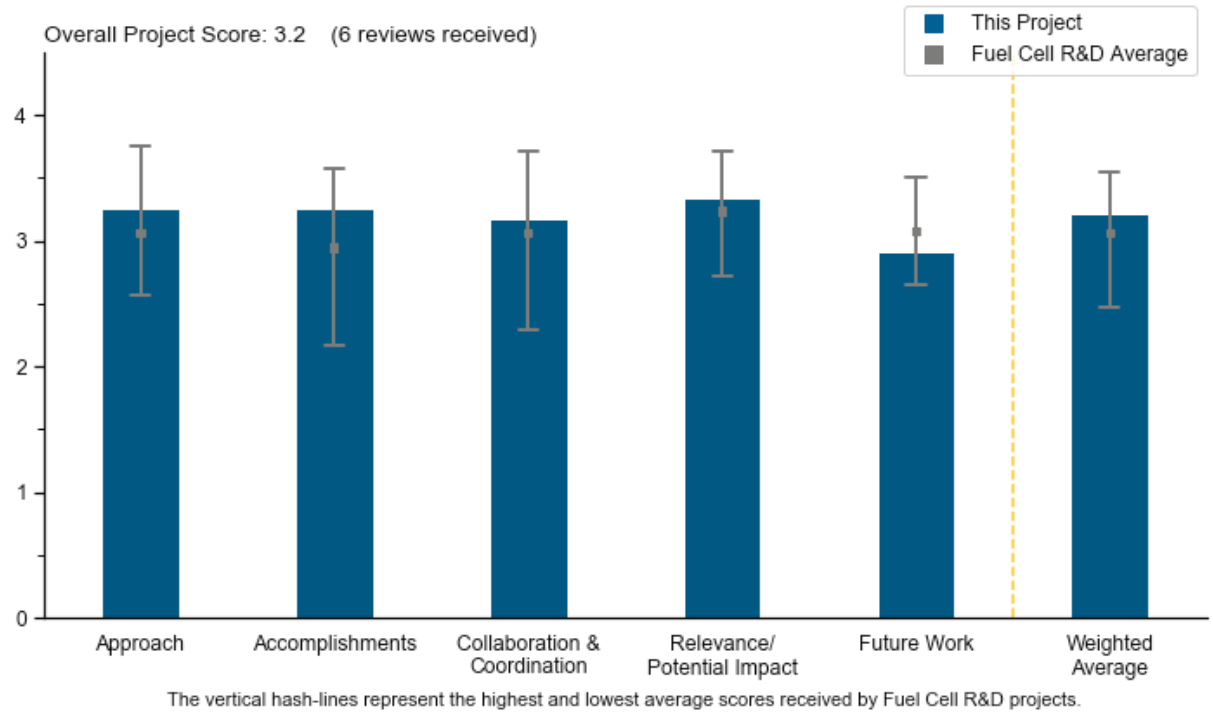
Project #FC-178: Lab Call Fiscal Year 2018 (Membrane): Spirocyclic Anion Exchange Membranes for Improved Performance and Durability

Bryan Pivovar, National Renewable Energy Laboratory

Brief Summary of Project

Alkaline exchange membranes continue to be challenged with cation degradation at high temperature and pH conditions. State-of-the-art trimethyl ammonium cations exhibit limited durability under fuel cell operating conditions. Research has indicated that cations with a spirocyclic structure have improved durability. The project is incorporating spirocyclic ammonium cations into alkaline exchange membranes to improve membrane durability. The project addresses the barriers of cost, performance, and durability by aiming to (1) synthesize novel spirocyclic ionomers to membranes and ionomers in alkaline membrane fuel cells (AMFCs), and (2) optimize performance to meet the U.S. Department of Energy's AMFC targets.

Project Scoring



Question 1: Approach to performing the work

This project was rated **3.3** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This project is positioned well to leverage related projects at the National Renewable Energy Laboratory (NREL). There is little chance that the synthesis will not work as planned, and the new spirocyclic materials can be tested using the same protocols used for other anion exchange membrane (AEM) projects. The objectives and barriers are clearly defined.
- The team is extremely competent and experienced. The fundamental approach is excellent. The development of tasks and sequence of work is clearly well thought out and well executed.
- The project's approach is to identify and produce spirocyclic, ammonium-cation-based AEMs that show increased stability in alkaline environments. The project is running parallel tracks: one to characterize membranes and perform accelerated aging, and the other to develop membrane electrode assemblies (MEAs) and test fuel cell performance.

- The current project is based on previous work done on multiblock copolymers of polydiallylpiperidinium segments in a polysulfone backbone, with the goal of scaling the synthetic procedure for the production of AEMs for long-time testing.
- This project is focused on evaluating polymer chemistries based on cations that have proven to have high stability. Unfortunately, the backbone design includes ether linkages that have been proven to be weak points for degradation and appear to be degrading as expected, based on color change and brittleness after hydroxide soak compared to the perfluoro (PF) AEM membranes. However, the team provided a good justification in that such polymers allow for the evaluation of electrode ionomer adsorption in different ways than other polymers; this should be the focus moving forward.
- Spirocycles may open up a new path for the development of a durable AEM. It appears, however, that the selected chemistry (i.e., the backbone) may not give the expected results; it has poor durability at higher ion exchange capacities (IECs).

Question 2: Accomplishments and progress

This project was rated **3.3** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The development of advanced AEMs is of crucial strategic importance in reducing the cost of fuel cell systems and meeting overall DOE goals for decarbonization and reduced U.S. dependence on fossil fuels. This group of spirocyclic anionic polymers is an important candidate in the portfolio and should be explored. The project has done an excellent job looking into these materials and characterizing their performance.
- The project has developed spirocyclic membranes with three different IECs. The team has measured greater peak power than existing Generation 2 PF AEMs with IEC 1.7, and the project nearly matched long-term stability with IEC 1.3. It would be great for the team to demonstrate longer life with IEC 1.7 power performance. The team also tested an MEA with low platinum group metals (PGMs) that meets performance goals.
- The spirocyclic materials have been made and tested, and the fuel cell performance meets the stated goals. The durability is highly questionable, and the inverse relationship between stability and IEC limits the power output because of the use of less conductive membranes. The possibility of using the spirocyclic polymers as electrode ionomers is an interesting idea, based on the initial results.
- Year 1 focused on the synthesis and evaluation of the polymer chemistry and evaluation of the MEAs. Given that it was follow-up work from the post-doc's chemistry work in graduate school, it was expected that most of the synthesis would have been done already, and in-depth MEA results evaluating the role of the different cations would have been the focus. However, this project can still be successful if this electrode work is the dedicated focus in year 2.
- A major milestone on MEA performance (0.6 V at 600 mA/cm^2 on hydrogen–air) was met, and another milestone was partially met on the durability target. However, after 500 hours of testing, the proposed spyrocyclic AEM with satisfactory initial area-specific resistance (ASR) demonstrated substantial loss of IEC and conductivity and became easily breakable. The mechanical properties have to be substantially improved.
- The first-quarter (Q1) milestone was not met; it was met at 500 hours instead of 1000 hours. The project team is testing some membranes at a relatively low temperature. Good peak power density (1.48 W/cm^2) was shown with AEMs of poor durability. When using the only reasonably stable membrane, the power dropped to 0.85 W/cm^2 at the same testing conditions. The proposed ionomer appears not to solve the problems laid out by DOE.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- Given the low scope of the project funding, it is difficult to expect much external collaboration. However, the principal investigator (PI) has wisely and excellently integrated the polymer into existing AEM efforts in order to benchmark against other polymers. This should continue.
- Though there is no direct collaboration, this project effectively leverages the AEM efforts of the Advanced Research Projects Agency-Energy (ARPA-E), the Membrane Working Group, and HydroGEN Advanced Water Splitting Materials.
- This is an NREL-only project, and outside partners are not needed at this early stage. NREL is well positioned to work with relevant partners when the time is appropriate.
- This is an NREL-only project, but there is good leveraging of existing related efforts at NREL.
- This is an NREL-only project, but there have been interactions with other institutions.

Question 4: Relevance/potential impact

This project was rated **3.3** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The development of advanced AEMs is of crucial strategic importance to reducing the cost of fuel cell systems and meeting overall DOE goals for decarbonization and reduced U.S. dependence on fossil fuels. This group of spirocyclic anionic polymers is an important candidate in the portfolio and should be explored.
- The project is designed to directly address DOE milestones for AMFCs. The proposed spirocyclic polymers offer a realistic opportunity to improve AMFC performance and durability.
- Spirocyclic AEMs show good promise and merit further development. There is great return on investment for this project.
- The project is highly relevant to DOE goals in the development of AMFCs that do not contain PGMs; the potential impact may be high.
- The project aligns well with the DOE Hydrogen and Fuel Cells Program and DOE research, development, and demonstration objectives, but the proposed ionomer chemistry will not work well.
- The polymer is useful in that it uses a different cation from most other membranes, but there probably would be more direct ways of evaluating this phenomenon, and the polymer itself has poorer stability than the PI's other PF AEM ionomer. It is not promising as a base stable ionomer, given the IEC loss.

Question 5: Proposed future work

This project was rated **2.9** for effective and logical planning.

- The planned next steps are reasonable, particularly the plan to further investigate the incorporation of spirocyclic ionomers into AMFC electrodes. Since the synthesis is limited to a single block copolymer structure, there is not much opportunity to mitigate durability issues by, for instance, adding spirocyclic cations to different backbones. Given the small budget, such synthetic options were probably never realistic.
- The PI plans to leverage the limited promise of the polymer to study it in electrodes, which makes good sense.
- The project should do more work on improving high IEC stability.
- The researchers have identified polymer stability as an issue, as well as the development of more advanced electrode ink formulations. However, there should be some fundamental analysis in the reduction of IEC capacity (and fuel cell performance) with increase in water uptake (as seen on slide 10). There are deep concerns about these spirocyclic materials: they have an inherently rigid backbone structure and form random three-dimensional fragments as they polymerize, which means they have to re-adjust to provide

continuous, directional ionic channels. In less rigid polymers, the polymer backbones are more flexible, and they re-adjust to enable functional groups to aggregate, forming more continuous channels. In addition, in spirocyclic systems, the backbones are so rigid that the channels cannot balloon as they imbibe more water. The rigidity is also an issue with long-term chemical stability. In any case, the reduction in ionic capacity with water uptake should be fundamentally studied. The project team should study why they are losing ionic continuity with greater water uptake.
- This ionomer has poor stability (at reasonable conductivity). It does not make sense to study it further; a new monomer structure with a spirocycle should be proposed.
- The proposed future plans are very vague and do not focus on the project's major issues.

Project strengths:

- This study is very attractive because of its ability to synthesize polymers with spirocyclic cations using a previously used synthetic route, and then subject the polymers to the same battery of evaluations already being used by NREL for other AMFC projects. This is an easy way to evaluate a promising new material set.
- The new chemistry was worth a shot and fit in well with existing work streams at NREL to study AEM polymers. Focusing on MEAs and understanding electrode phenomena is a good route going forward. The work overall helps the AEM space move forward.
- The spirocyclic functional groups look promising, based on their resistance to hydroxide attack; comparison to other ionomers will allow for selection of the best ionomer.
- This is an excellent, highly accomplished, very experienced research team. The work is of very high quality, and the team is well positioned to analyze this class of materials.
- There is great use of limited resources to probe the improved stability of spirocyclic AEMs.
- The project team has probably chosen the direction of the right cation type (spirocycle), but the specific chemistry has problems.

Project weaknesses:

- There is no inherent weakness. However, based on the results provided, there needs to be more fundamental work done to understand ionic conductivity in this class of materials. Results on slide 10 are counter to observations with other ionic polymers.
- The project's only weakness is the lack of synthetic flexibility. The spirocyclic cation was reported to be very stable, and it would be nice to be able to evaluate that stability on different polymer backbones, particularly ones without aryl ethers in them.
- The formation of solid residue during stability testing is an issue that points to instability in the polymer backbone. There needs to be an explanation for the absence of a correlation between IEC and conductivity losses for different ionomers. The interaction between the proposed AEM materials with catalysts remains unknown, though it is very important for making a decision to continue work in this direction. It is unclear how the addition of spirocyclic groups will affect the cost of an AEM or whether the performance improvements justify the cost increase. Strictly speaking, the best results have been obtained for cyclic, not spirocyclic functional groups, so the latter's advantages remain unproven.
- The project team needs to do more complete characterization of copolymer structure, blocks, and improving stability in high IEC.
- The authors did not answer key questions, including why increasing the IEC from 1.3 to 1.7 mmol/g caused significant durability issues or why the conductivity was poorly correlated with the IEC.
- The polymer chemistry is not terribly successful at being base-stable. It has limited promise as a candidate for widely distributed AEM use.

Recommendations for additions/deletions to project scope:

- The project needs to do more fundamental study of water uptake, ionic conductivity, and morphology of the polymer, as well as identification of what mechanisms result in the loss of ionic continuity with water uptake. The team may possibly need to do some work with reinforcements to reduce dimensional changes during water uptake and determine whether ionic continuity can be maintained.

- If it is not already planned, there should be an investigation into why the spirocyclic AEMs with higher IECs are less stable than the ones with lower IECs.
- The study of the interaction between the catalyst and spirocyclic polymer material should be accelerated, and the cost analysis has to be performed.
- The project team should try another backbone or ionomer structure with a spirocyclic cation.
- The team should leverage the unique cation structure to understand electrode phenomena.

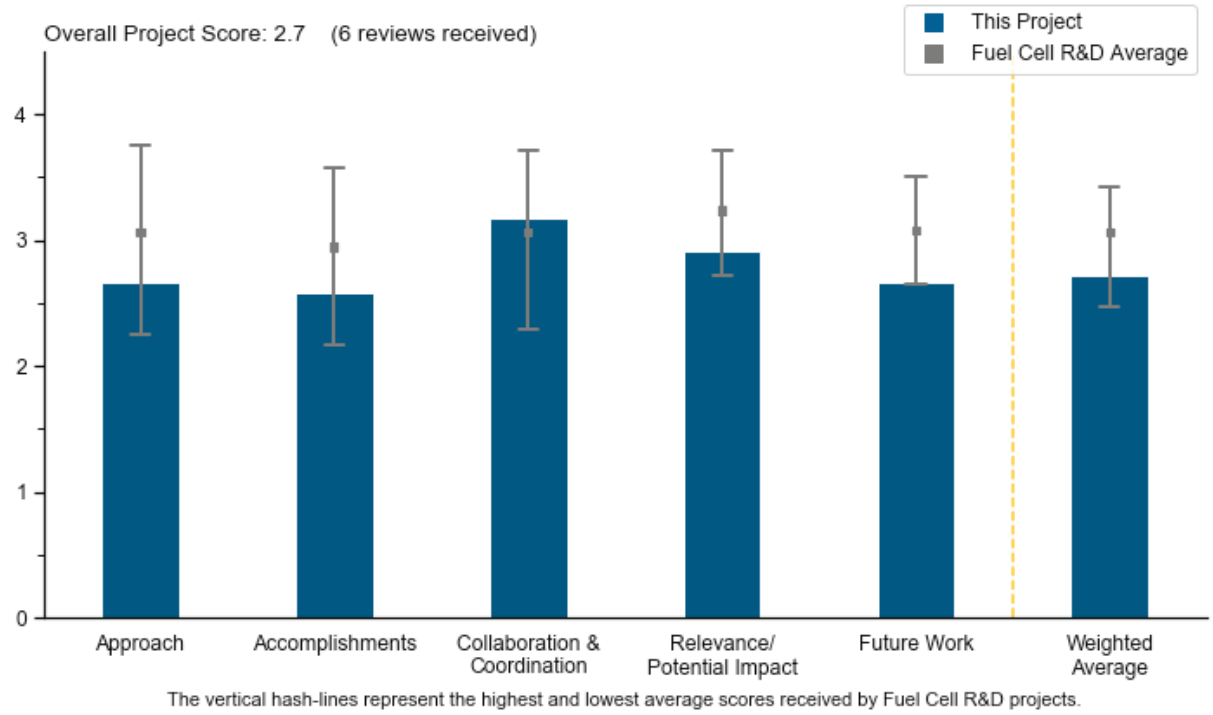
Project #FC-179: Lab Call Fiscal Year 2018 (Membrane): Stable Alkaline Membrane Based on Proazaphosphatranes Organic Super Base

Gao Liu, Lawrence Berkeley National Laboratory

Brief Summary of Project

Lawrence Berkeley National Laboratory (LBNL) is developing new alkaline membranes with improved stability and performance to enable a platinum-group-metal-free (PGM-free) alkaline-membrane-based fuel cell. In addition, proof of concept will be derived for proazaphosphatrane superbases in alkaline membrane applications. The project team has identified a stable and flexible polymer matrix constructed from these superbases; the material demonstrated superior stability in high alkaline conditions. The project has also developed a feasible process to synthesize the superbase-grafted ionomers by connecting the superbases to Kraton Corporation (Kraton) copolymers via methylene chloride modification. The project team plans to address membrane synthesis and membrane electrode assembly (MEA) development and testing in the next phase.

Project Scoring



Question 1: Approach to performing the work

This project was rated **2.7** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The approach of adding a superbase group to commercial polymer is a wise choice for this project. It is very good that quantifiable technical targets are stated.
- The project is based on the assumption that the stability and high alkalinity of a proazaphosphatrane organic superbase would result in highly conductive and durable AEMs. These superbase units were grafted to a stable polymer backbone to make them highly conductive and stable against thermal and chemical decomposition. The structure of proazaphosphatranes could be modified to exclude tertiary carbon atoms and thus improve stability.
- The team's approach of using a stable and highly basic organic superbase is excellent. Instead of quaternary ammonium cations, an organic superbase may work for anion exchange membrane (AEM) fuel cells.

However, proazaphosphatranes may not be an ideal superbase, as the molecules are bulky and possibly difficult to make. Tethering the superbase in all hydrocarbon (polystyrene-based) polymers is also a good approach. However, the reaction in the bromoalkyl group will produce the benzyl cation, which would make the polymer unstable.
    - There is not a clear pathway to reach the initial conductivity target (>100 mS/cm, which is better than an ammonium-based system). The pH of the proposed material (1.0 M) is 14, a level similar to ammonium (1.0 M); but the size of the molecule is much larger, which makes it hard to produce a high-ion-exchange-capacity (IEC) membrane.
    - The solvation energy of the cation will likely be low. Therefore, the conductivity of the AEM tethering this superbase will be much lower than that of an ammonium-based system. The principal investigator (PI) should provide a more specific approach to achieving the milestone, rather than just making a general statement based on the funding opportunity announcement.
- The approach is acceptable; however, the project team will not actually make membranes until the project is almost over. The previous work is important, but it is hard to see how the team is going to obtain sufficient results to motivate additional work.
- There are serious concerns about the wisdom of the work in general. Using cations with high molecular weight limits the IEC of a final membrane (such as in the work of Coates) and raises serious concerns about the promise of the membrane to be sufficiently conductive. The literature has suggested that smaller existing N-based cations are sufficiently stable and that attaching them to a polymer backbone in a way that makes them stable should be the focus of AEM work at this point. Also, many groups appear to be close; perhaps focusing on upstream applications of AEMs (such as how they degrade or affect electrode performance) should be more of a focus. However, given the idea, the approach has been generally good, if slow. Testing cation stability as a model system and then leveraging existing styrene copolymers is the right way to evaluate new cations.
- It is unclear why the organic superbase would have improved stability in an aqueous AEM application.

Question 2: Accomplishments and progress

This project was rated **2.6** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The route to a superbase-grafted polymer has been identified, and surrogate or precursor compounds have been synthesized. The team has studied potassium deuteroxide (KOD) stability with a small surrogate molecule. The project timeline may not allow much time for MEA characterization once the polymer is produced.
- So far, most of the work has been done on synthesis and the stability study of model monomeric compounds. The functional group loading that was achieved is too small. It was shown that the methyl-group-functionalized superbase is stable for four months (after that, some changes in the nuclear magnetic resonance [NMR] spectrum are visible), while the lifetime is less than two months for an isopropyl derivative. The methylene chloride modification to the Kraton polymer was successful, but the reduced polymers were either gels or extremely brittle films.
- The results shown were interesting, but the stability of the materials made to date does not appear all that exceptional. It is not clear that these high-cost materials are worth pursuing. Additionally, the team has not actually made any of the proposed polymers, even though the project is more than half over.
- The progress of this project is rather slow. All that has been accomplished is to check the alkaline stability in a small molecule study. The accomplishment is on the level of a preliminary result before starting the project. It is unclear why the PI spent so much time on the stability of proazaphospatrane derivatives. It turned out that more complex proazaphospatranes have lower stability. The project will not likely meet even the membrane stability target (2,000 hours). The PI will likely not be able to show a fuel cell performance of 620 mA/cm^2 at V = 0.60 V by the end of the project.
- The methyl stability results are confusing. The hydrogen NMR plots show shifts of peaks and new peak growth but are claimed to be stable. There is no clear way to rationalize this. As for the polymer stability, it is not clear that 10 eq $NaOCH_3$ is the best solution for an aging study. It is not clear what the details of that study were or if they were just used as a reference point. The team stated that the copolymer T_g is too low, but no data were presented to quantify the T_g. The details of the membrane casting were not given, so it is

not possible to quantify whether the processing could be causing poor membrane properties. The team tested the Kraton copolymer, but there are no data to suggest that this would be a better material than the initial copolymer tested. It is unclear whether the decomposition rate is good or bad.
- After well over a year of effort, no polymer has been made yet. This work should have been proceeding along with the model compound stability study, but the effort appears to have been sequential. Understanding the promise of the approach requires that evaluation be done in a polymer, especially given the bulky and high-molecular-weight cation; the lack of evaluation severely diminishes the project.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- This is a lab call project, so a minimum level of collaboration is expected. Obtaining materials from Kraton is a good thing for the project. Collaborating with other teams is also a good thing, although the other teams' efforts on this project are minimal so far.
- Despite the modest budget, the team has managed to include three groups at LBNL and a commercial partner (Kraton).
- The project team seems to have relevant partners at LBNL, though perhaps it would be wise to include Ahmet Kusoglu (of LBNL) for some membrane characterization, specifically given the mechanical issues that have been identified. Maybe Kusoglu is assumed to be a part of Adam Weber's team at LBNL.
- The collaboration between team members seems to be working, though a collaboration outside the project is desirable for cost evaluation of the proposed functionalized polymers.
- There is really only one partner (Kraton) that appears to be just a material supplier. However, this is acceptable for a lab call project. Kraton has supplied materials.
- The PI has done well to leverage external polymers as a platform to which to attach his cations, but little tangible progress has been made yet.

Question 4: Relevance/potential impact

This project was rated **2.9** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- If successful, the project will lead to the development of high-performance alkaline membranes and, based on those, fuel cell systems that would be competitive with current fuel cell systems using Nafion polymer electrolyte membranes. The use of alkaline membranes could lead to replacing Pt-based catalysts with PGM-free ones.
- It seems like there should have been more funding. The project team seems to be addressing current stability problems with available alkaline membranes.
- If the project is successful, then it will have a potential impact on several different areas of the DOE Hydrogen and Fuel Cells Program. The general approach of this project—using organic superbase instead of ammonium—has been tried by groups before, but it has never been successful because of the synthetic complexity and other reasons. It is very challenging to make a reasonable system using superbases. Based on the project's progress so far, the potential impact does not look great.
- The DOE should fund AEM fuel cell work, but it is not clear why this particular AEM approach was funded.
- The project aligns with the DOE's goals of supporting stable AEM research, but it may not be the most successful way to go about it.
- The benefits of using a superbase are unclear.

Question 5: Proposed future work

This project was rated **2.7** for effective and logical planning.

- The future work is focused on synthesizing and evaluating actual polymers, which are sorely needed in this project; this is the correct direction.
- It would be good to see an MEA performance evaluation.
- The team should strive to make membranes as soon as possible to obtain sufficient results to potentially motivate additional work.
- The stated goal is to increase the superbase loading to 30%. It is hard to tell if this is a reasonable goal. Assuming this is possible, the equivalent weight or expected IEC would still need to be determined. This would help quantify whether this 30% goal is adequate, too low, or too high.
- The future work is focused on the development of a superbase polymer based on a polystyrene-hydrogenated-polyisoprene backbone. The work plan is rather generic and based on milestones that have been missed or are yet to be met.
- Most of the proposed future work is unrealistic. The PI has not had any reasonable membranes from this project so far. The PI cannot finish all the tasks and milestones in the years remaining. The project team aims to continue with the stability of the superbase in 2M KOD in a D_2O solution, which has been done extensively during the past year. The PI should stop this task and focus on making AEMs with the superbase and measuring the target properties.

Project strengths:

- The researchers are taking a smart approach to this project in addressing membrane stability, which seems to be one of the primary challenges with alkaline membranes.
- The proposed proazaphosphatrane functional groups that are tethered to a stable hydrocarbon-only backbone could potentially lead to highly stable AEMs and durable fuel cells that use such membranes.
- Leveraging existing polymers to use as a platform for the new cations is a good idea. The evaluation of cation stability has shown some promise in terms of stability.
- The project's strength is in the concept. This is a new idea and a challenging project.
- The team is good, with relevant backgrounds and experience.
- This is a good team with an interesting idea.

Project weaknesses:

- The only weakness is that there has not been any investigation of membrane structure or mechanical properties.
- The proazaphosphatranes are expensive to make and hard to attach to a polymer backbone. Their bulkiness makes creating strong and flexible membranes very hard; this may be a fundamental problem that should be solved by selecting the right backbones. So far, the project has encountered multiple synthetic problems, such as a low level of modification of the styrene units and unsatisfactory mechanical properties of the films. There is no apples-to-apples comparison of stability between the proposed proazaphosphatrane groups and the known functional groups that are much less expensive. Therefore, it is unclear what benefits these groups could bring. The cost analysis of the proposed polymer materials should be done based on synthetic schemes.
- The project's progress is way behind. The PI did not accomplish the milestones for the third or fourth quarters (Q3 or Q4) for fiscal year 2018. There is not much hope for obtaining the rest of the milestones within the project's schedule and budget. The targets for the proof-of-concept project are inappropriate. The PI may not have enough capability to make polymeric materials.
- The work has shown no comparison with baseline benzyltrimethylammonium or other cations to evaluate the base stability of the designed cations. The progress on polymer chemistry has been slow.
- The team is not planning to work on any actual membranes until too late in the project.
- There is not much support (e.g., literature, models) for the advantages of the superbase.

Recommendations for additions/deletions to project scope:

- In the limited time remaining, polymer chemistry is the correct focus. Given the limited funds of the project, no additions/deletions are recommended.
- No additions are recommended. The PI should focus on making stable alkaline superbase AEMs and characterizing the AEM conductivity and stability for the rest of the project's time. The team should not waste more time on investigating the stability of the superbase. The project should not invest time in making MEAs or evaluating cell performance and durability. The team should not investigate more robust connections between the superbase and polymer matrix. If this team shows that the superbase can replace the ammonium-functionalized AEMs with comparable hydroxide conductivity, then this may be a valuable outcome for the project.
- The funding was limited, but the project would have benefitted from some additional modeling to show how a large superbase group would affect phase-separated morphology. Ahmet Kusoglu and the LBNL Advanced Light Source team would be able to look at this and should be considered.
- Cost analysis should be added to help with understanding the cost-competitiveness of the proposed AEMs. The next period of funding should be based on meeting the go/no-go milestone, which does not currently look achievable.
- The project team should accelerate the fabrication of the membranes.

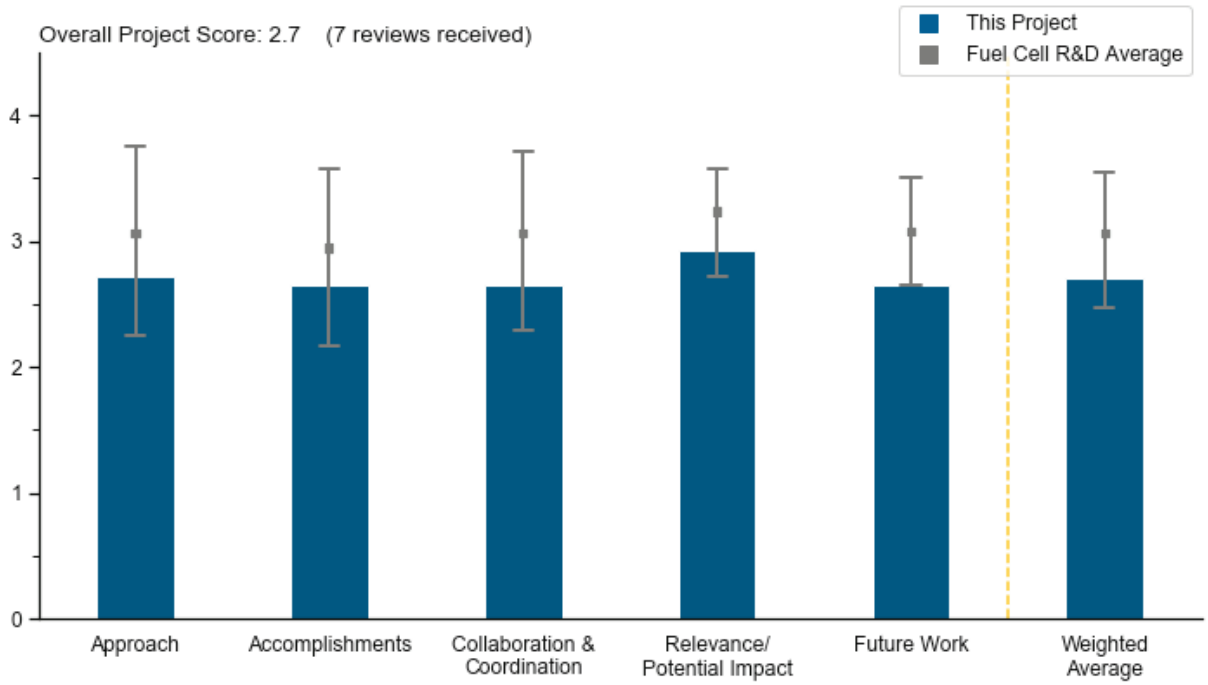
Project #FC-180: Lab Call Fiscal Year 2018 (Membrane): High-Performing and Durable Pyrophosphate-Based Composite Membranes for Intermediate-Temperature Fuel Cells

Cortney Kreller, Los Alamos National Laboratory

Brief Summary of Project

The project objective is to develop a membrane that enables system operation at higher temperatures (200°C) and low relative humidity (RH) with conductivity across the entire range of operating conditions for transportation applications. To do this, the project will tailor the composition of MP_2O_7 (metal pyrophosphates [MPPs])/polymer composite membranes to achieve the U.S. Department of Energy target for membrane area-specific resistance (ASR) of ≤0.02 Ωcm^2. The project addresses the barriers of developing an early-stage membrane concept that can (1) decrease system costs by operating at higher temperature (150°C–400°C), (2) achieve membrane ASR of ≤0.02 Ωcm^2, (3) achieve ASR ≤0.03 Ωcm^2 under low RH conditions, and (4) achieve sufficient conductivity (0.2 → 0.02 Ωcm^2) across the entire range of operating temperatures.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **2.7** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The approach is addressing mainly system cost reduction, thanks to the development of membranes for fuel cell operation at higher temperatures (about 200°C). It is basic research, based on a material identified a few years ago showing the capability to conduct protons at high temperature and ultralow humidity. The approach is clearly defined; the idea is to make a composite membrane that enables conductivity in the whole range from ambient temperature to 200°C. Nafion™ is a planned part of the composite; current work is focused on the high-temperature side, with the modification of the original material identified at the beginning of the project. SnP_2O_7 has been modified by replacing part of the Sn with various metal cations to improve conductivity. In and Sc have been selected. The integration of the new materials into composite

membranes should be done in the coming months. Collaboration for the integration of the membrane into a membrane electrode assembly (MEA) is considered in the approach. A major risk will be getting a thin membrane that does not present the required mechanical or electrical properties on the broad temperature range.

- The project's approach is to produce intermediate-temperature MPP proton conductors, evaluate trends in phosphorous-to-metal (P:M) ratio, produce membranes, and minimize thickness. This is a logical progression with the modest budget.
- The team has followed its research plan and executed it. They have synthesized the MPP materials and made membranes. They have begun to characterize performance.
- The project proposes to develop MPP or polymer-composite membranes with low ASR and the capability to work at 200°C. The main focus was on the effect of substitution on MPP conductivity. However, the data obtained showed that the unsubstituted material has one of the highest conductivity values.
- The objectives are clearly identified, although it is hard to see how this differs significantly from previous work putting other inorganics, such as heteropolyacids, into polymer membranes. If these can conduct protons without water present, that is potentially interesting, but achieving conduction through a matrix of dry Nafion may prove to be very difficult.
- It is not clear when the go/no-go should be completed. Since the project started in January 2018, it seems like the third quarter (Q3) and Q4 milestones should have been completed by the time the slides were due, but it was reported that they were 0% complete. It seems that the team is far behind schedule. It was nice that quantifiable metrics were presented. It seemed like the precipitation method is good because of low temperature.
- The justification for the MPP–Nafion composite membrane does not make sense. It is an interesting idea to have a composite membrane with different conduction pathways as a function of temperature, but the sulfonic acid moieties in Nafion (or another perfluorosulfonic acid [PFSA]) will start to degrade at 200°C, and the PFSA will flow significantly at more intermediate temperatures, such as 120°C and above. It is highly unlikely that such a composite membrane would survive hot and cool cycles without changing morphology, and no proof of concept has been demonstrated in more than a year of project funding.

Question 2: Accomplishments and progress

This project was rated **2.6** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Good progress has been made toward the project's objectives, with preparation and characterization of various materials going as planned, allowing the project team to select two new ones to be integrated into a membrane.
- If these membranes reach their technical targets, the project will actually contribute to cost reduction as targeted by DOE.
- There is good progress on a variety of metals materials produced. Conductivity has been measured, but there are no results on membrane fabrication.
- A variety of MPP materials have been synthesized and appropriately characterized. The Q3 milestone will be the real indicator of whether this approach is feasible.
- The particle component appears to have been well researched and optimized, but no work on the composite has been undertaken. The entire proof of concept is left unfulfilled at this point.
- It seems there is no trend in conductivity and P:M. Another issue that was identified is that it is not possible to compare dopants unless they are at the same P:M. Bulk pellet conductivity is the primary result related to the go/no-go. There is great concern that since the pellet conductivity is not that much higher than the go/no-go, it will not be possible to achieve the go/no-go when the MPP powder is blended into the polymer.
- As of yet, the main focus has been on the development of conductive MPP materials, with little success on MPP or polymer membrane development. Even the most conductive materials (which are actually not much better than the starting SnP_2O_7) could not achieve the project goals unless they were to be converted into thin composite membranes.

- The data provided was sparse, with only a few slides and limited data points. Based on the data provided, it is hard to determine whether this technology pathway has merit. The impedance and conductivity data appear to be sufficient to down-select a few promising candidates and perform additional testing.

Question 3: Collaboration and coordination

This project was rated **2.6** for its engagement with and coordination of project partners and interaction with other entities.

- There seem to be many team members on a project with a small amount of funding. The team members' roles were not discussed, so it cannot be determined whether all the team members are needed. The low level of funding clearly limits the project team's ability to collaborate with other organizations, so this being a Los Alamos National Laboratory (LANL)-only project seems reasonable.
- One national laboratory is doing the job; however, this is acceptable. UNM is interested in evaluating the membrane as a no-cost partner, and the project team is planning to solicit more partners once the proof of concept is established.
- All the work to date appears to have been done at LANL. The University of New Mexico (UNM) is a partner whose work, evaluating the materials for a specific application, occurs later in the project.
- This project is a laboratory-only call, but the team has a no-cost partner: UNM.
- The project has no partners yet, but this makes sense, given its pre-proof-of-concept status and the fact that it is funded through a lab call.
- Though this lab call project is open to national laboratories only, the project could greatly benefit from collaboration with material scientists and polymer chemists outside the project.
- No collaboration has been undertaken so far. UNM is on the list as a collaborator, but they are described as interested in testing the final product, rather than being meaningfully involved in its generation.

Question 4: Relevance/potential impact

This project was rated **2.9** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- High-temperature materials for proton-conducting membranes may play a huge role in reducing the balance of plant size and cost, if the ASR and durability targets are met. The proposed polymer electrolyte membrane (PEM) work is at 200°C, which is higher than the optimal temperature for automotive applications, so the system modeling should be done to understand these trade-offs.
- The project team is addressing the barriers that were outlined in the proposal call. The materials seem to have some advantages since they do not require phase change.
- The proposed membranes have the potential to achieve several DOE technical targets and could have an impact on fuel cell design for transportation.
- There is a strong need for intermediate-temperature fuel cells. The proton conducting is novel.
- The core aspect of the project aligns with the goal for system cost reduction. The impact will be positive if the technical targets are reached; the work will have to be continued to enable assessment of the actual potential impact.
- Identifying fuel cell materials that can perform at higher temperatures has inherent value. The question is, of course, whether this group of materials has inherent promise and can provide a pathway to achieving this overall technical goal. The data provided are inconclusive.
- The concept could perhaps be adapted, but as designed, there is no pathway to success that can be seen for a particle–Nafion composite membrane to perform as described.

Question 5: Proposed future work

This project was rated **2.6** for effective and logical planning.

- The researchers have clearly identified what they need to do in terms of making membranes and testing them going forward.

- The plans for next steps are consistent with the original project objectives and the results reached on the new materials during the first period. Materials selected will be implemented in composite membranes for further characterizations. The features identified for validating these membranes are conductivity and thickness; mechanical properties and permeation are missing and should be added to the characterizations to be conducted.
- The future work is satisfactory to attempt to prove the concept.
- The problems with dispersing powder in polymer are concerning, and this needs to be addressed. However, given what seems to be the low conductivity of the MPP powders, it seems that more effort should be focused in this area to achieve higher MPP conductivity, in anticipation that the composite membrane will have lower conductivity than the MPP pellets.
- Milestone 3 will be critical to demonstrating success. Fabricating composite membranes will be tricky, and the presentation does not say much about the anticipated challenges and mitigation strategies.
- It does not seem certain that the year 1 go/no-go criteria will be met.
- The proposed future work targets PEM development, which should be done in year 1, but no MEA work or testing has been done or is planned.

Project strengths:

- The strengths of this project are the good methods that have been applied with systematic analyses and conducted for the selection process, and the actual identification of materials that are potentially able to improve membrane conductivity at high temperature and ultralow humidity.
- The LANL team has tremendous experience in evaluating new materials for PEM fuel cells, and the proposed materials have potentially very advantageous properties.
- The proposed MPP demonstrated high conductivity, and an MPP–polymer composite membrane could potentially work at higher temperatures than conventional PFSA PEMs.
- The use of polyphosphate phases is an important area of research. LANL has the resources and is well placed to do this work.
- The project has a good approach and development of various metal-containing materials.
- The strategy to use materials that do not require a phase change seems like a wise approach.
- There is strong optimization on the inorganic piece of this project.

Project weaknesses:

- For 15 months of full-time activity, the total output seems low. It would have been nice to have more data on actual membranes in situ and in fuel cells to determine the viability of this technology. The project needs a big spur of activity in the next 7 months. It may be that the materials are inherently difficult to work with, and progress will be inherently slow, in which case, this needs to be mentioned. If not, in situ fuel cell data would be great to have as early as possible in the project. There is nothing comparable to in situ, real-life operating data.
- One issue is that after more than 15 months, no composite membranes have been made with new materials. This should be fixed soon. It could be difficult to reach the target of 100 mS/cm with a composite membrane when this is the level that was obtained with just the material alone in pellets. It is also not so obvious that the Nafion will ensure the required conductivity at low temperature with ultralow humidity. A major weakness is the risk of getting composite membranes with poor mechanical properties or permeation, but the project does not seem to consider this aspect.
- It seems that the results are lacking, and the project is behind schedule. It seems like it may be hard for the project to meet the go/no-go point with the MPP materials that have been synthesized so far. It seems there should have been a stronger focus on integrating materials with ionomers since that is part of the go/no-go, though that may only be clear now because of unanticipated challenges with blending.
- It is still unclear how to convert good material conductivity into practical, flexible, and durable composite PEMs with the target thickness and ASR, or how the catalyst activity will be affected by interaction with such membranes. As of yet, the major focus has been on material improvement, although issues with manufacturing the thin composite membranes were not addressed.

- It may be difficult to load Nafion with an inorganic material and get it evenly dispersed without compromising the mechanical properties of the membranes. There are many MPP formulas from which to choose, but discussion on how to prepare composite membranes is limited.
- There does not seem to be a clear strategy for producing the membrane. There may be binders or fillers that need to be added to facilitate sintering. It is not clear what the prospects are for achieving the desired thickness.
- There is a lack of understanding about the behavior of Nafion. There is no collaboration in the design of the experiments.

Recommendations for additions/deletions to project scope:

- The first recommendation is to speed up the process for making membranes with the selected materials. In addition, it is recommended that the team add mechanical properties and permeation as criteria for validating the composite membranes. The project team should also consider integration in MEAs as an important objective. If needed to save resources, the project team could decide to work with only one selected material to enable more membrane and MEA optimization and characterizations.
- The project could consider another (uncharged) binder that serves simply as a matrix for the particle material. This would limit conduction to higher temperatures but could be more successful than trying to find a membrane to conduct across a wide variety of temperatures. For example, hydrocarbon ionomers that could be more resilient to flow at high temperatures would require higher water content to be conductive at low temperatures.
- The scope is appropriate, but most of the effort should be focused on increasing the conductivity of pellets and on the challenges of blending MPP powder into polymer.
- If it is not already planned, the team should include a study of how water-tolerant the composite membranes are. It needs to be determined whether the MPP will leach out if the membranes are used in a fuel cell that operates briefly at high RH.
- The project team needs to identify a route to creation of the membrane, including strategies for barriers and mitigation.
- The project seems to be lagging behind the original schedule, and the go/no-go milestones were not met.
- The project team needs to make membranes and test them as soon as possible.

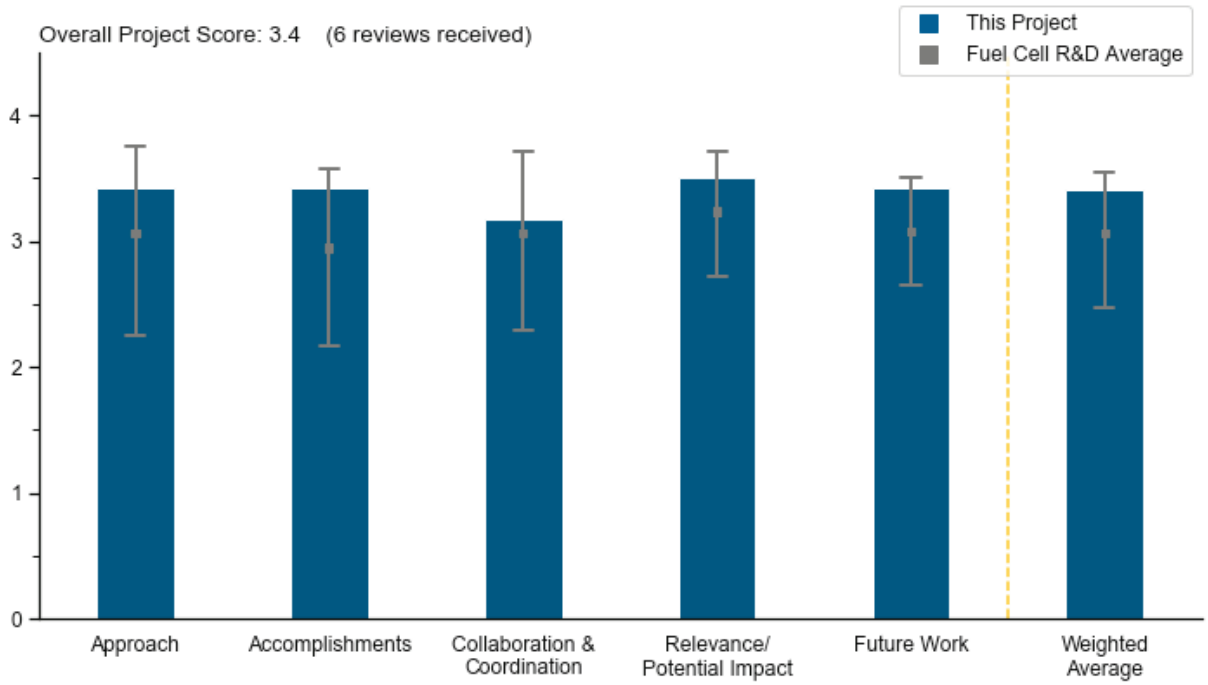
Project #FC-181: Lab Call Fiscal Year 2018 (Reversible Fuel Cells): Microstructured Electrodes and Diffusion Layers for Enhanced Transport in Reversible Fuel Cells

Jacob Spendelow, Los Alamos National Laboratory

Brief Summary of Project

The objectives of this project are to enhance the H_2O and O_2 transport for unitized reversible fuel cells (URFCs), as well as fabricate, test, and validate these devices for high performance and durability. The UFRCs use amphiphilic electrode structures, diffusion media, patterned gas diffusion layer (GDL) substrates, and microporous layers (MPLs) to address transport challenges. Patterned hydrophilic/hydrophobic GDL substrates, MPLs, and catalyst layers (CLs) are used to construct an amphiphilic membrane electrode assembly (MEA) to achieve improved fuel cell and electrolyzer performance and rapid switching between fuel cell and electrolyzer modes.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **3.4** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The team proposes a novel approach for designing electrodes to enhance the transport property of URFCs. The optimized electrode structure is made of hydrophobic and hydrophilic channels to address the challenges of water management, satisfying both electrolyzer and fuel cell modes. Electrode structures composed of micropatterned amphiphilic GDLs and MPLs are expected to bring enhanced transport of H_2O (through hydrophilic channels) and O_2 (through hydrophobic channels), which ultimately results in improved performance in URFCs that require rapid switching modes between electrolyzer and fuel cell.
- The approach proposed is sound and fundamentally relevant to DOE targets. The idea of patterned electrodes is brilliant and should be explored further. The main aim of the work is to improve performance of reversible cells. Cost and durability are not yet addressed in this year's work. It is vital to understand

which would be the most important degradation phenomena related to the mutual interactions of the hydrophobic and hydrophilic MPLs.
- The principal investigator's (PI's) approach to achieving high activity in water electrolysis and fuel cells is to locally pattern the GDLs and MPLs and electrodes to make them hydrophobic and hydrophilic in different regions. This approach is sensible and appears to be working well. The outcome of this project along with other approaches can create MPLs for oxygen evolution reaction on titanium-based electrode layers that will help with water management. This is cutting-edge work.
- The use of an amphiphilic electrode structure is a clever idea to balance the needs of reversible fuel cell technology. The use of a patterned electrode structure to achieve balance between hydrophilic and hydrophobic needs is also very promising.
- The project focuses on a fundamental issue of URFCs: absolutely different humidity requirements for fuel cell and water electrolyzer modes. The proposed approach is logical, and the preliminary results confirm the assumptions.
- The fixed-polarity approach has not been sufficiently justified. From a system design standpoint, the fixed-gas approach appears more robust. The approach of trying to develop catalyst supports to enable increased performance with decreased loading is good.

Question 2: Accomplishments and progress

This project was rated **3.4** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project is on track and making good progress. The atomic layer deposition of Ir and Pt on TiO_2 appears to be resulting in high-quality catalysts, but unfortunately, the hydrogen oxidation reaction durability was poor. It would have been good to have seen data on the effect of catalyst loading on performance and durability since the TiO_2 support may be helping in this area. The down-selected physical mixture of Ir and Pt black seems to be performing well, but this is a conventional catalyst for reversible fuel cells, so the result is expected.
- Hydrophobic/hydrophilic patterning of GDLs via polytetrafluoroethylene (PTFE)/Nafion™ has made significant enhancement in the fuel cell mode of URFC performance under different relative humidity (RH) conditions, and its impact is greater, particularly at highly flooded conditions. In electrolysis mode, the amphiphilic electrode structure also showed improved performance compared to hydrophobic MPLs treated with PTFE alone. DOE has funded a number of URFC projects in the past. Although optional, a comparison with previous funded projects would be helpful for the reviewers and audiences to understand the importance of this project's progress.
- Progress in making structured electrodes with amphiphilic structure is very good. The results are encouraging. The MPL design and fabrication progress is very good. This can help meet DOE goals for reversible fuel cells. The direct blade coating data look good.
- Project accomplishments are very good. Go/no-go criteria are met. Improved fuel cell performance is seen when TiN-treated Ti felt is used and when hydrophobic/hydrophilic treatments are applied to the GDL. Effects are especially significant in fuel cell mode at high RH, when flooding is likely. A greater focus on confirming the major role of water–gas transport in achieving these results, perhaps via computational models that were apparently used at the start of the project, would be helpful.
- The proposed hydrophobic/hydrophilic patterning improves URFC performance in the fuel cell mode, even at high RH, while not decreasing the performance in the electrolyzer mode. The go/no-go criteria have been met, though the round-trip efficiency is very low.
- Accomplishments are relevant to DOE goals, and the project is fairly on time with milestones.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- Collaborations with the Fuel Cell Consortium for Performance and Durability (FC-PAD) and others on electrode microstructure appear to be good.

- The collaboration with Argonne National Laboratory on catalyst development is quite useful.
- It was mentioned that there is collaboration with multiple DOE projects, but it would be useful to hear more elaboration on their roles and outcomes from the collaboration.
- An industrial collaboration would be beneficial to understanding the scalability of the idea.
- Industry partnerships are highly productive when used smartly. There is no industry partner so far.
- Collaboration with other organizations is limited and could be improved.

Question 4: Relevance/potential impact

This project was rated **3.5** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- Sustainable hydrogen production via water electrolysis and its use via fuel cells, the basic concept of URFCs, would be ideal to address this challenge, but device configuration satisfying both modes is not trivial. Because electrolysis and fuel cells require different levels of water management, their electrodes should be constructed differently. This project proposes an optimized electrode design satisfying both modes.
- Reversible fuel cells are very relevant for energy storage to support greater penetration of renewables. This project provides an opportunity to increase round-trip efficiency. It can also lead to new applications for hydrogen and fuel cells.
- The proposed method could be instrumental in making URFCs that work equally well in both operational modes. Such URFCs could find a wide application in long-term energy storage.
- The project appears to be having significant impact on improving URFC technology, particularly for low-temperature operation.
- Work on reversible fuel cells is extremely relevant to DOE multiyear goals.
- The fixed-polarity approach needs to be better justified to demonstrate the relevance of this work. However, the project is demonstrating good reversible fuel cell performance, which is relevant to the DOE Fuel Cell Technologies Office's increasing emphasis on H2@Scale.

Question 5: Proposed future work

This project was rated **3.4** for effective and logical planning.

- The proposed future work seems well planned. The approach of investigating separated Pt/TiO_2 and Ir/TiO_2 is good. The work on examining operating conditions and components such as diffusion media and flow fields should be helpful to the community.
- The use of neutron radiography for water management is a nice idea. The synthesis and testing plan is good.
- The project is on track, and the team is well aware of the challenges ahead and offered a number of mitigation strategies. Particularly, scalability of the technique is certainly of importance.
- The team is aware of what the current project lacks and has a detailed future work plan to tackle those weaknesses.
- The PI's proposed future work seems sensible and appropriate.
- The future work plan is satisfactory but not very specific, and it lacks technoeconomic analysis.

Project strengths:

- The project brings together significant expertise in creating and adjusting electrode microstructure to achieve desired behavior for the disparate needs of water electrolysis and fuel cell operation. The work with Ti electrodes is very important and likely to provide guidance to others working on related problems.
- Patterned electrodes and microstructural design are highly relevant for reversible cell performance improvement.
- There is good technical progress so far. The team met the go/no-go milestone. The dual-function electrode performance is good. Amphiphilic electrodes provide a wide range of operation.

- The project is focused on the right critical issue of URFCs, and the proposed approach seems to be working.
- LANL uses a novel approach with solid work progress.
- This is a strong team with a good background in both fuel cells and electrolysis.

Project weaknesses:

- The preliminary data look very promising. A comparison with other earlier DOE-funded URFC projects would be helpful.
- The project work presented at this Annual Merit Review lacked a significant modeling component, and because of this, most findings are interpreted somewhat qualitatively. Results demonstrating improved performance are claimed to be due to improved water–gas transport, but they could be explained by other factors. Now that the PI has data using patterned electrodes, he may find it useful to go back to the models and see if they offer predictive capability that would link specific experimental observations with specific physical mechanisms. In turn, this work could suggest pathways for further optimization.
- URFC performance in the two modes is not symmetrical. Using the proposed technology for energy storage will result in overdesign of the system to match the charge and discharge power and times. The effect of channel alignment/misalignment of GDL, MPL, and CL is unknown and should be studied. The UFRC modeling using real grid data should assist in the system design. The development of bifunctional electrocatalysts is not in the project scope, so cooperation in this area is highly desirable.
- The core approach of using fixed polarity has not been adequately justified.
- There is uncertainty in fabrication cost and degradation performance.
- There is a lack of industry partnership durability data, which would be helpful.

Recommendations for additions/deletions to project scope:

- Technoeconomic analysis of the proposed patterned MEA and stacks should be added to the work scope.
- It is recommended that the team investigate quality and durability of the local structure of amphiphilic electrodes after numerous cycles of electrolyzer and fuel cell modes.
- It is recommended that the team consult with an industrial partner.
- Adding durability data in the work plan will be useful.

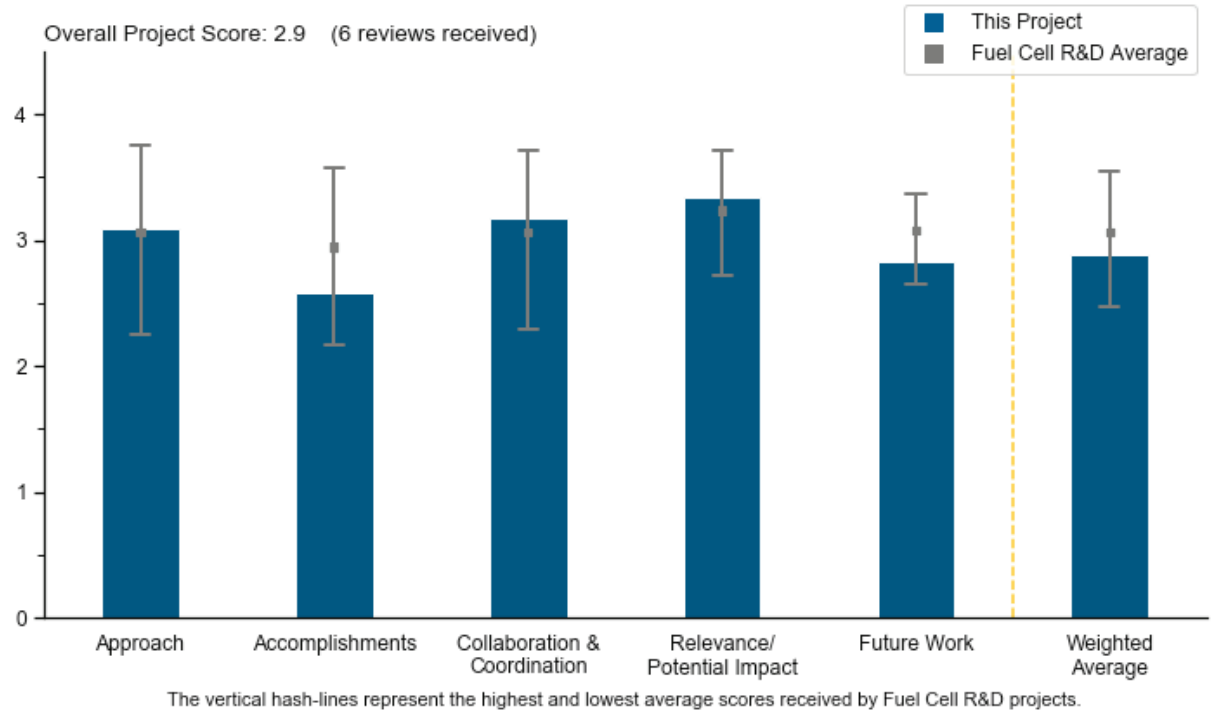
Project #FC-182: Lab Call Fiscal Year 2018 (Reversible Fuel Cells): Bipolar Membrane Development to Enable Regenerative Fuel Cells

Todd Deutsch, National Renewable Energy Laboratory

Brief Summary of Project

This project directly addresses the Fuel Cell Technologies Office's interest in developing reversible fuel cells (RFCs). The project objectives are (1) to fabricate a bipolar membrane (BPM) with a dual-fiber electrospun junction that can be employed in a stable, high-performance RFC membrane electrode assembly (MEA), (2) to fabricate and optimize the BPM junction with available materials (leveraging others' ongoing development), and (3) to obtain BPM device data in both fuel cell and electrolysis mode. The effort will focus on optimizing the BPM junction interface. The project addresses the barriers of cost, durability, and performance.

Project Scoring



Question 1: Approach to performing the work

This project was rated **3.1** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The use of a three-dimensional (3D) electrode structure is a very innovative idea; electrospinning and interspersing makes it unique. Higher efficiency targets can be reached using the 3D design. Additionally, using graphene oxide (GO) for water disassociation is synergistic.
- This is a unique approach to RFCs using a BPM, one that addresses issues with the BPM junction and interface by co-electrospinning the alkaline and acidic membranes. This approach should improve the interface and decrease the delamination of commercial BPMs that is observed when trying to operate at high current. Other than changing the catalyst content at the interface, it is not clear what parameters of the interface the project team plans to vary (e.g., the thickness of interface layer and gradient versus step change).
- The proposed approach is interesting, as it allows for the intimate mixture of polymer electrolyte membrane (PEM) and anion exchange membrane (AEM) polymer fibers with a water-splitting catalyst to produce

BPMs with different thicknesses (though the target thickness was not defined). However, it is unclear what benefits such a homogeneous mixture can provide compared to a conventional two-layer BPM, or how it will affect crossover through the membrane. This approach still requires platinum-group-metal-based (PGM-based) catalysts at both electrodes.

- The approach is focused on overcoming barriers for electrolyzer and fuel cell performance.
- The approach of the project is to demonstrate regenerative fuel cells using perfluorinated BPMs. The general approach is good; however, the detailed approach for making the proposed system affordable is not clear from the presentation. This is because the target milestone (500 mA/cm^2) is not a measure of the performance without the cell voltage level. If the target is merely 500 mA/cm^2, then commercial BPMs already met the milestone before the project began. In addition, other researchers have suggested the use of 3D structure. There does not seem to be much of an innovative idea except for putting together two available membranes.
- The team was not clear about why the junction should be porous. It was good that they are comparing against a commercial membrane, but they were not clear which one is being used or how it is structurally or functionally different from what they are making.

Question 2: Accomplishments and progress

This project was rated **2.6** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has made good progress but has fallen behind the original schedule. The team has been able to electrospin membranes and demonstrate a lower resistance of the water dissociation reaction by adding GO at the 3D interface. The team has demonstrated performance superior to that of a commercial BPM in an electrolyzer MEA and achieved current densities of 5mA/cm^2 in both fuel cell and electrolyzer MEAs. (This was not achieved with the same membrane, and higher pressure [311 kPa] is needed for a fuel cell.) The project is behind the original schedule, and the team has delayed the milestone of demonstrating an area-specific resistance of <0.2 ohm/cm^2 and achieving 500 mA/cm^2 in both electrolyzer and fuel cell modes. The issues with removing halides from the electrospun AEM could slow progress. It would be beneficial to find a different method or different copolymer to allow for electrospinning the hydroxide form of the alkaline ionomer. The characterization of the membranes, and specifically of the membrane interface, is lacking. It would be nice to see what characterization has been done, even if it would show only the morphology at the interface. If the GO catalyst is dispersed in just one of the ionomers, the team should be able to get some indication of what the interface looks like from the GO distribution.
- One of the intermediate targets—a current density of 500 mA/cm^2 for both fuel cell and electrolyzer modes—has been demonstrated, though for different cells, with high PGM loading for the fuel cell. However, the cell voltage at this current density is unsatisfactory, and it is not clear how durable the BPM produced by electrospinning is (the fuel cell voltage started to decline within 20 minutes). It seems that the major improvement was reached not because of the 3D architecture of the BPM but because of the introduction of GO. No data on membrane thickness have been given, so the comparison with commercial BPMs is not correct. No progress was made in improving the understanding of the electrospun BPM junction interface.
- Progress has been made toward the development of a 3D BPM that demonstrates initial performance characteristics that are close to those of a commercial BPM. The advantages of a 3D design are not very clear, since the voltage–current density curves look very similar. The DOE goals in terms of performance are not clearly defined.
- The go/no-go goal was met, but with two different membranes; this makes RFC application difficult. The complications of 2D-to-3D membranes need to be better understood.
- Demonstrating the electrolyzer and fuel cell performance was a good accomplishment. However, considering the funding level, the National Renewable Energy Laboratory (NREL) team should provide more data. It does not seem like the 3D structure is much better than the 2D structure in cell performance, in spite of the notably lower water-dissociation-reaction resistance. The electrolyzer performance should be compared with the performance of the 2D structure. There is no 3D structure performance for the fuel cell MEA mode; there are no data that show how the 3D structure has been optimized; there are no data on how

to optimize the electrospinning process; and there are no microscopic data on the morphology of the 3D structure. Overall, the progress of the project is disappointing.
- This would have received a "fair" or "poor" rating if not for the no-cost extension and personnel difficulty. The team has not done a sufficient job in characterizing the structure of the created interfaces. The team has not truly developed a baseline or control in this study. In addition to what has been done, the team should also create the electrospun junction without the GO to elucidate whether the improvement is from the project's electrospun polymer or the GO—or if the GO is doing anything at all.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- The partnership with 3M and the Colorado School of Mines is positive.
- The collaboration between groups within NREL appears to be going well. Collaboration with the Colorado School of Mines is not evident in the presentation. It is not clear what characterization has been done other than the electrochemical characterizations, which appear to have been done at NREL.
- This is a well-coordinated effort within NREL; however, contribution from the Colorado School of Mines is not noticeable. Collaborating with other institutions outside of NREL may help with membrane characterization and optimization.
- There is clear collaboration with industry and academia and inside NREL, mainly based on the use of materials for BPMs, fuel cells, and electrolyzers. However, there is no apparent coordination between the partners.
- The project needs more involvement from the outside collaborators, if possible. Using 3M's nanostructured thin film catalyst is advantageous for the project. NREL may want to provide data using other available catalysts for comparison, and also change the catalyst loading to see the impact of the catalyst on device performance. No data from the Colorado School of Mines can be seen. Leveraging the research efforts with other DOE projects is excellent.

Question 4: Relevance/potential impact

This project was rated **3.3** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The relevance and potential impact of this project is high. Bipolar cells could theoretically take advantage of the high PGM-free activity for oxygen reduction reaction in alkaline conditions, and the high activity for hydrogen oxidation reaction in acidic conditions, providing a better PGM-free cell. Issues at the bipolar interface have hampered BPMs; this approach could solve those issues.
- The relevance of the project to the interests of the DOE Fuel Cell Technologies Office is high. As the project's target is to demonstrate RFC technology with high round-trip efficiency, the potential impact of the project on fuel cell and electrolyzer technology may be significant.
- If successful, this technology could move toward the use of RFCs for long-term energy storage.
- The 3D design expands the possibility for operating range and higher efficiency.
- This project is aligned with DOE goals on the development of RFC technology.
- The overall project is directed toward the right goals, but the approach and execution could be improved.

Question 5: Proposed future work

This project was rated **2.8** for effective and logical planning.

- The proposed future work will focus more on instrument-centered research and performance measurement. While the project has shown some promising performance in the fuel cell and electrolyzer modes, more efforts on BPM development are needed. Particularly, the optimization of the 3D structure and the construction of desirable morphology and membrane formation are urgently required to warrant a successful outcome from the project.

- The future work correctly targets demonstrating durability operating in both individual modes (electrolyzer and fuel cell); however, the work focuses mostly on optimization of the current setup (catalyst loading, flow field, etc.) without finding the root causes of instability and low activity.
- The work plan looks good; a staffing issue can slow down progress.
- The proposed future work is more or less straightforward.
- The proposed future work is reasonable. Non-electrochemical characterization of the membranes and interface is lacking here; the team's efforts in characterization should increase. Increased efforts to characterize the interface, ex situ testing (relative humidity cycling) for interface stability, and mechanical testing, etc. should be included.
- The proposed future work is focused on overcoming barriers in terms of performance in fuel cell and electrolysis modes. The performance goals are not clearly defined. For example, 500 mA/cm^2 as a go/no-go decision should be complemented by an operating voltage. "High fuel cell and electrolysis performance" is not a very specific goal.

Project strengths:

- The strength of the project is in NREL's facilities and capability to do the research. Teaming with 3M is another strength. Leveraging project efforts with other existing DOE projects is particularly attractive for increasing the success rate of the project.
- The project team has shown good improvement versus the commercial BPM, especially on the electrolyzer side. Also, the team clearly has the expertise to be successful.
- The project's strengths include its electrochemical characterization and characterization techniques.
- The project's strengths include its novel method for manufacturing BPMs, which may result in the development of RFCs for energy storage.
- The project proposes a novel approach for the 3D design of a BPM.
- The 3D design is a good strength; the partnerships are good.

Project weaknesses:

- The project approach is rather empirical; the material selection for BPMs is limited by the available ionomers and catalysts that are sometimes not designed for selected materials. The leftover salt ions in the BPM electrolyte after water electrolysis seem to be a fundamental problem for the fuel cell operation. The BPM resistance is too high for practical applications. There are no cost estimations or a cost–benefit analysis of electrospun BPMs.
- The research progress is slow. For example, one of the objectives of the project was stated as "The key technical aspects of the project are focused on fabricating and optimizing electrospun 3-D junction… membrane characteristics such as composition, fiber diameter, and the incorporation of catalysts and particulates at the interfacial or junction." However, it does not appear that any of those have been accomplished at a satisfactory level. The project's principal investigator, team members, and the Colorado School of Mines need to spend more time and effort on this project.
- The project's weaknesses include the following: (1) The achievable current density is low. This may be a project-ending issue eventually. These bipolar systems are notoriously lower-performing than AEM- or PEM-only systems. (2) The team does not appear to show a single cell or cell condition in which both the fuel cell and electrolyzer mode were tested and efficiently evaluated. (3) The team did not show a clear protocol for how the unitized RFC would be tested or what the metrics for success are.
- Data for the reversible mode and cycling were not shown; the team needs to make sure that both modes are in good performance and that life data are shown. The efficiency pathway is also not shown.
- The project's weakness is in the lack of membrane characterization and the uncertainty of the project's metrics.
- The project's weaknesses include its membrane and interface characterization.

Recommendations for additions/deletions to project scope:

- Recommendations for the project scope include the following: (1) A more robust physical characterization of the electrospun material and interface is needed. (2) The team should create the electrospun interface

without GO and compare the water vapor gas constant value. The team should determine whether it is the catalyst or the structure that is having the effect. (3) In future presentations, the project team should try not to refer to the electrodes as "anode" or "cathode"; they should be "O_2 electrode" and "H_2 electrode." (4) The fiscal year 2018 go/no-go of 0.5 A/cm^2 should be achieved using a single cell. (5) It should be determined where the Cl goes when the AEM component is exchanged from the Cl- form to the OH- form during electrolysis mode. The team should determine if it evolves as Cl_2 and whether that has any safety ramifications.

- The project team needs to measure the round-trip efficiency and cycle life of the most optimized 3D BPMs. More characterizations of the 3D structure are also required. The progression of the performance improvement, depending on the BPM structures, needs to be presented.
- Modeling would help with the fundamental understanding of BPM performance and its optimization.
- The study of BPM structure and catalyst–ionomer interaction should be added to the project scope.
- Developing cycling data is critical, as is higher efficiency.

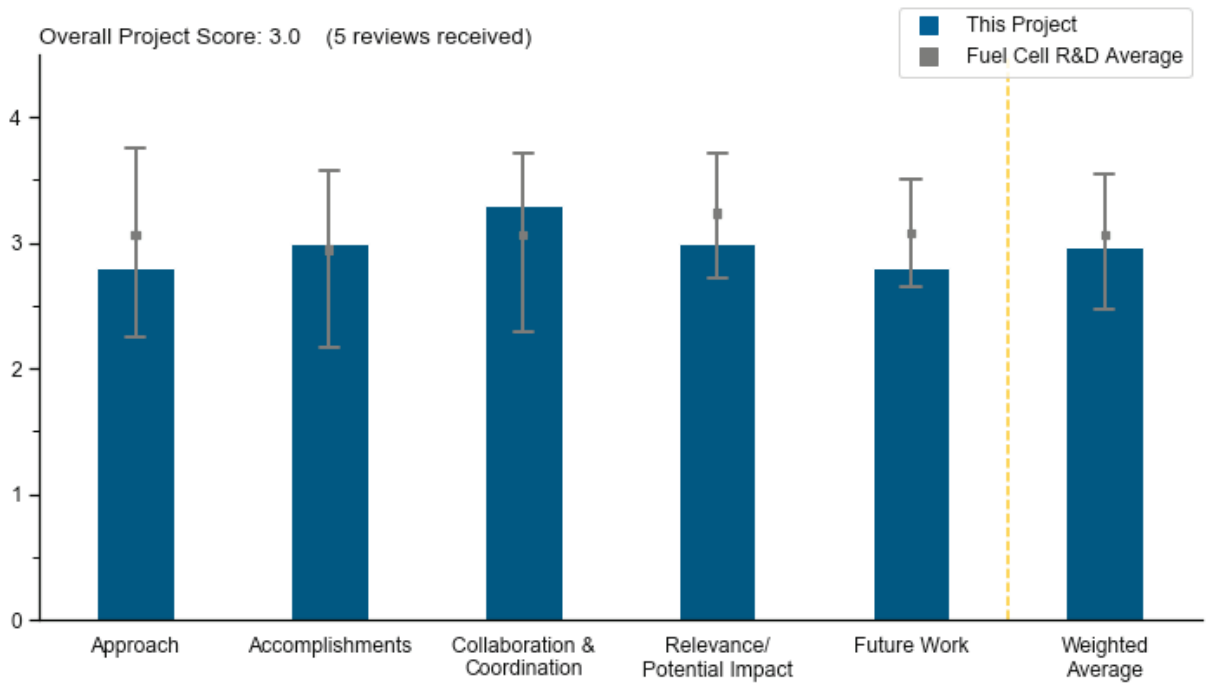
Project #FC-183: Lab Call Fiscal Year 2018 (Reversible Fuel Cells): Technology-Enabling Materials, Cell Design for Reversible Polymer Electrolyte Membrane Fuel Cells

Nem Danilovic, Lawrence Berkeley National Laboratory

Brief Summary of Project

Unitized regenerative fuel cells (URFCs) are energy storage devices that store electricity in the form of hydrogen and oxygen gas (electrolyzer mode) and produce electricity (fuel cell mode). The project will show the feasibility of fixed-polarity URFCs and an engineered bifunctional oxygen evolution reaction (OER, for electrolyzer mode)/ hydrogen oxidation reaction (HOR, for fuel cell mode) catalyst. To do this, Lawrence Berkeley National Laboratory (LBNL) will show the feasibility of the URFC approach in membrane electrode assembly (MEA) testing using state-of-the-art polymer electrolyte membrane fuel cell (PEMFC) and electrolysis materials and developing an application-relevant cycling protocol and accelerated stress test. Argonne National Laboratory (ANL) will demonstrate the feasibility of the engineered supported catalyst approach by (1) developing an atomic layer deposition (ALD) process and (2) characterizing the activity and stability of the supported bifunctional catalyst versus baseline materials for HOR, OER, and cycling.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Question 1: Approach to performing the work

This project was rated **2.8** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The approach is original and may be interesting if it fulfills all U.S. Department of Energy targets concurrently, not only at the MEA level but also at the system level. The investigated approach will need purge phases when switching from electrolysis mode to fuel cell mode and the reverse. This will induce some hydrogen and oxygen release and affect the system efficiency and system time operation.

- The project focuses on development of URFCs using an innovative concept of cell fixed polarity and an engineered bifunctional OER/HOR catalyst. This concept may provide a more effective use of bifunctional catalysts but requires more complexity from the UFRC operation.
- The approach leverages the skills of LBNL and ANL with private industry; collaborating with both 3M and Proton OnSite is a very productive combination. The fixed polarity mode concept is an interesting opportunity for regenerative fuel cells. The catalyst design strategy is good for yielding better MEA performance.
- The technical barrier being addressed is the operation of a URFC with "fixed polarity," keeping one side as the anode and the other side as the cathode for the two operating modes. Because of the dual operation, the project is addressing barriers identified for the fuel cell and water electrolysis but with targets being a compromise between the two applications. Thus, clear specific targets have been proposed. The most complicated part of the approach is to use the same electrode as a single anode for operating both a PEMFC and a polymer electrolyte membrane water electrolyzer. The project's core activity is to identify the right catalyst based on materials used for hydrogen oxidation or for oxygen evolution, which are Pt- or Ir-based. The work started with assessment at the rotating disk electrode (RDE) level, then at the MEA level. A major issue is the poor stability of the anode catalyst under voltage cycling (up to 1.6 V to simulate an OER situation, as requested), which is leading to strong losses with the HOR.
- The team does not make a compelling argument for why there would be synergy between the oxygen reduction reaction/hydrogen evolution reaction and/or the HOR/OER. In fact, despite the fact that one electrode is always doing a reduction or oxidation, the potentials are totally different, and the swings in the potential at both electrodes make it seem like it would be higher than that of a catalyst in a typical "fixed gas" URFC. It is unclear why the ALD is necessary, whether it is scalable, or how this process would help with cost.

Question 2: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The experimental data showed better performance of the proposed URFC in the fixed polarity, compared to the conventional fixed gas mode. The high (60%) round-trip efficiency demonstrated at the current density of 1 A/cm^2 is impressive, and the go/no-go milestone has been met. Interestingly, the down-selected bifunctional HOR/OER catalyst is just a physical mixture (9:1) of Ir and Pt black, which outperforms more elaborate structures.
- The cycling data are meaningful and promising. The go/no-go goal has been met. The test stand issue may cause delays. The results need to be illustrated with greater clarity and compared with the project's goals.
- The team has shown proof of concept early on, which is good. They do need to develop protocols more similar to how real URFCs would be deployed in the future.
- The work is ongoing, following most of the future work indicated in the previous year. Some aspects are missing. For example, the project team did not present varying flow fields with discrete MEAs or evaluations of the performance and durability of the traditional versus the proposed URFC concept. In addition, there is no information about the comparison of the different operation modes.
- New results have been provided. However, the stability of the investigated catalysts has not been demonstrated in both modes. No information is given regarding the purge effect or the time spent switching from fuel cell mode to electrolysis mode and the reverse. No information is given on the future stack and system pressure.

Question 3: Collaboration and coordination

This project was rated **3.3** for its engagement with and coordination of project partners and interaction with other entities.

- It appears that both members of the team are participating and that ANL materials are being deployed well at LBNL.
- There is a working collaboration between LBNL and ANL, as well as participation from industrial partners.

- ANL's expertise in catalysts is a plus. The project is leveraging LBNL's capabilities.
- The collaboration between LBNL, ANL, and the two industrial partners, 3M and Proton OnSite, appears correct.
- The collaboration between ANL and LBNL is clear and positive for the research side. The contribution of the industry partners is not obvious at this stage of the project and should be increased for the definition of assessment tests.

Question 4: Relevance/potential impact

This project was rated **3.0** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The proposed fixed-polarity concept could be instrumental in making URFCs work equally well in both operational modes. Such URFCs could find wide application in long-term energy storage.
- The investigation of reverse PEMFCs is of interest but challenging. Even if a relevant MEA is developed, system efficiency and total cost of ownership will be the key drivers when comparing architecture consisting of two systems or two stacks (i.e., fuel cell and electrolysis) to common balance-of-plant components.
- The project is considering several DOE objectives but addressing a very specific device; thus, it is not easy to draw conclusions for the expected impact. If the project's objectives are reached, progress will be made toward cost and performance objectives for PEMFCs and efficiency in hydrogen production.
- The project team needs to show how the materials contribute to higher efficiency on a round-trip basis and other cost benefits. Batteries are a major competitor, and no comparison with these is given.
- The project is focused on meeting DOE targets, but there are some remaining issues to making its possible impact high.

Question 5: Proposed future work

This project was rated **2.8** for effective and logical planning.

- The project's main next steps will be to find out how to improve the anode catalyst by changing the structure. There is a strong risk of failure that should be taken more into account. After that, selecting the best mode for actual validation of durability should be the priority.
- The proposed future work is well aligned with identified challenges and issues but lacks quantifiable metrics. There is no plan to explore decreased platinum-grade-metal loading.
- Everything that is proposed makes sense, but there could be more attention paid to the protocols for testing.
- The chosen operating cycle of four hours is acceptable, but it may be adapted to real duty cycles (i.e., fuel cell and electrolysis duration), depending on its applications. The operating single cell conditions should consider the targeted working pressure of 30 bars.
- The relationship to efficiency and cost goals should be included. Life-cycle improvements were not clarified.

Project strengths:

- Surprisingly, high performance was achieved with first-generation materials. Both team members are working well together.
- The idea of developing a single catalyst layer as a fuel cell and electrolytic anode is innovative and could be a strength, but demonstration is required. The proposal of a specific duty cycle for URFCs should be valuable for further studies on this type of device; it should be validated on the selected operation mode.
- The project has a good combination of component development work (i.e., catalyst and MEA) and technoeconomic analysis.
- The laboratory collaborations are a good strength, with ANL in particular. Industry feedback is important.
- The competence of the project partners is a strength of the project.

Project weaknesses:

- The poor stability of the anode catalyst under voltage cycling (as requested, up to 1.6 V to simulate the OER situation) is leading to strong losses for HOR. This is a major issue, and the approaches planned as mitigation, such as dissociating Pt and Ir, each on its support, are not convincing for solving the problem of Pt dissolution. Information such as the microstructure of the catalysts developed should be added in the characterizations and used in data analyses. At this stage, between fixed polarity, fixed gases, and vapor feed, the best-suited operation mode (i.e., the most stable and most efficient) should already have been selected for further evaluation of durability; this should be fixed soon.
- The switch between fuel cell and electrolyzer modes in the proposed concept is not trivial and may result in a dangerous mixing of hydrogen and oxygen; therefore, a purging procedure should be developed. The URFC's durability under switching operations is extremely important, and the team should pay more attention to this issue. The effect of water management on the URFC performance should be studied, and an optimization should be made.
- It is not currently clear what catalyst will concurrently meet the fuel cell and electrolysis targets and stabilities. The system operating conditions (e.g., output pressure) are not considered.
- The project's weaknesses include the following: (1) The team does not have a reasonable durability test in place. (2) It would seem that ALD will not be a scalable approach, and the team does not justify why it is necessary or what difference it will make at higher metal loadings in future systems. (3) The team does not address electrode development, particularly related to water management.
- Efficiency round-trip and degradation issues are not adequately addressed or correlated with materials used.

Recommendations for additions/deletions to project scope:

- The following are recommendations for the project: (1) In future presentations, the team should better detail the reacting environment (URFC, fuel cell, electrolyzer, RDE, etc.) on the slides. (2) It would seem that deployed URFCs would likely be operated in one mode for longer periods (not voltage-cycled, as was done here). Thus, a better stability test might be to operate these cells at constant current density (e.g., 1 A/cm^2) in fuel cell mode for ~10 minutes to allow for a steady state to be achieved, then switch to electrolyzer mode at 1 A/cm^2 for the same amount of time in order to equally "charge" and "discharge" the URFC. It also seems that controlling the rate would be better than voltage cycling, in which the total charge that is passed could be wildly different for the two cases. (3) Any metrics on performance or durability should be made with a single cell or MEA and materials set.
- Microstructure analyses of the anode catalysts should be added to see if such analyses could help the team understand and improve catalyst behavior. The team should make it a priority to select the best operation mode and complete actual assessment of one URFC on the long term, following the proposed duty cycles. Applying these cycles on "fixed gas" mode could be selected if the proposed approaches do not solve the anode issue for HOR in "fixed polarity" mode. If the issue of the bifunctional anode catalyst is not solved, the difference between the work planned and conducted within FC-313 should be clarified.
- Increasing efficiency is critical for energy storage. Batteries are already >80%, with good cost reductions. Cost and efficiencies should be addressed at a greater level.

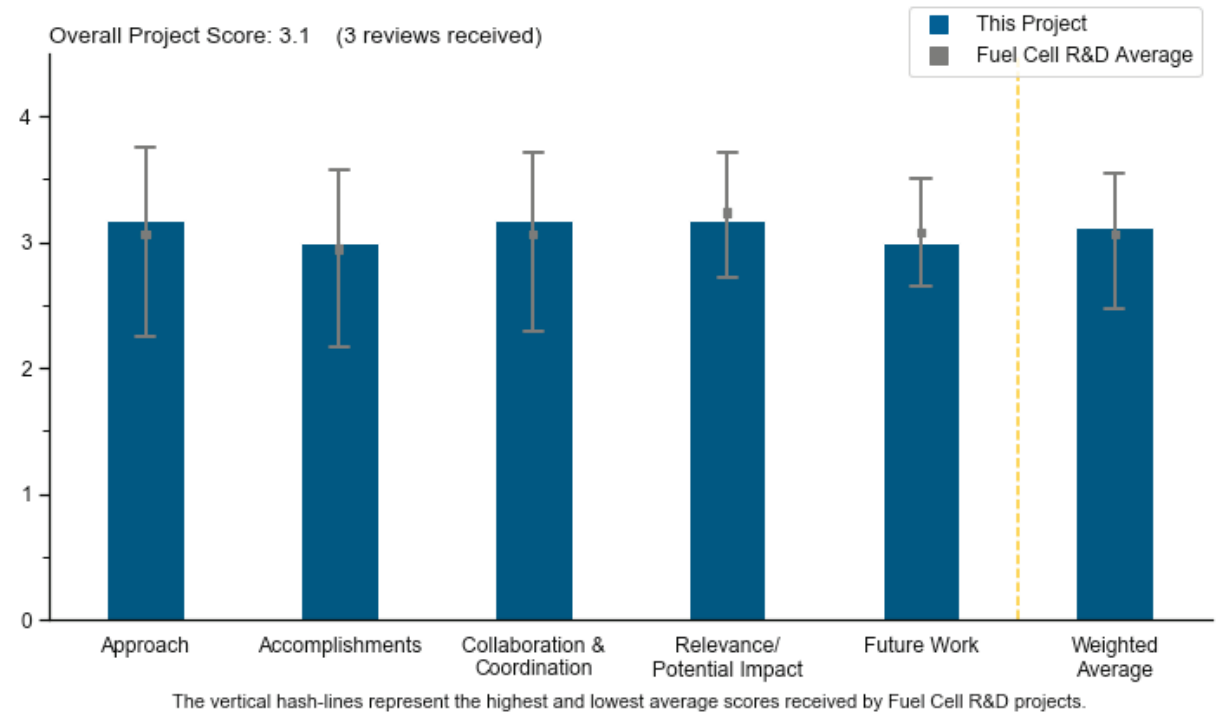
Project #FC-302: Developing Platinum-Group-Metal-Free Catalysts for Oxygen Reduction Reaction in Acid: Beyond the Single Metal Site

Qingying Jia, Northeastern University

Brief Summary of Project

Northeastern University seeks to develop platinum-group-metal-free (PGM-free) oxygen reduction reaction (ORR) catalysts with high activity and durability in polymer electrolyte membrane fuel cells (PEMFCs). These PGM-free ORR catalysts are developed via the following concurrent pathways: (1) M_x-N-C catalyst development featured with multiple metal centers (MMCs) and (2) M_x-N-C catalyst synthesis using surface deposition methods. These catalysts are developed to attain the following performance targets: (1) 0.035 A/cm^2 at 0.9 V in an H_2/O_2 PEMFC (1.0 bar partial pressure, 80°C); (2) loss in activity ≤40% after 30,000 square wave cycles with steps between 0.6 V (3 s) and 0.95 V (3 s), and (3) power density of 0.5 W/cm^2 in an H_2/air PEMFC with a membrane electrode assembly (MEA) size ≥50 cm^2.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.2** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The approach in this project is based on the use of multiple metal (apparently Fe) sites that should be incorporated in carbon materials to improve ORR activity and durability. This approach can be justified as much as all other PGM-free funded projects that are supposed to deliver carbon-based materials with some amount of low-cost transition metals, while maintaining high catalytic activity and durability. Considering that the Fuel Cell Technologies Office (FCTO) has recognized that as its main direction in catalyst development, the approach must be well justified.

- The proposed approach is focused on producing multi-metal active sites that can better facilitate the ORR. The key focus is on the synthesis characterization, but there are also MEA and modeling components to the project.
- In this PGM-free cathode catalyst development project, it is expected that multi-metal-site catalysts will perform better than single-metal-site catalysts. The success of this project hinges entirely on this hypothesis, and therefore a go/no-go decision would make a lot of sense for this project.

Question 2: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- This is the first year of the project, and it is challenging to evaluate what has been accomplished toward DOE goals considering that, so far, the principal investigators (PIs) have mainly managed to synthesize and characterize materials. No MEA data were presented that would support the main expectations of this project. For that reason, all claims from the approach fall into the category of predictions and are speculative. The PIs should be acknowledged for the materials that were made and for the fact that structural characterization is consistent with what was proposed. The main issue that remains is how to justify the idea in this approach. It is not clear why these systems should be more active and durable, as well as what the foundation is for such claims.
- It is the beginning of the project and too early to evaluate accomplishments. The initial activity of the MMC catalysts seems low, based on rotating-disk-electrode (RDE) half-wave potential. It will be useful to devise a means of estimating the MMC site density such that the turnover frequency can be evaluated to better quantify whether the MMC site is more active.
- On slide 6, the results should be shown with respect to reference and/or state-of-the-art materials. This project has just started, and the accomplishments are sparse, as would be expected. The initial results demonstrate the project's capability for the work to be conducted.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- The collaboration between the participants is well coordinated.
- There is collaboration from one academic institution, one national laboratory, and the Energy Materials Network. The technical back-up slides show how the consortium members (perhaps ElectroCat) are being utilized. More details on the roles would be helpful to understand the levels of interaction better. For example, it is unclear who is doing the MEA integration work, how the feedback loop from the testing is given, or how the preliminary evaluation is aligned with subsequent diagnostics.
- There is collaboration outside of Northeastern University with Lawrence Berkeley National Laboratory. The value of the mass transport modeling will be apparent only if the catalyst activity becomes sufficient; otherwise, there is too much uncertainty in future catalyst morphologies that will meet activity targets.

Question 4: Relevance/potential impact

This project was rated **3.2** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The project's topics are right on target with the interests of the FCTO in PGM-free catalysts for fuel cell systems. Although not mentioned in the presentation, these catalyst types could reduce cost significantly if they could achieve higher performance and lifetimes.
- The potential impact would be significant if it could be demonstrated that a multi-metal active site was more active and could be reliably synthesized at high density.

- The project's main issues were, are, and will be the insufficient activity and poor durability of PGM-free systems. A modest knowledge of material science and electrochemistry does not imply that carbon-based materials doped with Fe would ever evolve to the level that would enable application in PEMFC cathodes, considering that even Pt-based catalysts suffer from degradation. Nevertheless, keeping in mind that the goal of this project is to improve activity and durability, the relevance is satisfactory.

Question 5: Proposed future work

This project was rated **3.0** for effective and logical planning.

- The proposed work seems reasonable. Densifying the MMC sites is a key part of the future work.
- Overall, this is a sound approach for the work on the catalyst side. It is unclear why ion-beam-assisted deposition and electro-spinning are highlighted for MEA fabrication. Electrode integration and optimization should be an important aspect. Furthermore, some evaluation of catalyst degradation at the end of the project should be added to the scope.
- The future work should provide feedback that would clearly address the approach of this project.

Project strengths:

- The project's strengths include clear communication about what is targeted and what was accomplished. The project also has had successful synthesis and structural characterization, as well as well-executed collaboration.
- The project has an interesting approach that could have the potential to improve PGM-free catalyst performance significantly.
- The project's strength is its focus on an active site that is more novel than the common MN_4 site.

Project weaknesses:

- The project's weaknesses include the lack of evidence that the proposed approach can effectively address the activity and durability of PGM-free catalysts, as well as the lack of RDE durability measurements or MEA data.
- The project's early weakness is likely the density and verification of the MMCs.
- Everything is based on one hypothesis.

Recommendations for additions/deletions to project scope:

- It is recommended that the project have a go/no-go decision point that shows some indication that the hypothesis is true.
- MEA testing is required.

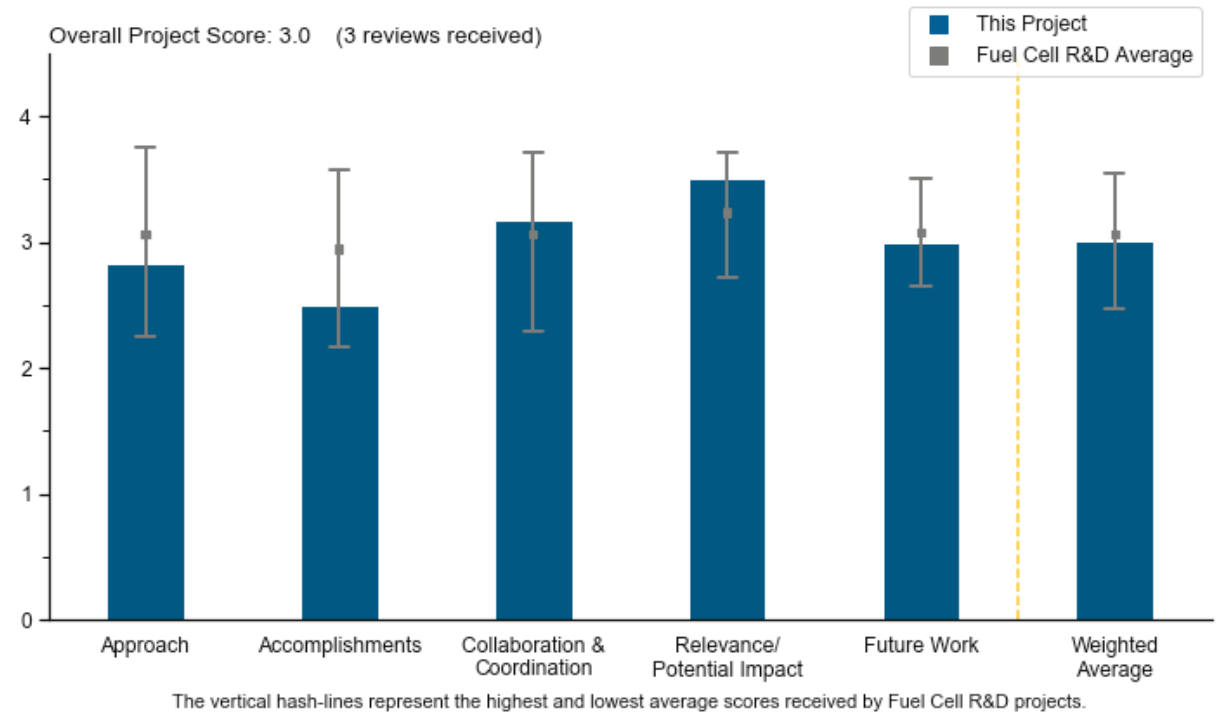
Project #FC-303: Mesoporous Carbon-Based Platinum-Group-Metal-Free Catalyst Cathodes

Jian Xie, Indiana University–Purdue University Indianapolis

Brief Summary of Project

Indiana University–Purdue University Indianapolis (IUPUI) will use controllable synthesis to design and develop advanced hierarchically porous carbon sphere (HPCS) M-N-C catalysts for precious-group-metal-free (PGM-free) cathodes in polymer electrolyte membrane fuel cells. The project team also aims to develop membrane electrode assemblies (MEAs) using the novel ionomer–catalyst interface by controlling the surface charges on catalyst particles to obtain improved catalyst activity, utilization, and high-current-density performance. The project goals are addressed on both intra-particle and inter-particle levels respectively via the following approaches: (1) develop high-performance PGM-free catalysts with mesopore structure and (2) construct an ideal Nafion™ ionomer–catalyst interface within a catalyst layer of MEAs.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **2.8** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This project has four different tasks. Task 1 is the development of HPCS-based M-N-C catalysts with high mass activity. This approach is very solid and led by the University of Buffalo. Task 1.3 addresses the stability of M-N-C catalyst on mesoporous carbon support (MCS@M-N-C). Some milestones need to be added to monitor the durability of these catalysts. Task 2 is the development of cathode layers using the HPCS-based catalysts. The approach to this task has some serious issues. The HPCSs are approximately 500 nm in diameter, and it is expected that most of the active sites would be buried in the smaller (micro- and meso-) pores within this large particle. The approach involves functionalizing only the surface of this

large particle and not the mesopores. Controlling only the ionomer coverage on the outside of these particles might not be enough. It needs to be made clear whether this project relies on just surface proton transport or liquid water to provide access to the active sites within the mesopores and micropores of the carbon particle. This needs to be addressed early in the project and will be key to achieving good MEA performance. Tasks 3 and 4 are MEA preparation, testing, and cost analysis; that approach is sound.

- This is a polymer electrolyte membrane low-temperature electrolysis technology project, as well as a catalyst synthesis and integration project, pursuing the development of HPCSs. From the images on slide 5, it is not apparent how this approach differs from traditional electrode structures. Slide 9 indicates that the hope is that improvement will be made with the particle itself and in its coating with ionomer. The images of slide 5 show a Pt catalyst system, while the project is aiming at PGM-free catalyst development. Images on slide 8 and the following slides are much more aligned with the description of the project, though from the images on slide 11, it is not clear how the ionomer thickness and distribution are being controlled.
- While the project is good in terms of scope, the approach is not well defined in terms of achievability. It seems a bit overly optimistic and ambitious. It is not clear that adding mesoporosity will increase the non-PGM mass activity, as it is unknown whether it is mass-transport-limited; what is really needed is an increase in the number of active sites. Similarly, the idea of an ideal ionomer–carbon interface is very good, but again, the approach is unknown in terms of its feasibility, as it relies on several understandings and assumptions that are not well known as of yet. For example, it is unknown what morphology or properties are wanted with the ionomer backbone interactions versus the side chains. Ink formulation by itself is perhaps a very large effort, and here it seems to be trivialized. The exact surface groups being examined are not well discussed.

Question 2: Accomplishments and progress

This project was rated **2.5** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Considering that this project is just starting, there has been great progress, especially in regard to Task 1. The University of Buffalo has already demonstrated $E_{1/2}$ of 0.82 V versus a reversible hydrogen electrode, which was a milestone. The team should look into and report on the durability of this catalyst in parallel with the electrode design tasks.
- No accomplishments and/or signs of progress were presented. This is understandable, as the project has not spent any money yet and obviously has not started yet.
- The project has not started.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- The project has a good team, with the University of Buffalo providing catalysts, IUPUI making electrodes and MEAs, and the United Technologies Research Center providing testing and cost analysis.
- The project has a good team that is wide-ranging in terms of tasks, but there should be more effort devoted to characterization. The understanding gained from discussions with a national laboratory would be helpful.
- The project has an academic lead, with collaborators from one university and one industry research center. Beyond listing the partners, it was not clearly laid out how the partners would communicate and/or interact with each other. It would be helpful if there were more information about the actual collaboration and some understanding of how the partners work together and/or exchange information.

Question 4: Relevance/potential impact

This project was rated **3.5** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The project topics are right on target with the advancement of the technology and the goals of the DOE Hydrogen and Fuel Cells Program. Reducing the costs of available cost-effective catalyst materials is important. It is unclear how well this seemingly complicated catalyst fabrication process could be scaled up, or what impact scaling up might have on the cost of the material. This will, however, be addressed in year 2, quarters 1–3, of the project.
- This project supports ElectroCat, and the milestones are well aligned with the consortium's goals. The team should work closely with the ElectroCat laboratories to determine the stability of these catalysts. The laboratories can provide key capabilities that would complement the team's ability to understand these materials and electrode structures.
- Nominally, the project is tackling very important and critical fuel cell issues. However, it is not clear whether the approach is the correct one to solve those problems.

Question 5: Proposed future work

This project was rated **3.0** for effective and logical planning.

- Overall, this is a very sound, comprehensive project plan. The role of the ionomer will be critical for the success of this project. There seems to be too little attention being paid to understanding the effect of ionomer loading on transport phenomena and, ultimately, performance. PGM-free catalysts face significant durability challenges. Some durability evaluation needs to be conducted at some point to put this system in context with other PGM-free systems.
- The proposed work is perhaps too wide-ranging and ambitious, as it relies on understandings that are still not well developed. There is concern about Task 2 in terms of how many functionalities can be examined and the need to develop full catalyst layers. Simpler studies and setups are recommended. For Task 1, it is unclear whether the system is mass-transfer-limited or whether the catalyst site density will be increased and activity per site maintained.
- The project's future work is almost identical to its approach since this project is just starting.

Project strengths:

- This project is tackling a very relevant problem from two related angles and has some interesting aspects. The functionalization of carbon is a good idea, although the team needs to ensure it is maintained and is positive in terms of increasing performance.
- The project has good teaming with clearly defined roles. The project team has novel ideas about M-N-C catalysts and HPCS development that show promise for making better PGM-free catalysts.
- Multiple approaches have been suggested for achieving the project's goal. Overall, there is very sound project planning.

Project weaknesses:

- The surface functionalization and ionomer incorporation are unproven and need to be demonstrated early in the project. There is not much science or understanding on how functionalization will affect ionomer distribution. To provide DOE with lasting value from this project, some fundamental studies of how functionalization affects ionomer coverage and thickness need to be added. With advanced characterization techniques from ElectroCat, these studies might be accomplished with a systematic study.
- It is unclear how the partners interact and help each other to achieve the project goals. It is also unclear how the ionomer thickness is controlled or to what extent ionomer will fill the pores and hinder gas transport. Understanding the effect of ionomer distribution and being able to control it will be key for the success of this project.

- The project relies on a good deal of understanding or empirical luck to occur quickly. It is not certain that the objectives can be met within the proposed time, scope, and budget.

Recommendations for additions/deletions to project scope:

- It is recommended that the team add more model-type studies and more characterization so that understanding can be gained before just trial and error. The scope should perhaps be readjusted to be more focused.
- The team should add a durability milestone, at least at the end of the project. This is a known issue with all PGM-free catalysts, and it needs to be explicitly addressed, at least toward the project's end.
- Results and/or signs of progress need to be demonstrated. The project has apparently not started yet. It is also recommended that the team do some long-term testing to probe the durability of this catalyst structure.

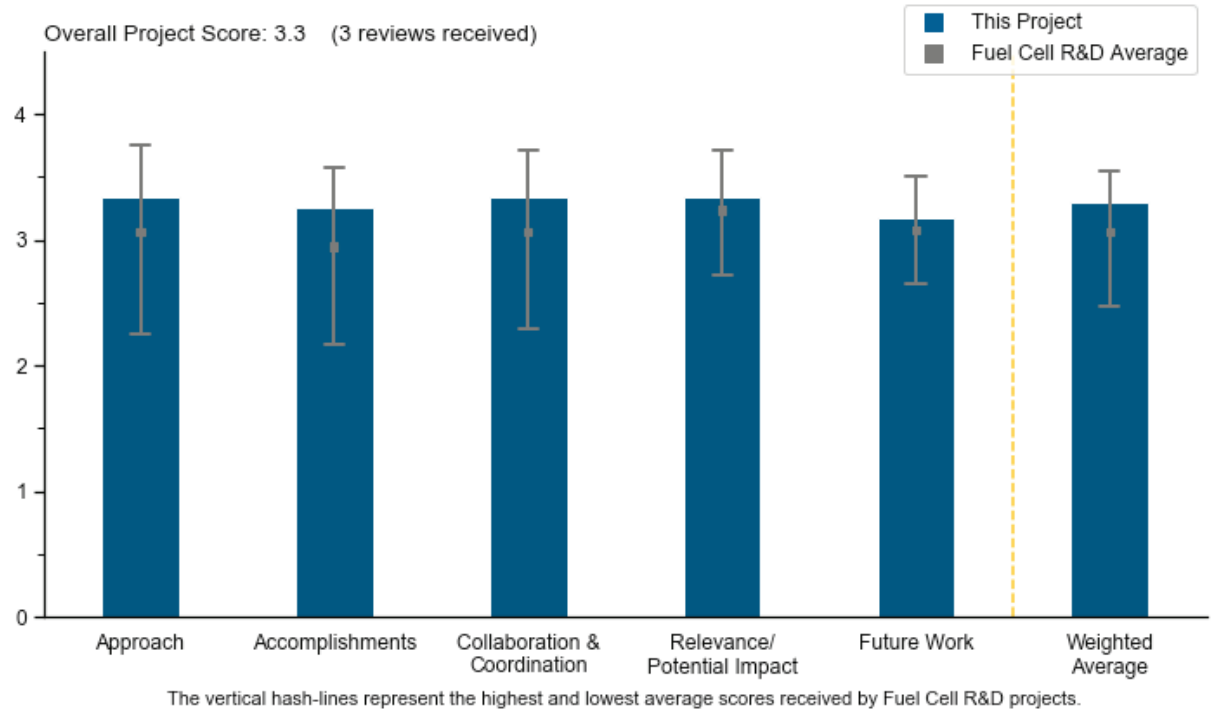
Project #FC-304: Fuel Cell Membrane Electrode Assemblies with Platinum-Group-Metal-Free Nanofiber Cathodes

Peter Pintauro, Vanderbilt University

Brief Summary of Project

This project will fabricate, characterize, and evaluate electrospun nanofiber mat electrode membrane electrode assemblies (MEAs) with platinum-group-metal-free (PGM-free) oxygen reduction reaction (ORR) cathode catalysts for H_2–air fuel cells. The novel electrospun electrode structure has potential to overcome shortcomings of conventional fuel cell electrodes (e.g., prepared by slot die coating) to improve MEA performance and durability. This is especially true for PGM-free ORR catalysts that have issues with catalyst durability (associated with metal leaching) and high cathode catalyst loading (i.e., thick cathodes, with the potential problem of significant O_2 and H+ mass transfer resistance). The project seeks to better understand and further improve the power output and durability of nanofiber mat fuel cell cathodes and MEAs with state-of-the-art (SOA) PGM-free catalyst powders (from project partner Pajarito Power). The project will also optimize electrospun nanofiber mat cathode composition and morphology for MEAs to meet U.S. Department of Energy performance and durability targets.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.3** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The project has a well-organized approach with a good understanding of the properties and requirements of electrodes. The principal investigator recognizes potential difficulties and has anticipated a research approach to resolve or test these difficulties.
- The project's approach is effective and contributes to overcoming most barriers. The objectives and critical barriers are effectively identified and are being addressed.

- The project's approach is to utilize nanofiber technology to enhance the performance of thick non-PGM catalyst electrodes.

Question 2: Accomplishments and progress

This project was rated **3.3** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- This is a new project. Past work demonstrated the ability to form and attach catalysts to nanofibers. This is key to developing a high-performance, PGM-free electrode. The work has demonstrated a break-in period for the nanofiber PGM-free electrodes, with increased performance after 50 hours.
- The project had just started upon slide submission, so few specific project results were available. The pre-project results shown indicate good initial feasibility.
- The project has just started; all of the results shown were prior results.

Question 3: Collaboration and coordination

This project was rated **3.3** for its engagement with and coordination of project partners and interaction with other entities.

- The technical, synthesis, and fabrication areas are covered by team members. The project may need additional help in cost analysis. There is extensive collaboration with national laboratories for testing nanofibers and catalyst performance. Collaborators have demonstrated technical capabilities in measuring electrode properties and performance. Industry partners were identified that could move the successful project toward the manufacturing of nanofiber PGM-free fuel cell systems.
- The project plan indicates good coordination between the formal project organizations and the Electrocatalysis Consortium (ElectroCat).
- There is good collaboration between the three partners and good interaction with ElectroCat.

Question 4: Relevance/potential impact

This project was rated **3.3** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- PGM-free catalysts have low power density, and the use of nanofiber mat electrodes offers an approach to increasing the nominal power density. However, there will be a question of catalyst utilization for the mat electrodes, and it is not clear that a ten-fold increase in power density will be feasible. If successful, the potential impact will be important.
- For PGM-free catalysts to be incorporated in commercial devices, questions of power output and durability— which this project seeks to address—must be investigated.
- The project has good prospects for advancing the performance of PGM-free catalysts. However, PGM-free durability is a severe fundamental challenge, and the project is not addressing this issue.

Question 5: Proposed future work

This project was rated **3.2** for effective and logical planning.

- The proposed future work is logical, based on the project objectives.
- The future work is reasonable, but it would be good to see a risk and mitigation table.
- The future work emphasizes catalyst agglomerate size. However, for the PGM-free electrodes to be successful, a three-dimensional electrode needs to be developed. The future work does not include studying the effects of mat thickness. The future work does not appear to evaluate catalyst utilization.

Project strengths:

- The project brings together an alternative PGM-free cathode catalyst with nanofiber fabrication to improve the power density of a PGM-free cathode and yield short-term stable performance (300 hours) at 0.5 V, after initial wetting of the mat nanofiber electrode.
- The project will utilize SOA PGM-free catalysts and nanofiber electrode technology, which has been shown to be very effective with PGM catalysts.
- This is a good team with experience in the areas being investigated.

Project weaknesses:

- The project addresses improved catalysts but does not address the varied thickness of nanofiber mat cathodes. A three-dimensional dispersion of PGM-free catalysts is necessary to get nominal power density at levels similar to electrodes that contain PGM catalysts.
- This is a large monetary award for a university-led project. The scope of the work seems small when considering this budget and the relatively low overhead at universities and companies.

Recommendations for additions/deletions to project scope:

- A recommended addition to the project scope includes the development of nanofiber, PGM-free mat electrodes of different thicknesses, with measurement of the catalyst utilization as a function of mat thickness. It could be anticipated that thicker mat electrodes could have lower catalyst utilization, similar to what has been observed with PGM electrodes. This would suggest that with thicker nanofiber PGM-free electrodes would have reduced effectiveness and present limits on increasing the power density of the fuel cell system and, therefore, an inability to achieve performance at a level competitive with PGM fuel cell systems.

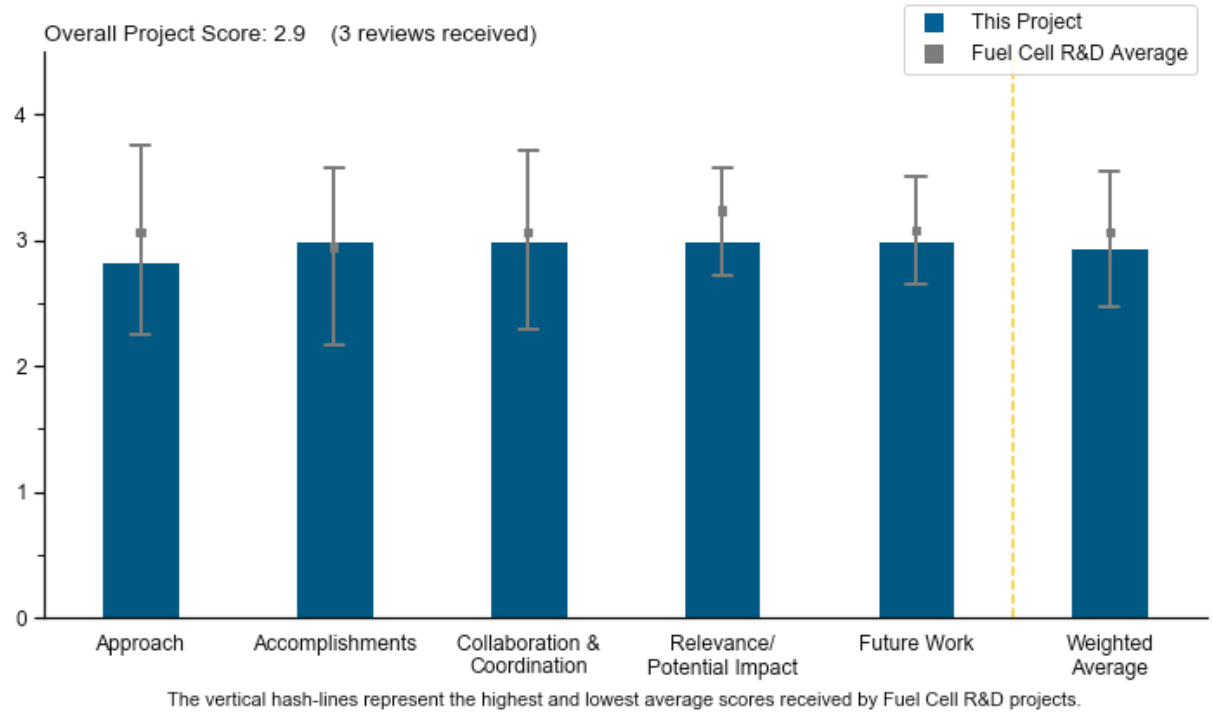
Project #FC-305: Active and Durable Platinum-Group-Metal-Free Cathodic Electrocatalysts for Fuel Cell Application

Alexey Serov, Pajarito Powder

Brief Summary of Project

The project objectives are to (1) develop platinum-group-metal-free (PGM-free) electrocatalysts for oxygen reduction reaction (ORR), (2) scale up production of the catalysts to 50 g batches, (3) integrate PGM-free catalysts into the industrial state-of-the-art membrane electrode assemblies (MEAs), and (4) comprehensively evaluate the catalysts using electrochemical methods. The project addresses existing barriers by (1) increasing the activity of PGM-free ORR catalysts, (2) decreasing the cost of PGM-free catalyst manufacturing, and (3) increasing PGM-free catalyst durability. Improved understanding of the electrochemical processes relevant to PGM-free materials in mass-produced MEAs will allow commercial manufacturers to develop inexpensive, highly active, and stable PGM-free ORR catalysts that demonstrate performance levels required by the U.S. Department of Energy.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **2.8** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- Utilizing sacrificial silica substrates has been a successful approach at small scale, but scaling up to 50–200 gr batches will be beneficial. The proposed use of probe molecules should provide good quantification of active sites. Use of more than one probe molecule should provide more information. The approach of optimizing MEAs with PGM-free catalysts using high-throughput screening is beneficial. Capability for spatially graded catalysts and ionomers, especially in the thickness direction for thicker PGM-free catalyst layers, may prove beneficial.

- The project approach is satisfactory and clearly specific to using morphology techniques to optimize catalyst performance.
- The approach is focused on large-batch manufacturing of a PGM-free catalyst, producing MEAs, and characterizing fuel cells. The approach does not include a significant effort to develop a new catalyst. The preliminary data are from a 2013 Annual Merit Review report.

Question 2: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- It is the beginning of the project, so it is too early to comment on accomplishments. The principal investigator (PI) and Pajarito Powder have made significant advances in large-batch synthesis of a PGM-free catalyst. The progress toward PGM-free targets depends on the ability to significantly increase activity while maintaining the present durability.
- The project began only in January, so not much progress is expected, and there is not much to show to date. Pajarito Powder has been able to show increased graphitization over the baseline. Performance of the graphitized samples has not been demonstrated yet. Initial stability (over a limited 12-hour hold) looks good, and the team has achieved the first milestone of 25 mA/cm^2 at 0.83 V iR-free.
- Although the project is in the beginning stages, the team has already achieved a performance milestone.

Question 3: Collaboration and coordination

This project was rated **3.0** for its engagement with and coordination of project partners and interaction with other entities.

- The project has just started, but collaborations between partners appear to be working well. Plans for collaboration with ElectroCat are not apparent. It is unclear whether the team will take advantage of ElectroCat's capabilities of—and if so, which ones—or if the interaction will just be supplying catalyst to ElectroCat.
- The team is well aligned, and the roles are assigned clearly; there is no duplication of effort. Verification testing at Hawaii Natural Energy Institute in the different cell architecture is essential.
- The partners are appropriate for MEA development and testing.

Question 4: Relevance/potential impact

This project was rated **3.0** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The project is relevant to DOE's fuel cell efforts and fits in with the recent emphasis on PGM-free catalysts and low-technology-readiness-level material development. The project seeks to address barriers of performance, cost, and durability for PGM-free catalysts and PGM-free-catalyst-based MEAs.
- The team set well-aligned performance targets with the DOE objectives; however, the durability targets of the project are less clear.
- The learning in mass producing PGM-free catalysts is valuable as more active catalysts emerge.

Question 5: Proposed future work

This project was rated **3.0** for effective and logical planning.

- The PI's plans for scale-up and testing are good. Effort to develop activation protocols is important, but it is not clear whether an activation protocol will be universal for all PGM-free catalysts, or even all $Fe\text{-}N_4$-type PGM-free catalysts, because of the different precursors and preparation routes used. It is not clear what changes are targeted for Generation 2 (Gen 2) or what properties will be changed to improve performance.

- Proposed work is relevant to the milestones and Hydrogen and Fuel Cells Program work scope.
- There should be a key focus on increased activity with the Gen 2/Gen 3 catalysts.

Project strengths:

- The strength of the project is the team, including catalyst and MEA developers, as well as performance verification by the third party.
- The strength is the experience in mass producing a consistent PGM-free catalyst with good durability.
- The project team is strong.

Project weaknesses:

- No weakness has been identified at this point.
- Gen 1 catalyst lifetime is questionable; therefore, the mitigation approach needs to be defined.
- The weakness is the current catalyst activity and performance in MEAs.

Recommendations for additions/deletions to project scope:

- The team should include specific stability and durability targets in addition to the performance milestones.
- More planned interactions with ElectroCat could be beneficial.
- No changes are recommended.

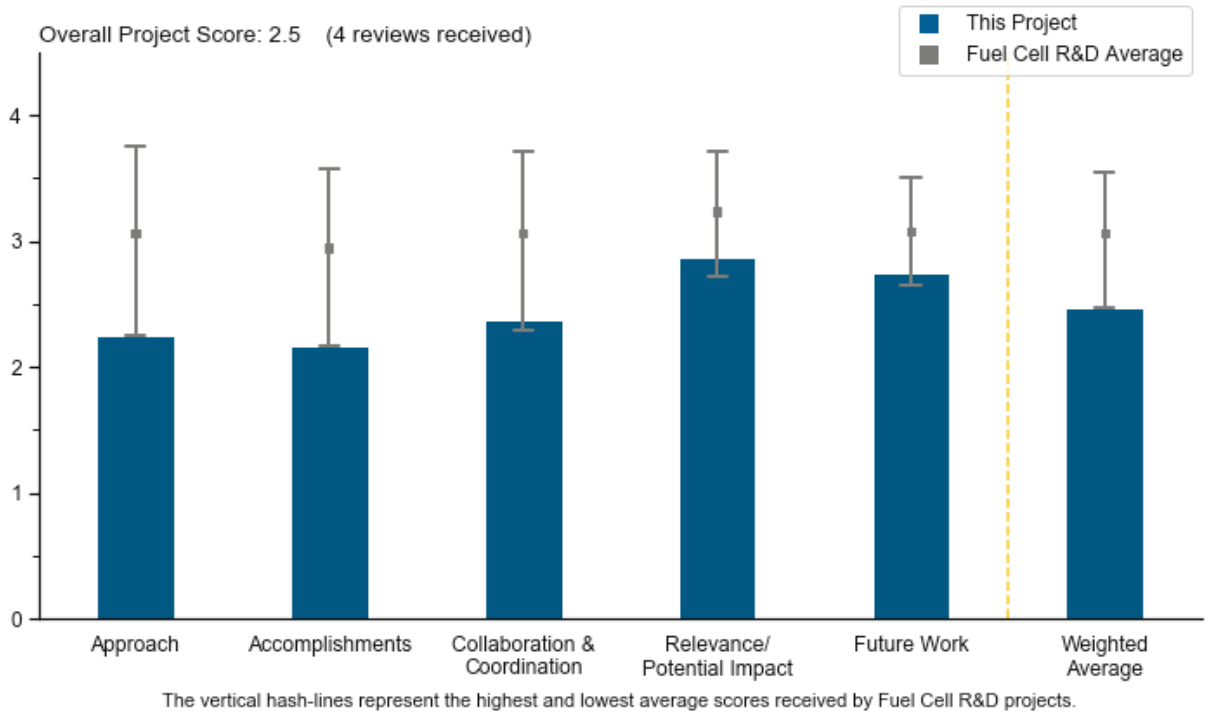
Project #FC-306: High-Performance Non-Platinum-Group-Metal Transition Metal Oxide Oxygen Reduction Reaction Catalysts of Polymer Electrolyte Membrane Fuel Cells

Timothy Davenport, United Technologies Research Center

Brief Summary of Project

The project's objective is to develop acid-stable non-platinum-group-metal (non-PGM) metal oxides and optimize oxide catalytic activity for oxygen reduction reaction (ORR) reactivity. The project will (1) utilize high-throughput computational methods to develop acid-stable complex doped transition metal oxides, (2) leverage high-throughput experimental electrochemical testing to optimize identified acid-stable oxides for ORR electrocatalytic activity, and (3) utilize a rapid development process to optimize ink formulation and optimize membrane electrode assembly (MEA) fabrication for metal oxide electrocatalysts. The project addresses barriers required to achieve U.S. Department of Energy 2020 targets for non-PGM MEAs: (1) reduce catalyst mass loss by 40%, (2) decrease performance loss, and (3) increase PGM-free catalyst activity.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **2.3** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This project applies metal oxides as ORR catalysts for polymer electrolyte membrane fuel cells (PEMFCs). It is different from most existing projects and represents a new approach, if perhaps not a new concept.
- The project is at technology readiness level (TRL) 1, the concept phase. It is quite ambitious to get to and be successful at TRL 4, which is component testing in the relevant environment with a small MEA, in only two years. It was assumed that this funding opportunity announcement was for getting a TRL 2–3 technology to TRL 4. For this to happen, the project must have a specific plan and approach and be

aggressive with the timing. These are lacking in this project. The Massachusetts Institute of Technology (MIT) will work on theory and propose target chemical composition for the catalyst. The United Technologies Research Center (UTRC) will evaluate the catalyst. However, it is not clear how or who will do the most important part: developing the catalyst. UTRC will likely produce the catalyst, but considering the capabilities and resources required, this appears to be troublesome.
- This project just started, and its objectives and critical barriers have been clearly identified. This is the only effort that aims to develop cathode catalysts based on complex metal oxides. The approach is based on published work in alkaline electrolytes, which had rather poor activity. It is hard to envision that these systems would be efficient cathode catalysts in PEMFCs.
- The theoretical approach is rather insufficient and lacks any specific data.

Question 2: Accomplishments and progress

This project was rated **2.2** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- If the principal investigator could provide some preliminary data, it would be really helpful for evaluating the approach.
- The project is in the beginning stage, and progress is limited.
- The project has just started, but the plan is not very specific.

Question 3: Collaboration and coordination

This project was rated **2.4** for its engagement with and coordination of project partners and interaction with other entities.

- The collaboration between industry and the university is a good start.
- The proposed collaboration is well justified.
- The project has just started, but it is not clear who will do the most important part or how it will be done: developing the catalyst.
- MIT's work is on a parallel path with UTRC's. It is unclear why the duplication of effort has been chosen.

Question 4: Relevance/potential impact

This project was rated **2.9** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- Transitioning away from platinum-grade-metal catalysts can reduce cost if the new catalyst is as active and durable. The project offers an alternative to the highly unstable metal nitrogen-doped carbon catalyst.
- If successful, this project might have significant impact on DOE technical targets.
- The project's chosen outline and approach are more appropriate for fundamental science, rather than applied science, and the impact of yielding new catalytic formulations by the end of the project seems quite remote.
- It is difficult to evaluate the impact without any preliminary data.

Question 5: Proposed future work

This project was rated **2.8** for effective and logical planning.

- Considering the project is starting from TRL 1, it will benefit from having frequent down-selections and implementing fast evaluation methods, commonly done during the TRL 2–3 stages.
- The go/no-go should include simultaneously demonstrating voltage performance and acid stability with the same formulation.
- The principal investigator has a good justification for the future work, but the preliminary support for the justification should come sooner.

- The proposed work relies on published work in alkaline electrolytes. There is no evidence that these systems would perform in acidic environments at a level that would be of practical relevance.

Project strengths:

- The project elaborates on the acid stability of transitional oxides and could find some stable formulations.
- The project has good teaming and a new approach that is outside of the current DOE portfolio.
- The project's strengths include its area of research, which is less clouded and less explored.
- The project's strengths include its team members.

Project weaknesses:

- The project's weaknesses include its catalyst synthesis capability and the demanding progress, milestones, and timing of a project funded by the DOE Office of Energy Efficiency and Renewable Energy. The project might be better suited to an Advanced Research Projects Agency–Energy or National Science Foundation setting.
- There is a lack of evidence that complex metal oxides can be active, stable, and conductive under the relevant conditions for PEMFCs.
- The idea is not entirely new and has been in the literature as one of the "not so promising" methods. Therefore, some of the previously known limitations need to be addressed as soon as possible.
- There is a lack of data for the chosen approach.

Recommendations for additions/deletions to project scope:

- An immediate electrochemical characterization would be more beneficial than lengthy combinatorial modeling.
- The project should clearly identify acid stability and include acid stability in the go/no-go decision point.
- The project should generate preliminary data as soon as possible.

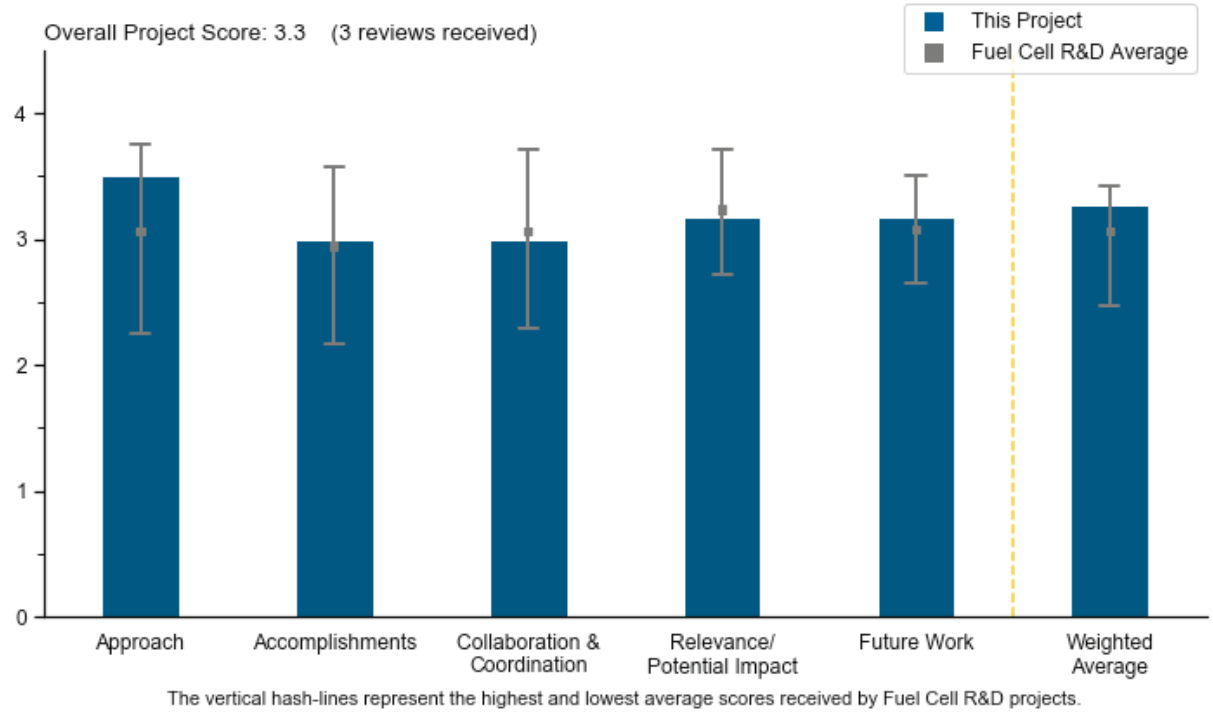
Project #FC-307: Cyclic Olefin Copolymer-Based Alkaline Exchange Polymers and Reinforced Membranes

Chulsung Bae, Rensselaer Polytechnic Institute

Brief Summary of Project

In this project, Rensselaer Polytechnic Institute (RPI) will develop a series of innovative ethylene–norbornene copolymer (ENC)-based alkaline exchange membranes (AEMs) that would overcome the challenges of the state-of-the-art AEM. Specifically, the project team plans to (1) develop ENCs with tunable backbone rigidity, (2) incorporate alkyl chain-tethered quaternary ammoniums (QAs) of different structures to the polymer by simple post-functionalization method, (3) impregnate the anionic polymers into a mechanically stable matrix (reinforced AEM), and (4) demonstrate the membranes' performance and durability in fuel cells using Pt-based and platinum-group-metal-free (PGM-free) catalysts. The reinforcement of AEM will allow RPI to produce thinner (e.g., 10–15 µm) membranes, affording lower area-specific resistance and better water management in MEAs, particularly with a PGM-free catalyst.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.5** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The technical approach is novel and reasonable, namely using no heteroatom (O or N) in the backbone for alkaline stability, using high molecular weight for good mechanical properties and dimensional stability, getting tunable rigidity by varying the ratio of ethylene and norbornene and cyclic olefins in the backbone, and using a pore-filling reinforcement framework for enhancing the durability and extending the lifetime of MEAs. Whether this is inexpensive remains to be seen. The more complex the synthesis in the commercial stage, the more impractical and expensive it will be to manufacture.

- The project addresses a number of issues with these polymers, including avoiding phenyl groups that poison the catalyst, as well as some chemical weaknesses in a system that should be fairly low-cost to produce.
- The project's approach is laid out in straightforward terms. The project team's partners are there to help with key parts.

Question 2: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has just started, so only the polymer has been synthesized. However, that is often done before the proposal, so the project might have gotten a little bit further done in the first tasks, especially some work toward the functionalization.
- This is a new project, but prior work indicates the team can do what they say.
- The project is around two weeks old; it is too early.

Question 3: Collaboration and coordination

This project was rated **3.0** for its engagement with and coordination of project partners and interaction with other entities.

- There is a good combination of collaboration from academics (led by RPI), industry (Xergy), and national laboratories (LANL), with well-defined tasks and complementary skills.
- Kim at Los Alamos National Laboratory (LANL) is one of the premier scientists in this area, though it is not completely clear what his role will be; if it is only for testing, then that is a waste. Also, expanded polyethylene is likely a poor choice for a support material, and the team advertises that the base material should be strong, so it is not clear that this is needed.
- It is too early to answer in much detail.

Question 4: Relevance/potential impact

This project was rated **3.2** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The project addresses a number of concerns for AEMs.
- The project offers some uniqueness of approach that opens up new possibilities.
- This relevance falls under a speculation on a speculation, which is not a good place to be. The authors have responded to the funding opportunity announcement, but the wisdom of this line of work (AEMs) is questionable. It is not certain that there is proof that AEMs will lead to making good and practical fuel cells and fuel cell power sources. These workers received a "good" rating for these reasons. Making these membranes is a bit speculative, and if there is success, then a good alkaline membrane is achieved—but it is not clear what good having a good AEM does for making a fuel cell and fuel cell power source. The former is the job of the research team; the latter is the responsibility of DOE management and remains to be seen.

Question 5: Proposed future work

This project was rated **3.2** for effective and logical planning.

- Durability is the last thing that the team studies, and it should be addressed much earlier; there is no point in pursuing these materials if they will not be stable. The other characterizations and testing are appropriate—it is just the order that is in question.
- The design of the experiment is reasonable for this new project.

Project strengths:

- The project's relevance is a strength, and the team is aware of the issues. As a collaborator, Kim should help and is a good choice for making MEAs. The approach is good, but it is doubtful that the project will get to MEA testing within 16 months from now; this schedule is a bit ambitious for a new polymer that has not even been functionalized.
- The project has good teaming. The academic team is good in small-scale synthesis, Xergy has proven support technology, and LANL is proven in assessing AEM technology.
- There are multiple opportunities for success in this project. The project seems focused in scope so as to better enable success.

Project weaknesses:

- The resistance targets seem quite ambitious. There is the additional difficulty of adding in reinforcement, which perhaps may dilute some from the core synthesis effort. Scaling up to larger batch sizes is listed as a challenge, but no clear path or process was given. There may be many challenges, including ionomer cleaning, separation, fabrication, testing, etc. It would be nice to see a few more details about this effort.
- Chemical durability should be moved ahead, and the project team should have some strategies for what to do next if the product is not stable; that is not addressed.
- There are no clear weaknesses at this stage of this new project.

Recommendations for additions/deletions to project scope:

- The real benefit of this work is to see whether this new synthetic approach leads to chemically and physically stable ionic membranes under fuel cell conditions. Once that is clear and a membrane gives satisfactory properties, then the next step is to see whether that membrane (or multiple membranes) is amenable to mass manufacturing (i.e., low cost).
- The project's tasks are all relevant, but moving chemical durability up earlier is suggested.
- The project team could perhaps provide a better breakdown of synthesis risks and mitigations.

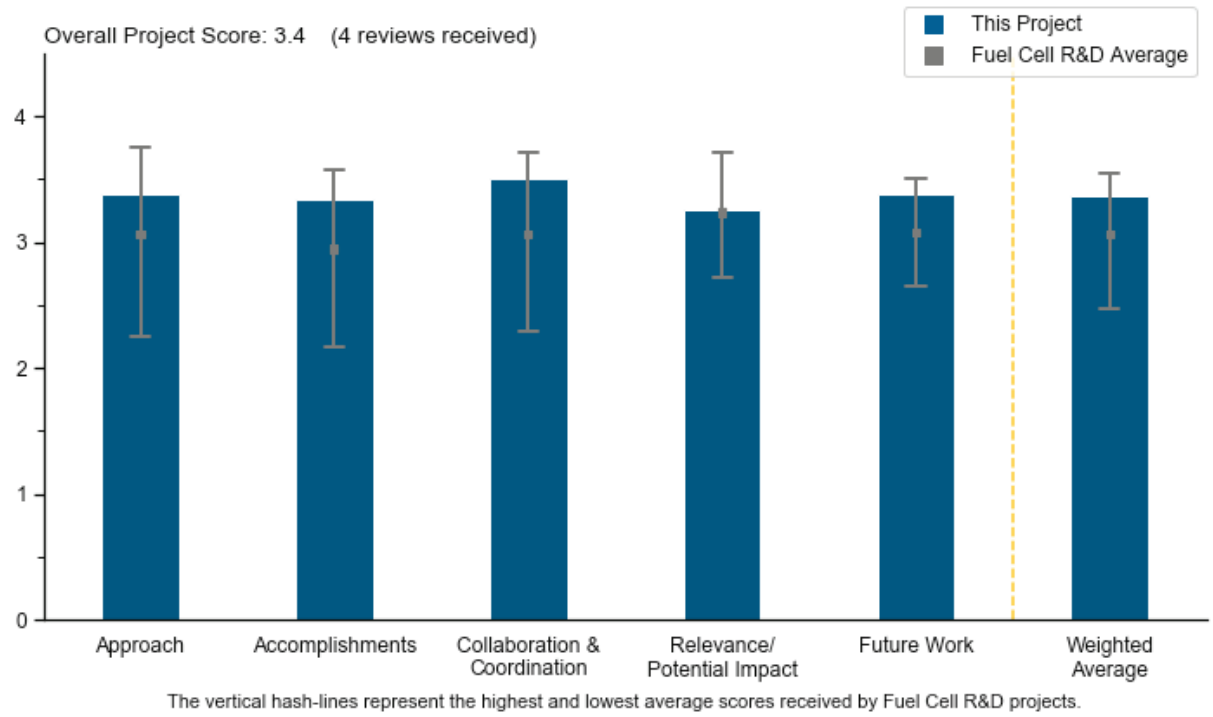
Project #FC-308: Advanced Anion Exchange Membranes with Tunable Water Transport for Platinum-Group-Metal-Free Anion Exchange Membrane Fuel Cells

Michael Hickner, Pennsylvania State University

Brief Summary of Project

This project will enable high-performance, long-lifetime, low-platinum-group-metal (low-PGM) (PGM loading ≤0.125 mg/cm^2) anion exchange membrane fuel cells (AEMFCs) through (1) synthesis and fabrication of novel thin, mechanically supported anion exchange membranes (AEMs) and electrode ionomers with validated outstanding water transport properties and stability; (2) integration of these new polymers with high-performing low-PGM and PGM-free catalysts and electrodes; and (3) precise control over the distribution of water in operating cells. What makes this project unique are the team's capabilities in new material synthesis to tune water transport and the world-leading knowledge in membrane integration with electrodes to achieve the current world-record performance in AEMFCs.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.4** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The two membrane metrics are reasonable and in the team's control: (1) develop novel poly(olefin) AEM chemistries with tunable water transport and (2) incorporate these novel ionomers into mechanical supports and integrate the resulting membranes into AEMFCs. The third performance metric seems beyond the scope of this project. For the third metric, the project title says "platinum-group-metal-free" (PGM-free), but the project talks about low-to-moderate Pt loadings. There is no path to PGM-free given, except on slide 12 of the presentation, which says that "risk will be mitigated by taking advantage of state-of-the-art catalyst for alkaline membranes that are reported." The project's approach is to develop novel poly(olefin)

AEM chemistries with tunable water transport. To facilitate high AEMFC performance, the fuel cells will have the following properties:

- OH- conductivities greater than 60 mS/cm at 60°C, 100% relative humidity (RH)
- Less than 10% degradation in conductivity after 5000 hours in 1 M NaOH at 60°C and 2000 hours in 1 M NaOH at 80°C
- A water diffusion coefficient $>5*10^{-6}$ cm^2/s (a 50% improvement over existing AEMs.

Additionally, the project team will incorporate these novel ionomers into mechanical supports and integrate the resulting membranes into AEMFCs. During operation inside the AEMFC, the membranes will have:

- Area-specific resistance values less than 100 mOhm×cm^2 over 2000 hours of operation
- Water flux greater than $2*10^{-5}$ mol $H_2O/cm^2 \times s$ to be able to back-diffuse 80% of produced + electro-osmotic water from anode to cathode at 600 mA/cm^2.

Finally, the project will demonstrate of all of the following DOE metrics in a single MEA with hydrogen and oxygen fuel:

- Greater than 2000 hours of AEMFC operation at 600 mA/cm^2
- An operating voltage greater than 0.6 V with less than 10% decay over 2000 hours
- Operating T ≥60°C and P ≤1.5 atm with PGM loading less than 0.125 $mgPGM/cm^2$

- There is very good focus on barriers relevant to AEM implementation. The poly (norbornene/olefin/long-chain quaternary ammonium) system appears to be well chosen, based on the principal investigator's prior experience and the rationale stated in the poster materials. The project has a target to more than double the water-diffusion coefficient from values in existing AEMs. The proposal does not provide the rationale for how the project team thinks they can accomplish that. One question is whether these polymers are more gel-like than existing AEMs. If so, it may be that the membrane mechanical properties are compromised by the liquid-water-like character. The information given in the poster is not enough to tell.
- The Pennsylvania State University (Penn State) is developing novel AEM chemistries with improved water transport. This will in turn result in improved AEMFC performance. The polyolefin-based chemistries hold promise for AEM membrane development. While the relationship between water transport and performance is clear, the link to durability is unclear and needs a better scientific basis. The team should add experiments to understand the link between enhanced water transport and improved durability.
- The approach of the project is to prepare polyolefin membranes for AEMFCs. Synthesizing polyolefinic membranes may improve alkaline stability, although this approach has been tried by one of the project team members and other researchers. The approach to preparing membranes with a high water-diffusion coefficient is not well justified, although there are not any major issues with it. First, the slide does not show a clear relationship between the water-diffusion coefficient and AEMFC performance. Second, the diffusion coefficient target ($2*10^{-6}$ cm^2/s) is rather arbitrary. Third, the presentation does not explain how the proposed polymer can have a high diffusion coefficient. The final issue is that, while the project title indicates these membranes are for PGM-free AEMFCs, the benefits of these membranes for PGM-free AEMFCs are not apparent. It is not clear how high water transport helps use PGM-free AEMFCs.

Question 2: Accomplishments and progress

This project was rated **3.3** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Slides 6 and 7 show syntheses done up to the 4 g level. This is very impressive for a new project that began a half year ago (in late 2018). If the team keeps up this pace of work, then the "nobornyl concept" for making stable and conductive water-transport AEMs should be checked during this work.
- This project is about 16% spent in 5 months. Given the early stages of this project, there is significant progress, especially from Penn State and the University of South Carolina (USC), with over 80 polymerization batches conducted, membranes developed, and initial fuel cell performance reported, albeit at very high loadings.
- Good progress has been made on synthesis, but in other areas, progress is hard to determine at this early stage. Lifetime testing will be especially hard to gauge until membrane fabrication is well along.
- It seems the project has started with some tryouts for polymer synthesis. However, it is too early to review the progress of the project at this moment.

Question 3: Collaboration and coordination

This project was rated **3.5** for its engagement with and coordination of project partners and interaction with other entities.

- The project's teaming is excellent, comprehensive, and complementary for four team members' making membranes. The work for low-PGM is reasonable; for no-PGM, the work is questionable. The four teams are (1) Penn State, which makes new polymer; (2) USC, which is responsible for electrode formulation, cell testing, and water transport studies, including neutron radiography; (3) National Renewable Energy Laboratory (NREL), which is responsible for lifetime testing and water balance studies; and (4) 3M Company (3M), which is responsible for membrane coating and supported membranes.
- The team is well organized. Penn State has been working in this area for a long time. USC is known to be an expert on MEA fabrication and fuel cell testing. NREL can accomplish the job as stated. 3M has extensive experience in synthesizing chain-growth polymers.
- This is an excellent team. Penn State is making the polymers and membranes, USC is developing electrodes, NREL is responsible for lifetime testing and water balance, and 3M is developing the supported membranes. This is a very experienced team.
- The first round of newly synthesized polymers have been sent to partners at NREL and USC, and the early-stage results for hydrogen and oxygen cells are in hand. This is good progress for only a few months in. There are no results from the collaboration with 3M yet; presumably that comes later.

Question 4: Relevance/potential impact

This project was rated **3.3** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The project has a goal to meet all DOE targets simultaneously (>2000 hours at 600 mA/cm^2 with V >0.6 V and <10% decay with T >60°C, P <1.5 atm, and PGM loading <0125 mg/cm^2). If the project meets these goals, it will have significant impact on the DOE Hydrogen and Fuel Cells Program.
- The project's relevance and potential impact are excellent.
- This work is commendable, but it remains a speculation (of whether the membranes can be made with suitable properties) on a speculation (that John R. Varcoe of the University of Surrey is correct that AEMs are good). Yet another speculation is that PGM-free catalysts will work. The only durable alkaline fuel cell is the one that is used in space, which uses pure oxygen and hydrogen, as well as noble metal catalysts. Once again, the DOE has "bid on questionable work" from outside the country. The team has responded with great relevance to the funding opportunity announcement. The potential impact for making a practical fuel cell and fuel cell power source system is questionable for the reasons stated above. The question of making a practical fuel cell and fuel cell system is the responsibility not of this project's research team but of DOE management.

Question 5: Proposed future work

This project was rated **3.4** for effective and logical planning.

- The two membrane goals are good. The catalyst goals are highly speculative and questionable. The first membrane goal in fiscal year (FY) 2019 is to synthesize larger-scale batches of polymer with vinyl-norbornene motif and to fabricate supported membranes. The team will also continue to optimize electrode structures and cell conditions to meet milestones and the year 1 go/no-go for performance and durability at required loadings. The team will also measure water transport in membranes using pulsed field gradient and nuclear magnetic resonance (PFG-NMR) and connect cell water transport observations using water balance measurements. Next year, the second membrane goal in FY 2020 is the fabrication of high-performance supported membranes and the optimization of electrode structures and cell conditions that will allow for progress on year 2 milestones. The project team will also develop strategies for increasing durability by modifying cell water transport. The team will also develop a holistic picture of water transport in AEMFCs using neutron radiography. The catalyst goal is that risk will be mitigated by taking advantage

of state-of-the-art catalysts (PGM-free seems improbable) for alkaline membranes that are reported. The major risks will be in approaching required (possibly PGM) catalyst loadings while still reaching durability targets. Particularly worrisome is the durability of PGM-free catalysts. If these existed, they would presumably be being used in the space program.

- All the 3M work and NREL work is proposed in FY 2020. Currently, all major milestones are related to AEMFC performance, which is very good for judging progress. However, it would be good to have some milestones associated with the water transport work and durability work and how those are linked.
- The project's proposed future work is good. More membrane characterizations beside the water transport measurement should be listed (e.g., ex situ membrane stability, conductivity, etc.) before putting the membranes in the MEAs. No pathway for incorporating PGM-free catalysts can be seen.
- The project's proposed future work seems reasonable.

Project strengths:

- The overall strength of the project is teaming for the project. The team is capable of preparing a series of membranes for optimum performance. Penn State has a good track record for delivering the products, so much progress is expected next year. The other strength of this project is that the performance milestone is challenging, which is very much appreciated. Just one suggestion would be that the go/no-go decision (in month 12) should include the catalyst loading target (0.3 mg/cm^2). If the team does not meet the interim loading target, then it is likely that the team will not meet the quarter 6 (Q6) target with loading <=0.125 mg/cm^2.
- The project's strengths include its good teaming. The team has a complementary and comprehensive set of skills, including Penn State's skills in new synthesis, USC's skills in characterization, NREL's skills in durability, and 3M's membrane supports. However, new catalysts are an "Achilles' heel"; it is doubtful a PGM-free catalyst will be seen in our lifetime.
- The project's strengths include its use of a robust polymer synthesis system and its choice of partners, which will enable the team to make and evaluate large amounts of AEM using methods suitable for scale-up and manufacturing.
- This is an excellent team with complementary strengths.

Project weaknesses:

- The overall weakness is that the project is based on the idea that water transport is the single most important factor for AEMFC performance and durability. It may be true, but in case this is not the decisive factor, then the whole project may go in the wrong direction. The membrane milestone is not challenging (40 mS/cm at 60°C). Those targets with quaternized polyolefinic polymers have been achieved by a couple of projects. USC has demonstrated over 2 W/cm^2 peak power density with its polyolefinic membrane and ionomers. It is unknown how much better performance can be achieved with highly water-permeable, "more advanced" polyolefinic membranes. It is understood that this is not an AEMFC project but an AEM project. However, if the project does not achieve better performance, better durability (>2,000 hours in fuel cell operating conditions), or low PGM loading (<0.125 mgPGM/cm^2), the advantage of using the proposed AEMs for AEMFCs may be too small. A clear pathway for achieving those challenging targets is not apparent.
- The project team needs to quantify "larger-scale" batches. The current loading (as seen on slide 8) is way too high and needs to be lowered soon. It seems like the cell has some transport issues, even in oxygen. A systematic study of those limitations and how they can be overcome with advanced membranes should be added.
- The linkage between polymer structure and the desired AEM property improvements has not emerged from results to date. This may reflect the fact that the project is still in a synthesis-heavy phase, with the first round of feedback from measurements not yet completed.
- PGM-free catalysts are highly speculative, and an especially speculative idea is that there are ones that are durable.

Recommendations for additions/deletions to project scope:

- A recommended addition would be to the go/no-go decision point: an interim total PGM-loading target is needed. Regarding deletions, the target for the later stage of the project is overly challenging. Either the 500-hour H_2/air (CO_2-free) or 2000-hour H_2/O_2 target can be deleted and modified to some progress measure, e.g., 100-hour H_2/air (CO_2-free) for Q7 and 500-hour H_2/air (CO_2-free) for Q8.
- The project team should forget about catalysts and do what they do best and check whether they can make durable membranes that operate under fuel cell operating conditions with low-PGM catalysts.
- The project is off to a good start. Seeing the progress in the coming years is gladly anticipated.

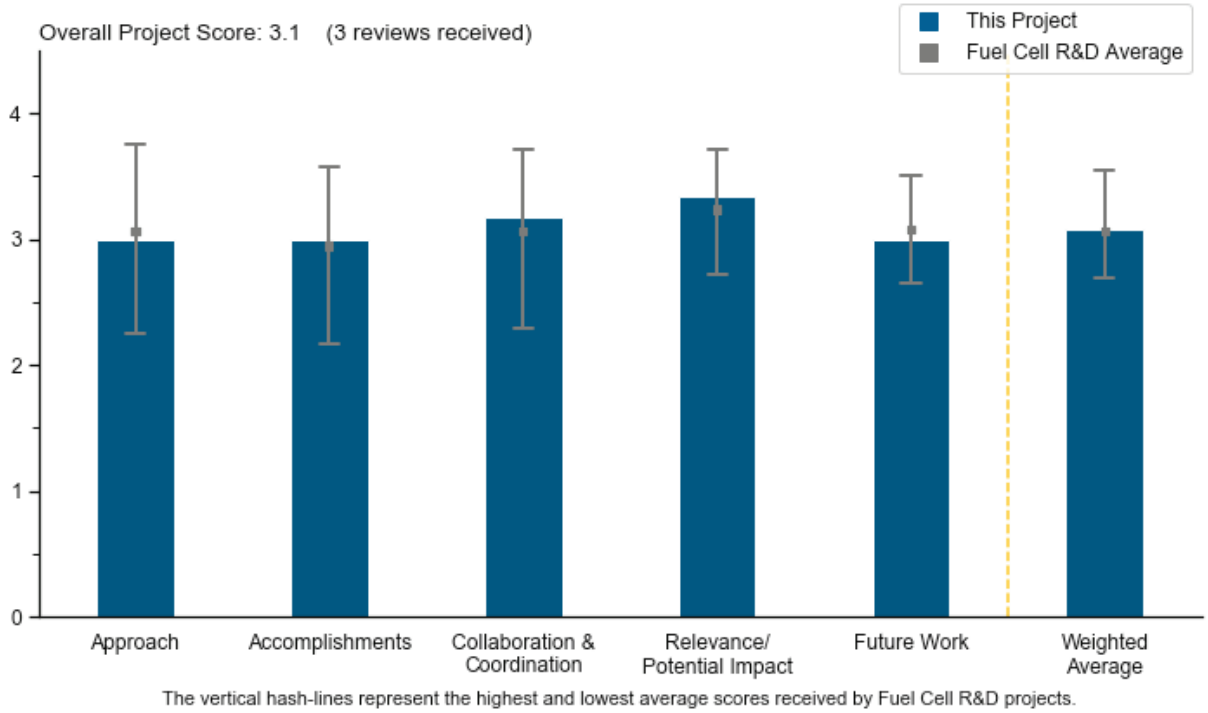
Project #FC-309: Polymerized Ionic Liquid Block Copolymers and Ionic Liquids (PILBCP-IL) Composite Ionomers for High Current Density Performance

Joshua Snyder, Drexel University

Brief Summary of Project

The project's goal is to develop a polymerized ionic liquid block copolymer/ionic liquid (PILBCP/IL) composite ionomer to replace traditional perfluorosulfonic acid (PFSA)-based ionomers and address their associated limitations. The expected outcomes include (1) development of a cathode that meets U.S. Department of Energy targets for low and high current density (HCD) and (2) improved understanding of how interface engineering affects HCD performance. The project will develop the PILBCP/IL ionomer and then develop and study membrane electrode assembly (MEA) performance and durability. The project addresses the primary technical barriers associated with (1) oxygen transport through ionomer thin films, (2) ionomer-specific adsorption onto catalyst, (3) inaccessible catalyst in porous carbon supports, (4) distribution and retention of IL in catalyst layers, and (5) humidity tolerance at HCD (Pt utilization).

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.0** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- PILBCP/IL is a good concept for enhancing catalyst mass activity and thereby achieving increased cell performance under low humidity conditions. Covalently bonding the IL is a great approach to immobilize the IL into the catalyst ionomer interface and provide consistent higher catalyst mass activity and proton transport. The team should address the issue of IL leaching during fuel cell operational conditions.
- Using protic IL additives to improve access to catalysts is sensible and has been shown to work in prior efforts. The present approach builds on this prior work using copolymers with immobilized ionic groups to

prevent the loss of IL. This approach may work, though it appears that gradual IL loss could occur if the ionic groups from the two parts of the polymer combine with each other to release their counterions. This situation could result in dissolution of bis(trifluoromethanesulfonyl)imide (HTFSI) into the fuel cell exhaust. If this happened, it could result in significant and irreversible long-term performance loss. The team should be on the watch for this. Also, the polymers the researchers are proposing may be subject to degradation during long-term fuel cell operation.
- Some groups have tried to use ILs in catalyst layers without much significant success, but this group's approach with PILBCP/IL may work. This project is also targeting key technical barriers.

Question 2: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- This is a new project that started in October 2018. The team has demonstrated increased cell performance and higher catalyst mass activity using IL [MTBD] [beti]. The team has also demonstrated higher electrochemical surface area and lower Pt dissolution using IL. The team has also shown intermediary IL thin-film formation with the Nafion™ ionomer.
- This project has started recently, and the team can leverage a lot on a previous General Motors Company (GM) project, which studied similar technical barriers. The team tried some IL deposition techniques and preliminary ex situ tests, which is also encouraging. Establishing a baseline or benchmarking against conventional Nafion should be the first step in MEA. This is planned, as outlined in future work.
- The project is at an early stage (7% complete, having started in October 2018), so few new results are expected. The poster mostly presented results from the prior funding period using [MTBD] [beti], which is intriguing. The new findings on the capacitive deposition of IL are intriguing but are at an early stage.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- The team is composed of excellent research institutions conducting effective research in fuel cells. The team is a good combination for pursuing the proposed work and includes the principal investigator, Drexel University, and sub-contractors GM, Texas A&M University, and the National Renewable Energy Laboratory (NREL).
- Collaboration among team members is key, and this team has all the relevant and capable members (from industry and academia) to make it successful.
- The team appears to be well chosen, but the poster did not clearly show how the work was distributed among team members.

Question 4: Relevance/potential impact

This project was rated **3.3** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- Improving HCD performance could result in cost reduction, so this project is well aligned with the DOE Hydrogen and Fuel Cells Program's (the Program's) research and development objectives. Improving HCD performance with a low Pt-loading catalyst has been a great challenge in recent years, so positive outcomes in this project would be very helpful.
- The project is relevant to developing a fuel cell cathode that meets the DOE targets for low and high current density and performs under low humidity conditions. The use of IL to boost specific and mass activities of the catalyst is directly linked to DOE's goal of catalyst performance enhancement.
- The project is targeting improvements in metrics relevant to the Program's goals.

Question 5: Proposed future work

This project was rated **3.0** for effective and logical planning.

- The team has listed and described the future work very well. All the relevant tasks needed for the success of this project have been captured.
- The future work looks like it would take lot of knowledge gained from past GM projects, which is a plus. Additionally, the team should try MEA testing first to make sure it works before spending too much effort on understanding the underlying fundamentals using ex situ tests.
- The future work seems appropriate. The project is still at an early stage; the team needs to make progress on the synthesis work on new polymers so that other project areas can move forward.

Project strengths:

- The team is very strong and capable of meeting the challenges of this project. The tasks are appropriately shared between the prime and sub-partners, as per their technical expertise. This approach is very effective in enhancing the catalyst activity under stressed operational conditions, such as low relative humidity. The inclusion of an industry partner (GM) and a national laboratory (NREL) allows the team access to a significant fuel cell network for accomplishing the project.
- The very capable team is the project's strength. Each team member has the respective expertise to make this project successful. GM's significant previous work can be leveraged by the team to move the project along quickly.
- The project provides a rational approach to improving electrolyte access to all catalyst particles in high-area supports, which should improve performance under high-rate conditions.

Project weaknesses:

- The project's weaknesses include cost; the DOE mandates low-cost fuel cell systems to meet commercialization targets, and IL is expensive. The incorporation of IL in block copolymer is also a very cost-intensive proposition. It is very unlikely that the proposed effort is going to be a cost-effective approach to the development of high-performance fuel cell MEAs. The project also faces weaknesses in terms of durability. To meet the desired cost objectives, DOE targets high-durability MEAs. Being hydrophilic, the IL additive is expected to leach out through the product water in the cell. This leaching will lead to the loss of catalytic activity and catalyst–ionomer interfacial proton conductivity and, therefore, an increase in cell impedance, resulting in poor cell performance and the loss of stack performance over time. Plug Power Inc. and the Rensselaer Polytechnic Institute team experienced the same phenomenon with their DOE-funded project for phosphoric-acid-doped polybenzimidazole membrane development. The copolymerized IL from the block copolymer may be stable and not leachable; however, the free IL has a very high chance of getting leached and affecting the stack performance durability. Also, the leached IL may contaminate the balance of plant, leading to unintended consequences. The project also has a weakness in terms of ink fabrication. The capacitive deposition of IL onto the catalyst surface adds increasing steps, which may increase the cost of ink fabrication. This increased ink cost will ultimately increase the total MEA cost, which is not aligned with DOE's mandate.
- The use of IL additives may suffer from a weakness of poor long-term durability due to eventual loss of IL in the exhaust, especially if the IL has high water solubility. The team should watch for this. Additionally, the polymer's chemical structure may suffer its own degradation, e.g., from hydroxyl radicals or other oxidative species known to be present in H_2/O_2 fuel cells.
- Based on some preliminary results, it is assumed that PILBCP/IL will work in MEAs, which can be very tricky. There are many unknowns with this material; Nafion in the catalyst layer systems has been studied extensively by various groups, so it is pretty well understood. Pt and Pt-alloy interaction with this new polymer IL is unknown, and its effect on durability is also unknown.

Recommendations for additions/deletions to project scope:

- The team should consider developing a means of monitoring IL loss into the output stream, perhaps using methods similar to those used to monitor the fluoride loss associated with polymer electrolyte membrane degradation.
- No additions or deletions to this project are needed. The project is fine as proposed.

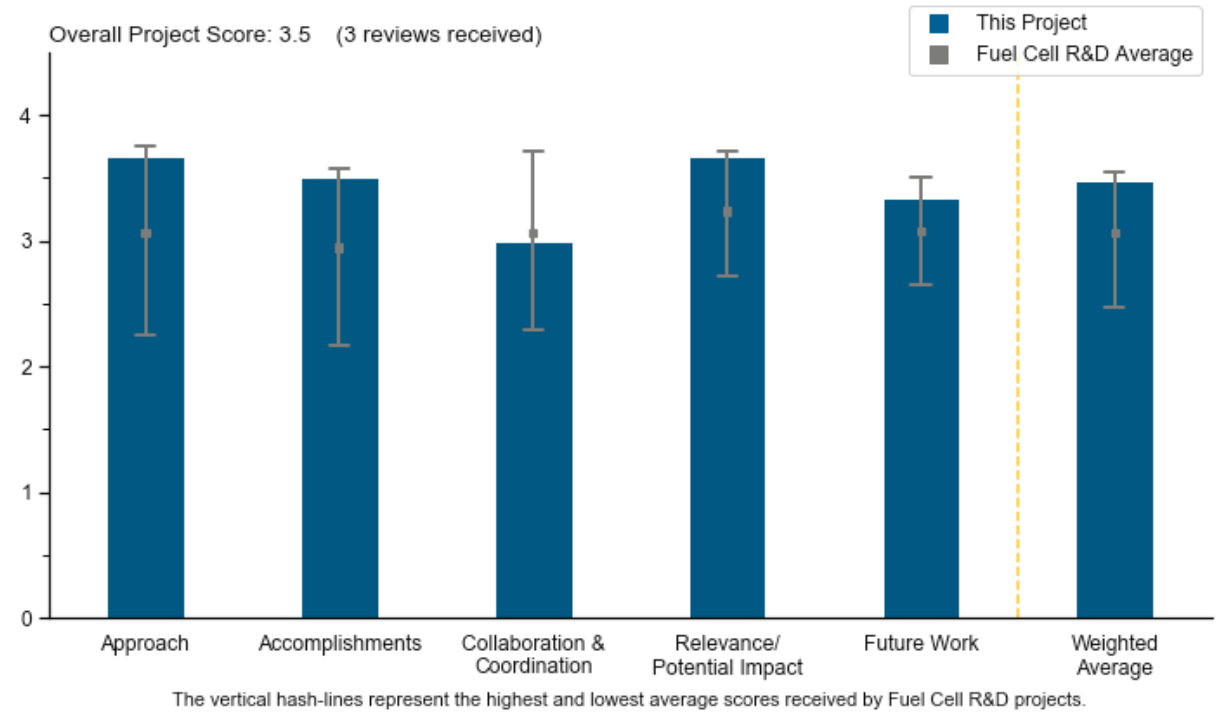
Project #FC-310: Composite Polymer Electrolyte Membranes from Electrospun Crosslinkable Poly(Phenylene Sulfonic Acid)s

Ryszard Wycisk, Vanderbilt University

Brief Summary of Project

The project objective is to fabricate a novel electrospun, non-perfluorosulfonic acid (non-PFSA) fuel cell membrane that meets all 2020 technical targets in the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan. The project approach is to develop/fabricate a robust, low-cost composite all-hydrocarbon membrane via dual fiber co-electrospinning of a crosslinkable poly(phenylene sulfonic acid) (cPPSA) and poly(phenyl sulfone) (PPSU) mixture mat, followed by mat densification via solvent-vapor-induced softening of PPSU fibers and thermal crosslinking. The project addresses the barriers of (1) the high cost of PFSA membranes, (2) low proton conductivity at reduced humidity (water partial pressure), and (3) performance drop above 80°C.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.7** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This project takes over from prior work performed at Case Western on similar composite membrane systems. The team has just started working on this project, and it is too early to assess performance. Generally speaking, the idea of combining fibers with ionomers remains very promising. In this project, the fibers and ionomer share similar chemical structures, which ensures excellent interfacial properties. The researcher has a strategy for eliminating voids in the structure, and it will be interesting to see cross-sectional views of the overall structure once membranes are produced. One concern with this technology is the viability of the chemistry, from a chemical resistance and durability standpoint, inside fuel cells and electrolyzers. Durability should be investigated. In addition, the principal investigator (PI) mentioned that

there is some issue with brittleness in prior structures produced at Case Western. Therefore, mechanical properties will need to be investigated. Nevertheless, this project is off to a good start.
- The approach and combination of electrospun and previous high-performing polymer is interesting and provides good synergy, assuming that it is processable.
- This is a polymer electrolyte membrane fuel cell technology project. The approach is membrane synthesis using a combination of electrospinning and subsequent densification (solvent vapor + thermal crosslinking). The project tries to combine individual successes of two previous projects into one material.

Question 2: Accomplishments and progress

This project was rated **3.5** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has started to get materials and equipment together. The first materials were successfully synthesized.
- It is too early to comment. Overall, this project is off to a good start. The PI has properly identified issues to study.
- The project just started, with minimal work accomplished before the slides were due.

Question 3: Collaboration and coordination

This project was rated **3.0** for its engagement with and coordination of project partners and interaction with other entities.

- This is an academia-lead project with one university. It was unclear how the partners would communicate and/or interact with each other. The roles of individuals was laid out, but no interaction between the institutions was presented that showed any material, sample, or information exchange. Slide 6 states that Los Alamos National Laboratory (LANL) is performing fuel cell testing in year 2, but it is unclear how LANL is tied into this project.
- The overall collaboration is sufficient, although further testing and characterization is recommended, perhaps at a national laboratory.
- It is too early to judge collaboration and coordination. Collaborations were not discussed specifically.

Question 4: Relevance/potential impact

This project was rated **3.7** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- Ionomers are inherently weak. Any approach to making new and different composites should be investigated in addition to fundamental work on ionomer synthesis. Both processes must be done. This is a novel approach, and it will be interesting to see progress reports in future Annual Review Meetings.
- The project topics are right on target with the technology's advancement and DOE's goals. Reduction of cost toward available cost-effective membrane materials is important. If at the same time the resistance can be improved, specifically at temperatures greater than 80°C, performance improvement could be achieved.
- The project aligns well with the membrane targets, assuming cost projections and durability are met. It has yet to be determined whether compatible electrode ionomers will be found, which could be an issue that minimizes impact.

Question 5: Proposed future work

This project was rated **3.3** for effective and logical planning.

- Overall, this is a very sound, comprehensive project plan.
- The project just started. The statement of work seems appropriate.

- There needs to be further property testing and full characterization of the membrane, including mechanical properties, uptake, conductivity, water diffusivity, etc. The focus on synthesis makes sense, although it is unclear how it is a true composite membrane.

Project strengths:

- This is a different approach to composite membrane production. It should be explored, and it should be determined whether the approach has merit. It is more about the approach than the specific chemistry of the project. The industry needs to develop "technology platforms" that can be useful with different chemistries for different applications. Some similar work has been reported at other institutions, and this area should be reviewed as part of an overall approach. The industry needs to fundamentally understand whether this approach has merit.
- The project has a strong background and previous success in similar projects, which makes this project very promising. Successfully developed materials would not be limited to one application but could be applied to multiple technologies.
- There is good synergy of a processing technique and a novel polymer that have both demonstrated good results independently.

Project weaknesses:

- It is unclear how the partners interact and help each other to achieve their project goals.
- It is unclear whether this chemistry is fundamentally stable in actual applications and whether the final mechanical properties meet application requirements. Both research questions need to be tested as part of this project.
- The project seems too empirical and relies on the need to address several key issues, such as carrier polymers, solutions, heat treatment, etc., by formulation processes.

Recommendations for additions/deletions to project scope:

- It would be good to see more knowledge gained in terms of key processability and transport parameters that can be used for other polymer systems. More experimental characterization of polymers would be helpful.
- In situ (or application-oriented) testing must be done. Perhaps the PI needs industrial partners that can assist with this effort. That should be added as part of the scope of work.
- So far there are not significant results or progress, which is expected since the project has just started.

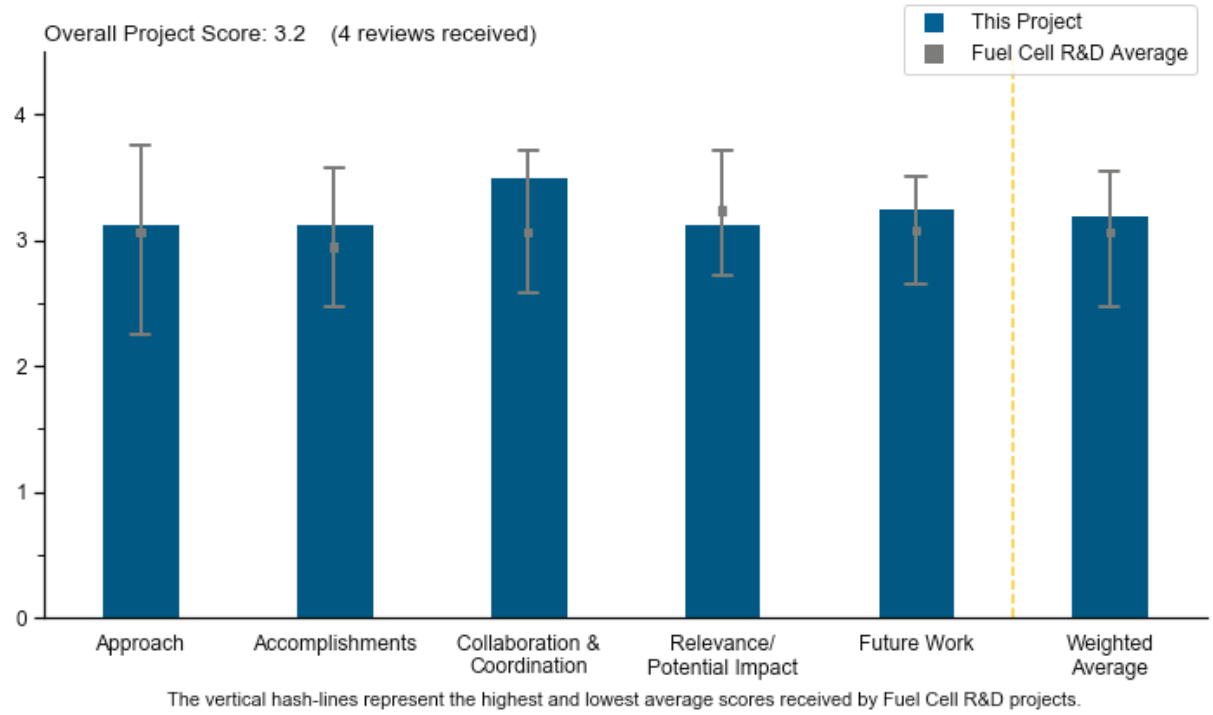
Project #FC-311: Novel Non-Perfluorosulfonic Acid Proton Exchange Membrane for Fuel Cell Application

Taoli Gu, Xergy, Inc.

Brief Summary of Project

This project seeks to develop a novel composite polymer electrolyte membrane (PEM) using uniquely designed hydrocarbon-based aromatic sulfonated polymers and reinforcement technology to meet the U.S. Department of Energy's durability, cost, and performance targets for PEM fuel cell electric vehicles (FCEVs) and provide enhanced characteristics over state-of-the-art PEMs. The project will synthesize a novel BP-Ar_3 ionomer, incorporate the ionomer into porous support materials, characterize a membrane electrode assembly (MEA) based on the technology, and conduct a cost analysis.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.1** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- Xergy, Inc. (Xergy) has an interesting and novel approach for hydrocarbon membranes. One of the long-standing challenges of aromatic sulfonic acids is the low conductivity at low relative humidity (RH). The side chain structure might change the morphology enough to improve this property. Comparisons of conductivity as a function of RH with traditional perfluorosulfonic acids (PFSAs) would be valuable and a true measure of advancement over the existing hydrocarbon options.
- The synthesis of a partially fluorinated, aromatic sulfonated hydrocarbon membrane (called BP-Ar_3 PEM) and the optimization of the sulfonation of ionomer precursor to synthesize the BP-Ar_3 PEM ionomer are reasonable new membrane technology. The questions to be answered are about physical stability (i.e.,

shrink and swell) and conductivity as a function of humidity. These can be solved but certainly need to be optimized.
- The approach of using multiple proton-exchange sites in a single side chain is not new. In the past, 3M has demonstrated this strategy with PFSA-based ionomers to achieve higher proton conductivity. This approach is expected to provide higher proton conductivity per unit area, with the possibility of increasing the performance density of the fuel cell stack.
- The approach is interesting in that it will result in highly aggregated functional groups, which has been shown to be very beneficial for low-RH conductivity. The concept completely ignores years of history and learning in sulfonated aromatics, and there is no reason to believe that these polymers will have better durability than those membranes. There are numerous vulnerable spots for OH to attack, on both the main and the side chains, and especially the functional group itself. Also, a post-sulfonation of the aromatic groups will leave a sulfonate group that can easily be removed, as McGrath showed conclusively. A review of the history of sulfonated polystyrene/divinylbenzene, as well as the Ballard membrane and many other membranes that have been tried over the years, would make one hesitate before going down this path. Finally, using porous polyethylene will be very difficult, particularly if the team wants to hot press electrodes at some point, as that will erase the polyethylene porosity.

Question 2: Accomplishments and progress

This project was rated **3.1** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project started in March 2019, and the team has developed, optimized, and scaled up the synthetic pathway of the BP-Ar_3 PEM ionomer precursor. The proton nuclear magnetic resonance (NMR) shows that the team is able to make pure BP-Ar_3 PEM ionomer precursor.
- Slide 6 shows that polymer has been made on a small scale at Rensselaer Polytechnic Institute (RPI). Slide 9 shows that support of the membrane is available from Xergy. Characterization in fuel cells can be done at the University of Delaware (UD), according to slide 10.
- The project has just started, so it is hard to know; the team has made the polymer, so that is a good start.
- This project is in its first quarter. The plan looks good, but expectations for progress are minimal at this time.

Question 3: Collaboration and coordination

This project was rated **3.5** for its engagement with and coordination of project partners and interaction with other entities.

- The project has good teaming and division of labor. RPI is doing polymer synthesis, Xergy supported the membrane-making, and UD is doing MEA and fuel cell testing.
- The collaboration between the principal investigator (Xergy) and subcontractors (UD and RPI) makes sense. With their unique expertise, all three entities are complementary to each other in running this project.
- Xergy, RPI, and UD are all well-respected organizations. The project looks to have a good balance of skills.
- The partners are well qualified but not yet engaged, so the collaboration cannot be judged at this time.

Question 4: Relevance/potential impact

This project was rated **3.1** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The development of a novel PEM that uses hydrocarbon ionomer and low-cost reinforcement is needed to meet DOE's performance, durability, and cost targets for the commercialization of FCEVs. The proposed project addresses the key issue of increasing fuel cell power density via an increase in membrane conductivity, which will help FCEV manufacturers make miniaturized, high-power-density fuel cell stacks.

High-density fuel cell stacks would also make a big impact on miniaturizing the balance of plant of fuel cells in the vehicle.
- This project could lead to low-cost fuel cell (BP-Ar_3 composite) membranes, but the durability of the novel BP-Ar_3 composite membrane is unknown and remains the greatest challenge.
- This project might develop some knowledge regarding structure and property relationships, as these polymers should have very high local charge densities. There is no reason to believe these polymers will have appreciable lifetimes in a fuel cell operating at relevant temperatures.
- While a low-cost, high-performing membrane is always desirable, it is unclear that this approach can truly compete with well-established PFSA technology. A lower cost alone will not be enough unless the performance and durability match, or exceed, that of PFSAs.

Question 5: Proposed future work

This project was rated **3.3** for effective and logical planning.

- The team has clearly described the project's future work for fiscal year (FY) 2019 and FY 2020. The future work has been adequately divided between the team members on the basis of individual team expertise.
- This project has the potential to advance the state of the art in hydrocarbon membranes. The project's future plans look reasonable for evaluating this approach.
- As the greatest question with these polymers is of durability, this should be addressed early. The investigators recognize that this is the largest challenge for these films, but they address only mechanical durability, not chemical.
- The researchers have restated their goals, but there are no design-of-experiment details for how they will meet these goals. The team is to scale up in 2019 and develop a membrane in 2020. The team sounds confident, but this seems improbable. It is unclear how the team can scale up and make membranes when the team members do not even know the stability of the optimized polymer membrane yet.

Project strengths:

- The project team is very strong and knowledgeable in the field of the proposed work. The tasks were appropriately divided between the team members on the basis of their technical expertise.
- The main strength of this project lies in the unique aromatic sulfonic acid side chain. If this structure can improve on the conductivity at low RH compared to traditional, backbone, aromatic sulfonic acids, then the project will be a success.
- The project has a good division of labor, with RPI on synthesis, Xergy on support, and UD doing testing.
- The polymer structure addresses the need for high localized charge density.

Project weaknesses:

- A major weakness for all hydrocarbon membranes is oxidative stability. The team has a good plan for mechanical stability through reinforcement, but chemical stability may still be insufficient for many applications. If the conductivity as a function of RH is the same as with other hydrocarbon ionomers, then this approach has not much advanced the state of this class of membranes. However, the project is well suited to answer this question.
- The approach of using multiple proton-exchange sites that are anchored to the single side chain has been attempted in the past by 3M in a DOE-funded project. That project suffered significant issues with swelling and high water retention (MEA flooding), despite the expanded polytetrafluoroethylene (ePTFE) reinforcement. The current team's similar approach may encounter similar hurdles.
- The project completely ignores scores of years of experience that says that these chemistries are not stable in a PEM environment.
- Very little detail was given on how the project's goals will be achieved.

Recommendations for additions/deletions to project scope:

- No addition or deletion is needed in this project. The project is fine as proposed.
- As the team states on slide 9, the "durability of the novel BP-Ar_3 composite membrane is unknown and the greatest challenge. Team will systematically identify failure modes." This is the top priority, as this is really the main thing this project offers.
- The project needs to add a chemical durability component, as well as ways to mitigate this issue. This may perhaps be partial fluorination. Ce has been shown to be ineffective in these systems.
- Comparison to well-established PFSAs and traditional hydrocarbon ionomers (if available) would highlight the potential (or lack thereof) for this approach.

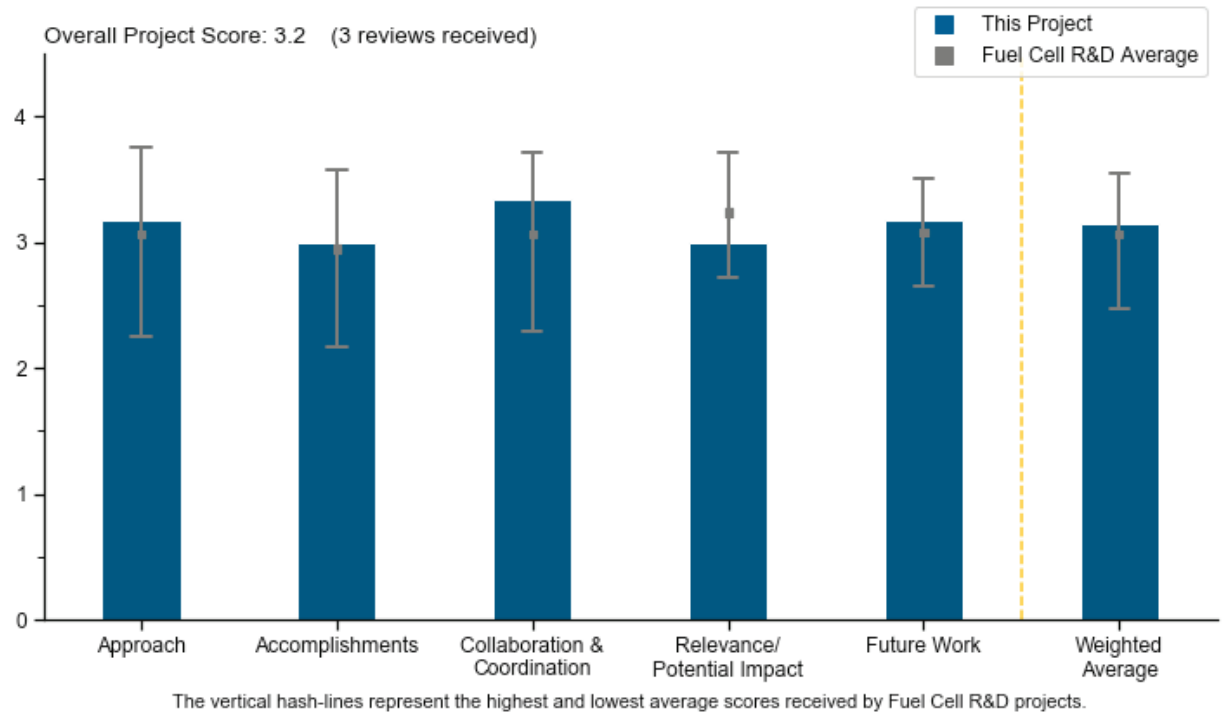
Project #FC-312: Molten Hydroxide Dual-Phase Membranes for Intermediate-Temperature Anion Exchange Membrane Fuel Cells

Patrick Campbell, Lawrence Livermore National Laboratory

Brief Summary of Project

This project will demonstrate the performance of molten hydroxide dual-phase membranes as anion exchange membranes (AEMs) in intermediate-temperature, air-oxidant compatible hydrogen fuel cells. The dual-phase molten hydroxide/ceramic support membrane being developed utilizes steam-based carbonate management and works at intermediate temperature to enable superior fuel cell performance and simplified operation. The project aims to (1) achieve ionic conductivity of greater than 600 mS/cm, (2) produce a complete fuel cell assembly with a membrane area greater than 50 cm^2, (3) validate that a steam-based carbonate management approach is effective during long-term operation, and (4) demonstrate high performance across an intermediate temperature range of 150°C–400°C.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.2** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The approach of molten hydroxide/ceramic support membranes utilizing steam-based carbonate management has been demonstrated to work at intermediate temperatures. This is expected to enable superior fuel cell performance and simplified operation due to efficient carbonate management.
- This is an interesting approach to alkaline fuel cells, utilizing a porous ceramic support for molten hydroxide ionomer. The method for carbonate management is novel and has the potential to reduce the impact of CO_2. The project has achieved high ionic conductivity so far, but the membranes are thick, and the area-specific resistance (ASR) is high at 0.5 ohm cm^2, more than an order of magnitude higher than

polymer electrolyte membranes (PEMs). The thickness of the membranes and supports needs to be <100 microns to get close to the ASR for polymer AEMs or PEMs. This will require a membrane 5x–10x thinner than what the team is making. The durability of porous ceramic membranes that are less than 100 microns thick could be an issue. Mechanical durability does not seem to be addressed in the approach.
- The project's approach involves dual-phase molten hydroxide/ceramic support membranes with steam-based carbonate management. The dual-phase membrane approach seems reasonably straightforward and sound. However, there is very little detail on the steam-based carbonate management. It is not clear what the mechanism is or whether peroxide is formed from the water to convert the carbonate to CO_2. This needs to be better demonstrated in this project; a detailed mechanism should be proposed and validated. The approach should also target thinner membranes to get to lower ASR.

Question 2: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has achieved good pore size distribution with a majority of pores <100 nm. The approach of dip coating for edge sealing has promise, but the team needs to demonstrate that it is gas-tight. The test station is assembled, which allows for testing. The project has just begun and is only about six months along.
- Good progress has been made, given that this project started in October 2018 and is only 20% spent. The team has already made porous supports and incorporated 1:1 NaOH–KOH into those. However, the ASR of 0.5 ohm cm^2 is still too high. The 50–80-micron-dense yttria-stabilized zirconia (YSZ) coatings over the sealing area seem to work. It will be good to see fuel cell results from these soon.
- The team has successfully demonstrated the fabrication of <500-µm-thick porous membrane support with no defects. However, in typical AEM fuel cells, the electrolyte thickness is <50 µm to achieve low cell resistivity and high power density. Porous membrane supports that are <500 µm thick may be adequate for the solid oxide fuel cell (SOFC) architecture, as it operates at much higher temperatures (>800°C), but it is not clear how such a thick electrolyte will perform at intermediate temperatures. Preliminary electrolyte conductivity measurements with 1:1 NaOH–KOH at 200°C showed a conductivity of 196 mS/cm, which is lower than the project goal of >600 mS/cm. The electrolyte thickness needs to be decreased to achieve such high electrolyte conductivity. The team may also need to determine the conductivity improvement that the present electrolyte could achieve by increasing the temperature to 300°C or 400°C.

Question 3: Collaboration and coordination

This project was rated **3.3** for its engagement with and coordination of project partners and interaction with other entities.

- The team has a very good commercial partner, OxEon Energy LLC (OxEon), who will be providing fuel cell and electrolyzer expertise that is needed for this project. The team also has the right collaborating partner for design consulting and testing capabilities. FCA US LLC is well known for providing support to these aspects.
- Lawrence Livermore National Laboratory (LLNL) has a technology and has partnered with OxEon for this project. LLNL also has three patents and/or applications on this technology. The team should clarify whether OxEon plans to license these patents if this project is successful.
- The initial efforts suggest that the collaboration between partners is working okay.

Question 4: Relevance/potential impact

This project was rated **3.0** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The success of this project will help to achieve orders of magnitude of improvement in ionic conductivity compared to existing intermediate-temperature AEMs. Present AEM technologies are unable to achieve such high electrolyte conductivity. Validation of the proposed steam-based carbonate management

approach will be very effective during long-term operation. The carbonate mitigation strategy is already built in the electrolyte technology, and it will help the electrolyte to sustain long-term fuel cell operation. The formation and demonstration of such dual-phase membrane electrolytes in a >50 cm^2 cell will validate the viability of this technology in cells with larger footprints in the future.
- The project has high relevance and the potential for a new type of intermediate-temperature fuel cell that could potentially be low-cost. This could potentially provide a platinum-group-metal-free intermediate-temperature fuel cell.
- This project needs quantitative milestones to be more relevant to DOE. For example, the first go/no-go is "satisfactory membrane and cell component performance." These need to be quantified. The project should aim to achieve DOE's membrane ASR targets and to get close to DOE's target of 600 mA/cm^2 at >0.6 V at some temperature. The only quantitative target listed on slide 5 is >600 mS/cm. This needs to be significantly improved with more quantitative milestones and ASR and fuel cell performance targets.

Question 5: Proposed future work

This project was rated **3.2** for effective and logical planning.

- The team has adequately described the future work for fiscal years 2019 and 2020. The team has identified and defined different phases and milestones of the project very well. The team has also correctly identified the go/no-go steps to judge the actual progress of the project. The team has maintained a true balance between the scientific challenges and the feasibility of technical success in the project.
- The proposed future work addresses the challenges of sealing and shows efforts to prove the carbonate mitigation strategy. The proposed work does not seem to be directed at making thinner membranes, which is important to achieving good fuel cell performance. The supports need to be one-fifth to one-tenth as thick as current supports. That will take significant effort.
- The plan seems sound and will result in 50 cm^2 fuel cells and an eventual commercialization plan. The project needs more quantitative targets; also, the mechanism for the carbonate mitigation using steam needs to be determined.

Project strengths:

- The principal investigator, team, and collaborators of this project are strong. The team has a good industrial partner in OxEon, who has great insight into this problem. The team has a good fuel-cell-hardware collaborator in FCA US LLC, who can guide the team with appropriate fuel cell hardware and setup conditions to mitigate testing and hardware-related issues that may become an obstacle for the project.
- This is a good team and a nice concept. A laboratory technology is being transferred to industry.
- The project has a novel idea and concept.

Project weaknesses:

- No weaknesses are identified at this point.
- Sealing is a big challenge in high and intermediate SOFCs that possess ceramic materials. The team did not explain much of the project's sealing strategy with the proposed porous-ceramic-support-based electrolyte. The team has proposed using "glue" as sealing material; however, there are many other issues with traditional glue, including leaching and creep issues. The team did not specify the humidification and carbonate mitigation strategy, so it is not known how the team will manage these two complex issues.
- There is a lack of quantitative milestones.

Recommendations for additions/deletions to project scope:

- The whole project, as presented, is fine and does not need any additions or deletions.
- The team should determine the mechanism of carbonate mitigation via steam operation.
- There are no recommendations at this time.

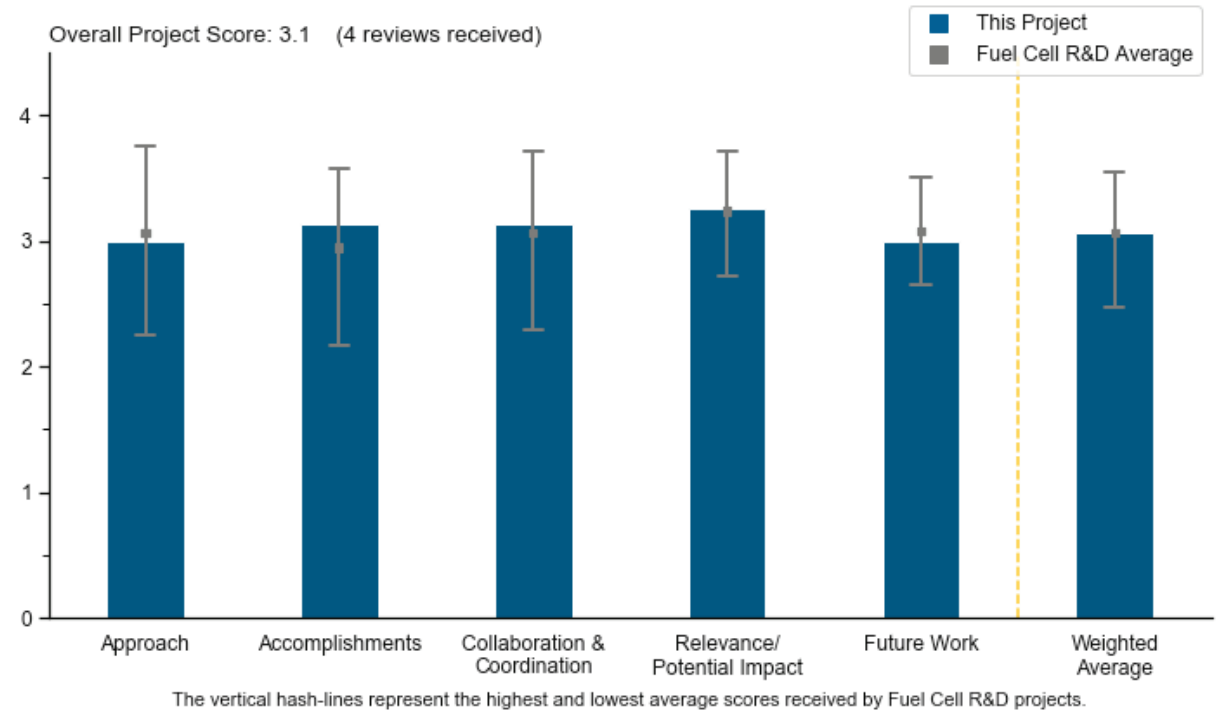
Project #FC-313: Novel Bifunctional Electrocatalysts, Supports, and Membranes for High-Performing and Durable Unitized Regenerative Fuel Cells

Nem Danilovic, Lawrence Berkeley National Laboratory

Brief Summary of Project

The main focus of this project is to demonstrate a highly efficient and stable unitized regenerative fuel cell (URFC) achieved through a novel membrane and supported electrocatalysts. The goal is to achieve 50% round-trip efficiency utilizing advanced membranes and bifunctional oxygen evolution reaction/oxygen reduction reaction catalysts on engineered supports. Project tasks include (1) developing membrane/ionomer and catalyst supports, (2) integrating the membrane into a membrane electrode assembly (MEA) and integrating the bifunctional catalyst onto supports, (3) demonstrating MEA performance and durability in electrolysis testing, and (4) demonstrating MEA performance and durability in fuel cell testing.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.0** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The proposed approach addresses the performance and durability of reversible fuel cells (RFCs). The team has identified major project risks and has appropriate mitigation strategies for them.
- The objectives are clearly articulated and clearly address the barriers that are identified. A strong multi-sectorial team has been assembled to tackle the various problems. The major technical barrier being addressed is to manage a huge breakthrough in the URFCs. Because the approach is logically based on developments for both fuel cells and water electrolysis, the project is addressing barriers identified for the two of them but considering specific targets. The targets are outstanding compared to baseline URFCs but are less challenging than individual targets for each technology. Only durability is expected to be

demonstrated at much lower levels compared to fuel cells or electrolyzers. The targets are particularly challenging for the water electrolysis mode: it is difficult to get how the approach on materials will allow for long-term operation with thin membranes at such loadings. Specific targets are identified, and the project plan is to reach them thanks to the work done on core materials, the membrane, and oxygen-side catalysts for improving both fuel cell and electrolyzer efficiency. The lack of information about the development routes prevents proper assessment of the approach. However, the overall approach is simple and consistent, starting with materials development, followed by their implementation in MEAs and testing in dedicated cell devices for fuel cell, electrolyzer, or URFC modes. Only one partner seems to actually be involved in the URFC.

- There are issues in this project that need to be addressed. (1) This project is confusing. It is regenerative, but in barriers it states "no regenerative-fuel-cell-specific barriers." (2) The proposal is quite broad, with membranes and supported catalysts all in one project. (3) It would be nice to see a flowchart of how the contributors are interfacing. (4) It was stated that key personnel may have changed, which has impacts on the approach. (5) The challenges list the gas diffusion layer (GDL) and porous transport layer (TPL), which seem to be out of scope. (6) There is no mention of what will be unique about the membranes; they seem to be just commercially bought.

Question 2: Accomplishments and progress

This project was rated **3.1** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Lawrence Berkeley National Laboratory (LBNL) has made reasonable progress in the short time the team has been active. Baseline MEA fabrication has been completed, and initial baseline testing has been done at LBNL. Subcontracting to partners is still not completed.
- The work started recently; it is too early for proper ranking. However, baseline materials were defined, and the first results are available as planned.
- Despite subcontract negotiation issues, which are common to these collaborative efforts, good progress has already been made.
- It is early, but it seems that a lot must be done in two years.

Question 3: Collaboration and coordination

This project was rated **3.1** for its engagement with and coordination of project partners and interaction with other entities.

- Coordination and plans for coordination are good. Bringing in Pajarito Powder, LLC (Pajarito) as a subcontractor should improve the project. Coordinating with Max Wei (LBNL) for URFC targets should be beneficial.
- This is a highly collaborative project with participants from industry (U.S. and foreign), national laboratory, and university sectors.
- Pajarito was added at the beginning; this seems relevant and required. Therefore, it is not clear why this role was not planned at an earlier stage. There are overlaps between partners for manufacturing and testing. Only one partner is actually involved in the URFC, even though this is the core objective of the project. The link with Solvay is indicated as a collaboration, but Solvay seems to be a supplier. If a common development with Solvay is planned, this should be mentioned clearly, including how they are collaborating and on what aspect of the membrane.
- Based on discussions at the presentation, there seems to be some flux with the contributors. There perhaps needs to be more clarity on what exact formulations will be made and why (e.g., how much Ir with Pt).

Question 4: Relevance/potential impact

This project was rated **3.3** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- RFCs are highly relevant to DOE's H2@Scale efforts. The potential impact of this work is high, and participation by Ballard Power Systems, Inc. (Ballard), Proton OnSite, and Pajarito should increase the probability that the project will have an impact.
- The milestones and targets are aggressive and could significantly advance RFC technology.
- The project is considering several DOE objectives but addressing a very specific device; thus, it is not easy to conclude on the expected impacts. If the objectives are reached, progress will be made toward cost and performance objectives for polymer electrolyte membrane fuel cells and toward efficiency on the hydrogen production side.
- Using existing commercial membranes does not create much new impact. Pt–Ir systems have been around for a long time. The main impact, it seems, is the Washington University in St. Louis's supports for stability.

Question 5: Proposed future work

This project was rated **3.0** for effective and logical planning.

- The proposed work should advance RFC performance. The proposed supports should provide improved durability, while the bifunctional catalysts should improve performance.
- Good risk mitigation plans are presented.
- In year 1, the next developments are planned for the catalyst's support and integration, but nothing is mentioned for the membranes; this should be justified, considering the title of the project, including the membrane aspect. MEA optimization should focus on the reversible operation; performance for discrete operation could be second in the order. This is the same for the accelerated stress test (AST); it would be better justified to focus on defining the right duty cycles for URFCs or a specific AST for URFCs, if needed. In year 2, scale-up and discrete testing are planned; it should be that scale-up and reversible testing are planned for validation.
- The GDL and PTL seem to be out of scope. It seems there is flux in the project.

Project strengths:

- The collaboration between industry and laboratories is a positive aspect, allowing for combined interest in innovative developments and applicability. The targeted technical improvements, if they are reachable, are project strengths. The idea of selectively combining fuel cell and electrolyzer catalyst materials for the manufacturing of the reversible electrodes is the most interesting aspect of the ongoing work.
- The project has assembled a team with large industrial participation.
- The project's strengths include the new supports and the breadth of the team.
- A strong team has been assembled to address several barriers.

Project weaknesses:

- The complementarity between partners is not always clear, particularly for MEA manufacturing and testing; the choice to double some actions should be justified. More efforts seem to be put on the separate assessment in fuel cell or electrolyzer modes instead of pushing URFC tests, which is the core scope. There is a lack of technical information about developments concerning the membrane in particular. Concerning catalysts, results could have been shown (electrochemical analyses or microstructure observations could have been mentioned, if available). The differences in the level of information expected from 25 cm^2 and 50 cm^2 is not obvious; the choice of considering these two areas for evaluating up-scaling should be clarified. For the validation task, the reason that Proton OnSite and Ballard are performing fuel cell or electrolyzer testing only is not justified. Validation should be done at larger scale for actual URFC cases.

Duty cycles are defined as a challenge; this should be considered more of a required objective for final validation.

- The project has a broad, unfocused scope. Out-of-scope challenges are listed. The project has confusing targets and seemingly arbitrary baselines. Using a 175 µm membrane as a baseline needs explanation as to why it is so thick. Also, the team is in flux.
- Contracting issues have delayed the work.

Recommendations for additions/deletions to project scope:

- It is recommended that the project clarify whether the membrane development aspects are still needed for the project outcomes and, in this case, what the routes explored and next steps are and what the possible role of Solvay might be. Focusing on the assessment and validation of materials and devices in URFC conditions with the definition of proper duty cycles would add value to the project (instead of focusing on discrete validation), since other projects are already considering the specific developments for fuel cell or electrolyzer materials and improvements. For the next review, more details should be given and more results shown.
- The project should eliminate or clarify the membrane part and flesh out the catalyst development paths and reasoning. Out-of-project components should be eliminated.
- There are no recommendations at this time.

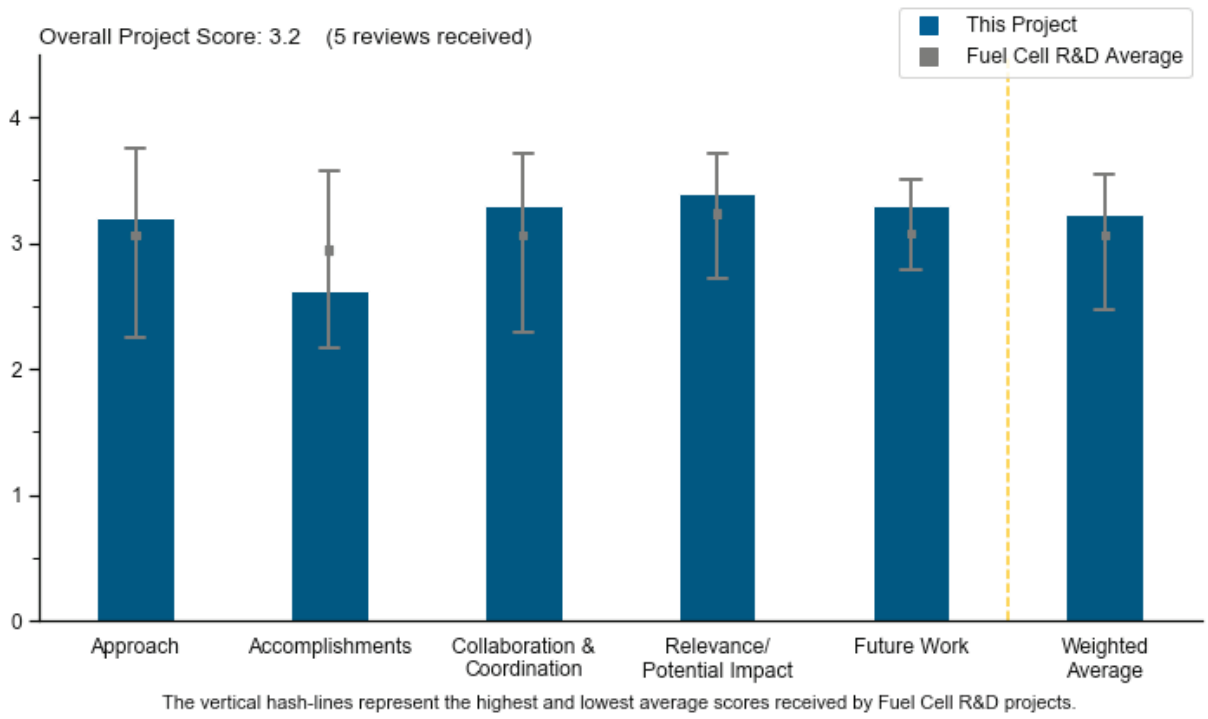
Project #FC-314: Efficient Reversible Operation and Stability of Novel Solid Oxide Cells

Scott Barnett, Northwestern University

Brief Summary of Project

This project will develop and test reversible solid oxide cells (ReSOCs) for electrical energy storage applications, including system concepts for high efficiency. The ReSOCs will be designed to operate efficiently and durably in both fuel cell and electrolyzer modes. The project approach employs novel high-temperature cells with the potential for high power density, long-term stability, and high round-trip efficiency. The ReSOCs will be designed to achieve the low area-specific resistance (ASR <0.15 Ωcm^2) required for high round-trip efficiency at high current density (>1 A/cm^2). One focus is on durability improvement via a combination of materials development, mechanistic degradation model development, and accelerated stress testing to determine the factors that affect long-term stability, including reversible operation cycles. Multiple cell designs will be investigated. The team will also fabricate and test large-area cells and determine the effects of pressurized testing. System modeling (system concept development and technoeconomic analysis [TEA]) will inform designs that can achieve cost and efficiency targets for renewable electricity storage.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.2** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This is a novel concept: high-temperature solid oxide fuel cells (SOFCs) capable of achieving high power density, long-term stability, and high round-trip efficiency in ReSOCs. The project will fabricate and test large-area cells based on current button cells. The team will focus on the relationship between long-term stability and operating conditions.

- The technology barriers for durability, cost, and performance are clearly identified. Project objectives are to develop, fabricate, and test large ReSOCs; determine the effect of operating conditions on cell performance and durability (including operation at high pressure); assess systems efficiency; and validate technology viability using TEA.
    - There is a significant emphasis on the degradation studies. The team needs to be careful not to test cells under unrealistic conditions (e.g., overly high cathode/anode overpotential, temperature, local oxygen partial pressure) to force earlier and more pronounced degradations.
    - When reporting ASR or degradation rates, including those in the go/no-go milestone (slide 6), the specific test conditions (temperature, pressure, cell voltage, etc.) should be provided. It would be beneficial to compare to the state-of-the-art performance and list the target values.
    - The round-trip efficiency is typically discussed for a device or system because it ties into storage rather than for a single button cell, as in this work. From thermodynamics, cell efficiency is always high for a high-temperature cell, but there are penalties for storage and compression. The efficiency itself may not be as important as the cost per stored amount of energy. Efficiency can also be measured in many ways; how it was calculated should be explained.
- In general, the approach is good. It appears that the work is very focused on small-scale tests and materials characterization. This is cost-effective, but the project could benefit from access to samples taken from larger systems. This may simply not be possible, as there are not a large number of solid oxide electrolyzer cells (SOECs) and systems in operation, even at a laboratory scale.
- The approach and down-selection to look at three different cell designs is appropriate. The use of a 3D-printed cell design has some potential for improvements. The thermal energy storage concept could provide advantages and ease thermal stresses during transitions between SOEC and SOFC modes. The round-trip efficiency target of >70% is aggressive.
- The approach to addressing performance and durability is very good, but it is limited to small cells. It appears that work on scale-up will not occur until quite late in the project. Since making cells of a practical size is a major challenge for SOFCs and will undoubtedly be required to meet the cost targets for the intended application, this major challenge should also be a focus of the project. A similar comment can be made with respect to fabricating a robust cell stack, which is another major SOFC challenge.

Question 2: Accomplishments and progress

This project was rated **2.6** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project is just starting. Models for high-temperature reversible fuel cell (RFC) systems have been established. Desired operating conditions have been determined.
- A number of results were presented, and it appears the project is generating some reasonable outcomes.
- The project contract was just recently finalized, so no work has been done yet.
- This project was only recently funded. It contains no new results, and it is not possible to evaluate progress.

Question 3: Collaboration and coordination

This project was rated **3.3** for its engagement with and coordination of project partners and interaction with other entities.

- The team members will utilize their best strengths and support each other. Northwestern University will develop and fabricate cells, test and characterize them, and provide experimental data to Colorado School of Mines (CSM) to perform stack and system modeling. CSM will provides input to Northwestern University regarding desired cell characteristics and operating parameters, ensuring that test results are relevant.
- Collaboration between Northwestern University and CSM should be good. The combination of modeling and experiment should be beneficial.
- It appears that the team has clear roles and a good plan on how they will collaborate throughout the project.
- The team is collaborating with CSM. CSM will perform stack and system modeling and TEA.

- There are only university partners in the project. It can be challenging to do fundamental research on materials degradation with industrial partners, but the team should establish some link with an organization that manufactures and is currently developing or producing SOFCs or SOECs. This would not need to be a paying partner, just an adviser who could suggest whether the approach taken and materials selected are relevant to what will likely be a commercial product in the future.

Question 4: Relevance/potential impact

This project was rated **3.4** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- It is still an open question whether energy storage using RFCs is going to be viable since the round-trip efficiency is typically much lower than other options (e.g., batteries). However, the cost of the storage media is low relative to the cost of battery-active materials, so RFCs may be viable for long-duration storage. SOFCs can potentially enable higher round-trip efficiency than lower-temperature fuel cells, so this does appear to be a promising technology for the intended application. Of course, in addition to performance, substantial durability and scale-up challenges must be overcome.
- SOEC is an important technology, and little work is done on degradation, so the project will likely have significant impact.
- The project has high relevance and is applicable to the Fuel Cell Technologies Office H2@Scale efforts. If the target round-trip efficiency of 70% is reached, it will have a large impact.
- Development of RFCs is very important for advancing energy storage and power generation technology.
- This project is related to an existing project on hydrogen production. The overall objective is to develop a system with high round-trip efficiency and low storage cost.

Question 5: Proposed future work

This project was rated **3.3** for effective and logical planning.

- The proposed future work is relevant and addresses issues of degradation and thermal effects from cycling between modes.
- The project seems to be well planned and organized.
- The proposed future work is reasonable, though no details on specifics of cell fabrication, testing, characterization, or scale-up approaches are given.
- Identifying realistic performance targets is helpful.
- The future work will be directed at achieving low ASR (<0.15 Ωcm^2) for high round-trip efficiency at high current density (>1 A/cm^2). The project will carry out tests to determine factors that affect long-term stability. Future presentations may better illustrate the methodology and approach to achieve stated objectives.
- More focus on scale-up earlier in the project would be a good addition.

Project strengths:

- The principal investigator has useful experience in identifying cell degradation at high current density and the possible benefit of cyclic potentials. Use of multiscale modeling tools for system design, analysis, and optimization may help in further understanding the degradation mechanisms.
- The project will develop and test novel, efficient, and stable ReSOCs. Teaming is excellent, and the work is well coordinated.
- The combination of system modeling and cell development is a strength.
- This is a good fundamental study of materials degradation.
- The project focuses on performance and durability.

Project weaknesses:

- A weakness is not evident at this time.

- There is no significant weakness.
- It is really tough to work on the degradation of a technology still in development. It is a simple fact of life that no one knows what materials will be in an SOEC in the future, so there is a chance that this work may not be relevant to future systems. That should not be taken as a reason not to do the work, but it is a potential weakness.
- What makes the team's SOFC technology different—i.e., why this should be better than other SOFC systems—is not really clear. Also, there is not enough focus on scale-up of cell size and stack technology.
- It is not clear what is "novel" in novel ReSOCs. The materials set listed is well known and has been previously tested and characterized.

Recommendations for additions/deletions to project scope:

- There are no recommendations on the project approach; the approach is very reasonable.
- An industrial partner might be beneficial for stack-level testing.
- The project should have some form of industrial engagement, even in a fairly informal way.
- At the next Annual Merit Review (AMR), the team should communicate the following:
    - What differentiates the team's SOFC technology
    - What the issues are with making larger cells, and how these risks will be reduced in this project
    - What the issues are with making cell stacks, and how these risks will be reduced in this project

  If the answers to the above questions are known but were just not clearly communicated during the AMR, then the team should also modify the plan.
- It is too early to change directions or add tasks.

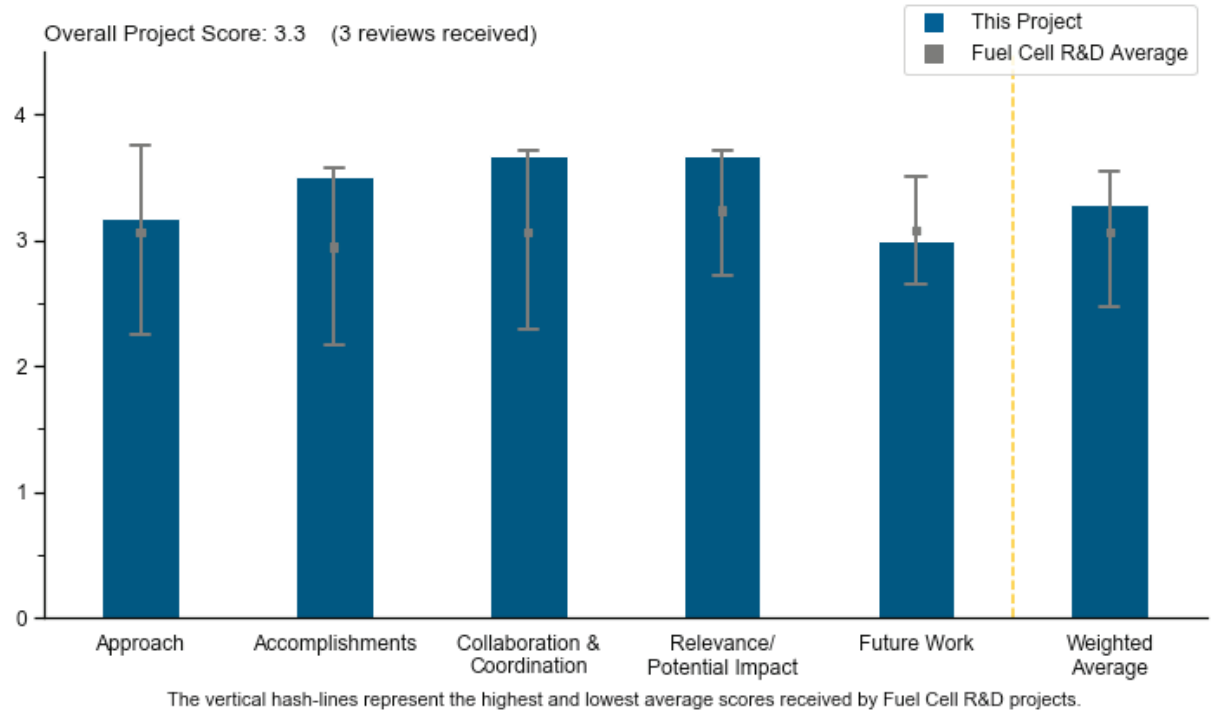
Project #FC-315: High-Efficiency Reversible Alkaline Membrane Fuel Cells

Hui Xu, Giner, Inc.

Brief Summary of Project

Reversible fuel cells can store renewable energy as batteries do, with one advantage: the ability to accommodate much higher energy density. An alkaline membrane system may reduce the associated capital costs, thanks to adopted inexpensive materials (e.g., catalysts and bipolar plates). The project team is investigating the following technical approaches to enable reversible alkaline membrane fuel cells (AMFCs) for stationary energy storage: (1) bifunctional catalysts for hydrogen oxidation reaction (HOR)/hydrogen evolution reaction (HER), (2) bifunctional catalysts for oxygen reduction reaction (ORR)/oxygen evolution reaction (OER), and (3) high-performance alkaline membranes (with high OH- conductivity, oxidative resistance, and mechanical stability). This project will demonstrate, for the first time, a reversible AMFC with greater than 50% roundtrip efficiency at 1 A/cm^2 without introducing any salt or base in the aqueous feed.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.2** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This project is extremely ambitious. The work scope includes advances in membranes and bifunctional catalysts at each electrode, with project funding of only $1.25 million over only two years. This seems like a good deal to tackle. However, the project team is excellent and comprises leaders in each relative area. The principal investigator and the project team have among the best records of broad collaboration in the electrochemical space, and a unitized reversible test cell is a big step forward for this type of work.
- The project team members are very experienced in the development and testing of electrolyzers. They understand that there are many benefits to using OH- membranes but equally understand that these

membranes have many stability issues. The combination of materials experts from university and industrial partners with device expertise is clearly beneficial.
- The general technical approaches to using hydroxide exchange membrane (HEM), HER/HOR and OER/ORR catalysts are suitable. However, it seems all components used for the reversible fuel cells have already been developed for fuel cell or electrolyzer applications. There is no clear explanation of how these materials produce the best reversible fuel cell performance. High-temperature AEMs >100°C is something new, but that is a risky approach. No AEMs have demonstrated excellent stability at >100°C with water vapor, even in ex situ conditions. This approach likely will fail.

Question 2: Accomplishments and progress

This project was rated **3.5** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- This project has only just started; the project team is making reasonable progress, but it is still early in the project.
- It is too early to see progress in this project, as work has been ongoing for only three months.
- The project is at an early stage. Therefore, not much progress is reported.

Question 3: Collaboration and coordination

This project was rated **3.7** for its engagement with and coordination of project partners and interaction with other entities.

- Leaders in ORR/OER and HOR/HER bifunctional catalysts have been selected, along with a very successful AEM chemist. The project is well designed for collaboration; now the project team needs to deliver.
- The project is at a very early stage, but it appears that both partners have been assigned clear, complementary tasks to complete.
- The project's collaboration exists. Giner, Inc., is managing project coordination.

Question 4: Relevance/potential impact

This project was rated **3.7** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- Reversible fuel cells are probably best suited with an AEM, given the wider array of catalysts possible in a basic environment. This moonshot approach is well suited for the current DOE emphasis on early-stage research and development (R&D) work. Success in this project would be very impactful in the electrochemical device field.
- The project target aligned well with the DOE Hydrogen and Fuel Cells Program. Developing platinum-group-metal-free catalysts for reversible fuel cells is relevant to the DOE approach to reducing device cost. Applications for regenerative fuel cells exist. If the project is successful, the product of this project will find markets and have impacts on them.
- The development of a stable OH- system could potentially reduce electrolyzer and fuel cell costs, but it also has the potential to allow for more novel electrochemical devices to be manufactured.

Question 5: Proposed future work

This project was rated **3.0** for effective and logical planning.

- The correct work has been proposed. The project hits all areas of reversible fuel cell R&D necessary to be successful.
- The project plan seemed reasonable.

- Similar or the same poly(aryl piperidinium)-based AEMs can be seen in multiple AEM-related projects. The base membranes have been reported in the literature. All milestones related to the AEMs seem similar to what the University of Delaware already has. The project target is >100°C under vapor conditions; the fuel work should align with the device condition, not 80°C conditions. Likewise, the catalyst development milestone should not target xx grams of catalysts; it should target xx activity. Having inactive catalysts with quantity will not help the project. The participants seem aware of this issue, so they need to need to rewrite the milestone. The go/no-go milestone needs some more description. For example, it needs to be known if the reversible fuel cell will use caustic solvent and, if not, what voltage limit it will have, what the target temperature is, and how many cycles will be analyzed. Milestone 4.2 should also carefully define the degradation rate of <1%; the voltage, temperature, and other conditions should be defined specifically. Also, the diagnostic works of the MEA should be included.

Project strengths:

- One project strength is that Giner, Inc., has experience in reversible fuel cell technology. The other strength is that all critical reversible fuel cell components will be developed in one project, in an organized manner. By the end of the project, the project team may bring results that identify the bottleneck of alkaline HEM reversible fuel cells.
- The unitized cell is a big advance in studying these MEAs. The team is excellent and broadly capable of delivering on the ambitious targets.
- Overall, this is a sensible, solid project for developing a new set of materials that could benefit many different electrochemical devices. However, reversible fuel cells offer very limited benefits over a separate fuel cell and electrolyzer combination. Clearly, DOE and many in the field disagree, but it would still be better to test the materials developed in this project in separate devices (fuel cell and electrolyzer) rather than in a combined device that offers little benefit in terms of cost or performance.

Project weaknesses:

- This is a challenging project. It will be very difficult to have to have an efficiency of about 70% of the performance of acid reversible fuel cells, even if all those proposed materials work extremely well. This project does not provide good metrics to evaluate component performance. This type of project should be supported in the innovative concept category with a lower funding level.
- There is little value in combining a fuel cell and an electrolyzer. That device is called a metal hydride battery, and significant compromises must be made in the way the oxygen and hydrogen are stored because of the different balance of plant for each of these two devices. There is some value in doing this with high-temperature devices because the round-trip efficiency is higher, but there is no value in doing this if the device operates below the boiling point of water.
- The project is hugely ambitious and may not have enough funding to be successful. The reduction–oxidation (redox) reaction cycle and oxidation concerns in AEMs is a new idea, and the materials did not explain well why it is important.

Recommendations for additions/deletions to project scope:

- A recommended addition would be SMART (Specific, Measurable, Achievable, Relevant, and Timely) milestones for each component. The AEM performance needs to be measured at the relevant device conditions, including stability at >100°C at a reduced relative humidity. The HER/HOR and OER/ORR catalysts need to be evaluated in rotating disk electrode testing with control sample performance.
- This is very valuable work for the fuel cell and electrolyzer fields, but it would be better to focus on the development of a high-performance fuel cell or electrolyzer. There is very little value in combining the two. In reality, this would make little difference to the project, as the team would still make the same cell and test it first as a fuel cell and then as an electrolyzer. However, it is important to recognize that either a good fuel cell or a good electrolyzer would be valuable individually, whereas a combined device is likely always to be a compromise that might simply not be worth the effort.
- It would be desirable to see some more focus on explaining the concerns about AEM oxidation within the reversible fuel cell concept.

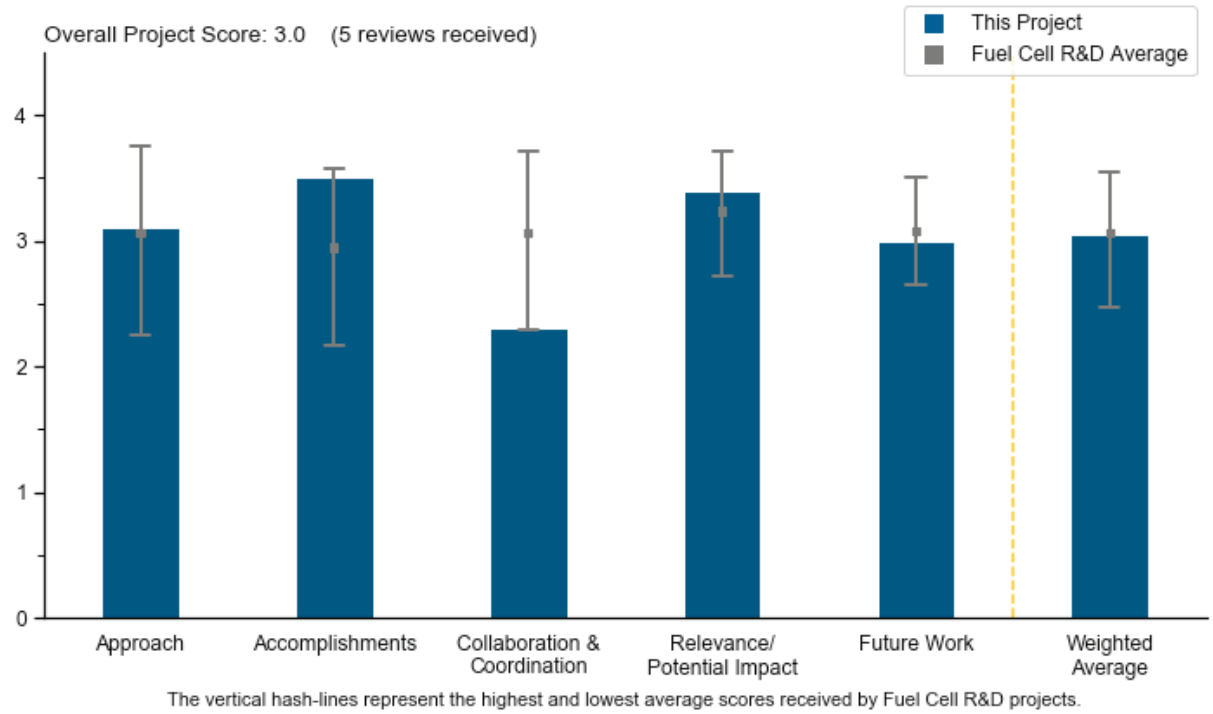
Project #FC-316: Durable, High-Performance Unitized Reversible Fuel Cells Based on Proton Conductors

Meilin Liu, Georgia Institute of Technology

Brief Summary of Project

This project is developing a robust, highly efficient, and economically viable high-temperature unitized reversible fuel cell (URFC) based on proton conductors for large-scale co-located energy storage and power generation. Project activities focus on two main areas: (1) gaining better understanding of the degradation mechanism of cell and stack materials and interfaces, using various in situ, ex situ, and operando measurements guided by theoretical analysis; and (2) integrating nanostructured components into the cell design and interfaces between electrodes and electrolyte, which will be modified with active bi-functional catalysts and protection coatings to achieve >70% roundtrip efficiency at 1 A/cm2 in both operating modes.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.1** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This is an excellent approach; the principal investigator (PI) is experienced and focused on U.S. Department of Energy goals for high-temperature URFC technology.
- The approach to develop high-temperature, high-performance reversible fuel cells (RFCs) based on proton conductors has some advantages, as it may produce pure or dry hydrogen without downstream separation or purification. According to the PI, the system can tolerate sulfur impurities at a parts-per-million level.
- Understating the degradation mechanisms in RFCs is essential for designing a robust, reliable system. Thus, the fundamental focus of this project is very important and in line with Georgia Institute of Technology's (Georgia Tech's) strengths. There are a few comments to consider.

  - There is some disconnect between the technology barriers—listed as (1) capital cost, (2) system efficiency and electricity cost, and (3) operations and maintenance—and the project's immediate objectives and approaches. It seems that the barriers are more related to a large-scale system, while the objectives and approaches (i.e., to gain a profound understanding of the degradation mechanism of electrolyte, electrode, and catalyst materials) are related to the cell development. Thanks to the high operating temperatures, cell efficiency will be high, but the system efficiency is unknown. Another barrier that was listed, capital cost, is not discussed in the poster.
  - There is significant emphasis on the degradation studies of all three cell components—electrode, electrolyte, and catalyst—without stating the issues with each of these components. It would be helpful to introduce the limitations of the state-of-the-art materials first. The approach suggests that there are issues with charge transfer and mass transfer in the electrode materials associated with the reversible operation of a fuel cell. It is not clear if this is the issue with all (or any) of the materials, or only with those (state-of-the-art-materials) that were down-selected for this study, or with those developed at Georgia Tech.
  - This project aims to test the proton-conducting reversible cells at very high temperatures, 700°C–850°C. Typically, such cells offer an advantage over oxygen ion conductors when operated at lower temperatures, such as 500°C–650°C. At >650°C, the oxygen ion conduction in BZY-based materials becomes significant, as most of the "proton conductors" are in fact mixed conductors. Degradation is also expected to be lower if operated at lower temperatures, especially since the use of nanoparticles was proposed here to avoid their coarsening.
  - It is not explained why the new coating materials for the electrolyte are needed or how they would affect the cell resistance, performance, and cost. It would also be very helpful to list the targets for durability to understand what "required durability" means.
  - The roundtrip efficiency is typically discussed for the device or system, rather than for the single button cell, as in this work. Giving the Faradaic efficiency instead is probably more reasonable.
- The approach to addressing performance and durability is very good, but it is limited to small cells. It appears that work on scale-up will not occur until quite late in the project. Since making cells of a practical size is a major challenge for solid oxide fuel cells (SOFCs) and solid oxide electrolysis cells (SOECs) and will undoubtedly be required to meet the cost targets for the intended application, this major challenge should also be a focus of the project. Although the team does have a target of 10 mm cells, these are still a very long way from the size that will be required in a future product. A similar comment can be made with respect to fabricating a robust cell stack, which is another major SOFC/SOEC challenge.
- The communication skills of the scientist who was presenting the poster were very poor. It can be tough when English is a second language, but in discussion, it was hard to understand what experimental work had been conducted. A simple diagram of the proposed system is strongly recommended. Good spoken English should not be a requirement for any scientist, but an image that can be pointed at to explain the principles of the system would be a valuable communication aid. This can be useful, especially when presenting to a group that does not speak English as a first language.

Question 2: Accomplishments and progress

This project was rated **3.5** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has made good progress, particularly in cell fabrication and microscopy characterization. This work appears to be a continuation of something the PI has been doing for a long time. The present results are reasonable for a project that has been in place only since October. One project goal is to unravel the mechanisms of charge and mass transport; this appears not to be happening quite yet, as the results are not yet discussed in terms of mechanism or of why certain materials and processes show improved behavior. Presumably, that comes at a later stage.
- This is a new project. To date, it has developed a new electrolyte coating material that has high conductivity and high stability against water and CO_2. The project has evaluated the electrochemical performance of single cells and has already demonstrated a roundtrip efficiency of ~72% at 1 Acm^{-2} at 700°C.
- This project was only recently funded. Thus, most of the data presented are likely from previously performed fuel cell activities. Some good progress has been made since the beginning of the project in

January: the team has synthesized electrode materials and fabricated and tested small cells with <1 cm diameter.
- Many results are presented here, considering that the project has just started.
- The project has early initial results.

Question 3: Collaboration and coordination

This project was rated **2.3** for its engagement with and coordination of project partners and interaction with other entities.

- The PI does not appear to have any partners involved in the work; the project resides solely at Georgia Tech. The PI says he is looking for an industrial partner.
- This project does not yet have a formal partner. Georgia Tech would need to identify an industrial partner for scaling up the cell and developing a roll-to-roll manufacturing concept.
- This project would be greatly enhanced if it included an industrial partner, since it is not clear that the team has much experience on what is required to potentially enable a viable product.
- As far as can be seen, there is little or no collaboration in this project.
- So far, there are no outside collaborators or contributors.

Question 4: Relevance/potential impact

This project was rated **3.4** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The development of RFCs is very important for advancing energy storage and power generation technology.
- There is significant potential to develop a URFC system with >72% roundtrip efficiency.
- The project's prospects for advancement are excellent.
- It is still an open question whether energy storage using RFCs is going to be viable, since the roundtrip efficiency is typically much lower than it is with other options (e.g., batteries). However, the cost of the storage media is low relative to the cost of battery active materials, so RFCs may be viable for long-duration storage. Proton-conducting SOFCs can potentially enable higher roundtrip efficiency than lower-temperature fuel cells, so this does appear to be a promising technology for the intended application. Of course, in addition to performance, there are substantial durability and scale-up challenges that must be overcome. A key advantage of proton-conducting SOFCs is supposed to be lower operating temperatures, which can enable improved durability and lower-cost sealing materials; however, the team does not appear to be focused on operating temperatures significantly lower than those used with conventional SOFCs.
- High-temperature systems offer much higher roundtrip efficiencies, which has the potential to allow for better economics. It would be simpler to operate the SOEC as an electrolyzer and then use the hydrogen in a PEM, as the same high efficiency could be achieved with a simpler and potentially much cheaper system.

Question 5: Proposed future work

This project was rated **3.0** for effective and logical planning.

- The proposed future work is clearly outlined: (1) synthesize electrodes and electrolyte powders with desired properties and (2) complete the fabrication and/or microstructure modification of electrode support with targeted 30%–40% porosity before reduction.
- The researchers have a plan, and they are following it. Once there are more results, there will be an opportunity to refine the plan.
- The proposed future work is reasonable.
- The team should focus more on the key challenges of scaling up the technology and develop methods to reduce these risks as much as possible during the project.
- The proposed future work is very extensive and covers all of the aspects of cell fabrication, testing, in situ characterization, scale-up, and the development of manufacturing concepts for mass production. This is

perhaps too broad and not realistic for the budget. The goal of testing cell performance under "various operating conditions" seems ambiguous. Instead, identifying realistic performance targets would be more valuable.

Project strengths:

- This is a good start to the project, as electrode and electrolyte materials with desired particle size and morphology have been synthesized. The team has made good initial progress in fabricating symmetrical cells and single cells.
- Georgia Tech has an excellent reputation in developing new fuel cell materials and in situ characterization of fuel cells. The same approach will be used to assist in developing high-performing materials for reversible cells.
- This is quite a simple project, with the objective of making a relatively simple cell. This is appreciated, as there is a culture of trying to oversell projects. This project will achieve its objective and add value to the field.
- The high-temperature approach is attractive, and the PI has much experience in this area.
- There is a major focus on performance and durability with small cells.

Project weaknesses:

- The project's focus so far seems to have been mostly on presenting findings but not proposing the rationale for why improvements are seen over existing materials and devices. Going forward, it would be good to focus on understanding the mechanisms and relationships that the materials and structures have with device performance.
- The project needs to put more emphasis on scale-up issues. The results that were shown are not consistent. For example, the claimed fuel cell performance is based on results obtained at 700°C, and the durability results shown are at 650°C (as seen on slide 12). Similarly, the claimed cycling performance is based on results obtained at 700°C, and the durability results shown are at 650°C (as seen on slide 13). The project needs to clarify what temperature is the focus.
- There is no collaboration or any real path for the work to deliver impact. This is acknowledged, but with no partner pushing to get results, this project will continue trying to achieve harder and harder performance goals, without ever getting to the point where a system is built.
- There is an inadequate understanding of degradation mechanisms. Precise control of the morphology, composition, and thickness of the catalyst coatings remains a challenge. Scaling up from button cell to large-area cell may be a challenge.
- The scope is too broad; the team is attempting to solve any potential problems without clearly starting the problem.

Recommendations for additions/deletions to project scope:

- At the next Annual Merit Review, it is recommended that the team communicate the following: (1) what the issues are with making larger cells, and how these risks will be reduced in this project; and (2) what the issues are with making cell stacks, and how these risks will be reduced in this project. If these answers are not obvious (i.e., known by the team but not clearly communicated in the presentation), then the team should also modify the plan.
- The team should focus on the solid oxide electrolyzer aspect. If it works as a fuel cell as well, then that is great, but the team should optimize for the solid oxide electrolyzer. Hydrogen fuel cells of good quality are already available, so the focus should be on producing low-cost hydrogen. The team should find someone who will want the technology beyond a cell.
- The team should split the scope into smaller, well-described tasks and clearly explain why each task is needed and what will be achieved.

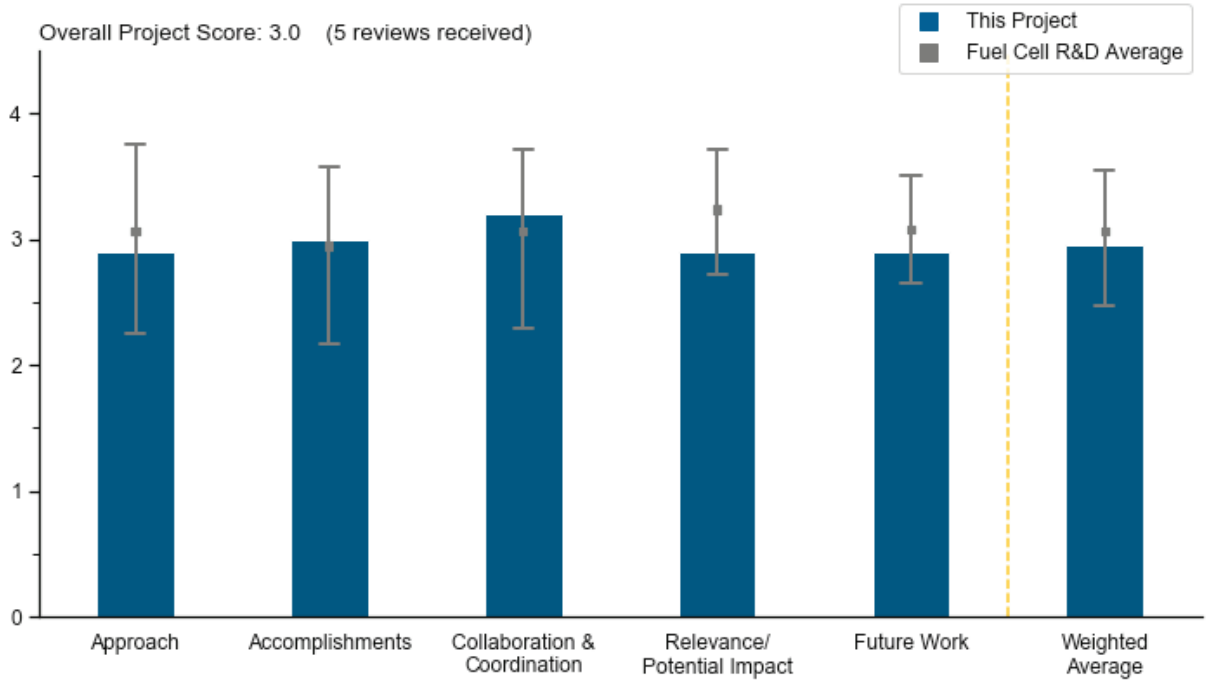
Project #FC-317: Stationary Direct Methanol Fuel Cells Using Pure Methanol

Xianglin Li, University of Kansas Center for Research, Inc.

Brief Summary of Project

The project goal is to develop stationary direct methanol fuel cells (DMFCs) using pure methanol as the fuel. The project will address three critical challenges from material to system levels: (1) reduce noble catalyst loading and cost, (2) enhance cathode tolerance of methanol poisoning, and (3) decrease methanol crossover. The end-of-project goal is to deliver a 50 cm^2 membrane electrode assembly (MEA) and prototype that produces peak power density of ≥300 mW/cm^2 with total loading of ≤3 $mgPGM/cm^2$. The project addresses the barriers of high-platinum-group-metal (high-PGM) catalyst loading, catalyst poisoning by methanol, and high fuel crossover.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **2.9** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The project combines an aligned carbon nanotube as anode and a PGM-free catalyst as cathode. Both approaches are known in previous and current research sponsored by the U.S. Department of Energy (DOE). Putting them together for a DMFC is a good idea.
- The project is in the startup phase, so the approach has not been tested yet. It is innovative to apply a PGM-free cathode to a DMFC system. If the project is successful, the impact will be high. There is good organization between the team members, who specialize in anodes, cathodes, and systems.
- This project integrates resident institutional expertise in PGM-free cathode catalysis (the University at Buffalo); supported anode catalysis (Kansas State University); electrode fabrication, characterization, and optimization (Carnegie Mellon University); and multiphase mass transfer (University of Kansas). The project includes an early go/no-go decision in fiscal year 2019.

- The team is taking a fairly systematic approach to developing a new fuel cell. One issue is that the motivation of the work does not seem to be clear, and what are suggested as the objectives do not quite match the deliverables. The stated project objectives are to reduce noble catalyst loading and cost, enhance cathode tolerance of methanol poisoning, and decrease methanol crossover. All objectives are using pure methanol, but the deliverable is for 3 M methanol or greater, 250 mW/cm^2, with a 4 mg/cm^2 catalyst loading. Some electrode characterization is also mentioned. Neither the objectives nor the deliverables are wrong, but they do not match. There is no measure of "tolerance to methanol poisoning" or "methanol crossover." There is a catalyst loading target, which is a start. The principal investigator (PI) should consider revising the objectives or the deliverables.
- The approach relies on replacing a Pt catalyst in the DMFC cathode with a PGM-free catalyst, which effectively makes this a PGM-free catalyst development project. From the electrocatalysis point of view, this is another DOE Electrocatalysis Consortium (ElectroCat) effort. The odds for this research to be successful are minimal, given the magnitude of the PGM-free catalyst development challenge clearly demonstrated in ElectroCat projects (several already involving the University of Buffalo group, which is responsible here for PGM-free catalyst development).
    - There are numerous fundamentally flawed assumptions in this project. The first one is the importance of reducing methanol crossover to make DMFCs viable; it is grossly exaggerated by the team. Controlling the feed concentration of methanol can minimize crossover losses (they are of concern only at the highest DMFC voltages anyway). Notwithstanding the ubiquitous lack of PGM-free catalyst stability, the proposed approach is almost certain to result in low PGM-free catalyst activity, which will dominate the overall cell performance. Lower performance than that of state-of-the-art DMFCs is likely going to be the outcome. The importance of methanol poisoning has been entirely misstated in the project. Methanol does not poison Pt at potentials (and temperatures) at DMFC cathode voltages that guarantee reasonable fuel conversion efficiency. The title of the project is misleading. In practice, most, possibly all, DMFC concepts assume operation on "pure methanol" in a water-balanced system, with water required for the anode process, and the same is true of this effort.

Question 2: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Although it is early in the project, key early elements have been met. PGM-free cathode catalysts are to be tolerant to methanol. Growth of vertical supports for vapor deposition is shown. Modeling has begun.
- The project just started. Early accomplishments include development of a fuel cell test platform, initial fabrication of anode and cathode electrodes, and rotating disk electrode (RDE) measurements.
- The team appears to be on track.
- It is too early to assess accomplishments and projects. More performance demonstration is needed at either electrode.
- The part of the effort devoted to the development of vertically aligned conical carbon nanofiber structure is interesting. The catalysis part has been derived largely from the University of Buffalo effort in ElectroCat-funded projects. Methanol tolerance of virtually all oxygen reduction reaction (ORR) PGM-free catalysts is a universal property, so methanol tolerance data are merely restating the obvious. All the data showing poor methanol tolerance of Pt catalysts once methanol concentration is made sufficiently high is also not novel; this has been demonstrated countless times before. Electrode characterization appears premature for as long as a promising PGM-free catalyst for the DMFC cathode has not been developed yet. DMFC performance demonstrated by the team to date has been very low, much below the DMFC state of the art. No advantage of high feed concentration of methanol has been demonstrated so far. It may prove to be an unattainable task, given zero-order kinetics of methanol oxidation at concentrations higher than 0.3 M or so.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- One strength of this team is a well-coordinated mix of specialists focusing on each subsystem of the MEA. The University of Kansas team is apparently responsible for MEA fabrication, which is also a strength. This part should be emphasized, as it will not be trivial to integrate two different electrode technologies.
- There are good collaborations among partners, with clearly defined roles. The University at Buffalo will develop a methanol-tolerant and low-cost PGM-free cathode catalyst. Kansas State University will develop vertically aligned carbon nanofibers with an ultralow-PGM-loaded anode catalyst. Carnegie Mellon University will fabricate and characterize electrodes. The University of Kansas will conduct modeling and system integration.
- The team members seem to be collaborating well. There are no external collaborations yet.
- The project has a good inter-institution collaboration.
- There are four partners. It is not 100% clear what each partner's role is, but there are results from each university partner. A summary of each partner's tasks and capability is suggested.

Question 4: Relevance/potential impact

This project was rated **2.9** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- A highly efficient and low-cost methanol fuel cell is an important component in the fuel cell technology portfolio.
- It is good to see methanol not forgotten as an intermediate green fuel on the way to a hydrogen economy. The project addresses key barriers to DMFCs: PGM loading and activity.
- The project has the objective of improving methanol fuel cell performance and reducing PGM loading for lower cost.
- The project is relevant to the development of DMFCs for portable power. However, none of the proposed pathways toward improvements to DMFC performance and cost promises to be successful.
- DMFCs have their uses, but this technology is not critically important for any sector. A comparison with the advantages of this approach to a reforming methanol fuel cell would be valuable, as would a suggested use case of the technology.

Question 5: Proposed future work

This project was rated **2.9** for effective and logical planning.

- The future plans generally build on past progress and will contribute to overcoming identified barriers. The critical step is to synthesize high-performance, PGM-free ORR catalysts that are stable and durable.
- The team has a good unit of experimental and modeling efforts. The team should include benchmarking on standards.
- If the objectives matched the deliverables, it would be easier to comment more on the relevance of the future work. The team will likely achieve the milestones, but it is unclear whether this will mean that the project has met its objectives.
- There are a couple issues that the PIs need to address quickly. (1) Catalyzing vertically aligned carbon nanofiber has been previously investigated and presents a challenge. The PIs should review the literature more extensively to help their work. (2) A PGM-free catalyst, though more tolerant to CO/methanol poisoning, is not stable under polymer electrolyte membrane fuel cell operating condition. The stability issue needs to be addressed at an early stage.
- There is very little in the proposed future work that stands a good chance of overcoming known barriers of DMFC performance and helping to achieve the very ambitious goals of this project. The plan lacks new ideas, which may not be surprising given the maturity of the DMFC concept. Methanol tolerance aside, the ORR development part does not promise a breakthrough, which depends entirely on the progress in the

development of PGM-free catalysts in ElectroCat projects anyway. The correlation between RDE and fuel cell performance, while interesting, can be (and has been) performed in one of the University of Buffalo ElectroCat projects. The outcome should be just applicable to DMFCs as it is to hydrogen–air systems. Catalyst scale-up is unnecessary as long as an active and stable catalyst is not found. Sputtering a standard PtRu anode catalyst is unlikely to improve anode electrocatalysis, which requires new formulations to overcome an approximately 0.2 V gap between the thermodynamics and practical PtRu-based catalyst systems.

Project strengths:

- This project combines the strengths of PIs at four institutions, and there are clearly defined tasks.
- There is good teaming in integrating some of the previously developed catalyst technology.
- Project strengths include innovative anode and cathode approaches and the use of modeling to help guide critical system operation with pure methanol.
- The proposed milestones are SMART (Specific, Measurable Attainable, Relevant, and Timely).
- There are very few project accomplishments, but the team deserves the benefit of the doubt, given the limited time project has had to prove itself worthy. This project may need major re-scoping to stand any chance of meeting its very difficult targets.

Project weaknesses:

- Since water transport is critical, the team should add a membrane screening component—especially some of the new thin commercial membranes. There will be great pressure to have good cathode performance at ambient pressure, which could limit performance.
- There is uncertainty in developing stable and durable PGM-free catalysts in the presence of crossover methanol. Perhaps the team should wait for real progress in the much larger ElectroCat project.
- The team needs to address the anode catalyzing issue (low risk) and instability of the PGM-free cathode catalyst (high risk) as soon as possible.
- The main weakness of this project is that none of the proposed approaches stands a good chance of meeting the very challenging targets of this project. In particular, this concerns the unwarranted belief that introducing a methanol-tolerant but poorly performing catalyst is going to result in high power densities. At the DMFC current densities required to generate high power, crossover has no impact on DMFC cathode performance. However, a poorly performing PGM-free ORR catalyst, additionally requiring thick electrodes to reach the required catalyst loading, is certain to have a negative impact on the power output relative to any Pt/C catalyst. Overall, the team's biggest weakness appears to be a lack of DMFC expertise.
- It is unclear whether the researchers know why they are doing what they are doing. From a technical perspective, the researchers are fine, but there is no clear reason a direct methanol fuel cell is needed. There are many good reasons why one might want to develop a methanol fuel cell, but without an application, uncertainty will remain around the objectives, which will lead to milestones and targets that relate more to what can be achieved rather than what needs to be achieved. This may not be a problem at this stage, but the answer to the question "why?" needs to be addressed.

Recommendations for additions/deletions to project scope:

- If this project is to have a chance of meeting its goals, the team needs to concentrate on improvements to the anode, rather than ill-conceived reductions in methanol crossover and methanol tolerance and unsubstantiated benefits of the use of high methanol feed concentration (there are some benefits, but overall, they are quite minimal). Improving the anode is easier said than done, but it is the right primary focus of any research aimed at changing DMFC status in a major way. The way the PGM-free catalyst development has been carried out to date appears not to be DMFC-specific and is largely a duplication of the effort in other projects. Deletion should be considered.

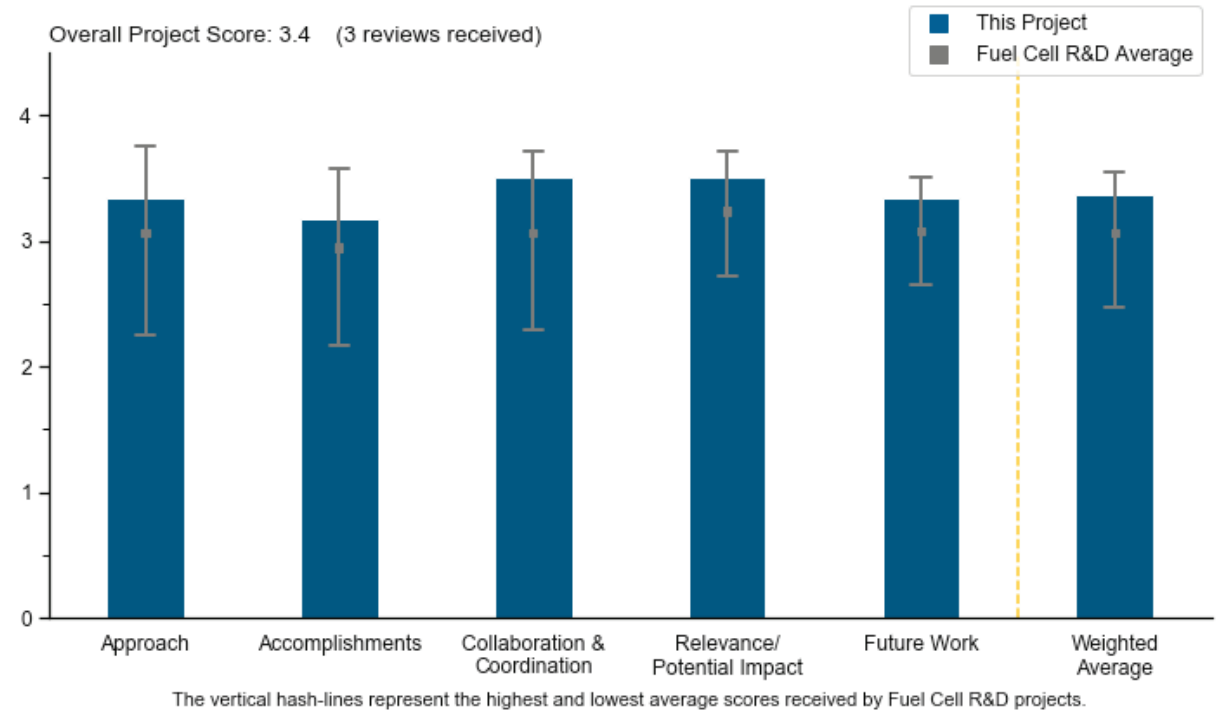
Project #FC-318: Lab Call Fiscal Year 2019: Accessible Platinum-Group-Metal-Free Catalysts and Electrodes: ElectroCat

Jacob Spendelow, Los Alamos National Laboratory

Brief Summary of Project

Platinum group metal (PGM)-free cathodes are much thicker and coarser than PGM cathodes. Model calculations suggest significant effects of electrode thickness on oxygen transport that, in turn, cause losses of several hundred millivolts at high currents. Similarly, H+ resistance for ~5 µm PGM-based cathode catalyst layer causes losses of ~20 millivolts, while a first approximation of the required ~100 µm PGM-free cathode catalyst layer results in losses of several hundred millivolts. The project targets improvements in electrode transport and catalysis of PGM-free catalysts to achieve U.S. Department of Energy target power densities. The project addresses the barriers of durability, cost, and performance.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.3** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This is an interesting approach to improving accessibility through electrode structure design. This is being tried for Pt-based catalysts, but it should be more effective here with thicker PGM-free electrodes.
- The focus on membrane electrode assembly (MEA) testing is appropriate. Robustness testing and operation of the full range of conditions will be important.
- This is an innovative approach to addressing the limitations of electrode design for PGM-free catalysts. The modeling portion will be important to guiding electrode structure development.
- The approach is based on catalyst and electrode structuring to improve transport in PGM-free electrodes. The project has little emphasis on catalyst synthesis.

Question 2: Accomplishments and progress

This project was rated **3.2** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has just started. There is good preliminary data on electrode structuring. There is some uncertainty as to what catalyst is going to be used in the project.
- This project is in the start-up phase, so it has only been a short period of time. The project team has started developing proton channels with encouraging utilization numbers.
- This project is just starting, so there are no accomplishments to date.

Question 3: Collaboration and coordination

This project was rated **3.5** for its engagement with and coordination of project partners and interaction with other entities.

- There is a good assembly of collaborators to reduce project risk. Advanced characterization from the DOE Electrocatalysis Consortium (ElectroCat) will provide good analysis to support hypotheses.
- The project will be integrated with ElectroCat and likely leverage the PGM-free catalyst expertise at Los Alamos National Laboratory.
- The plans for collaboration are good and include collaborating with and taking advantage of ElectroCat's capabilities.

Question 4: Relevance/potential impact

This project was rated **3.5** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The proposed effort is highly relevant to the implementation of PGM-free catalysts in fuel cells. Succeeding in decreasing the mass transport barriers will have a high impact on the ultimate reduction of fuel cell MEA costs.
- The impact will be determined by the performance and durability of the catalyst used in electrode development.
- The project is relevant and related to Fuel Cell Technologies Office efforts in PGM-free catalysts and ElectroCat.

Question 5: Proposed future work

This project was rated **3.3** for effective and logical planning.

- The effort is just beginning. The proposed plan is complete. It is suggested that the project team focus on one "best in class" PGM-free catalyst, versus the implied multiple catalysts.
- The project's proposed future work is directed toward improving the accessibility of PGM-free active sites.
- The proposed future work looks appropriate for the project goals.

Project strengths:

- The project concept should improve the accessibility and performance of thick catalyst layers. The proposed interactions with ElectroCat should leverage the consortium's capabilities in order to accelerate project accomplishments.
- The approach purports to address both proton and oxygen transport and the ability to change electrode architecture to maximize each. Similarly, the approach seems very flexible in being able to change the gas and proton channels.
- The strength of the project is in the principal investigator's experience in evaluating MEA performance and past accomplishments in microstructuring cathodes for PGM catalysts.

Project weaknesses:

- There is no major project weakness in reference to the electrode focus of the project, but there was a lack of consideration of the catalyst.
- No weaknesses are evident at this time.

Recommendations for additions/deletions to project scope:

- *Note: Reviewers did not provide comments in response to this question.*

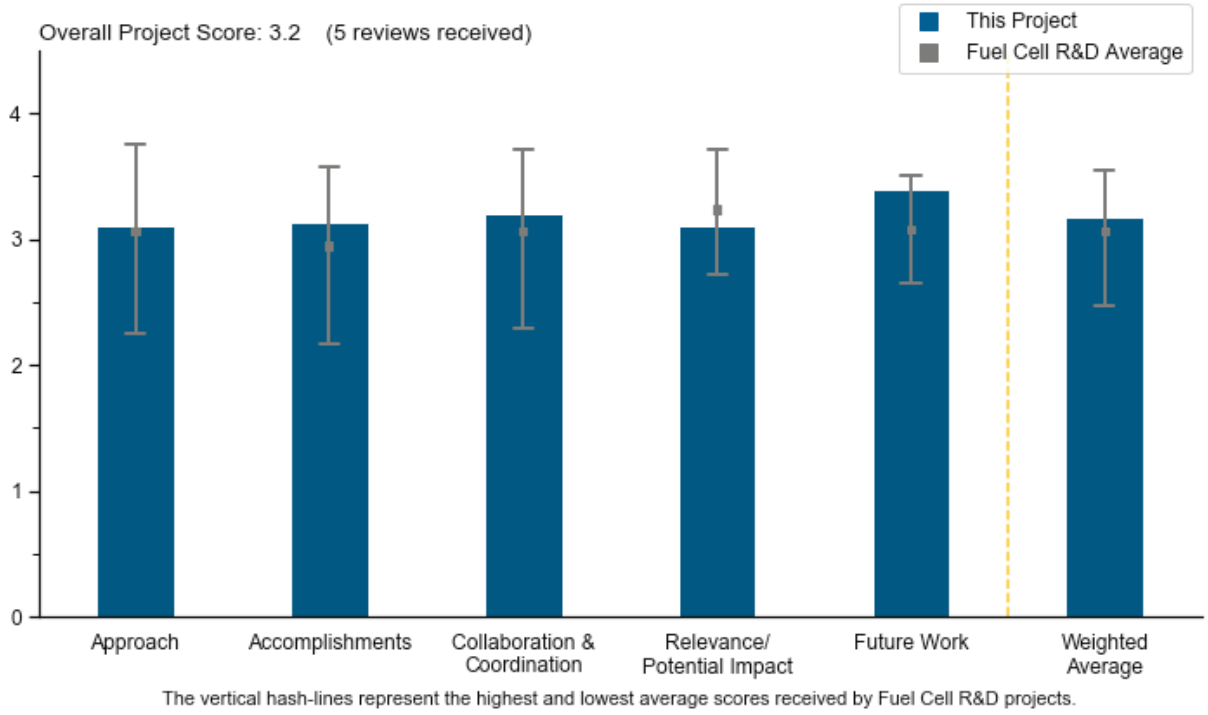
Project #FC-319: Lab Call Fiscal Year 2019: Low-Cost Gas Diffusion Layer Materials and Treatments for Durable High-Performance Polymer Electrolyte Membrane Fuel Cells

Rod Borup, Los Alamos National Laboratory

Brief Summary of Project

The objective of this project is to reduce the cost of gas diffusion layer (GDL) materials and improve GDL performance. To reduce GDL cost, the project will use low-cost fibers, use lower carbonization temperatures to reduce processing costs, and develop low-cost gas phase surface treatments to replace Teflon treatments, thereby lowering manufacturing costs. GDL performance will be enhanced through improved water management. The project will develop super-hydrophobicity coatings, which will prevent water flooding and transport losses, and incorporate hydrophilic pathways separate from hydrophobic domains to provide a pathway for water removal.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.1** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- Reducing cost for GDLs and replacing the Teflon-coating process for GDLs is a good approach. If lower-temperature carbonization materials show improved performance over traditional GDL materials, then it is worth it.
- The proposed approach of concurrently considering materials and processing to reduce the GDL cost while raising the performance appears relevant. Durability aspects should not be forgotten.
- The critical barriers being addressed are cost and performance toward membrane electrode assemblies (MEAs) with a focus on the GDL; the aim is to decrease component cost by introducing new raw materials (i.e., natural fibers) or cheaper processes for post-treatments, while improving performance by better

controlling water- and gas-transport-related properties, thanks to the management of hydrophobicity and hydrophilicity within the structures. It is not clearly mentioned if the strategy is considering both cathode and anode sides. If only the cathode is being addressed, this should be clarified and justified. If both sides are being considered, this should also be clarified, as well as how their differences are being handled. The approach is GDL-based only, and it does not directly address durability. The cost reduction based on cheaper fibers seems clear, but it is not obvious how the treatments proposed to replace Teflon would reduce cost. It should be clarified what the cheaper processes might be. In addition, the cost reduction at the GDL level should be huge to have a strong impact on the stack cost; the expected decrease could be clarified.

- More detail would be appreciated on whether or how this approach is different from or builds on previous work done by partner Oak Ridge National Laboratory's Carbon Fiber Technology Facility. Some questions remain on the ability of low-temperature carbonization treatments to meet the electrical conductivity target (which is of growing relevance, with ever-higher current densities). Teflon is relatively inexpensive, so the gas phase treatment has to at least match the material cost. Gas phase treatments are usually reactive, so stability over time may be a concern.
- This is a new project that is just getting started. Some of the approaches could be more clearly delineated, such as the type of superhydrophobicity that is to be pursued, including the biomimetic approach. Several sources of low-cost fibers are being considered, but the approach to choosing and evaluating these materials could be more systematic, including evaluation of the chemical content (e.g., nitrogen and others).

Question 2: Accomplishments and progress

This project was rated **3.1** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has just begun, so it is hard to judge the accomplishments. A few alternative materials have been synthesized, which is promising.
- It is very early in the project, so it is difficult to fairly assess the accomplishments and progress. The team has had some promising initial results.
- This is a new project, but some progress has already been made, including choosing and characterizing carbon precursors.
- The project has just started some months ago. The first results that were presented appear promising.
- The project is at too early a stage for ranking. The project's activities are going on as planned with the implementation, characterization, and testing of new fibers.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- Los Alamos National Laboratory is known as a leader in GDL and mass transport studies, Oak Ridge National Laboratory is a leader in imaging, and the National Renewable Energy Laboratory has good testing and synthesis capabilities. This is an excellent team to accomplish the work.
- The project team has a very good level of knowledge for investigating this topic. The team's contacts with GDL industry suppliers will be useful for investigating the processability of the most promising solution(s), as with Strategic Analysis, Inc. (SA) for evaluating the projected costs.
- The team's activity-sharing and coordination with three main laboratories is appropriate. A link with industry is planned in the next steps, and industrial products are being used as the baseline, which is consistent with the goal. Consulting manufacturers about upscaling the capability of the developments is planned but should be confirmed.
- There is good vertical collaboration with other Fuel Cell Consortium for Performance and Durability partners. Collaboration outside of the national laboratories would be beneficial.
- The team could benefit from interaction with a GDL supplier and/or original equipment manufacturer (OEM).

Question 4: Relevance/potential impact

This project was rated **3.1** for 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 is of high relevance for achieving the cost, performance, and durability targets of the Fuel Cell Technologies Office, in particular with the current trend of producing MEAs from gas diffusion electrodes.
- Reducing GDL cost, primarily by reducing material cost and processing, is a significant enabler of fuel cell commercialization, and thus this work is relevant and potentially impactful.
- If the project's objectives are reached, the potential impact will be important in terms of MEA cost due to the GDL, but at the moment, the project has only just started. It is too early for any conclusions to be drawn about actual potential impact. However, it will naturally be limited to GDL cost and performance.
- Cost reduction at material (or GDL) level is a good approach only if the new low-cost material matches current GDL performance. If not, then the MEA and eventually the stack cost may be more with lower performance. An analysis of the cost and performance tradeoff will be helpful in directing this research.
- The potential cost reduction impact is high, but it is probably not critical to the success of fuel cell technology. The performance compromises from low-cost materials are a particular concern.

Question 5: Proposed future work

This project was rated **3.4** for effective and logical planning.

- This is a well-laid-out plan, but emphasis should be placed on the relationships between the structure and composition, function, and cost.
- Because of the very recent start, the planned future work corresponds mostly to the project's initial work plan. This is adequate.
- The proposed work is well in line with the project targets. Mechanical characterizations of the developed GDL could be explicitly added.
- The team needs to add a target for thermal conductivity and assess the performance against it. This is important for membrane and electrode durability at high current densities and grade-climb high temperatures. Assuming the beginning-of-life targets are met, the team should develop a plan to address coating stability.
- The proposed future work is good, but the project is aiming at developing low-cost GDL materials, and there are no tasks for estimating cost or understanding cost and performance tradeoffs.

Project strengths:

- The project brings to bear significant resources on an important cost and performance component for fuel cells. There is strong potential for the discovery of new, naturally resourced, low-cost materials and hopefully scientific understanding of material advantages and process optimization.
- The project's technical strength is in its clear approach, with several new materials already identified and tested as fibers for the preparation of carbon papers for their further study as possible GDL components. The team is also a positive, as it is conducting the developments proposed with good skills for manufacturing, characterizing and testing of new components.
- The project's strengths include its team and technical topic. This topic is important but somehow has been neglected for many years by the research community in general.
- The project appears well structured, with a high-level project team.
- The project's partners have strong diagnostic and testing capabilities.

Project weaknesses:

- Details are missing about the technical process and the justification for cost reduction of the new hydrophobic treatment that is planned. It should be clarified whether and how the strategy is addressing both cathode and anode sides. All GDL properties should be considered in the characterizations planned. It is not clear from the presentation how durability is being addressed by the developments planned;

information is also missing on how the selected routes will enable better stability, but this was not a major objective, and the project is only starting.
- There is relatively limited understanding of structure-and-composition–function–cost relationships in this area, and the project could be improved by introducing a framework for understanding these relationships.
- A cost estimation from a third party (SA or another group) is missing; an analysis of lower-cost materials versus a performance tradeoff is also missing.
- The project would benefit from collaboration with a GDL supplier and/or an OEM.
- No weakness is identified at this early stage of the project.

Recommendations for additions/deletions to project scope:

- The team should explain the reasons (based on technical evidence) for expecting cost reduction with the new hydrophobic treatments. More details should be given about how new, lower-cost component structures should lead to improved properties and/or performance. The team should clarify whether the strategy is addressing both cathode and anode sides, and how. The team should also clarify in more detail how durability is being addressed, including whether there is a possibility to maintain or improve water or gas transport properties and their stability, as well as what the risk is regarding the hydrophilicity/phobicity stability of the new materials.
- It is recommended that the team add a thermal conductivity target, something necessary to mitigate potential high-temperature, high-current-density MEA degradation. The researchers should pursue coating stability testing if they successfully identify a candidate that meets initial life targets.
- The team should add a task for cost analysis based on the final performance of the developed GDL. Otherwise, this whole activity is meaningless.
- The project could be improved by introducing a systematic framework for understanding, along with complete characterization.

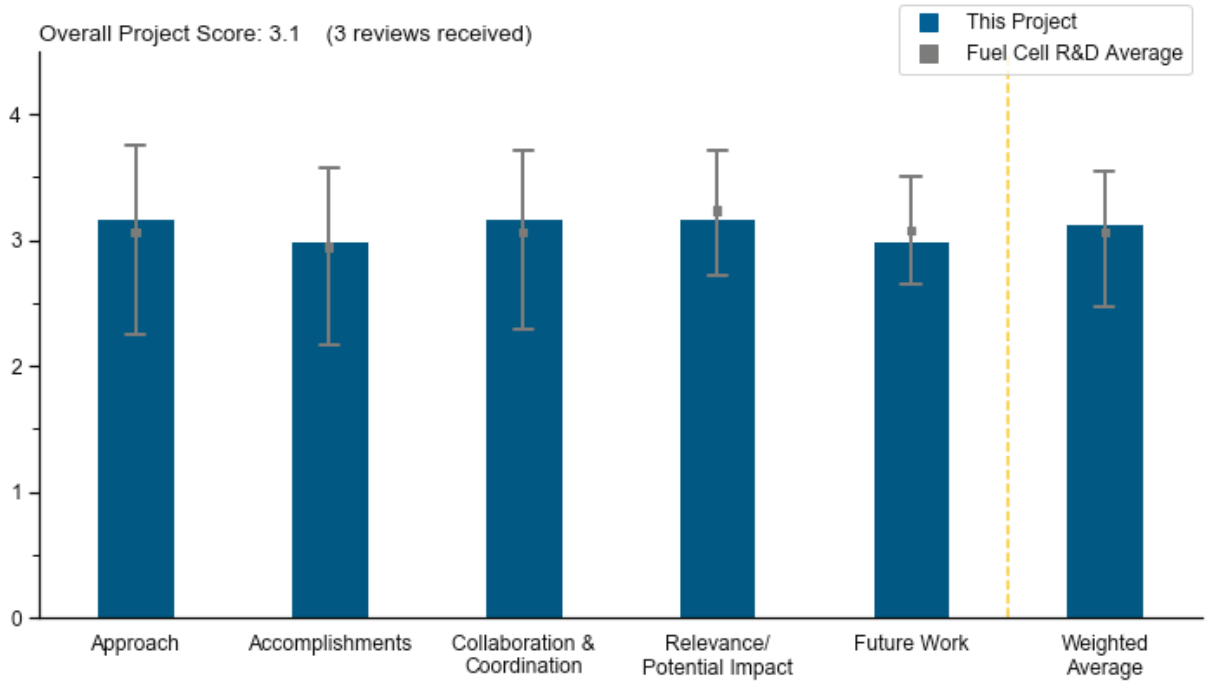
Project #FC-320: Lab Call Fiscal Year 2019: Electrode Ionomers for High-Temperature Fuel Cells

Michael Hibbs, Sandia National Laboratories

Brief Summary of Project

Sandia National Laboratories (SNL) seek to synthesize durable ionomers and demonstrate their use in fuel cells that can operate at temperatures of 200°C–300°C. These ionomers could reduce costs of future fuel cell technologies by enabling operation at high temperatures without humidification and at low-platinum-group-metal (low-PGM) loading. The project team plans to achieve >500 mW/cm^2 peak power density under hydrogen/air conditions, total PGM loading of <0.125 mg PGM/cm^2, and performance decrease of <5% after 1,000 hours of operation at 200°C.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Fuel Cell R&D projects.

Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.2** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This very early research and development (R&D) work is aimed at a relatively wide-open area, which is electrode function in intermediate-temperature fuel cells. It is forward-thinking to integrate the promise of phosphoric acid membranes in covalently functionalized polymers and their evaluation in electrodes, and the work addresses industry concerns about the next phase of fuel cell R&D.
- Phosphoric acid fuel cells (PAFCs) have had many challenges over the years. This approach offers a new perspective over the traditional or gel electrode (polybenzimidazole [PBI] or phosphoric acid [PA]) strategies. While the project is just starting, the team has some reasonable preliminary data that support the benefits of this type of membrane.
- The critical barriers being considered are cost, performance, and durability, focusing on the electrodes and high-temperature polymer electrolyte membrane (HT-PEM) operation only. The specific targets proposed

are low compared to low-temperature (LT)-PEM cases, but the targets are also lower than the state-of-the-art (SOA) HT-PEM cases with 500 W/cm² peak power and 5% performance loss after 1,000 hours of aging. Only the PGM loading that is being targeted is in the same range as that of LT-PEMs, with 0.125 mg/cm², which seems far too challenging for HT-PEM operation. The work is focused on the ionomer; hence the project team should consider linking with other projects that are focused on catalyst and membrane electrode assembly (MEA) development, at least for final implementation and assessment. The approach was initiated because of the general advantages of operating a fuel cell system at higher temperature, with lower costs thanks to humidification removal and easier heat management. However, at the highly innovative level of this work, it would be important to also mention the risks of getting performance at the targeted temperature that is too low or poor start-up management at ambient temperature. The approach aims at replacing the proton conductors within the catalyst layers with phosphonic groups that are less sensitive to the leaching issue. In the activities presented, the results are compared to previous innovative developments but not to the SOA components of the HT-PEM fuel cells, which should be part of the approach.

Question 2: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project is in very early stages, and while little tangible progress has been made, the team has started well and appears to be on the right track. Leveraging previous polymer chemistry probably gives the best chance for success.
- The project has just begun, and therefore, expectations for progress are minimal. However, the preliminary data look reasonable at this stage and show good progress toward the ultimate DOE Hydrogen and Fuel Cells Program goals.
- To assess the project's accomplishments toward DOE objectives, the results of the fuel cell tests that are planned for later will be needed. Regarding the project objectives, the progress is fair. The synthesis of at least one new material allowed for a comparative test to be performed with a previous material that was developed by Los Alamos National Laboratory (LANL) for HT-PEM operation as well. This material presents specific behaviors with better performance at 200°C compared to 160°C, but the performance is very low. More materials need to be prepared and tested to properly assess progress.

Question 3: Collaboration and coordination

This project was rated **3.2** for its engagement with and coordination of project partners and interaction with other entities.

- It is a good idea to integrate directly with LANL and indirectly with Rensselaer Polytechnic Institute to leverage other polymer chemistries besides those poly(phenylene) polymers available at SNL. LANL will be a great partner for evaluating polymers in a fuel cell.
- Both SNL and LANL are well regarded; however, it would be best to have an academic or industrial partner with expertise in PAFCs. It is acknowledged that this is a lab call project, but engaging a researcher with PAFC capabilities at some point would go a long way in validating the concept.
- Two national laboratories are involved; this seems relevant for the actions planned. A link should be envisioned with other projects focused on other MEA components, at least for implementation and/or assessment purposes, including protocols. It will also be necessary to plan links with industry for further development and for relevance in regard to scalability.

Question 4: Relevance/potential impact

This project was rated **3.2** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- Intermediate-temperature fuel cells potentially have much higher efficiency than PEM fuel cells, and their research is very promising. Few other laboratories are looking at this type of chemistry, and this project is potentially impactful far beyond its timeline. For such exploratory and high-risk work, this project is well designed.
- Fuel cell operation between 100°C–200°C could be very advantageous. If successful, this project has the potential to advance the phosphoric acid electrolyte field.
- At the system level, it should be clarified whether the gain that is expected thanks to temperature increase (>200°C) would still be valuable if performance loss is too high compared to conventional components. However, this analysis is hard to do at this stage in the project because more progress is needed. At the material level, evaluating the potential impact would require the team to get more data in actual fuel cell conditions.

Question 5: Proposed future work

This project was rated **3.0** for effective and logical planning.

- The polymerization of new polymers with this functionality and testing them in an MEA is good initial work. Having several synthetic schemes reduces the risk of failure for synthesizing new polymers.
- The work proposed for both membrane and electrode–ionomer synthesis looks to be reasonable and well designed.
- The project's future work is still very open regarding the formulations that are to be synthetized; this is maybe not clear enough when considering very short-term objectives (sooner than June 2019). For longer-term plans, low-PGM durability is mentioned; this seems much too ambitious a goal and is maybe not so relevant. Performance with the usual PGM loadings should be strongly improved first. The possibility of bridging this new route with old PA-doped, PBI materials could be considered, as well as other baseline materials for membranes or catalysts to fill the gap with SOA for initial performance.

Project strengths:

- This project has the potential to advance the PAFC field. The idea of strongly binding the PA to a quaternary amine could solve one of the main issues of PAFCs: PA loss at high temperatures. The fully aromatic structures look to be stable in the preliminary tests that have been run to date.
- The main strength of this project is the initial idea of proposing a new molecule that is expected to compete against the weaknesses of previous materials due to acid leaching. The type of material proposed presents interesting conductivity properties and original behavior during fuel cell testing. The team has identified possible obstacles and proposed means for mitigation.
- The project's strengths include its broad polymer synthesis and its good leveraging of collaborators. This project is doing potentially high-impact work in an important area.

Project weaknesses:

- It is unclear how the team intends to overcome some of the traditional issues with PA or PBI/PA fuel cells. For example, there are issues with start-up/shut-down, liquid water, PA catalyst poisoning, etc. Even with the fully aromatic backbone polymer, there are still reservations about long-term operation at elevated temperatures (~150°C–200°C).
- The performance level is low. The gap with conventional HT-PEM fuel cells should be considered and discussed, major causes should be identified, and solutions should be proposed. No hydrogen/air data were presented. The stability target does not seem consistent with the current status of the project and may be too ambitious.

Recommendations for additions/deletions to project scope:

- Comparison and positioning should be done regarding the SOA components of HT-PEM fuel cells. The team should discuss and explain the justification for the performance difference with previously developed PA-ADAPP and PPFS when the temperature is increased from 160°C to 200°C, particularly when considering the limited differences seen in the conductivity data. Selecting one route and improving the initial performance with conventional PGM-loading electrodes should be prioritized.
- It would be helpful to include some start-up/shut-down and durability testing data, as well as a comparison with traditional PBI/PA systems. Even a comparison with literature results would help to show the differentiation of this approach in those areas.

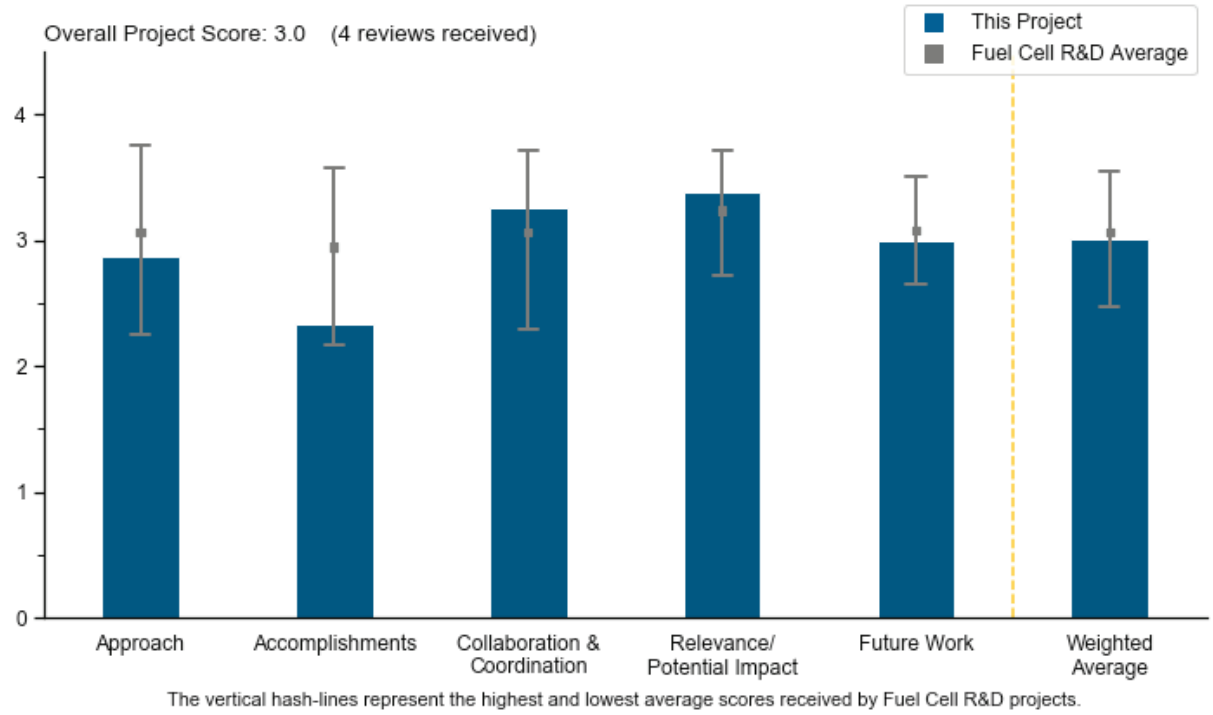
Project #FC-321: Lab Call Fiscal Year 2019: Solid Phase Processing for Reduced Cost and Improved Efficiency of Bipolar Plates

Ken Ross, Pacific Northwest National Laboratory

Brief Summary of Project

The goal of this project is to develop and demonstrate methods to fabricate bipolar plates (BPPs) to meet or surpass all 2020 U.S. Department of Energy (DOE) technical targets for BPPs, including a cost target of less than $\$3.00/kW_{net}$. The fabrication concepts with the greatest potential to meet the technical targets will be identified and ranked using best practices of product design and development. Among the manufacturing approaches to be evaluated is high-velocity cold spray coating, which is orders of magnitude faster than currently used vapor deposition techniques. Top-ranked concepts will undergo process development, and coupons will be tested to validate performance. The project will fabricate and test full-scale prototypes and conduct economic analysis to validate achievement of cost and performance targets.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **2.9** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- The overall approach appears to be good since the team is utilizing cost models to help screen alternative concepts, which should help ensure that the project develops a BPP that meets both the challenging technical targets and the cost targets.
- The project has identified pathways to identify the problems with achieving BPP targets and the solutions to those problems. The project's approach does not explain how the team will fabricate ribbed BPPs. The principal investigator said the method was proprietary, which makes it very difficult to evaluate.
- It would be beneficial to see more detail on the approach, but the general outline appears sound.

- It is unclear from this poster what is driving the cost of BPPs; it could be raw materials costs or processing. Knowing these details would help with evaluating whether the approach is well designed to meet the 2020 DOE targets. It would also be helpful if the slides presented how current, state-of-the-art BPP technical properties compare to technical targets; this information would help with understanding where improvements need to be made or what would be the most impactful. It is stated that cold spraying should be lower in cost than chemical vapor deposition (CVD). It would be beneficial to include some generic cost comparisons, even if not for the materials used here, to demonstrate that the approach used in this project should result in cost savings. Since Strategic Analysis, Inc. (SA) has participated in the project, it seems it would be possible to attain these data. However, since a breakdown of BPP manufacturing costs is not provided, it is not possible to determine whether improving production rates by "orders of magnitude" would reduce costs by orders of magnitude, 10%, or 1%. The presenter mentioned that cold spraying should result in fewer pinholes than CVD, but it is not clear why this would be the case or if there are any data to support this.

Question 2: Accomplishments and progress

This project was rated **2.3** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- This ranking tool seems like it could be helpful, but it also seems like this analysis should have been done in the proposal stage to identify that the proposed strategy had merit. While the specifics of the fabrication categories may be proprietary, some generic data should be presentable, such as that Category 1 is expected to reduce cost by XX% or improve conductivity by YY%. The lack of any details makes it hard to know how much of an impact any of the concepts might have on cost or technical performance. Based on the milestone table, it appears that coupons should have been generated, but no data were presented, and the presenter stated these had not yet been prepared, so it is assumed that this milestone was not met on time.
- No details on results to date are provided, so it is hard to assess. It is also not clear how much work has been done since the only indication is that the project started in October 2018. The work to date appears to be the completion of a concept down-selection process, but the details are sparse.
- It is early in the project, so the accomplishments and progress are not easy to assess.
- This is a new project; the activities are just initiating.

Question 3: Collaboration and coordination

This project was rated **3.3** for its engagement with and coordination of project partners and interaction with other entities.

- The project has identified important coating technology and a company with experience in fabricating coated BPPs. SA provides excellent guidance on cost.
- The project's collaboration and coordination is positive; the partner has experience in addressing BPP goals. The project could benefit from engagement with an original equipment manufacturer (OEM).
- The team has good partners with the right experience and interest in this project. It is too early to comment on the actual collaboration.
- The inclusion of industrial partners is beneficial for allowing researchers to better understand what processes may be feasible in a manufacturing setting. It seems that data from SA has not been used as well as it could be.

Question 4: Relevance/potential impact

This project was rated **3.4** for supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- A high-rate fabrication method for producing stable BPPs is needed, and this approach has the potential to meet that need.
- BPP cost and durability are significant enablers for the DOE Hydrogen and Fuel Cells Program.

- This project is definitely focused on a technology that still needs to be developed to meet the automotive targets.
- Given that current BPPs do not meet cost targets, it seems innovation is needed in this area. A higher score would have been given if it had been clear how well current BPP technologies address cost and performance targets.

Question 5: Proposed future work

This project was rated **3.0** for effective and logical planning.

- The proposed future work is light on details, but the approach seems sound. For Al to be viable, a perfect pore-free coating with good adhesion will be needed. The team should clarify what the surface pretreatment is, as well as how likely cold spray is to meet formability and pore-free requirements.
- The proposed development of three selected fabrication concepts provides multiple pathways to success. The team will need to demonstrate a method for forming BPPs without developing punctures in the plates.
- The go/no-go is missing one key metric, which is measuring the contact resistance between the BPPs and a typical gas diffusion layer (GDL). This interfacial resistance can be large with some materials and carbon GDLs, and it should be assessed early in the project to rule out surface coatings that have high contact resistance.
- The work seems to be following the project plan, but it is behind schedule. It is not clear from the slides if the project is on track to meet the go/no-go.

Project strengths:

- This is a good approach with a low-cost process and cost focus. The project has a good team with the right experience, capabilities, and background.
- Strengths include the potentially high throughput of cold-spray processing.
- The new approach to coating the BPP may give high-rate deposition.
- Given that the project was only six months old when the slides were due, it is not surprising that only initial results were available. Cold spraying is a novel approach to BPPs.

Project weaknesses:

- The team claims to have a proprietary method for forming BPPs that eliminates stamping and the high cost of processing equipment; however, the researchers will not discuss the method, so it is very difficult to evaluate. If DOE wants an evaluation of this proprietary forming method, they will have to find an alternative evaluation method.
- The presentation was light on details, so it is difficult to gauge. The team needs to clarify what the surface pretreatment is and how likely cold spray is to meet formability and defect-free requirements.
- The lack of any details presented in this poster makes it difficult to assess this project. Estimates of the potential of cold spraying were not presented.
- The technical milestones do not include all of the key technical targets.

Recommendations for additions/deletions to project scope:

- The project scope is acceptable.
- The team should measure all of the metrics established by DOE (i.e., the list shown on slide 3). In addition, the team should measure the contact resistance of BPPs and GDLs and compare that to the baseline BPP.
- The team should consider interacting with a stack developer or OEM.
- The team should discuss the forming method so it can be evaluated.

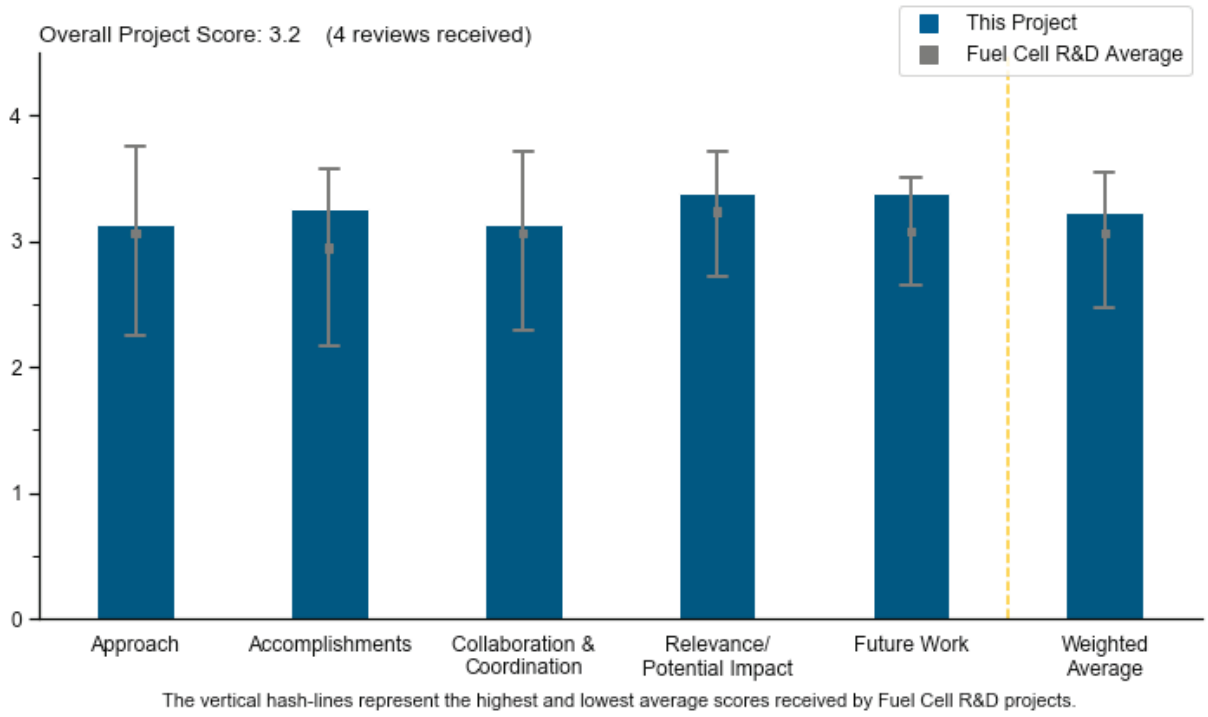
Project #FC-322: Lab Call Fiscal Year 2019: Polymer Electrolyte Fuel Cell Electrode Structures with Encased Catalysts to Eliminate Ionomer Adsorption on Catalytic Sites

Deborah Myers, Argonne National Laboratory

Brief Summary of Project

This project intends to address the lack of sufficient surface power density at rated power for an automotive polymer electrolyte fuel cell (PEFC) stack and the lack of sufficient performance durability due to direct contact between the proton-conducting and electron-conducting components of the PEFC cathode catalyst layer with the platinum catalyst surface. The approach is to protect Pt and Pt_xCo_{1-x} nanoparticle cathode catalysts from direct contact with the proton-conducting and electron-conducting phases while also maintaining sufficient oxygen, proton, and electron transport to the catalytic sites. This is achieved by encasing Pt and Pt alloy catalyst particles in the cages and pores of zeolites. The new catalyst powders will be synthesized and characterized, and membrane electrode assemblies (MEAs) using the catalysts will be fabricated and tested.

Project Scoring



Note: This is a new project in 2019. Reviewers were given the option not to evaluate Accomplishments. In such instances, the other criteria were re-weighted to total 100% (see the Introduction section for details).

Question 1: Approach to performing the work

This project was rated **3.1** for identifying and addressing barriers, project design, feasibility, and integration with other relevant efforts.

- This is an excellent, but very challenging, approach. If successful, it will boost the performance (areal power density) and durability too, as Pt nanoparticles are not in contact with the ionomer directly.
- The project has a logical approach to address key issues with low-platinum-group-metal (low-PGM) electrodes. The work started with some prior Argonne National Laboratory (ANL) results and learnings.
- The approach is very logical and focused. The project has spoken to and planned for key challenges.

- There are many questions or uncertainties that would have to work out for this approach to work. First, the zeolites have insufficient conductivity, and placing the catalyst within an insulating cage is a dubious start. The encaged catalyst's having electrical and ionic connectivity through the catalyst layer will be very difficult to achieve. Even for rotating disk electrode testing, the team needed to add carbon for electrical conductivity. Finally, if the team is able to achieve all of that, the question as to whether caging the catalyst will lead to decreased dissolution is completely open.

Question 2: Accomplishments and progress

This project was rated **3.3** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- It is impressive to see that 62 Pt catalysts were synthesized. However, getting Pt or PtCo in zeolite cages will be very challenging. Really good progress has been made so far, considering the project has just started.
- At this point, the researchers are just getting started; they have been able to make a number of catalysts, although they are quite far from demonstrating sufficient performance.
- It is six months into the project. Although struggling to get reasonable oxygen reduction reaction activity, the team is recognizing the problem and revising work accordingly.
- It is early, but there are materials in hand already. Many samples have been made.

Question 3: Collaboration and coordination

This project was rated **3.1** for its engagement with and coordination of project partners and interaction with other entities.

- This is a relatively modest project that should not need multiple collaborators. Having said that, the other members of the Fuel Cell Consortium for Performance and Durability are doing what they do best: K.C. Neyerlin (National Renewable Energy Laboratory [NREL]) with MEA studies and Karren More (Oak Ridge National Laboratory [ORNL]) with microscopy.
- The team members cover all of the required tasks.
- This is a small but focused team.
- Between ANL, NREL, and ORNL, this is a good team, but communication with other or outside teams is not clear.

Question 4: Relevance/potential impact

This project was rated **3.4** for 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 is a critical and well-known issue; many different approaches, such as various types of ionomer and ionic liquid, have been tried without much success. If this works, it will be the ultimate solution to the problem.
- The impact could be high in reducing Pt dissolution and migration away from the catalyst. The key concerns would be processing cost and quality control.
- This project may improve the performance and durability of low-PGM electrodes. If successful, the cost reduction could be substantial.
- This is hard to judge without knowing if these can ever be made economically. They have so many challenges that it is hard to believe that they will be able to meet them all, certainly not in this first project.

Question 5: Proposed future work

This project was rated **3.4** for effective and logical planning.

- The team's focus on electrical continuity and cycling stability is perfectly appropriate. The cycling stability is the entire *raison d'être* and should be shown first.
- The work is well laid out. More needs to be discussed about how electrical conductivity might be maintained. Carbon inclusion is laid out, but the seemingly random inclusion could still lead to significant metal deactivation. This seems to be a key risk in the project. Secondly, the impact of water management should be discussed.
- All proposed future work is targeted toward achieving the project goals. The most challenging thing will be to avoid Pt deposition outside or on the zeolite cage.
- There is a clear plan.

Project strengths:

- It is a unique idea. If, for some reason, caging a catalyst will stop it from dissolving, this will have broad applications for fuel cells, direct methanol fuel cells, and electrolyzers.
- The project has a novel idea that will offer significant learning and chance for improvement. The goals are focused.
- The approach to solve the issue is a major strength, along with the team players.
- This is a small and focused project. The challengers are understood.

Project weaknesses:

- Zeolite has a proton conductivity of around 0.1 S/cm, which may hamper the performance. Also, carbon needs to be integrated into the catalyst layer to provide electron conductivity. Additionally, Pt needs to be inside the cage, so it is a very complex system to make work. This is not really a weakness but a challenge.
- The synthesis team is not as experienced as other groups. This is a topic that would be better executed by industry.
- There is a risk with the carbon conductivity and water management. There is concern over the cost of the process.
- There are too many leaps of faith. It must be believed that placing the catalyst in a zeolite will stop dissolution. Then it must be believed that once it is in the cage, it will have heady access to ions, electrons, and reactants. Finally, it must be believed that it will be economical to manufacture. Any one of these is a fatal flaw.

Recommendations for additions/deletions to project scope:

- The team should down-select a few catalyst candidates and work with those only.
- Although the chances of these catalysts do not seem favorable, the go/no-go performance metric seems to be incorrect. The important thing to show would be the decrease in catalyst dissolution while being able to communicate with the catalyst. If the researchers can show this, they should be given the chance to try more effective catalyst blends.
- Considering the developmental stage of the project and similar development that has already been done in the industry, this stream of work will not add much value.