Fuel Cell Technologies – 2021

Subprogram Overview

INTRODUCTION

Fuel cells convert the chemical energy of hydrogen or other fuels into electricity and are a key element of a broad portfolio for building an affordable, resilient, and clean energy economy. The Fuel Cell Technologies (FCT) subprogram applies innovative research, development, and demonstration (RD&D), with the main goal of developing a diverse portfolio of low-cost, durable, and efficient fuel cells that are competitive with incumbent and emerging technologies across applications. The subprogram seeks a balanced, comprehensive approach to fuel cells for the near, mid, and long term.

The subprogram's RD&D strategy is target-driven, with technical targets developed for different fuel cell technologies, specifically considering end-use requirements. In this holistic approach, the subprogram develops targets based on the ultimate lifecycle cost of using fuel cell systems in diverse applications. While the subprogram has previously developed comprehensive technical targets in areas such as light-duty vehicles (LDVs), it continues to develop and refine additional targets for emerging and high-impact applications. These include heavy- and medium-duty vehicles (HDVs and MDVs), maritime applications, stationary power generation (primary and back-up), and reversible fuel cells for energy storage. The specific focus on HDVs—which have more stringent durability requirements than LDVs—will also offer transferrable benefits for light-duty, medium-duty, and stationary power fuel cell applications.

The subprogram strategically addresses crosscutting challenges for fuel cell development through focus on materials and components (especially low-platinum-group-metal [low-PGM] and PGM-free catalysts and electrodes); systems and manufacturing (design, standardization, improved supply chains); and analysis and modeling.

GOALS

The FCT subprogram's goal is to develop fuel cell technologies that are competitive with incumbent and emerging technologies across diverse applications.

Specific objectives of the subprogram include:

- Developing fuel cell systems with an emphasis on systems that are highly durable, efficient, and low-cost, while meeting the needs and constraints of varied heavy-duty transportation applications for the near to mid-term.
- Developing new materials and components for next-generation fuel cell technologies for transportation, distributed power, and long-duration grid-scale energy storage, emphasizing innovative mid- to long-term approaches.

KEY MILESTONES

By 2030:

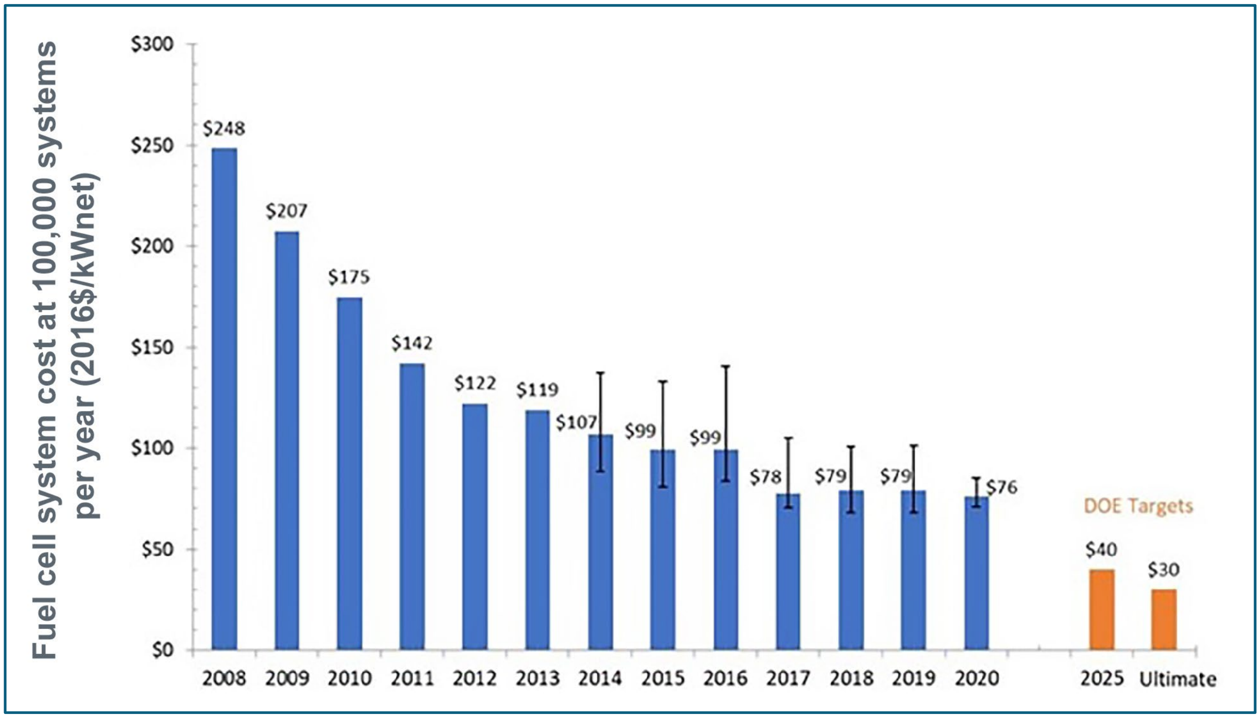
- Develop a 68% (ultimately 72%) peak-efficient, direct hydrogen fuel cell power system for heavy-duty trucks that can achieve durability of 25,000 hours (ultimately 30,000 hours) and be mass-produced at a cost of $80/kW (ultimately $60/kW).
- Develop medium-scale distributed generation fuel cell power systems (100 kW–3 MW) that achieve 65% electrical efficiency and 80,000-hour durability at a cost of $1,000/kW.
- Develop reversible fuel cells for energy storage applications that can achieve 40,000-hour durability at a cost of $1,800/kW ($0.20/kWh levelized cost of storage).

Fiscal Year 2021 Technology Status and Accomplishments

One of the most important metrics used to guide the FCT subprogram's RD&D efforts is the projected high-volume manufacturing cost for automotive fuel cells, which is tracked on an annual basis (see the chart below). The subprogram is targeting a cost reduction to $40/kW by 2025. Long-term competitiveness with alternative powertrains is expected to require further cost reduction to $30/kW, which represents the subprogram's ultimate cost target.

The 2020 estimated cost of an 80 kW_{net} automotive polymer electrolyte membrane fuel cell (PEMFC) system (based on next-generation laboratory technology and operating on direct hydrogen) is projected to be $76/$kW_{net}$ when manufactured at 100,000 units/year and adjusted to meet 8,000 hours of durability.[1] These cost reductions were achieved through factors such as reduced Pt catalyst loading (30% since 2008); increased cell power density (>70% since 2008), which allows for smaller stacks; optimized balance of plant (BOP) components and system design; and innovative manufacturing processes for BOP and stack components.

70% Cost Reduction from 2008 to 2020



Cost status (2008–2020) for 80 kW_{net} automotive PEMFC system (at 100,000 units/year) vs. targets (2025 and ultimate)

Fuel cell electric buses (FCEBs) continue to demonstrate high levels of durability. Fuel cell bus durability was determined to be 17,000 hours with less than 20% degradation (8,500 hours with less than 10% degradation), approaching the U.S. Department of Energy (DOE)/U.S. Department of Transportation interim fuel cell bus target of 18,000 hours (with less than 20% degradation). This status is based on real-world fuel cell bus data collected between 2011 and 2017, and newer FCEB power plants currently undergoing evaluation are expected to be more durable. In fiscal year (FY) 2021, 12 FCEBs surpassed the ultimate target of 25,000 hours, with one FCEB reaching over 32,000 hours. Four of these fuel cell power plants have been retired from service because they no longer

[1] On-road technology is not warrantied for the ultimate lifetime target of 150,000 miles (8,000 hours), suggesting a shortcoming in stack durability that would likely be addressed by replacing the stack. To account for this in the cost of a fuel cell system, a single stack replacement is included in the durability-adjusted cost.

provided the power necessary to meet service requirements, including for the vehicle that operated for over 32,000 hours. The remaining fuel cell power plants have continued service.[2]

In FY 2021, the subprogram also continued its efforts on cost analyses of direct hydrogen fuel cell systems suitable for MDVs and HDVs, aiming toward achieving the 2030 target of $80/kWnet and the ultimate target of $60/kWnet. Recent analysis estimates the 2021 cost status for medium-duty systems at $170/kW and heavy-duty fuel cell systems at $185/kW at volumes of 100,000 units/year.[3]

SUBPROGRAM-LEVEL ACCOMPLISHMENTS

Reversible Fuel Cells for Energy Storage

Performance, cost, and durability targets were developed and published—based on extensive stakeholder engagement and industry input—for unitized reversible fuel cells for electric energy storage applications. These include targets for both low- and high-temperature technologies at both the cell/stack and system levels with the same stack operating in both fuel cell and electrolyzer modes.[4]

Key 2030 system-level reversible fuel cell targets established include the following:

- Uninstalled capital cost of $1,800/kW (on a power basis) and $250 kWh (on an energy capacity basis)
- Roundtrip efficiency of 60% (high temperature) and 40% (low temperature)
- 40,000-hour durability with <10% degradation at end of life
- Levelized cost of storage of $0.20/kWh

Ultimately, innovative RD&D will be needed to improve roundtrip efficiency and durability, decrease levelized cost of electricity/storage to <$0.10/kWh, and meet long-term system capital cost targets by power and energy capacity of less than $1,300/kW and $150/kWh, respectively.

L'Innovator Cooperative Research and Development Agreement (CRADA)

The L'Innovator CRADA was fully executed between industry partner company Advent Technologies, Inc., and three national laboratories: the National Renewable Energy Laboratory, Los Alamos National Laboratory, and Brookhaven National Laboratory. This effort aims to move fuel cell technologies closer to commercialization by bundling state-of-the-art lab intellectual property, lab manufacturing expertise, and the industrial partner's market experience.

PROJECT-LEVEL ACCOMPLISHMENTS

Million Mile Fuel Cell Truck Consortium (M2FCT)

The mission of the M2FCT consortium is to advance PEMFC efficiency and durability and to lower PEMFC cost, thereby enabling PEMFC commercialization for heavy-duty vehicle applications. A "team-of-teams" approach is being used, featuring teams in analysis, durability, integration, and materials development. The objective for fuel cell development efforts under this consortium combines efficiency, durability, power density, and (implicitly) cost in a single metric: 2.5 kW/g_{PGM} power (1.07 A/cm^2 current density at 0.7 V) after 25,000 hour-equivalent accelerated stress tests (ASTs).

An Accelerated Stress Test Working Group (ASTWG) was formed to recommend test protocols and performance targets for fuel cells in HDV applications. M2FCT has developed a membrane electrode assembly (MEA) durability

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[2] DOE, "On-Road Transit Bus Fuel Cell Stack Durability," DOE Hydrogen and Fuel Cell Technologies Program Record #20008, September 15, 2020, https://www.hydrogen.energy.gov/pdfs/20008-fuel-cell-bus-durability.pdf.

[3] Brian James, "2021 DOE Hydrogen and Fuel Cells Program Review Presentation: Fuel Cell Systems Analysis," prepared for DOE by Strategic Analysis, Inc., June 9, 2021, https://www.hydrogen.energy.gov/pdfs/review21/fc163_james_2021_o.pdf.

[4] DOE, "Reversible Fuel Cell Targets," DOE Hydrogen and Fuel Cells Program Record #20001, April 16, 2020, https://www.hydrogen.energy.gov/pdfs/20001-reversible-fuel-cell-targets.pdf.

AST, incorporating relevant degradation mechanisms for catalyst, support, electrodes, and membrane in a single AST. The AST is currently being validated in coordination with the ASTWG.

ElectroCat 2.0

The mission of the ElectroCat (Electrocatalysis) consortium is to develop durable PGM-free catalysts for PEMFCs and for low-temperature electrolyzers as low-cost alternatives to PGM catalysts, addressing critical mineral supply challenges. In FY 2021, the activity of PGM-free catalysts was improved more than twofold compared to the 2016 baseline (16 mA/cm^2). Performance of 38 mA/cm^2 exceeded the FY 2021 catalyst activity target (35 mA/cm^2). These improvements were achieved with a NH_4Cl-treated "single-zone" Fe-C-N catalyst. A total of 193 unique catalysts were synthesized, with 30% enhancement in oxygen reduction reaction (ORR) activity performance improvement over the highest ORR activity reported in FY 2020.

NEW PROJECT SELECTIONS

Fuel Cell Systems for HDVs

- Cummins Inc. – Polymer Electrolyte Membrane Fuel Cell System for Heavy-Duty Applications
- Plug Power Inc. – Domestically Manufactured Fuel Cells for Heavy-Duty Applications

Membrane Development for HDVs

- Nikola Motor Company – Advanced Membranes for Heavy-Duty Fuel Cell Trucks
- 3M Company – Extending Perfluorosulfonic Acid Membrane Durability through Enhanced Ionomer Backbone Stability
- The Lubrizol Corporation – Additive Functionalized Polymers for Extended Heavy-Duty Polymer Electrolyte Membrane Lifetimes
- University of Tennessee, Knoxville – A Systematic Approach to Developing Durable Conductive Membranes for Operation at 120°C

Anion Exchange Membrane (AEM) Fuel Cell Development

- Los Alamos National Laboratory – Advanced Anion Exchange Membrane Fuel Cells through Material Innovation
- National Renewable Energy Laboratory – Advanced Ionomers and Membrane Electrode Assemblies for Alkaline Membrane Fuel Cells

Consortia

- ElectroCat 2.0 – A relaunch of the ElectroCat consortium, co-led by Argonne National Laboratory and Los Alamos National Laboratory, to develop durable PGM-free catalysts for PEMFCs as low-cost alternatives to PGM catalysts, addressing critical mineral supply challenges. ElectroCat2.0 expands the scope to include PGM-free catalysts for low-temperature electrolyzers.
- M2FCT – The mission of the M2FCT consortium is to advance PEMFC efficiency and durability and to lower PEMFC cost, thereby enabling PEMFC commercialization for HDV applications.

FY 2021 FUNDING OPPORTUNITY ANNOUNCEMENT SELECTIONS

Fuel Cell RD&D for Heavy-Duty Applications – Low-Cost, Durable Bipolar Plates

- Plug Power, Inc. – Fully Unitized Fuel Cell Manufactured by a Continuous Process
- Neograf Solutions, LLC – Development of Low-Cost, Thin Flexible Graphite Bipolar Plates for Heavy-Duty Fuel Cell Applications
- General Motors, LLC – Fuel Cell Bipolar Plate Technology Development for Heavy-Duty Applications

- TreadStone Technologies, Inc. – Development and Manufacturing of Precious-Metal-Free Metal Bipolar Plate Coatings for Polymer Electrolyte Membrane Fuel Cells
- Raytheon Technologies Research Center – Low-Cost, Corrosion-Resistant Coated Aluminum Bipolar Plates by Elevated Temperature Formation and Diffusion Bonding

Fuel Cell RD&D for Heavy-Duty Applications – Innovative, Low-Cost Air Management Components

- Caterpillar, Inc. – Leveraging Internal Combustion Engine Air System Technology for Fuel Cell System Cost Reduction
- Eaton Corporation – High-Efficiency and Transient Air Systems for Affordable Load-Following Heavy-Duty Truck Fuel Cell
- R&D Dynamics Corporation – Foil-Bearing-Supported Compressor–Expander
- Mahle Powertrain, LLC – Durable and Efficient Centrifugal-Based Filtered Air Management System and Optimized Balance of Plant

Fuel Cell Cost and Performance Analysis

- Strategic Analysis, Inc. – A system-level cost analysis annually updated for technological advancements in fuel cells provides insight into the most impactful research directions to help lower fuel cell manufacturing cost. This project supports cost projections for fuel cell auto, trucks, marine, and rail that will have positive impacts on the industry by quantifying the impact of promising fuel cell manufacturing techniques and designs, charting annual progress, and identifying areas with RD&D/manufacturing deficiencies.

Small Business Innovation Research/Small Business Technology Transfer Research (SBIR/STTR)

FY 2020 SBIR Phase II

- pH Matter, LLC – Multi-Functional Catalyst Supports (IIA)
- Tetramer Technologies, LLC – Improved Ionomers and Membranes for Fuel Cells

FY 2021 SBIR Phase I

- Giner, Inc. – Durable High-Efficiency Membrane and Electrode Assemblies for Heavy-Duty Fuel Cell Vehicles
- Pajarito Powder, LLC – Fine Gradient Electrode and Microporous Layer Structures for Improved Heavy-Duty Fuel Cells
- Celadyne Technologies, Inc. – Nanocoating for Increased Nafion Membrane Durability and Efficiency

FY 2021 STTR Phase I

- Ionomer Solutions, LLC, with The Pennsylvania State University – Optimizing Liquid Free Ionomer Binders for High-Temperature Polymer Electrolyte Membrane Fuel Cells for Heavy-Duty Vehicles
- Energy 18H, LLC, with University of Delaware – Development of a Direct Fuel Cell for the Perhydrodibenzyltoluene/Dibenzyltoluene Fuel Pair

BUDGET

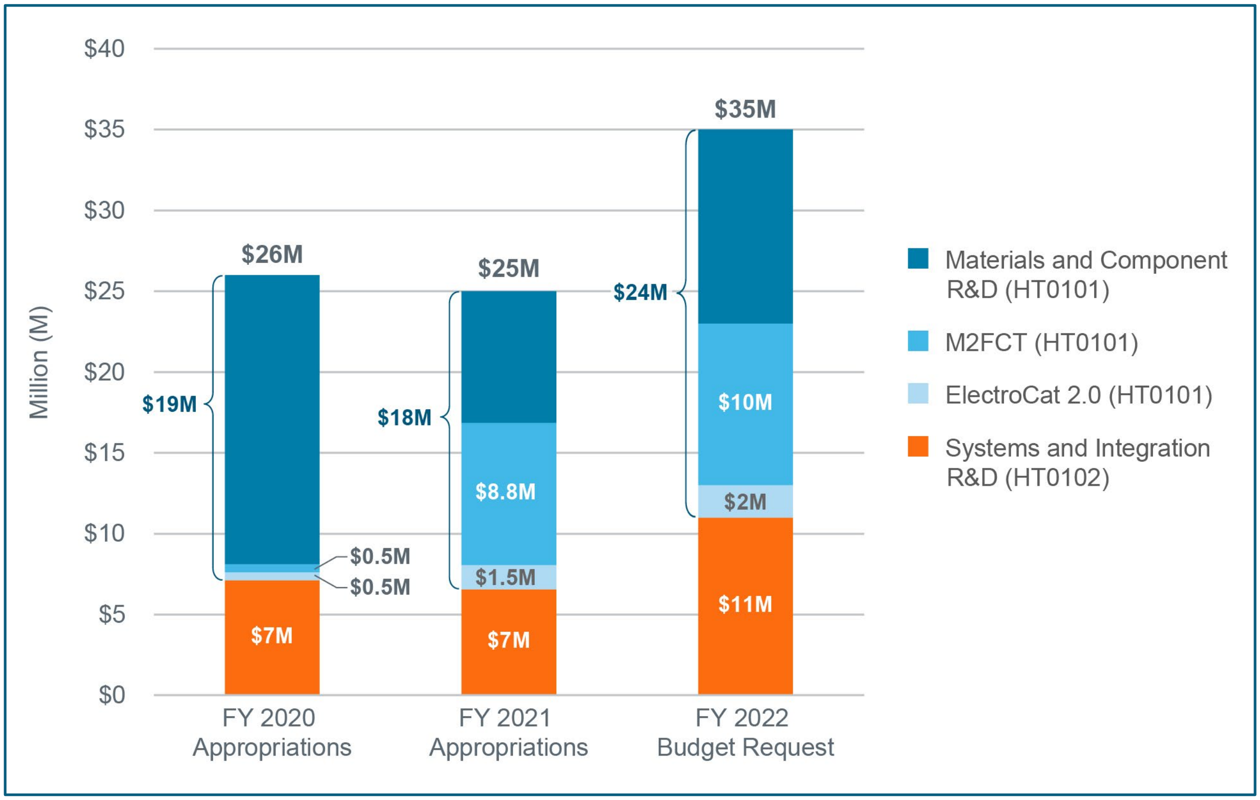
The FY 2021 appropriation was $25 million for the FCT subprogram. In FY 2021, the subprogram funded RD&D efforts in two key areas: materials/components and systems integration. Funding was dedicated to the two national laboratory consortia, M2FCT and ElectroCat 2.0, with M2FCT receiving a majority of the consortia funding (see the chart below).

Funding for research into fuel cell materials and components focused on areas such as low-PGM MEAs and MEA components with enhanced durability, PGM-free catalysts/electrodes, bipolar plates, and advanced manufacturing.

Funding for research into fuel cell systems integration focused on stacks, BOP components (for air management), systems analysis, and advanced manufacturing.

The FY 2022 budget request for the FCT subprogram is $35 million. Activities planned for FY 2022 include continuing RD&D of low-PGM and PGM-free catalysts and membrane durability, initiating development of gas diffusion layers and power electronics, continuing activities of the M2FCT consortium, and meeting durability-adjusted HDV fuel cell cost targets**.**

Fuel Cell Technologies Subprogram Budget Breakdown



Annual Merit Review of the Fuel Cell Technologies Subprogram

SUMMARY OF FUEL CELL TECHNOLOGIES SUBPROGRAM REVIEWER COMMENTS

Reviewers said that the focus on HDVs—with so many high-impact commercial applications—was appropriate and a great opportunity to decarbonize using hydrogen and fuel cells. However, some reviewers felt that it was somewhat premature to reduce focus on fuel cells for LDVs as a consequence of the increased focus on MDVs and HDVs. It was also suggested that the subprogram prioritize end-use demonstrations and do more to enable deployment in general.

Review panels commended the RD&D leading to PEMFC stack costs decreasing to less than $80/kW, as well as RD&D investigating fuel cell durability through both materials and system design lenses. These achievements were seen as significant milestones on the path to reaching the goals of the H2@Scale effort. The design for manufacturing and assembly approach used yearly to forecast the cost of transportation fuel cell systems—including the recent focus on MDVs and HDVs—was lauded for continuing to refine and expand as needed. Moreover, this approach was seen as key to helping DOE determine relevant goals and progress toward these goals. Reviewers encouraged closer collaboration with M2FCT to integrate the consortium's findings into the cost analyses.

Both ElectroCat and M2FCT were regarded as well-developed consortia and strongly supported by experienced researchers and companies.

ElectroCat was viewed as a well-coordinated and strong consortium, consisting of top-notch researchers and institutions who are collaborating to advance state-of-the-art PGM-free catalysts. The consortium's progress—especially milestones being exceeded—was specifically praised. More involvement of non-national laboratory teams, and especially collaboration with original equipment manufacturers (i.e., end users) was suggested. Increased industry involvement (with more dynamic dialogue between competitors) and linking consortia with state and regional stakeholders were seen as efforts with potential to increase value.

The M2FCT consortium was seen as an effective platform and complementary to other existing consortia. The "team-of-teams" approach used by the M2FCT consortium was commended for having recognized experts and being the most effective way to collaborate on key challenges. However, some reviewers also cautioned that the consortium may be too large for effective management, also noting that it may be better to separate out some of the work for separate reviews. Reviewers also commented that improvements seemed incremental, and thus suggested that the subprogram have stricter requirements for projects funded under this consortium (perhaps even including an advisory board review). It was further advised that the subprogram consider opening up M2FCT to international organizations. Reviewers also commented that the consortium has an opportunity to contribute to standardizing the process of testing and integrating new materials for PEMFCs.

The emerging areas of AEM fuel cells and the use of fuel cells for aviation were suggested for further attention. Reviewers regarded AEM fuel cells as having potential for significant cost reduction in catalysts and membranes and therefore encouraged increasing overall research scope and elevating this research area's priority. A note of caution was that use of catalysts with high PGM loadings would provide substantially different characteristics that are not representative of the desired end product (where PGM-free catalysts are used). Thus, reviewers suggested that more focus should be placed on using PGM-free catalysts.

Regarding the second emerging area, fuel cells and hydrogen are of interest for decarbonizing aviation. Requirements for electric power systems in conventional and emerging markets in aviation, such as drones and urban air mobility, are different from for those in LDV and HDV applications, with weight and specific power density being prime concerns.

Some reviewers regarded projects examining MEAs as high-risk and warranting closer tracking of progress before decisions are made regarding continued funding. Reviewers thought that scalability should also be considered and that there should be more collaboration with industrial partners for membrane performance validation.

The development of durable PGM-free catalysts and research progress on the subject was praised and seen as a key area contributing to reaching DOE goals. Reviewers noted that non-PGM catalyst activity and performance had improved. However, they saw the stringent durability requirements of HDVs as likely to make it extremely challenging to reach levels needed for commercial application.

Developing efficient, stable reversible fuel cells was seen as an enabling technology for energy storage. Reviewers encouraged more focus on catalysts and diffusion media, rather than on membranes. In addition, longer durability testing in electrolysis mode and more cycling between electrolyzer and fuel cell mode were recommended.

Thirty-eight FCT projects were reviewed, receiving scores ranging from 2.2 to 3.6, with an average score of 3.2. Project reviewers were impressed with specific project-level highlights and accomplishments. Following this subprogram introduction are individual project reports for each of the projects reviewed. Each report contains a project summary, the project's overall score and average scores for each question, and the project-level reviewer comments.

Project Reviews

Project #FC-117: Fiscal Year 2018 Small Business Innovation Research Phase IIB: Ionomer Dispersion Impact on Advanced Fuel Cell Performance and Durability

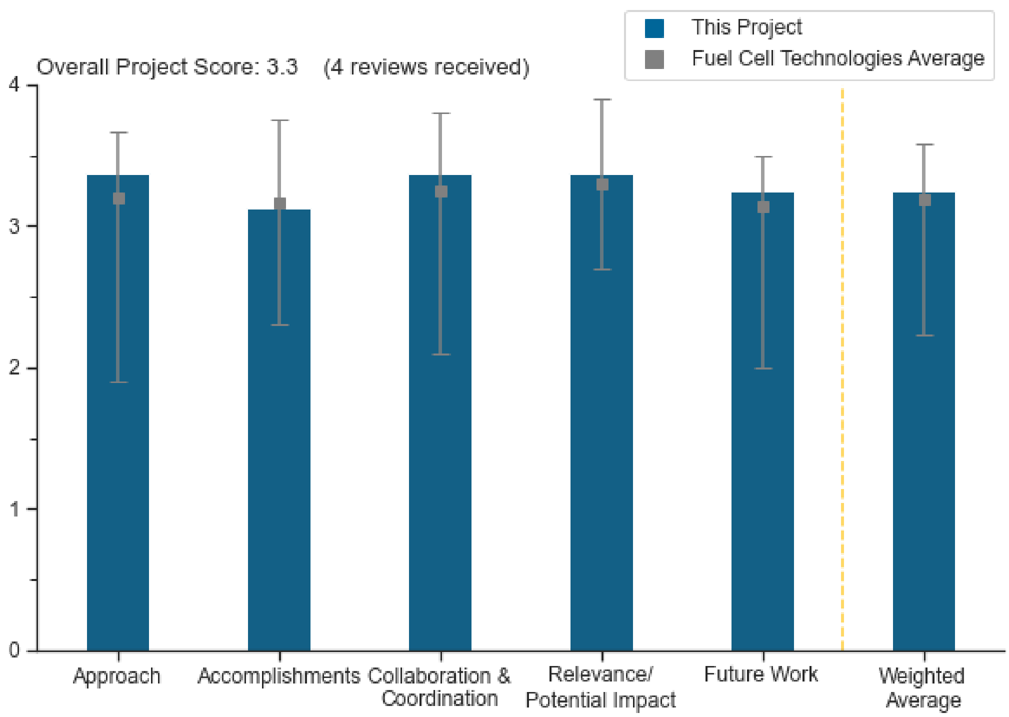
Hui Xu, Giner, Inc.

DOE Contract #	DE-SC0012049
Start and End Dates	8/27/2018 to 8/26/2021
Partners/Collaborators	Los Alamos National Laboratory, National Renewable Energy Laboratory, University of Connecticut
Barriers Addressed	• Polymer electrolyte membrane fuel cell and electrolyzer performance and durability

Project Goal and Brief Summary

This project aims to elucidate solvents' impacts on ionomer dispersion morphology and the associated changes to electrode structures and performance. The research team will design light- and heavy-duty fuel cell membrane electrode assemblies (MEAs) that are mechanically and chemically durable, and researchers will establish a correlation between catalyst ink properties, electrode structures, and MEA performance. Findings from this project could help develop processable and scalable MEA fabrication platforms and identify pathways to commercializing MEAs with enhanced durability via roll-to-roll (R2R) production.

Project Scoring



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

Question 1: Approach to performing the work

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

- The technical approach is reasonable and well-considered, with focus on correlating catalyst ink properties with electrode structure and fuel cell performance, which can lead to improvements in R2R manufacturing. Starting from investigation on different solvents, the combination of rheology/particle size analysis with microstructure characterization and MEA performance and durability evaluation creates a straightforward path to project implementation.
- The structural changes when casting Nafion using a high boiling solvent such as ethylene glycol during solution casting of membrane layers have been known since the early work of Charles Martin in the 1980s and workers from Virginia Tech in the late 1990s. There is a phase change at 140°C that leads to this morphology transition to microbiphasic, like that structure found in the heat melt of fluorides. It is important to follow up on this in R2R MEA making.
- The approach is good, and solvent variations would likely be a big factor in catalyst layer performance. It is unclear what the mechanism is for improved durability, which was one of the principal goals of the solvent modifications; it could be some more post-mortem analysis to decipher the mechanism of durability improvement. Many other unnecessary variables were introduced into the study, such as different gas diffusion layers (GDLs) and gas diffusion electrodes (GDEs) versus a catalyst-coated membrane (CCM) construction approach, making it difficult to follow the effects of the variations. As a Small Business Technology Transfer project, this should have a good commercial outcome/target, but it looks like technology development just goes full circle; it starts at Los Alamos National Laboratory (LANL), then there is some work at Giner, Inc. (Giner) and then it ends up at the National Renewable Energy Laboratory (NREL) for scale-up.
- The approach contains useful characterization techniques. However, additional detail, such as the methodology of analyzing particle size, would be appreciated. Many techniques that show particle size require very dilute solutions. The ink may have to be diluted to determine the average particle size, and it is not certain that the homogenization techniques used would affect the ink in the same way if the ink were more concentrated. The project should provide additional information regarding this technique and justification that the results are applicable to the concentrations of inks used in preparation of real MEA electrodes. Furthermore, there may be additional analyses that can help to improve the understanding of the materials. As this is a narrowly focused project, a deeper dive into all the properties of the ink may be useful. For example, perhaps it is possible to quantify the magnitude of effect of solvent–ionomer interaction versus solvent–catalyst interaction on the final catalyst layer microstructure and homogeneity. Relatedly, a paper from the Tokyo Institute of Technology (*Journal of The Electrochemical Society* 166 (2) F89–F92 [2019]) has shown that certain solvents may degrade in the presence of catalyst. It would be interesting to know whether the team sees the same degradation with ethylene glycol and butanediol.

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 does a very nice job by using constant stoic polarization curves with representative gas flow rates; it gave a good measure of progress toward DOE targets. It is surprising that solvents did have more of an impact on performance. It would be nice to compare the polarization curves and durability with the IPA (Nafion in 2-propanol/water) solvent MEAs since those catalyst layers were more significantly different in structure, as shown in the transmission electron microscopy images of slide 6. The EG/NPA (Nafion in ethylene glycol/Nafion in 1-propanol/water) images and polarization curves were actually very similar.
- The effects of solvents and mixing time were optimized under the guidance of rheology and particle size analyses. The project team examined MEA fabrication via both CCM and GDE and chose GDE for easy commercialization. Small MEAs prepared by Giner met several DOE 2020 targets on performance and durability, including mass activity, MEA performance at 800 MV, loss in catalyst activity, and performance at 0.8 A/cm^2. However, the rated power performance target was not met. While such a gap can be partially

attributed to the membrane, the property and structure of ionomer in the catalyst layer should be still the main contributor to relatively low current density, which needs further study and optimization. Some successes were seen for scaling up the process from small lab preparation to R2R.

- The formation of good films cast at high temperatures is noteworthy, but the film processing is not optimized. Treating at 180°C is above the temperature at which sulfonic groups come off of Nafion polymer, as can be seen by infrared spectroscopy of Nafion heated at different temperatures (see Peter Faguy et al. in the 1990s in the *Journal of Applied Electrochemistry*).
- The first project goal listed is to "Elucidate how solvents impact ionomer dispersion morphology, thus changing electrode structures and performance," which has useful data that are yet to be connected. For example, the solvent's effect on ionomer dispersion morphology has been partly studied through rheology, zeta potential, and particle size analysis. However, the results of the ink morphology have not yet been correlated directly to electrode structure and performance; for example, it would be useful to plot the results of the ink morphology studies with some quantifiable electrode structure metrics (e.g., electrochemical surface area [ECSA], gas transport resistance, total porosity, sulfonate poisoning of catalyst, catalyst layer ionic resistance, and current density at several cell voltages). Once each of these parameters is plotted, a sensitivity analysis may be performed to determine which characteristics of the ink are truly the most important in affecting the final electrochemical parameters. Additionally, while elemental mapping is interesting to view, subjective claims without a quantifiable direct relationship to performance should be made with caution. As an example, on slide 6, one of the bullet points says "better ionomer and Pt dispersion with EG and BUT" (Nafion in butanediol). It is unclear whether increased dispersion will necessarily lead to better performance; this is a matter of (among other effects) balancing an advantageous increase in protonic access to Pt sites and ionic conductivity of the cathode layer (through reduced tortuosity of the ionomer's water channels) and a deleterious increase in sulfonate–Pt interactions that may have negative impacts on both catalyst activity and oxygen transport. If claiming that a particular electrode structure is more advantageous, the project should indicate the mechanisms by which it is. The project should also avoid using speculation, such as, on the same slide, "likely associated with higher elastic and viscous components of catalyst inks." If this is the case, the project should provide evidence. For the goal of scalability, the methods to determine final ink structure at the small scale may not correlate to the large scale because of time limitations, such as milling for days and drying for days.

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 project teaming is well-considered and organized. LANL is responsible for ionomer dispersion preparation characterization because the ionomer material was originally synthesized there. Giner takes the main responsibility of investigating ink properties, fabricating, and testing MEAs, while the University of Connecticut supports the conduct of electrode structure characterization, and NREL helps with scale-up. The strengths of team partners are complementary and nicely arranged to achieve the project goals.
- Giner's and the national laboratories' work is well-coordinated and complementary.
- The results of the collaboration with the University of Connecticut are not quite clear in the presentation slides. It would be useful to outline exactly what kind of electrode characterizations are to be performed by this collaborator. The interactions with the Million Mile Fuel Cell Truck (M2FCT) national laboratories seem appropriate and useful. The results so far are a good foundation for work in the project's future, especially NREL's R2R capabilities.
- The team did have NREL as team member, but the work looked to be a repeat of the study using a different equipment. Some optimization needed to be done to obtain the same results. The project could have benefited from a collaborator with more analysis or modeling capabilities that could have given the team some correlation between observed performance differences and the structures and/or modeling.

Question 4: Relevance/potential impact

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

- The project is set to develop a new way to prepare catalyst ink to promote the application of a new ionomer. In general, new ionomer development and application have been quite difficult, but it is going to be very critical for future polymer electrolyte membrane fuel cell (PEMFC) development to meet DOE's goal for both passenger and commercial vehicle applications. To this end, this project indeed takes on high risks and challenges in a difficult topic with big impact.
- The project is quite relevant and has potential to provide a large impact on performance of PEMFC MEAs.
- Ionomer dispersion in both inks and electrodes is no doubt important. Some background evidence was provided to justify that solvent choice alone can affect fuel cell MEA electrode structure. However, there was relatively little discussion about how these structural changes specifically might affect efficiency and durability of the resultant MEA as a whole. Furthermore, for the purpose of establishing relevance, it would be useful to show how changing solvent alone might affect a voltage–loss–breakdown analysis. In other words, it is unclear which sources of overpotential are affected (and to what degree, specifically) by solvent choice.

Question 5: Proposed future work

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

- Overall, the proposed future work is reasonable, considering the project status. It would be great to dedicate some additional effort to (1) investigating why the high current/power performance is poor and exploring potential countermeasures and (2) understanding how the solvents and fabrication methods might have affected catalyst durability (composition or ECSA change). The result could provide hints for further improvement toward heavy-duty applications.
- Future work appears to address the concerns outlined in the presentation.
- The optimization is needed, but the temperature range of curing needs to be between 140°C and 170°C.
- Unfortunately, there is not much time left for future work on this project, so the future work outlined is much more than can be accomplished by August 2021. The scope of future work quickly expanded to an all-encompassing catalyst layer optimization study rather than the original scope of studying the impacts of the solvent on the ionomer interaction with the catalyst layer.

Project strengths:

- Evidence is presented to show that solvent has an effect on both electrode structure and resultant MEA performance. Collaborations with M2FCT national laboratories are useful and appropriate for the strengths associated with each laboratory's capabilities. Obstacles were noted (such as cracking in the electrode structure at particular thicknesses and electrode drying times), and the project plans to overcome these obstacles and offers at least one method to do so (e.g., reduce thickness of the catalyst layer on the GDL).
- The project has a strong team with enough capabilities to perform the planned tasks and reach the project goals. It has a fairly straightforward technical approach.
- An important topic for making a good catalyst layer is catalyst film that is durable and like a bulk membrane, and this project addresses this.
- The project has an important approach—looking at solvent interactions with a catalyst layer—that has a potential for large impact.

Project weaknesses:

- The project states that part of the procedure is to ball mill inks for between three and five days. In addition, part of the procedure is to dry the electrode structure in a vacuum oven for five days at 150°C. This protocol is unrealistically time-consuming for scalable electrode fabrication, so the resultant structures may have a questionable relationship to real electrodes for use in industrial applications. The ink analysis was not stated to be at relevant concentrations (e.g., it seems that the ink may have to be diluted to be analyzed in the zetasizer). This may not be a weakness if the concentrations of ink used were the same that were employed during fabrication, but the project should indicate which is the case.

- The project focuses only on ink and MEA preparation processes, not covering the modification and improvement of the ionomer material itself. This may limit the opportunity to further improve performance and durability.
- The project quickly diverged from the study of solvent interaction to an all-encompassing study of catalyst layer construction.
- The researchers are not well-versed in the effects they are studying and need to examine the literature better.

Recommendations for additions/deletions to project scope:

- Relatively few solvents were chosen for study in this project. There is significant additional literature that addresses other solvents used with perfluorosulfonic acids that may be useful to consider. The project should add more specific electrochemical analyses (ECSA, gas transport resistance, total porosity, sulfonate poisoning of catalyst, catalyst layer ionic resistance, and current density at several cell voltages) versus particular ink and electrode parameters (particle size, ink viscosity, zeta potential, etc.) and perform sensitivity analysis on these parameters. As an additional note, acoustic particle size analysis can be a powerful tool for measuring particle sizes, even in relatively concentrated dispersions, and may be something to consider as an additional tool to confirm the results.
- The project should do a literature search of solvent casting with ethylene glycol (Martin), phase changes (Virginia Tech), and thermal stability (Peter Faguy).
- The project should consider adding countermeasures to improve high-power performance.
- There is very little budget and time left to act on any recommendations.

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

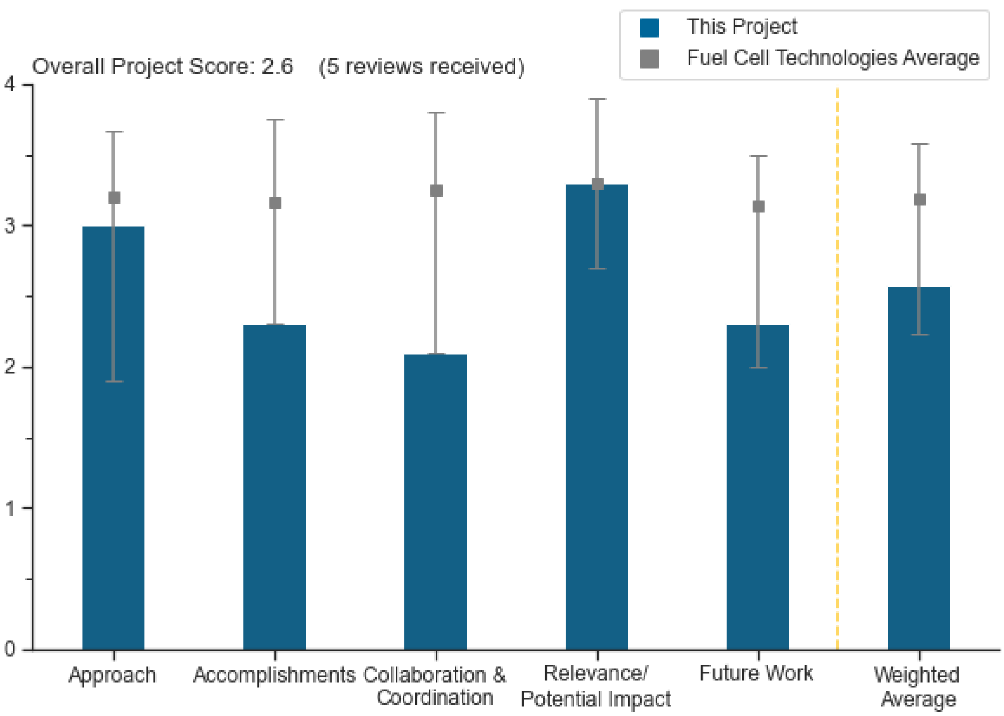
Peter Pintauro, Vanderbilt University

DOE Contract #	DE-EE0007653
Start and End Dates	1/1/2017 to 12/31/2021
Partners/Collaborators	Nissan Technical Center North America
Barriers Addressed	• High-current-density performance of polymer electrolyte membrane fuel cell membrane electrode assemblies is low for cathodes with low platinum loading

Project Goal and Brief Summary

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 containing various ionomer and blended polymer binders.

Project Scoring



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

Question 1: Approach to performing the work

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

- Using nanofibers of various configurations in the catalyst layer of MEAs is a novel and good approach, with excellent potential to improve performance over state-of-the-art catalyst layers. The project would benefit from some modeling work so that the mechanisms of the reported performance improvements could be better understood.

- Preparing structured electrodes through nanofibers can provide an optimal combination of protonic and mass transport resistances. The principal investigator (PI) has used platinum loadings that are relevant for automotive and heavy-duty fuel cell systems. The single-nozzle approach is not scalable. It would have been better if the project leaders had investigated options to scale up this technology by using multiple spray nozzles.
- The overall approach to improve catalyst layer performance is good. However, there are a number of areas of inconsistencies that make it hard to understand the technology improvements. For example, there is little understanding toward the effect of pre-compaction for 0.2 mg/cm^2 loading vs. 0.1 mg/cm^2. These loadings are not evaluated at the same compaction; hence, the benefits of this process cannot be understood. Similarly, a new binder is used in this year's process—one that is different from ones used in previous years—which does not adequately provide fundamental understanding of the binder's benefits.
- The project can still benefit from further systematic variation of compositions and process parameters, combined with Fuel Cell Consortium for Performance and Durability (FC-PAD) diagnostic and characterization tools, to correlate effects and enhance understanding of the underlying physics of the processes.
- Because there is no development of the basic materials (i.e., catalyst, catalyst support, or ionomer), the approach relies on assumptions that conventional methods of forming the catalyst layer create flaws (e.g., transport limitations) that warrant an alternative approach, and that the electrospinning approach would address that. There are several flaws in these assumptions. Also, using electrospun nanofibers brings its own challenges, such as higher contact resistance, pores and a residual carrier that take up valuable electrode volume, and potential higher processing cost.

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.

- After showing initial promising performance in a relatively short period, it seems like progress has been more modest on the nanofiber approach. Interesting results continue to be shown at very low (40%) relative humidity (RH) at high current densities. Superior water retention due to the pore nanofiber structure was suggested (and apparently previously demonstrated by neutron radiography results). It would seem more work may be warranted to characterize this (and should include the high-frequency resistance and electrode conductivity measurements). On the same low-RH performance issue, more emphasis should be put on power density at usable voltages rather than "max power density," where the cell potential (V_{cell}) is too low to be practical. New work seems to focus on the new additive binder, which demonstrated improved performance on a conventional spray approach (not the project's nanofiber approach), raising performance to be very similar to that with the nanofiber electrodes. If this is so, perhaps there are no longer clear benefits to the nanofiber morphology.
- The maximum reported power density is better than the project's internal baseline. However, the MEA performance is not best-in-class compared to other DOE-funded projects.
- Progress toward DOE goals is almost impossible to determine since the operating conditions of the fuel cell tests are not those specified in the DOE targets. All of the polarization curves are operated at constant gas flow rates, not stoichiometrically controlled testing conditions. The stoichiometry at the project's reported maximum power point of 1.5 A/cm^2 is 4 × for air and 2.2 × for hydrogen—all this on a small, 5 cm^2 MEA—and results from testing procedures (i.e., performance duration at each data point) are not given. At a reasonable air stoic of 2.5 ×, all the polarization curves show less than 0.2 V of cell voltage. Nissan (shown on the budget slide) is a good teaming partner from a system level, but no polarization curve data from Nissan was reported.
- Progress toward overall DOE goals is harder to evaluate because the fuel cell tests are performed at constant flow and not at stoic. Any mass transport issue in the nanofiber is not apparent under the flow conditions.

- Unfortunately, there has not been much progress in the last two years. The project still cannot say for sure whether the approach provides any kind of benefit over the state-of-the-art method. The choice of baseline electrode (hand-sprayed electrode) is poor, with likely poor quality and reproducibility. MEA tests are done with small-active-area cells, and yet the project chose an operating condition that limits evaluation of high current density. The measurement quality and repeatability are unclear. The project should have followed DOE recommendations for testing and collaborated with an external partner to get proper evaluation and comparison with a state-of-the-art electrode, if the project team is not capable of that. The fact that there is no difference in performance or durability between electrospun fibers and hand-sprayed electrodes with hydrophobic additive further discredits the benefit of electrospun fibers. There is no characterization update of any insight regarding electrode structure after fuel cell operation, nor of what causes the claimed benefit.

Question 3: Collaboration and coordination

This project was rated **2.1** for its engagement with and coordination of project partners and interaction with other entities.

- The Oak Ridge National Laboratory imaging results are appreciated, but these are all beginning of life. Addition of post-accelerated-stress-test (post-AST) imaging would also be interesting.
- This project could benefit from a fuel cell system partner. Nissan is shown on the budget slide, but no validation data from Nissan are reported. Also, a partner that could model the transport mechanisms of the improved catalyst structures would be beneficial.
- It would have been better if the project had collaborated with companies to pursue the pilot-scale multiple-needle demonstrator, per the team's response to the 2019 reviewer comments.
- Collaborations outside of the national laboratories are not apparent. Since the electrospinning collaboration has ended, it is not clear how the project can scale up this technology. There is no apparent interest in testing the MEAs at locations other than Nissan.
- There appears to have been some collaboration in prior years but none this year. The low score reflects the low collaboration in spite of being one of the FC-PAD (Million Mile Fuel Cell Truck [M2FCT]) projects. The project should have plenty of access to national laboratory resources and yet did not utilize them.

Question 4: Relevance/potential impact

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

- High-performance, stable, low-platinum-group-metal (low-PGM) loaded robust electrodes are key to programmatic success. Further progress is still needed in the nanofiber electrode high-power-density performance, but the electrode has shown some 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 $Q/\Delta T$ (inlet temperature difference) metric (driving higher temperature, which results in reduced RH). The project should further identify the reasons for this exhibited low-RH performance.
- High efficiency and durability remain the priorities of fuel cell research and development. This is an approach that potentially addresses the challenges.
- The concept of incorporating nanofibers into the catalyst layer is a good one and does have potential.
- The project supports overall DOE goals to improve performance and durability of MEAs for light- and heavy-duty vehicle applications. However, testing should be beyond the understanding of subscale MEAs under constant flow. Even the catalyst layer structure analysis stops with scanning electron microscopy (SEM) at high magnification. There are no high-magnification transmission electron microscopy (TEM) data to help researchers understand the catalyst–nanofiber interactions and structure. Similarly, there are no fundamental data to help researchers understand the fiber–membrane interactions. The focus seems to be only on evaluating different ink compositions and coating methods. A more comprehensive study to understand the benefits and interactions of the nanofiber with catalyst layer components in the MEA should be the focus for the last part of this project.
- The impact of this project toward the DOE goals is limited. The performance of these MEAs is subpar compared to best-in-class, and there is currently no active work on scaling up this technology.

Question 5: Proposed future work

This project was rated **2.3** for effective and logical planning.

- Little time remains in the project for what seems to be a rather lengthy list of proposed future work. Half of that focuses on the new binder additive with sprayed electrodes. This additive does appear interesting, but the work on sprayed electrodes does not appear to be in scope. If it is, it may be worth demonstrating the repeatability of the results, as this spray method may show high variation. It would be good to see more systematic parameter and process variation, combined with FC-PAD diagnostics, to further identify nanofiber mechanistic impacts on performance, RH dependence, and voltage stability with both beginning-of-life and end-of-life samples.
- Future work should focus on comprehensive analysis of catalyst layer interactions between ionomer–binder–catalyst for an identified set of materials. Any further investigation on optimizing additive content, new catalysts, and new coating methods should be stopped until a full understanding of existing material is published.
- The project should report fuel cell performance under constant, DOE-specified stoichiometry operating conditions.
- The project should focus on a binary alloy catalyst for cathodes and the pilot-scale multiple-needle electrospinner.
- Very little time is left in the project. No clear plan exists to identify the approach's benefit or the source of the benefit, which will leave the project ending without much value to the community.

Project strengths:

- The project has a novel concept in tuning properties of catalyst layers, which does have good potential to improve conductivity and mass transport in catalyst layers.
- The PI has a strong background in nanofiber technology. The project can still take greater advantage of FC-PAD's characterization capability, which is a key attribute.
- The project understanding of the electrospinning process is a strength.
- Moderate success equals moderate strength.
- The project strength is the approach to catalyst layer deposition using a nanofiber spray method. However, the project should have focused on understanding the benefits of this method by limiting the material set that was investigated. Changing the ionomer additive every year without getting a full picture of the catalyst layer interactions was not beneficial to the community. A trial-and-error approach is not a valuable return on investment for DOE.

Project weaknesses:

- The project did not involve other national laboratory partners or the greater research community to understand the benefits of the nanofiber technology. The project team should not have given an industrial-style optimization effort a "go" without understanding the benefits of this approach. Fuel cell testing at constant flow is a weakness and does not allow the community to evaluate the progress for light- or heavy-duty vehicle application. This year's progress does not seem to include any contribution from Nissan Technical Center North America.
- The team has the capability but has not yet used it sufficiently to fully characterize performance (local O_2 transport resistance, mass transport, and electrode and membrane conductivity). Although much work has been done to improve the spray electrode performance, how these electrodes perform compared to state-of-the-art electrodes remains unclear. Process variation is also still in question.
- The project did not follow DOE test recommendations. The team did not have the capability to evaluate materials properly and did not reach out for help. Collaboration was limited.
- There is no active scale-up pathway.
- Determining the progress toward DOE targets and goals is difficult.

Recommendations for additions/deletions to project scope:

- It would be good to see greater characterization of nanofiber (and now additive, binder, and spray electrode) morphologies and linkages to their observed performance. More systematic parametric variation is recommended to elucidate nanofiber electrode mechanisms and benefits.
- DOE should stop all future optimization work on this project, and the PI should focus only on extensive catalyst layer structural analysis and interactions of the additive and ionomer in the nanofiber with the catalyst and membrane. Future projects should identify a limited set of materials and focus all resources on understanding the fundamental interactions within that limited set of materials and comparing findings with the state-of-the-art MEA.
- The project should provide a polarization curve at fixed stoichiometry on an MEA with an active area of 50 cm^2.
- Optimizing additive content should be deleted from the scope. Working with original equipment manufacturers to conduct stack-level testing should be added, if possible.

Project #FC-160: ElectroCat 2.0 (Electrocatalysis Consortium)

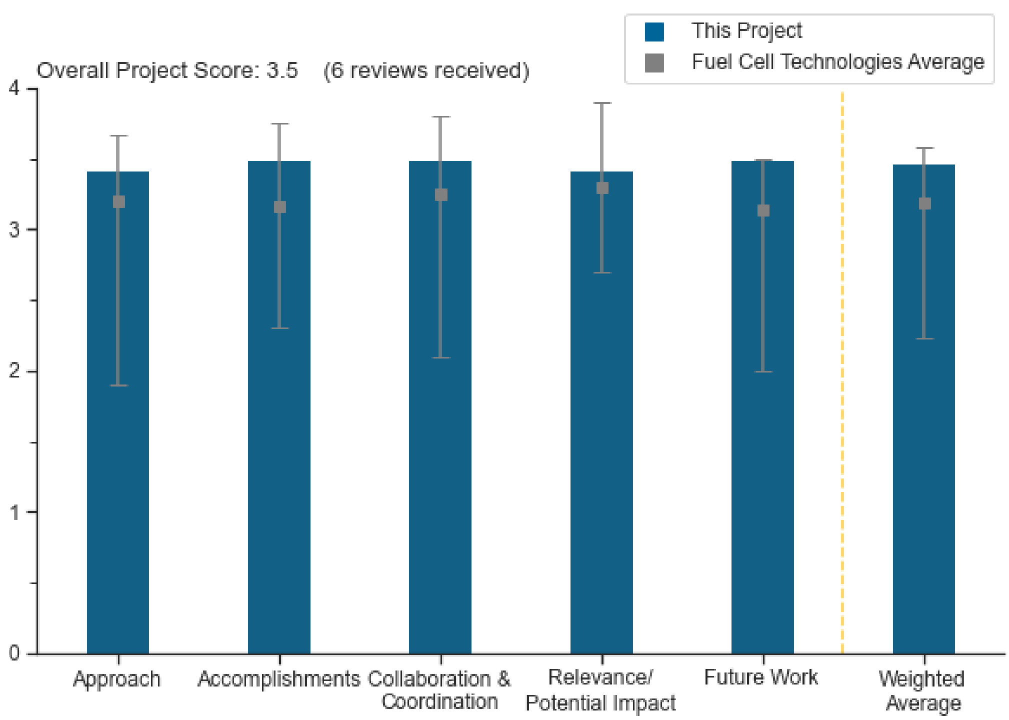
Deborah Myers, Argonne National Laboratory, and Piotr Zelenay, Los Alamos National Laboratory

DOE Contract #	Multiple
Start and End Dates	10/1/21–9/30/24
Partners/Collaborators	National Renewable Energy Laboratory, Oak Ridge National Laboratory
Barriers Addressed	• Cost (catalyst) • Activity (catalyst, membrane electrode assembly) • Durability (catalyst, membrane electrode assembly) • Power density (membrane electrode assembly)

Project Goal and Brief Summary

The Electrocatalysis Consortium (ElectroCat), created as part of the Energy Materials Network, aims to accelerate the development of next-generation catalysts and electrodes that are free of the platinum group metals (PGMs) currently required for good performance and durability. ElectroCat has focused its efforts on oxygen reduction reaction (ORR) catalysis for polymer electrolyte membrane fuel cell (PEMFCs) and has established a portfolio of unique synthesis, experimental, characterization, and modeling capabilities. ElectroCat 2.0 will increase focus on improving catalyst durability, investigating electrode engineering, and further advancing high-throughput catalyst synthesis and characterization capabilities coupled with machine learning while still working to improve catalyst performance. The Consortium has also expanded its catalyst portfolio to include the development of PGM-free catalysts for low temperature electrolysis.

Project Scoring



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

Question 1: Approach to performing the work

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

- The ElectroCat 2.0 team has continued the very well-integrated, thorough, and structured approach established in the first ElectroCat project. The combination of the combinatorial and high-throughput approaches with machine learning and density functional theory (DFT)/medium-duty modeling is very powerful. The use of extensive characterization approaches and the creation of new ones support a relatively strong pace in increased understanding. The neural network modeling and U.S. Department of Energy approach has been used to achieve significant results, with almost a doubling of the mass activity. Nevertheless, it is interesting that, despite several years of this structured approach, the catalyst system is still capable of significant surprises. For instance, the dual-zone process effect on performance is surprisingly poor, and durability is surprisingly good.
- The high-throughput and machine learning tasks have made significant advancements, and it is encouraging to observe the framework solidifying to the point where there appear to be sufficient data to generate useful predictions. It would be encouraging in future years to see whether the machine learning can predict synthesis/catalysts with performance/durability metrics that improve over the parametric studies used in generating the training data. Given the importance of the alkaline ionomer and membrane conductivity, the project would benefit from partnerships with multiple polymer electrolyte developers. The alkaline electrolyte membrane (AEM) electrolyzer effort would benefit from a greater amount of technical-assessment-driven targets and material-specific objectives based on performance and durability analysis, rather than cell performance goals. The x-ray spectroscopy, electron microscopy, and DFT modeling continue to be key assets to this project.
- ElectroCat 2.0 is (cautiously) welcome to address low-cost catalysts for electrolysis; however, there is very little publicity on this new effort. The Office of Energy Efficiency and Renewable Energy public relations does not mention ElectroCat 2.0, nor does the ElectroCat website. The integration of machine learning with experimental efforts is a good strategy. There is tightly coupled multiscale modeling in this effort to provide physical interpretations of optimization. A new LANL–NREL effort may contribute here. The direct comparison of the four high-throughput systems would be valuable. The table on slide 31 is difficult to map to Systems 1–3b on slide 15. Validation of DFT predictions with experimental data is limited. The fact that there are some experimental motivations is understood, but the results need to come back to these to demonstrate causality, for example, catalyst durability decay constant as a function of pH.
- The project appears to be on track and making important advances in the field of PGM-free catalysts. In particular, optimization of both activity and durability in parallel form an important approach to ensure that useful catalysts are developed, and the team is clearly adopting that strategy. A minor criticism of the work is that it is not clear how the fuel cell and electrolysis aspects of the projects are related or how they leverage each other; it seems these are entirely separate projects.
- The project is structured to focus on hydrogen–oxygen performance for heavy-duty transportation and electrolyzer stacks. Performance goals were met for hydrogen–oxygen, but decay is very high after 20,000 cycles. Development progress and spending are on schedule, and milestones have been met for the hydrogen–oxygen effort. The hydrogen–air tests are a significant concern because of the significant degradation after 10,000 cycles. The ultimate requirement is a hydrogen–air membrane electrode assembly. The team performed significant new machine learning efforts. The catalyst performance in the electrolyzer mode is very poor.
- There is a good mixture of performance-oriented work and fundamental scientific investigation and methods.

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 work is structured via well-defined milestones that are on track or have been exceeded, with excellent progress since 2019. The team has already exceeded the September 30, 2021, target. The durability improvement of the dual-zone catalyst is impressive, and understanding this will undoubtedly lead to further improvements. The team is making some good progress on a hypothesis that is still to be proven. The establishment of the Pourbaix-like diagram is a very good addition to the knowledge base and provides an effective approach to understanding stability of the catalyst systems. The oxygen-limiting current method for characterization of bulk-electrode transport is a useful addition to the characterization techniques.

- The team has made excellent progress here in terms of new catalyst synthesis techniques and new high-throughput data generators, coupled with artificial intelligence (AI) and physical models. The dual-zone and post-treatment catalysts should provide a wealth of information about a mechanism that can be studied using high-throughput systems, AI, and physical models. The dual-zone catalyst appears to have higher resistance than the single-zone catalyst. Since this does not appear in the high-frequency resistance measurements, it may be localized to the electrode. It is unclear whether this is the topic of "reduced cathode proton resistance," as mentioned for the future work on slide 41. If NH_4Cl is a pore former, then a before and after study of micro- and meso-porosity is of interest.
- Overall, the project appears to be making progress toward the end-of-consortium goals, although the gap on durability is probably most significant at this point. The publication and dissemination of work appears to be excellent.
- The project is on track with its proposed goals based on the data presented. The dual-zone discovery seems to be a potential breakthrough in this field if a better understanding of the mechanism for improved durability can be elucidated.
- The fuel cell performance of the current project catalysts currently lags behind several other projects, as well as prior-generation catalysts from the team. The activity metrics have excessive catalyst loadings (7 mg/cm^2) that are not viable for generating satisfactory power density. Excellent stability has been demonstrated, but with a low-activity catalyst. The insights from the high-throughput testing and machine learning will yield significant advancements in site density and stability. Initial progress has been made in setting benchmarks for PGM-free oxygen evolution reaction (OER) catalysts for AEM electrolyzers.
- Significant progress has been made in PEMFC development, but there are weaknesses that need to be addressed—particularly, performance of the catalyst in the electrolyzer setting. The cell voltage of 2.0 V at 600 mA/cm^2 is too high. Much more focus should be put on the study conditions for the electrolyzer performance if the project is ever going to achieve a goal of an 80,000-hour lifetime. An early understanding of the fundamental mechanisms associated with the IrO_2 anode's operation in the electrolyzer should be used to guide the catalyst development.

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 final team, facilities, and equipment required to complete this project are in place, ready, and available. The personnel are qualified, as demonstrated by the successful completion of previous projects and by their 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 consortium is a strong group of top-notch researchers and institutions. The research and development (R&D) plan uses the strengths of each institution. It would probably be impossible to assemble a better R&D team for this project or a team with better capabilities; however, the consortium seems to lack involvement of "end users" for the catalysts. This may be understandable because the PGM-free technology is far from commercialization outside of niche applications, but other DOE consortiums have more involvement from fuel cell original equipment manufacturers (OEMs).
- The project has a great deal of collaboration and provides critical support to the ElectroCat funding opportunity announcement (FOA) projects. Within the team, there does not seem to be very much coordination between the catalyst development and the electrode development. It would be good to have more coordination between those efforts.
- The work of the national labs in the consortium appears to be well-coordinated. ElectroCat is engaged in numerous collaborations.

- The team appears to be working with a variety of partners and organizations. For the electrolysis work, the team should consider working with some of the other (non-national-laboratory) teams developing AEMs: the University of Delaware, Georgia Tech, Rensselaer Polytechnic Institute (RPI), and others.
- The mention of external collaborators was very sparse in this presentation (granted, 2020 was a challenging year for collaboration). It would be of interest to see the way that external FOA projects have contributed to the core ElectroCat mission.

Question 4: Relevance/potential impact

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

- This is the leading effort to obtain a low-cost electrocatalyst for ORRs, and now OERs, in the United States. For this reason, this project is crucial to the success of fuel cells in low-cost applications. ORR is much more important than OER for mobile applications, and stationary applications may be better suited to high-temperature systems. For this reason, the original mission to develop ORR catalysts should be given as much priority as possible.
- Although progress has been achieved in PGM-free catalysts, allowing the eventual applicability to high current density and high power density, durable applications are still in doubt. PGM-free catalysts are now at the point where they may find application areas, but the relevance to transportation applications is still to be determined. It is, however, necessary work and should be continued. The addition of the electrolyzer activity is also relevant.
- It is important to have this effort on low-technology-readiness-level materials and technologies that could be very impactful in the long term. The transition to greater emphasis on hydrogen production is timely. At the same time, the overall momentum in the advancement of PGM-free PEMFC catalysts within this project and the other ElectroCat projects is still high, and significant efforts should continue to be devoted to them.
- Reducing PMG loading is critically important to the successful commercialization of PEMFCs and electrolyzers, as well as the reduction of IrO_2 on the anode in the electrolyzer.
- The work is addressing the challenging area of PGM-free catalysts for ORR fuel cells and OER electrolytes.
- The PGM-free fuel cell catalyst development aligns well with DOE's long-term goals for fuel cell cost reduction, but the potential results from this project are likely a long way off with respect to achieving the million-mile performance targets of 1.07 A/cm^2 at 0.7 V. Most truck manufacturers are pursuing quite high PGM loadings, and PGM-free catalysts do not currently appear to be of interest for heavy-duty transportation. However, since the purpose of the consortium is more focused on R&D and discovery, the team is making significant potential impacts in that regard. For the electrolysis part of the consortium, it was difficult to understand how the work performed was related to the electrolyzer stack goals.

Question 5: Proposed future work

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

- The proposed future work is clear and makes sense. The use of probe molecules for characterization could yield further interesting results on active site structure and mechanisms for degradation. It will be interesting to see further characterization of the dual-site catalyst and the team's discoveries.
- Emphasis on durability and durability mechanisms for fuel cell ORR is important and well-placed. Researchers on the electrolysis work should consider collaborating with some of the other (non-national-laboratory) teams that are developing AEMs: the University of Delaware, Georgia Tech, RPI, and others. Also, the team should more clearly define the gaps for alkaline OER catalysts and state whether the main issue is integration with membranes. It seems the hydrogen evolution reaction may be a bigger challenge than OER for alkaline systems.
- In the dual-zone-synthesis technique, it appears that a mitigation strategy has been developed before a comprehensive understanding has been achieved. The project should marshal all of its resources to take advantage of this good fortune.

- The future work is good; however, more effort should be dedicated toward understanding perovskite material operation under OER.
- The proposed future work appears well-planned.
- The future work on the AEM electrolyzers was not well-defined and missed specifics and a clear framework/organization.

Project strengths:

- The R&D team is very strong. The project is well-managed and appears to be on track with the proposed milestones. The project has uncovered scientifically interesting discoveries that have the potential to significantly improve the understanding, performance, and durability of PGM-free fuel cell catalysts.
- There is excellent progress in synthesis, modeling, and characterization of new catalysts, as well as good synergy across all components. The project has excellent capabilities for addressing OER catalysts.
- This project is very systematic and comprehensive, and it has aligned efforts to improve the catalyst activity and durability/stability. Extensive characterization and catalyst-level modeling are incorporated. Overall, this is a very strong team.
- The high-throughput synthesis and testing, coupled with machine learning, are key areas of innovation and strengths of the project. The characterization and analytical methods are also key strengths.
- This project has many strengths, but there is much to do. This project group is capable of conducting the R&D for PGM-free catalysts. Scale-up and stability verification are likely to present significant challenges.

Project weaknesses:

- It would be encouraging to see more OEM involvement with the consortium. The electrolysis part of the project seems to integrate poorly with the fuel cell work.
- The key weakness is the current fuel cell performance of the project’s catalyst, requiring high loadings to achieve activity targets.
- There is concern that the study of OER will be a distraction from the central goal of a low-cost ORR catalyst. The synergy between the two efforts seems minimal. There would be two potential reasons to pivot to OER: (1) ORR is very successful, and OER is a spinoff project, or (2) progress toward an ORR catalyst is slowing down, or there is no foreseeable pathway to commercialization. The former does not appear to be the case, and it is to be hoped that the latter is also not true. Finally, the integration of external FOA projects could be improved.
- The stability of the catalyst needs to be significantly improved.

Recommendations for additions/deletions to project scope:

- A recommended addition to the project would be more OEM involvement to help guide targets for potential applications. The electrolysis catalyst development is important, but it should probably be a separate project.
- The project should consider bringing in additional team members who have a good understanding of electrolyzer performance.
- Caution is recommended in adding OER to the scope of this consortium.

Project #FC-163: Fuel Cell Systems Analysis

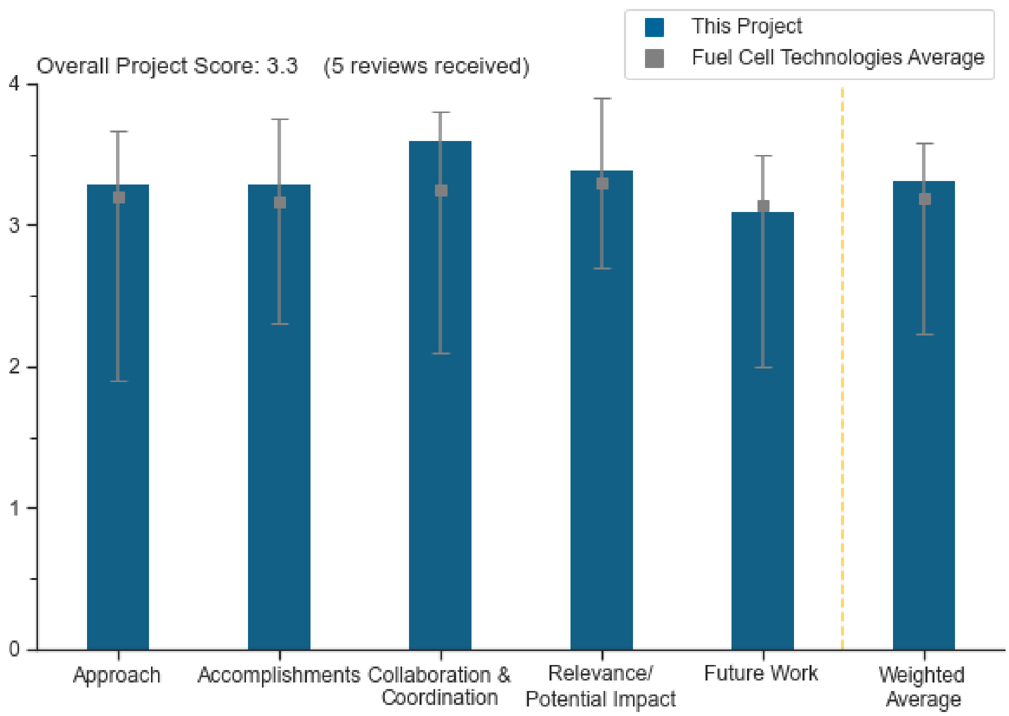
Brian James, Strategic Analysis, Inc.

DOE Contract #	DE-EE0007600
Start and End Dates	9/30/2016 to 9/30/2021
Partners/Collaborators	National Renewable Energy Laboratory, Argonne National Laboratory
Barriers Addressed	• System cost. Demonstrates impact of technical targets and barriers on system cost

Project Goal and Brief Summary

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. Analyses conducted project the impact of technology improvements on system cost, identify low-cost pathways to achieve U.S. Department of Energy (DOE) transportation fuel cell cost goals, benchmark fuel cell systems against production vehicle power systems, and identify fuel cell system cost drivers to facilitate Hydrogen and Fuel Cell Technologies Office (HFTO) programmatic decisions.

Project Scoring



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

Question 1: Approach to performing the work

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

- The approach is solid, working closely with Argonne National Laboratory (ANL) on the systems modeling and the incorporation of information as it becomes available. The project appears to extensively "validate" the approach and the numbers through one-to-one interactions with manufacturers. In some areas, the analysis may suffer slightly from not enough visibility into manufacturers' approaches. Each manufacturer will have a different combination of tradeoffs that the company uses to achieve results, and this analysis is only one view that may not reflect any approach actually used. On the whole, however, the model seems to come out approximately "correct." The stack oversizing approach is one area that does not seem like the best approach. More model analysis is required to justify or change this approach. Incorporating the graphite plate as a validated option is a good approach; it might also be good to run a sensitivity analysis that incorporates a potentially viable metal plate option. It will be important to include other degradation modes, such as start-up and shutdown. The removal of a humidifier may not be the most cost-effective approach when looking at the cost tradeoff (assuming an impact on performance and durability).

- Strategic Analysis, Inc.'s (SA's) design for manufacturing and assembly (DFMA) approach has been shown to be sound, and the team is working with relevant stakeholders and the fuel cell community to properly address the DOE-provided project goals. While not working directly on technologies that could reduce the barriers, the project's cost analysis plays a necessary role in assessing HFTO's progress toward overall goals.
- SA's approach remains appropriate and good. A continuous close interaction with component suppliers and system developers is key to fairly accurate cost estimations.
- The SA team attempts to create a comprehensive fuel cell model to predict where costs are most intensive and where researchers should focus development costs to save money. The model is also useful for setting DOE performance goals. The model seems to be appropriately detailed for accurate analysis. The durability predictions could be very important, but the degradation mechanisms are not fully understood, so it is hard to model. The modeling of the cell monitoring systems could also be very useful to manufacturers. The major problem with the modeling approach is that it cannot be validated because it is based on high-volume manufacturing that has yet to occur.
- The project takes a consistent approach to annually update and indicate durability complications in the cost analysis. Research and development (R&D) focus has shifted to heavy-duty vehicles (HDVs). Therefore, project resources should also focus on HDVs, rather than light-duty vehicles (LDVs). HDV customers are conscious of total cost of ownership (TCO) rather than the cost of the fuel cell system. It is recommended that the project use TCO to identify cost actions for fuel cell designs.

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 main focus in the past year was the DOE-driven request to assess the cost impact of durability, a difficult task. SA did as well as could be expected, given the uncertainties and relatively immaturity of fuel cell use in HDVs. In addition, the team updated all of the LDV, medium-duty vehicle (MDV), and HDV fuel cell statuses and performed cost assessments of various durability enablers (e.g., cell voltage monitoring, high-frequency resistance, and total harmonic distortion). The team completed quite a list of accomplishments and progress for a year.
- The SA team was able to incorporate extensive pricing details into the model and appears to have worked hard to bring accuracy to the numbers (the back-up information is impressive). The work is somewhat discouraging, as it predicts an increase in costs per kilowatt for longer endurance. The results almost suggest that additional innovations are needed to lower costs.
- The work done on HDVs is a real need. Unlike LDVs, TCO of HDVs is the key driver and thus should be the focus. Moreover, as the market share is less than that of LDVs, information on the corresponding market share, in addition to the number of produced systems, could be provided. For clarity, it would be good to be reminded as to whether the annual production rates refer to the total of fuel cell systems produced in the United States or to a single fuel cell system manufacturer, which would lead to even more difficult cost targets that need to be achieved. The final 10% cost decrease titled "miscellaneous" may be better detailed.
- This work is essential to guiding the overall DOE work on achieving cost-effective fuel cell systems.
- The durability-adjusted cost analysis shows more appropriate development direction, such as the tradeoff between Pt loading reduction and durability.

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.

- SA worked effectively with the National Renewable Energy Laboratory (NREL) and ANL to obtain information for the SA model. The researchers were also able to populate their model with information from companies such as Celeroton. The project team got feedback on the model from multiple sources, including Gannon & Scott and Cell Centric Canada. The overall approach to collaboration and coordination is thorough.
- The team has excellent collaboration with NREL and ANL. There are a good number of collaborators providing pertinent input and review, as well as a very extensive list of additional collaborations for each component.
- SA is doing a great job of collaborating, mainly with the ANL systems analysis project and NREL, but also expanding with many component and system developers, suppliers, and end users. The team's interaction with the Million Mile Fuel Cell Truck (M2FCT) effort is expected in the next period.
- SA's close interaction with the ANL systems analysis project is important and effective. The team also provided an extensive list of collaborations, as required for project effectiveness.
- There is effective collaboration on the fuel cell system analysis, including durability impact.

Question 4: Relevance/potential impact

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

- This project provides a powerful tool to forecast the costs of fuel cell systems for LDVs, MDVs, and HDVs. The tool allows systematic updates based on achievements of other projects, industry interviews, and modeling refinements. It contributes to identifying fuel cell system cost drivers to help DOE refine and update the critical issues to be addressed in future funding opportunity announcements.
- The project plays a necessary role in providing a status update to DOE on the key cost metric. Thus, the project is of greater benefit and relevance to HFTO and subprogram managers than to the direct advancement of the technology. The project also has indirect value in identifying promising technology enablers or areas and proposals of limited potential benefit.
- This project is essential to finding feasible solutions to meet DOE targets.
- The model clearly points to technology areas where improvements are needed to meet future cost targets for 2025 of automotive, bus, and truck fuel cell systems. The key unknown is whether the model is correct and whether it is possible to reach the projected cost targets; however, the study provides a useful direction. The disconnect with the industry might be whether industry would use this model or whether any company making 100,000 fuel cells a year has its own model.
- The R&D focus is now shifting from LDV to HDV applications.

Question 5: Proposed future work

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

- Given the significant cost impact of durability, the intent to continue to investigate ways to incorporate durability into cost models is appropriate. The project should continue to assess the potential cost impact/benefit of the various fuel cell projects funded by the U.S. DRIVE Partnership. Advanced coating of bipolar plates, membrane electrode assembly deposition techniques, and manufacturing control processes were mentioned, but should also be extended where relevant to any new U.S. DRIVE projects. Given the large cost sensitivity of various requirement-driven targets (e.g., thermal, power, end-of-life degradation, efficiency), it is thought to be worthwhile to more broadly explore the requirement-driven space and develop a more general and flexible cost vs. "x" functional relationship that can be more readily applicable to high-level TCO and technoeconomic studies. It is clear that a simple "$/kW" metric does not adequately describe this. Sensitivity studies are useful but, alone, are not likely sufficient.

- The proposed future work is reasonable. In this project, there will always be additional work to be done to improve the model and incorporate new approaches. It would be difficult to fully outline these.
- The proposed future work is in line with the project objectives.
- It is appropriate to update manufacturing quality processes for the MDV/HDV system cost.
- The future work is reasonable. The goal is to essentially continue to refine the model. There are some concerns about investigating technologies such as aluminum bipolar plates, as research along these lines has been attempted for years with no success (aluminum is tough).

Project strengths:

- The project has an excellent approach to providing current and future cost projections through great collaboration with ANL and NREL teams, who have broad expertise. The project has employed a stable calculation methodology, as well as transparency on its approach and data.
- This is a rigorous detailed model, with accurate inputs from industry. It can be used to target areas for cost reduction for manufacturing at a high volume.
- The project has provided consistent cost analysis updates. New approaches are properly implemented (e.g., durability and manufacturing adjustments).
- The team's extensive fuel cell cost analysis history, expertise in cost assessment, and extent of collaborations provide for success.
- The analytical collaborative approach is the project's greatest strength.

Project weaknesses:

- In the past, it would have been recommended that the project have access to state-of-the-art materials, but that was effectively addressed with the Fuel Cell Consortium for Performance and Durability (FC-PAD) partner projects (and continuing with M2FCT) and strengthened collaboration with ANL's system analysis project. Still, the team would benefit from greater feedback on durability, especially from fuel cell applications that have demonstrated high durability.
- The model is not validated and has no way to be validated, as that would require manufacturing at 100,000 units/year, plus full cooperation from private companies making 100,000 units/year. It is also not clear that a company with enough resources to make 100,000 units/year would use this model, rather than having its (the company's) own modeling resources. The model also has no way to incorporate truly disruptive technologies, such as membranes that do not require humidification.
- The project may not always have "true" data with which to work, owing to confidentiality issues.
- The main project weakness is the lack of comparison with real performance and cost data of systems.
- There are no significant weaknesses.

Recommendations for additions/deletions to project scope:

- The focus of the project could remain mostly on HDVs/MDVs in the short term by extending to additional applications, such as coaches and trains using comparable fuel cell systems. Additionally, close collaboration with M2FCT is recommended to rapidly integrate that consortium's findings into this cost analysis.
- The project would benefit from researching factors such as the effect of catalyst loading on durability, instead of stack oversizing; start-up/shut-down degradation; freeze-start requirement cost implications; precious metal commodity price sensitivity studies; recycling requirements and greenhouse gas emissions vs. material costs implications; and cost implications of materials that may be restricted in the future because of negative environmental impacts.
- The project should increase focus on a general functional cost model that can be readily used for high-level TCO/technoeconomic analysis—for example, cost as a function of end-of-life power, effective overall efficiency, lifetime, thermal rejection, or technology progress.

- For fuel cell system cost analysis of HDVs, TCO should be used for design suggestions, e.g., proper Pt loading and efficiency.
- It is most important for SA to continue cooperation with industry partners to refine the model.

Project #FC-167: Fiscal Year 2020 Small Business Innovation Research Phase IIA: Multi-Functional Catalyst Support

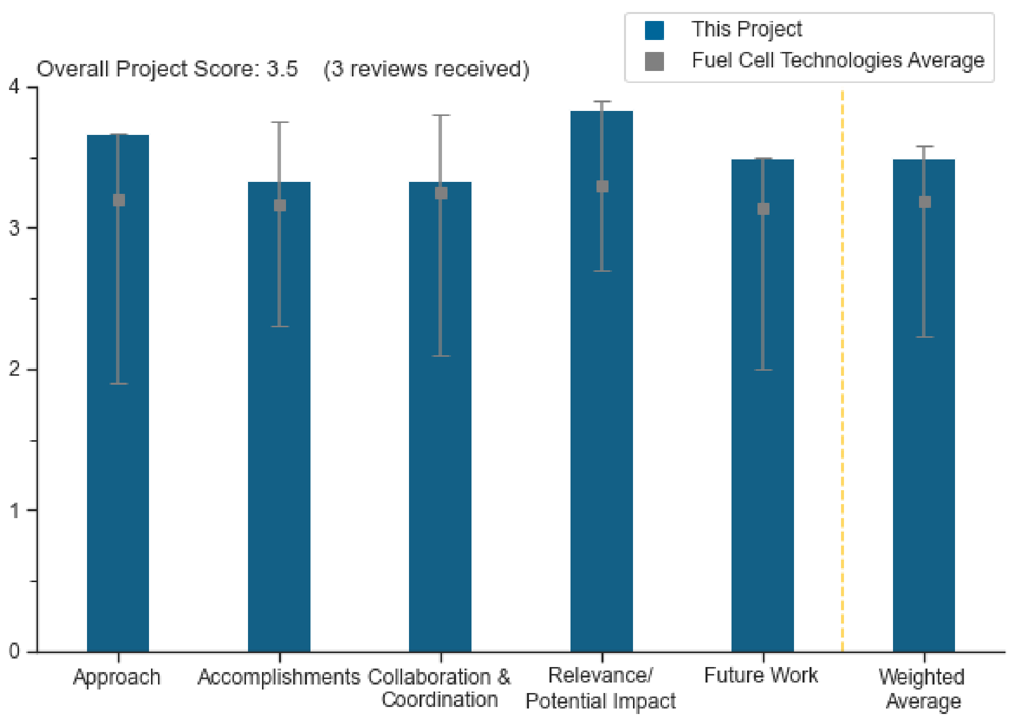
Minette Ocampo, pH Matter, LLC

DOE Contract #	DE-SC0017144
Start and End Dates	05/22/2018 to 05/20/2022
Partners/Collaborators	Giner, Inc., National Renewable Energy Laboratory, Shyam Kocha Consulting
Barriers Addressed	• Cost: enhance the platinum catalyst activity (and durability) to reduce its loading levels • Durability: optimize the interaction between the catalyst and the support material to improve chemical and thermal stability. • Performance: demonstrate improved performance in membrane electrode assemblies

Project Goal and Brief Summary

In Phase II of this project, the research team developed a multi-functional catalyst support (MFCS) approach for high platinum-group-metal (PGM) loading. In the present phase, researchers aim to use this approach to further develop catalysts and to demonstrate catalyst performance and durability requirements in membrane electrode assemblies (MEAs) for heavy-duty polymer electrolyte membrane (PEM) fuel cell applications. The project will optimize the MFCS to enable higher platinum loading, higher power, and extended durability performance for heavy-duty applications. Heavy-duty accelerated stress testing will be performed to evaluate catalyst durability and demonstrate the U.S. Department of Energy's (DOE's) 2025 Million Mile Fuel Cell Truck (M2FCT) target of 1.07 A/cm^2 at 0.7 V.

Project Scoring



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

Question 1: Approach to performing the work

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

- This project takes a rational approach to achieving good activity and durability in a catalyst. The project has a larger batch size and more reliable data than most research and development projects.
- Developing durable PGM catalysts is an essential step to achieving DOE's goals for transportation fuels. The general approach of optimizing the porosity of the carbon support is a widely used method to decrease catalyst loading due to agglomeration and migration. The details of the technology development are proprietary, so it is difficult to extensively evaluate the fine details of the research and development approach.
- This project leverages previous project results to address DOE heavy-duty vehicle PEM targets.

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 project tested a large number of catalysts and supports under several DOE protocols and made progress in achieving the DOE targets for durability and performance. It is difficult to assess whether a single formulation meets the DOE targets, as so many catalysts were evaluated.
- The project has demonstrated catalysts with reasonably good performance and durability in MEA testing. However, it is not clear whether the intended features of the catalyst have been achieved. More characterization focusing on those benefits would be useful. The absolute performance is still trailing commercial Pt/high-surface-area carbon catalysts.
- Within the first year of the project, the team has nearly met DOE's 2025 M2FCT MEA targets.

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.

- Collaborations are excellent, as they include Giner, Inc., National Renewable Energy Laboratory, and catalyst manufacturers. The research is well collaborated.
- Collaboration among project partners appears sufficient, although there is not much collaboration with other groups. More collaboration with national laboratories may help to obtain better insights about the catalyst structure and performance.
- Not much collaboration was reported during this project period.

Question 4: Relevance/potential impact

This project was rated **3.8** for supporting and advancing progress toward the Hydrogen and Fuel Cell Technologies Office goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The project is very relevant to the DOE Hydrogen and Fuel Cell Technologies Office mission, as catalyst development is key to reducing the cost of fuel cell systems. The potential impact is moderately high, but there is tremendous competition from larger catalyst manufacturers.
- Higher-activity and durable cathodes using low-PGM catalysts are a high priority for PEM fuel cells.
- This project is on track to meet DOE's 2025 M2FCT MEA targets.

Question 5: Proposed future work

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

- The proposed future work is reasonable. Extensive future testing in realistic field conditions is necessary to evaluate the long-term durability and performance of the catalysts. The researchers should also further optimize the catalyst loading.

- The proposed future work is appropriate. The researchers should seek more collaboration with national laboratories for characterization to better understand the catalyst structure and features. Providing samples to MEA integrators would help bridge the materials to commercialization.
- This project is tightly focused on preparing high-performance MEAs for commercial deployment.

Project strengths:

- This project has excellent teaming—good collaboration between catalyst developers, fuel cell manufacturers, and national laboratories—and a logical development approach.
- Strengths include efforts to scale the catalyst process and the team's know-how.
- This project is meeting aggressive project targets.

Project weaknesses:

- It is not clear whether the graphitized supports will be cost-effective or easy for the project team to scale.
- The project does not include characterization to identify and confirm features.
- It is difficult to evaluate the materials development approach, as it is proprietary.

Recommendations for additions/deletions to project scope:

- The catalysts should be benchmarked against newer formulations and highly graphitized supports.

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

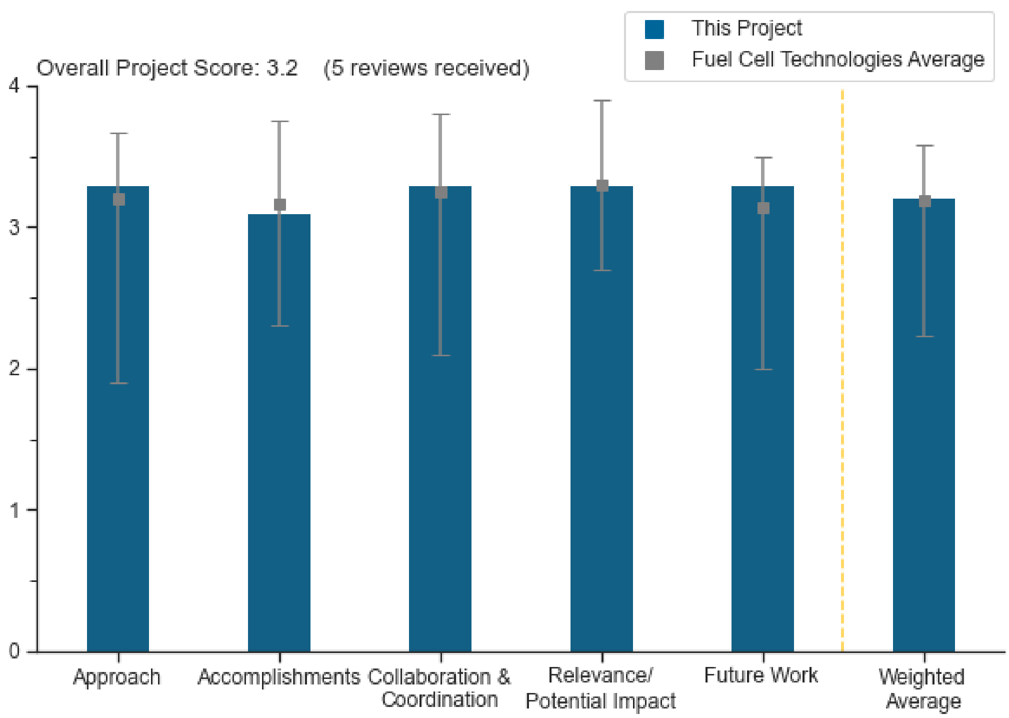
Hui Xu, Giner, Inc.

DOE Contract #	DE-EE0008075
Start and End Dates	10/1/2017 to 8/30/2021
Partners/Collaborators	University of Buffalo, University of Pittsburgh, General Motors, Indiana University, Compact Membrane Systems, Northeastern University
Barriers Addressed	• Durability (catalyst, membrane electrode assembly) • Cost (catalyst, membrane electrode assembly)

Project Goal and Brief Summary

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 and lower 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 Technologies projects.

Question 1: Approach to performing the work

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

- The project objectives are clearly outlined; the technical approaches are scientifically solid and appropriate for achieving the project objective. The identified critical barriers are being effectively addressed. The tasks and experiments are well-designed and are integrated with other relevant efforts.
- The team approach toward developing Mn-based non-PGM oxygen reduction reaction (ORR) catalysts is good to excellent. The researchers are using an appropriate balance of modeling, physical characterization, and electrochemical characterization to guide their work. Additionally, the team is appropriately emphasizing durability in MEA under relevant conditions, a key barrier for commercial viability of non-PGM ORR catalysts.
- Overall, the approach seems appropriate. The team is developing better catalysts, testing them in appropriate platforms, and doing work on fundamental understanding of the catalysts' performance and degradation.
- This is a good, solid approach for developing PGM-free catalyst materials with new development strategies. It is a step-by-step approach. It remained a little bit unclear what iterative process may be used if the envisioned steps are not as successful as expected.
- The approach is focused on achieving U.S. Department of Energy metrics on activity and durability.

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.

- It seems that the team has made excellent advances toward the outlined project objectives (and contributed to the DOE Hydrogen Program [the Program] goals as well) through well-defined and measurable performance indicators (such as electrocatalytic activity and durability). For example, it has been demonstrated that the designed Mn-based PGM-free catalysts have met the DOE catalyst activity target of >0.044 A/cm^2 at 0.9 V (iR-free) in an MEA test with the durability extended by 50%. The accomplishments to date suggest that the project has made considerable progress in addressing critical barriers to achieving DOE goals.
- The team has continued to make good to very good progress toward advancing the activity and hydrogen–air performance of the Mn-based non-PEM catalysts, with significant year-over-year advancement from 2020 to 2021. Determination of the importance of using forming gas to enable reduction of MnO toward better-dispersed single-atom Mn catalysts and space confinement (ordered structure) seems to be a key advancement.
- The project has met, or is on track with regard to, targets and milestones. The project made good progress on relevant catalyst development parameter studies and performance metrics. It is unclear what the flow conditions were for the in situ experiments. Stoichiometric flow rates or giving information about fixed flow rates together with cell size when showing current density would be more meaningful. Most important for comparison with PGM-based MEAs are the 150 kPa data sets. There is strong dependence of the catalyst on cell size (i.e., likely channel length) and relative humidity (RH). There is progress of degradation through post-treatment. Specifically, the durability of the material within an MEA structure will need improvement.
- The team has made consistent and excellent progress on the beginning-of-life catalyst activity. However, clear challenges for durability remain at this point as the project nears its end.
- The team has made excellent progress toward activity goals in comparison to where the project started. In particular, an excellent improvement in activity was achieved as a result of implementation of new synthetic procedures. Significant progress has been made toward fundamental understanding of the origin of the catalyst's electrocatalytic activity. However, the catalyst has not met the DOE activity target (0.044 A/cm^2) in MEA tests. Unfortunately, improvement in durability via the catalyst post-treatment with benzimidazole led to a loss in activity. The most serious durability problem is related to Mn oxidation due to Mn affinity to oxygen. This will be difficult to mitigate in the presence of air.

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.

- There appears to have excellent collaboration and coordination among a strong technical team. Of note is the good alignment between electrochemical characterization and physical characterization and synthesis efforts.
- An excellent collaboration has been established, both within the team and with external partners, such as ElectroCat, Indiana University–Purdue University Indianapolis, Northeastern University, and Compact Membrane Systems.
- The project management team effectively engages and coordinates the activities of the four project partners: Giner, Inc., University of Buffalo, University of Pittsburgh, and General Motors. Well-coordinated collaboration has been demonstrated between the partners.
- The core project consists of a combination of industry partners and four universities. The project uses the ElectroCat consortium well.
- It appears the involvement of partners and work with ElectroCat is sufficient.

Question 4: Relevance/potential impact

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

- Some of the barriers that face the Hydrogen and Fuel Cell Technologies Office are the high cost, limited availability, and durability of PGM catalysts. The development of durable, PGM-free catalysts is critical to the Program and has potential to significantly advance progress toward DOE research, development, and demonstration goals and objectives. The project supports and advances progress toward Program goals and objectives and also supports the HydroGEN Consortium mission. The project has good potential to advance the discovery and development of novel materials for efficient water-splitting systems, which will enable meeting the DOE ultimate hydrogen production goal of \$2/kg H_2.
- The relevance and potential impact are fair. While this project had made excellent progress toward addressing activity and performance, durability remains a significant challenge with non-PGM catalysts. Demetallation of highly active Fe or Mn atomically dispersed catalysts under relevant cyclic testing in air remains a key barrier. Oxidative stability of the carbon-based matrix also remains a fundamental and perhaps insurmountable barrier for these types of non-PGM ORR catalysts. These durability barriers are certainly challenging in light of passenger vehicle application requirements and are even more so with the evolution of focus of polymer electrolyte membrane fuel cells to heavy-duty vehicles. These concerns are not unique to this project but are related to the non-PGM ORR approach in general.
- This project has high potential impact; PGM-free ORR catalysts are needed to significantly reduce the cost of fuel cells.
- This program aligns well Program objectives. However, it is unlikely that this project will be a game-changer for DOE. The project makes contributions to understanding how materials work, which is also important.
- The team is directly addressing the challenge of PGM-free catalysts for ORR.

Question 5: Proposed future work

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

- The project's future work is directly aligned at addressing the key barrier of simultaneously having high activity/performance and durability. The approach of further understanding the degradation physically and mechanistically is appropriate. The stated strategy of "reduc[ing] Mn oxidation and demetallation" is appropriate, but it is unclear how this would be accomplished.

- Degradation and MEA performance in hydrogen–air were adequately picked as the key topics for future work. Finding the right strategies for improving performance and durability is a common challenge with PGM-free catalyst materials.
- The project will end on August 31, 2021. The planned activities for the next few months are adequate to complete the remaining tasks and, if successfully implemented, to achieve the project objectives.
- The future work is focused on achieving durability and activity targets simultaneously.
- This project appears to be coming to a close. If it were not, a more detailed plan for reducing catalyst degradation would be important.

Project strengths:

- The development of PGM-free catalysts is an important topic. It may have great impacts on the cost and the commercial viability of the technology. The concept of the new catalysts was well-conceived, and the technical approaches are scientifically sound. The tasks, experiments, and milestones are designed well and in a logical manner by incorporating appropriate decision points and considering potential barriers to achieving project goals. Also, alternate pathways are provided to mitigate potential risks.
- The project strength is in the combination of fundamental tools, such as density functional theory and extensive experimental validation, to provide fundamental insights into the catalyst's performance. The team is very creative in solving problems.
- The team consists of highly capable researchers with an excellent balance of fundamental material development, characterization, and modeling. The team is following a unique approach of focusing on Mn rather than Fe as the active center, which is important for maintaining membrane durability.
- The project has a good systematic approach that considers important aspects of catalyst design and MEA integration.

Project weaknesses:

_The project work, accomplishments, team, and approach are all very good. The primary weakness is that, owing to the long-standing durability concerns, the prospects of non-PGM ORR catalysts in commercial applications remain remote.

- MEA performance seems to be very dependent on RH and cell size. Understanding these barriers and counteracting them seems very important to further advancing the performance of this catalyst system.
- The weakness of this project is in selecting a Mn-based catalyst, which is susceptive to oxidation in the presence of oxygen.

Recommendations for additions/deletions to project scope:

- Operation of MEAs at stoichiometric conditions would allow more relevant comparison to realistic operation.

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

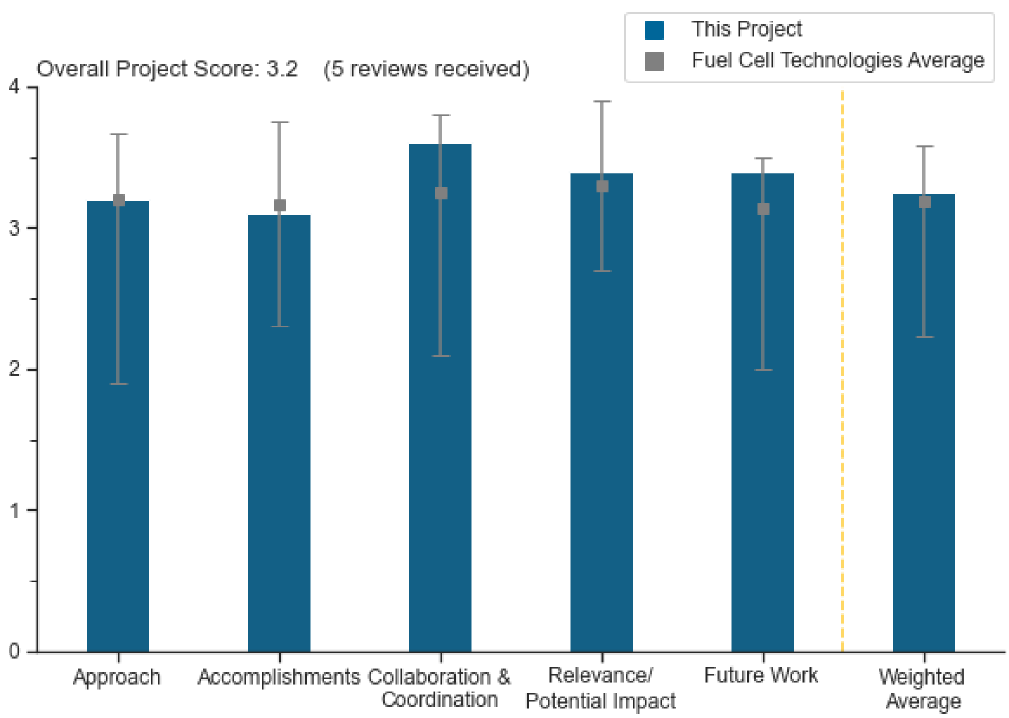
Yuyan Shao, Pacific Northwest National Laboratory

DOE Contract #	DE-EE0001647
Start and End Dates	10/1/2017 to 9/30/2022
Partners/Collaborators	Washington University in St. Louis, University of Maryland, Ballard Power Systems Inc., Oregon State University, Electrocatalysis Consortium (ElectroCat)
Barriers Addressed	• Cost (catalyst) • Activity (catalyst, membrane electrode assembly) • Durability (catalyst, membrane electrode assembly)

Project Goal and Brief Summary

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 catalysts developed 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



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

Question 1: Approach to performing the work

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

- This is a generally sound approach that uses the creation of a knowledge base for the development of a new catalyst material. In this case, the team attempts to identify mechanisms for degradation of non-Fe-PGM-free catalysts. With this fundamental understanding, the group wanted to identify non-Fe-based dual active sites that improve durability. This is a feasible approach, given the right diagnostic techniques are used and the required information can be acquired.
- Dual active sites, a non-Fe-PGM-free catalyst, and a radical scavenger are good approaches. A non-Fe-PGM-free catalyst is a critical path forward for PGM-free catalysts to avoid the Fenton reaction on the membrane. The scavenger can help to enhance the stability of the PGM-free catalysts.
- Developing an understanding related to the performance and durability of PGM-free catalysts is important to their long-term development and use. The incorporation of radical scavengers into the PGM-free catalysts seems to show a benefit. Developing non-Fe-based catalysts may be important to membrane durability.
- The project differentiates itself from other work funded by the U.S. Department of Energy by putting more effort into understanding durability of PGM-free catalysts, including development of less active but more stable cobalt-based materials. In particular, in situ characterization of degradation, such as measuring carbon dioxide evolution during operation, is an important and insightful approach. The only negative aspect of the approach is that it seemed to be limited in terms of the modeling effort. The team reported demetallation of Fe versus Co, but it seems that a larger number of potential active sites and methods of oxygen attack or oxygen species could have been examined.
- This is a well-organized approach with systematic identification of the milestones and the go/no-go events.

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 team had a successful demonstration of synthesis of clean FeN_X. There was a systematic evaluation of degradation with a focus on radical attack and demetallation, as well as quantified demetallation with acid attack and effect of hydrogen peroxide formation on degradation (artificially introduced hydrogen peroxide). The team's application of x-ray photoelectron spectroscopy (XPS) is very informative. The Co-N-C development (non-Fe-PGM-free catalyst) is successful, but prepared samples had poor activity. Co-N-C has superior durability to Fe-N-C. The team members explained why Co-N-C is more resistant to demetallation. They determined radical scavenger $TaTiO_X$ and identified optimum composition. Finally, they found a way to use Fe-N-C and benefit from its superior activity compared to Co-N-C, which appears to be a very important finding.
- The team presented some nice results to show the effect of demetallation and oxygen attack on ORR performance, as well as the improved stability with cobalt metal centers and radical scavengers. This information will be very useful to future researchers who are developing the next generation of PGM-free catalysts. The only negatives regarding the accomplishments were that there could have been a greater modeling effort and that characterization could have included adsorption and spectroscopy of active site markers, such as NO. This could have provided more information about degradation, the nature of the active site, and how it changes over time.
- Progress was seen in developing higher-durability catalyst materials when compared to Fe-based catalysts. The milestones were either completed on time or are still in progress. The analysis focuses on diagnostics and ex situ work. More device testing, specifically at stoichiometric flow rates, would be of interest in understanding the state of membrane electrode assembly (MEA) integration and potential of the catalyst materials.
- The project demonstrated that the catalyst layer picks up oxygen during the acid treatments. It would also be nice to measure the relative hydrophobicity change of the catalyst, as PGM-free catalyst layers are typically thick, with more hydrophilic layers; and the thicker they are, the more important flooding will be.

The durability improvement of Co-N-C is good, whereas the poor durability of Fe-N-C should possibly be compared to other developers in the area that are showing much better durability. It is important to know whether this difference is due to the fact that Fe-N-C is a low-durability catalyst or whether the durability of Co-N-C has in fact improved. The use of $TaTiO_X$ is interesting. This compound should potentially be examined in higher-technology-readiness-level (TRL) systems (e.g., Pt/C) to see whether it has a good effect on catalyst and membrane durability.

- Although good progress was made for the Co-N-C catalyst, achieving 22 mA/cm^2 at 0.90 V in H_2/O_2, it is still far from meeting the DOE target of 44 mA/cm^2, 150 mA/cm^2 at 0.80 V in H_2/air. The stability is even worse, with no MEA stability results provided—only rotating disk electrode (RDE) results. The dual catalyst synergistic effect was not shown in the presentation, and it was unclear why no MEA stability results were shown if the scavenger approach was working. The new scavenger $TaTiO_X$ developed showed good RDE performance but no MEA performance.

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.

- There is a sound mix of national laboratories, industry, and universities, supported by the ElectroCat consortium. There is a large team with distinct capabilities and clearly defined roles.
- The project is making excellent use of the ElectroCat capabilities. The project collaboration is an excellent set of partners: Pacific Northwest National Laboratory, Ballard Power Systems, Inc., University of Maryland, Washington University, and Oregon State. It is unclear what the no-cost partners are providing.
- The rating would have been outstanding if an industrial partner were included. The national laboratories, academic researchers, and no-cost partners all bring excellent technical strengths to the project.
- There are more collaborators involved in the project this year, and the team worked closely with ElectroCat.
- The project had a strong and diverse team, and there were no obvious weaknesses with the collaboration.

Question 4: Relevance/potential impact

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

- Relative to other ElectroCat projects, this project has a greater potential impact because of the focus on understanding durability for PGM-free catalysts. Poor durability is probably the largest hurdle that PGM-free catalysts need to overcome to be more useful in real-world systems. This project also included an industry participant, Ballard Power Systems, Inc., that is interested in commercializing PGM-free electrodes, which helps the project stand out in terms of potential impact. The only negative is that PGM-free catalysts' relevance to DOE's new focus on heavy-duty vehicles was not clearly communicated.
- This project has high potential impact, as reducing the cost of fuel cells significantly will require PGM-free ORR catalysts. It is difficult to compete with Pt for these systems, so higher loadings and thicker catalyst layers are required. The system currently lacks the performance and durability to be able to compete with PGM systems. This project specifically targets improving degradation aspects by designing a new type of PGM-free catalyst.
- PGM-free catalysts are critical for polymer electrolyte membrane fuel cell (PEMFC) commercialization since a PGM-free catalyst can significantly reduce the cost of PEMFC systems.
- The project is developing more active, more durable PGM-free electrocatalysts and appears to be making good progress. Performance is still far away from being competitive with Pt.
- This is an extremely important project if PGM-free catalysts are to be successful.

Question 5: Proposed future work

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

- There are good future work plans related to the precise identification of various degradation mechanisms and relationships to operation conditions, as well as demonstration of new radical scavengers in single fuel cells to improve durability.
- The future work addresses the missing links in the previous work. Future work aims at moving toward the demonstration of the novel catalyst materials in MEA devices through performance and degradation testing. Furthermore, the testing of additional radical scavengers may improve degradation behavior.
- The team proposes designing experiments to differentiate acid leaching from radical attack; however, there were no details regarding how these experiments would be done. More details would have increased confidence that the future work will be fruitful. The MEA experiments were proposed to achieve milestones, so adding the new radical scavengers to MEAs will be interesting.
- Each type of FeNx moiety was surprising, but discussion of these moieties was not found in the presentation. An explanation is needed as to why different FeNx moieties are important. It would also be good to know where the large MEAs are going to be tested and what Washington University's capabilities are.
- The project should focus on the mass activity and 0.80 V performance to meet the project goal.

Project strengths:

- This project had strong diagnostics and ex situ analysis. The team showed proof of principle that the degradation behavior of MEAs can be improved through the chosen approach.
- The main strength of this project was the focus on durability and understanding of PGM-free catalyst degradation.
- The project's strength is developing the fundamental understanding related to the degradation mechanism and understanding the importance of a radical scavenger.
- The dual active site (M-N-C and scavenger) is a unique approach. The use of non-Fe catalysts, which can avoid the Fenton reaction, is a plus. The work on the degradation mechanism is critically needed.
- There are excellent research capabilities at all the universities. There is a detailed approach to understanding issues. The project is well-organized.

Project weaknesses:

- Seeing more in situ test results that are comparable to the methods that are used by other ElectroCat collaborators would be beneficial. The proposed future work seems to promise this, which is great.
- The progress on both activity and degradation mechanisms seems slow and does not meet the milestones yet. The reported work on degradation mechanisms is not very clear as to the understanding of the degradation root causes.
- PGM-free is far from being competitive with Pt. The path to competitive performance is unclear.
- The project needs an industrial partner and/or a testing laboratory that works at an industrial scale, e.g., 10 cell stacks or greater.
- The project did not seem to put extensive effort into the modeling of degradation mechanisms.

Recommendations for additions/deletions to project scope:

- Using the radical scavenger with higher-TRL materials (Pt) and incorporating it into membranes to see its effect would be a nice addition to the project. It is unclear whether this radical scavenger is durable over long periods.
- A greater modeling effort could be useful for improving degradation mechanisms. Also, adsorption and spectroscopy of active site markers, such as NO, could provide more information about degradation and the nature of the active site and how it changes over time.

- The team needs to take a systematic approach to studying the degradation mechanism. The team also needs to focus on the new scavenger, $TaTiO_X$.
- More MEA device work should be included, as promised in the Proposed Future Work section.
- The project should get an industrial partner.

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

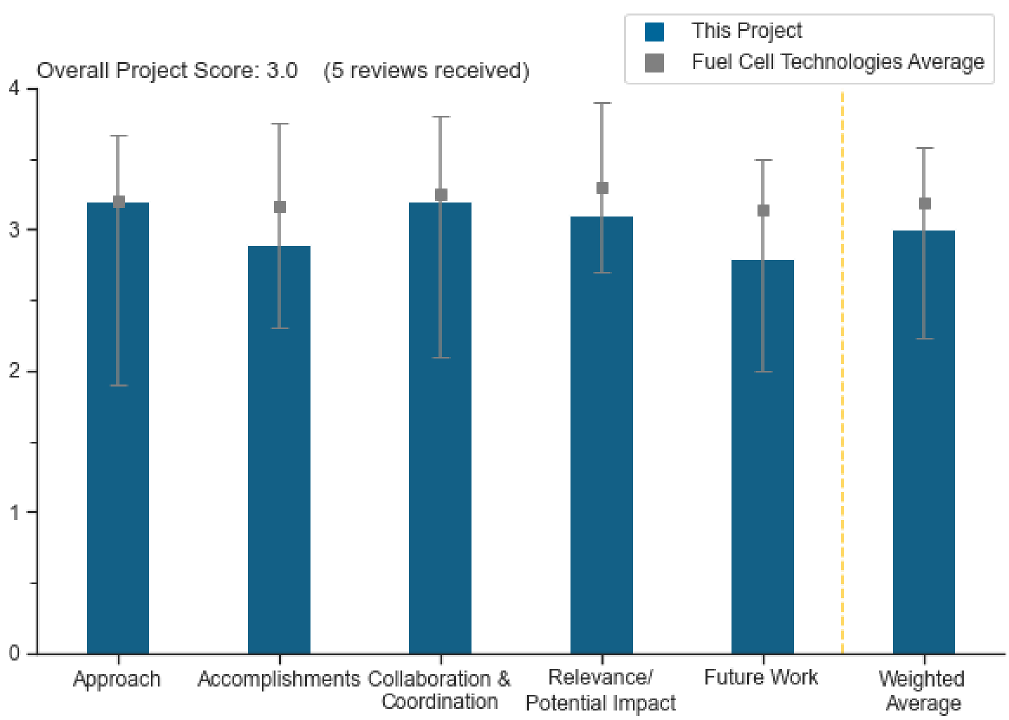
Qingying Jia, Northeastern University

DOE Contract #	DE-EE0008416
Start and End Dates	10/1/18 to 9/30/21
Partners/Collaborators	Lawrence Berkeley National Laboratory, Northeastern University
Barriers Addressed	• Performance in polymer electrolyte membrane fuel cells • Durability in polymer electrolyte membrane fuel cells

Project Goal and Brief Summary

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 hydrogen–oxygen 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 a hydrogen–air PEMFC with a membrane electrode assembly (MEA) size ≥50 cm^2.

Project Scoring



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

Question 1: Approach to performing the work

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

- The project is developing novel strategies for active site formation via chemical vapor deposition (CVD), strategies that form active sites in accessible surfaces of the supports for higher Fe utilization and potentially high site density. The MMC approach is intriguing. The half-wave voltage of the cobalt version provides encouraging results. The project's focus on active-site quantification is good and part of a scientific approach rich with characterization. It is good that the project is exploring alternative, high-risk approaches such as the ion-beam-assisted deposition (IBAD) concept for active site formation, although it was not successful. The MEA modeling of the catalyst kinetics with two Tafel slopes is questionable and requires additional validation. It is easy to conflate mass transport overpotential with a Tafel slope change, especially with high loadings. It is unclear whether the modeling would be coupled to improvements in catalyst morphology or electrode fabrication.
- The project uses a wide array of synthesis techniques, characterization techniques, and site-density-measurement methods to develop PGM-free catalysts. The plan and methodology are strong in this regard. The only negative with the approach is that it seems that significant effort went into developing and characterizing high-activity materials that have extremely poor durability. It would be beneficial if more effort were placed on durable catalysts or on understanding the degradation mechanism.
- The synthesis approach uses multiple techniques, which is nice to see; however, this appears to be mostly a shotgun approach, as opposed to being based on scientific hypothesis and theory. The project is using characterization techniques to understand the fundamental reasons for improving activity, which is good.
- This project has a good approach to increasing the activity of PGM-free catalysts. It aims to create MMCs to increase activity through high turnover frequency and active-site density. It is unclear why it is beneficial to avoid pyrolysis. In any case, the group plans to use several alternative synthesis routes to achieve the project development goals, which may increase the chances of reaching the research goals.
- The demonstrated project approach of screening multiple synthetic routes to achieving high-performing and durable PGM-free catalysts, rather than an application program toward MEA performance, is consistent with basic science. It is not clear why the targets specified by the project are lower than the DOE targets.

Question 2: Accomplishments and progress

This project was rated **2.9** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project had successful synthesis at a low temperature. Fe-metal-based materials show the highest performance. There was also a successful wet impregnation with MMC precursors. The team completed a detailed analysis to understand the nature of the resulting catalyst materials, had a successful transmetalation of Fe with Zn, and achieved high catalyst activity, although it was somewhat unclear from the presentation how this translates into MEA performance. It would be good to see more standardized MEA testing at various operating conditions, such as relative humidity (RH) and stoichiometric flow rates in hydrogen–air. Finally, there was very interesting voltage breakdown modeling for PGM-free MEA material sets.
- The team clearly conducted a large amount of work and, based on slide 19, is very close to the project goals listed. However, it was difficult to understand whether the project is on schedule and achieving milestones in a timely manner. A Gantt chart or table showing milestones is needed to communicate that. There were technical and audio issues during the presentation that may have contributed to difficulty with understanding whether the project is on track.
- The team demonstrated a CVD synthetic route to achieving highly active FeNC catalysts with high site density as an approach toward achieving performance goals; however, stability of obtained catalysts is not sufficient. The CVD process is not described well specific to its pros and cons, particularly, uniformity/ geometry spacing in the CVD chamber, which often results in non-scalable or non-uniform products.
- The project's progress has been reasonably good in terms of exploring various accessible approaches to active-site characterization and quantification. The catalyst activity is becoming good, achieving a first scan

oxygen current of 33 mA cm^{-2} at 0.9 V high-frequency-resistance-free, although with a high loading of 6 mg cm^{-2}. Several of the project targets have not been evaluated, and the fuel cell characterization of the catalyst appears to be limited (it is likely that these are outcomes of COVID-19 challenges).

- MMCs do not appear to show competitive activity with other PGM-free catalysts (e.g., MN_4). In fact, they are far from competitive. Unless a path is known for better performance, it seems like this path is not going anywhere. The FeNC-CVD shows poor durability. Understanding the degradation mechanism might be important. Improving site density might give better beginning-of-life (BOL) performance; however, unless the rapid degradation is prevented, the improved site density is not going to make a catalyst that is useful. The team should improve the site density after a stable catalyst is developed. The modeling seems fine, but without a more active and durable catalyst, it is not helping this project make a commercializable material. The isotherm degradation appears to give a more stable catalyst, but it appears still to lose significant activity from the other materials and in the initial cycles. It seems like the modeling and characterization should be applied to the post-cycled material to understand the stable structure.

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 is adequately leveraging several collaborations within the Electrocatalysis Consortium (ElectroCat) and externally.
- The project has an excellent team that contributed strengths of each individual institution to make the team strong. Slide 16 communicates this very clearly.
- The collaboration consists of Northeastern University and Lawrence Berkeley National Laboratory (LBNL), with ties to industry and contribution from ElectroCat.
- The team made use of ElectroCat, plus other partners.
- The partnership with LBNL seems limited. Other collaborations are not presented, or work is isolated to Northeastern University or an IBAD subcontractor.

Question 4: Relevance/potential impact

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

- This project has a high potential impact, as PGM-free ORR catalysts are needed to significantly reduce fuel cell costs. It is difficult to compete with platinum for these systems; therefore, higher loadings and thicker catalyst layers are required. If this approach leads to higher activity in PGM-free systems, various barriers to reducing costs of fuel cell systems may be overcome.
- The project's focus on novel approaches to creating active sites is critical to advancing PGM-free catalysts. Advances in increasing active-site density are necessary for realizing adequate PGM-free performance to compete with PGM catalysts.
- The project is helping to advance state-of-the-art PGM-free catalysts. PGM-free catalysts could help lower the cost of PEMFCs with further development. The only weakness is that DOE has shifted focus to heavy-duty fuel cells, and the PGM-free materials seem a long way off from the performance required for that application. Furthermore, original equipment manufacturers do not seem interested in PGM-free catalysts.
- PGM-free is far from competitive with Pt, and the path to competitive performance is unclear. Since a stable material is made from the isothermal carbonization, it seems like developing that material at BOL with a high site density is a preferred path.
- As presented, this project has minimal visibility in the short-term impact on advancing progress toward the DOE research, development, and demonstration durability goals.

Question 5: Proposed future work

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

- The proposed future work is logical for achieving most of the project objectives. It is not clear whether the project objectives will be obtained concurrently with the same catalyst. For example, the team is close on the activity target, but the performance lasts for only three cycles. It was not communicated clearly how many cycles the project needs to achieve or which of the catalysts could achieve both activity and durability targets. Also, more details regarding how the team will improve understanding of the $Fe\text{-}N_4$ degradation mechanism would be useful.
- The future work aligns fairly well with the relevant and listed remaining challenges and barriers. However, the future work may need to be expanded to include an MEA integration aspect. Moving from the development of a catalyst material to a functional electrode requires the consideration of many variables. This process is highly important for novel PGM-free catalyst materials to the device level. This is currently missing entirely from the proposed work.
- The ongoing work on the MMCs with Fe is worthwhile, but it is unclear whether the work can yield a single FeCo active site or two separate sites with Co and Fe.
- The researchers should focus on achieving stability of the resulting formulation and batch consistency, and they should specify their approaches in greater detail, providing justifications toward every approach. There are too many global statements in slide 17, resulting in questioning of the reality of these far-reaching goals. Additional background should be provided on the demonstrated exchange of experiences with project partners and collaborators in choosing one or another route and justifying the focus of the next budget period.
- The proposed work on understanding the degradation mechanism seems important. Increasing the site density might make for a better BOL catalyst, but unless the performance can be maintained by improving the durability, that seems like a fruitless goal.

Project strengths:

- The project has a strong team and is using a wide array of synthesis techniques, characterization techniques, and site density measurement methods to develop PGM-free catalysts.
- The project uses the Northeastern University team's extensive experience in surface modification techniques and catalyst synthesis. A variety of options are demonstrated, with thorough assessment of resulting formulations.
- The project is developing more active and durable PGM-free electrocatalysts and appears to be making good progress. The strength of the project is developing stable materials post-cycling (isothermal carbonization). The team should use extensive characterization to understand that structure, then synthesize that as an initial material.
- The clear strength of the project is the innovation in catalyst synthesis and active-site characterization.
- This project has a strong approach, backed with extensive diagnostic capabilities and sound methods.

Project weaknesses:

- The project does not have any obvious weaknesses, although the translation of the catalysts to electrodes in fuel cells could be given more consideration in the future.
- The engineering challenges of the proposed synthetic routes are not worked out specific to uniformity, cleanliness, etc. The project runs as if it were funded by the Office of Basic Energy Sciences and not the Hydrogen and Fuel Cell Technologies Office.
- The main weakness with the project is the communication of the milestones and whether they are being achieved on time concurrently with the same catalysts.
- The project is missing a device-level proof of concept and development.
- The performance is still far away from being competitive with Pt.

Recommendations for additions/deletions to project scope:

- It is recommended that the project drop the IBAD scope; focus on decreasing activity loss after the accelerated stress test with one transition metal, not two; and clarify the role of porosity with obtained formulations.
- The project should add an MEA integration aspect that makes strong use of the modeling capabilities combined with hydrogen–air performance at relevant stoichiometric flow rates to understand limitations and optimize an MEA-level device that contains the developed catalyst materials.
- The isotherm degradation appears to give a more stable catalyst, but it appears still to lose significant activity from the other materials and in the initial cycles. It seems like the modeling and characterization should be applied to the post-cycled material to understand the stable structure.
- More details and introduction of creative techniques to improve the understanding of Fe-N_4 degradation mechanisms are recommended.

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

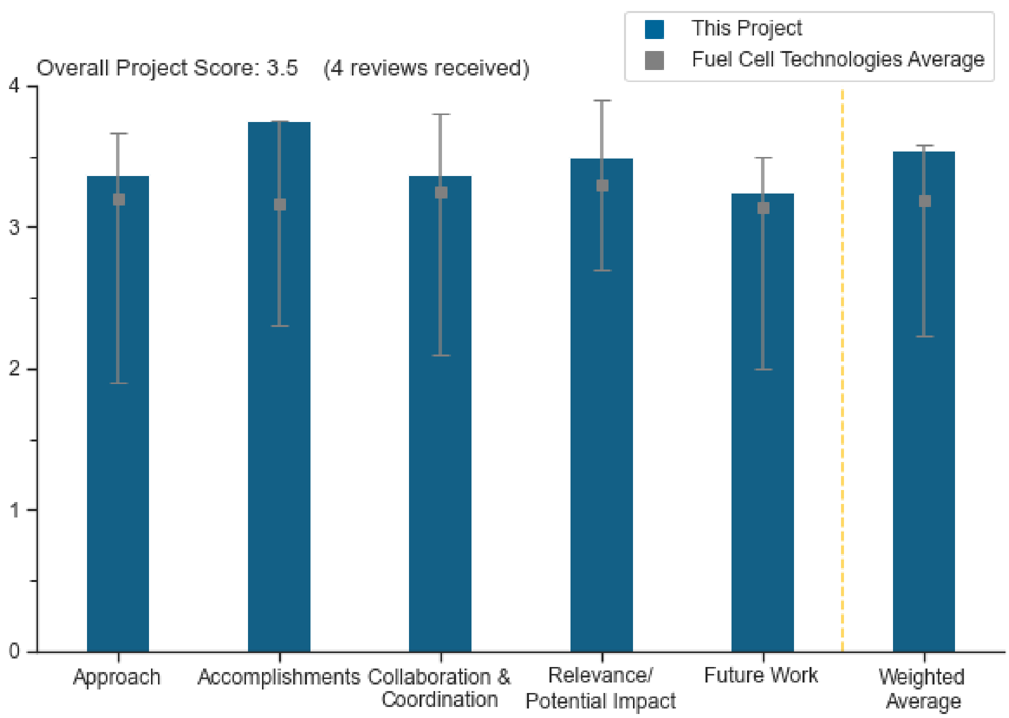
Jian Xie, Indiana University–Purdue University Indianapolis

DOE Contract #	DE-EE0008417
Start and End Dates	10/1/18 to 12/31/21
Partners/Collaborators	University at Buffalo, United Technologies Research Center, Electrocatalysis Consortium
Barriers Addressed	• Performance • Cost • Durability

Project Goal and Brief Summary

Indiana University–Purdue University Indianapolis (IUPUI) will use controllable synthesis to design and develop advanced hierarchically porous carbon sphere M-N-C catalysts for platinum-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 a mesopore structure and (2) construct an ideal Nafion™ ionomer–catalyst interface within a catalyst layer of MEAs.

Project Scoring



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

Question 1: Approach to performing the work

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

- The approach is very good in overcoming some of the most important barriers that exist for PGM-free catalyst material sets. The thickness and lower activity of the catalyst layer, when compared to Pt catalysts, create a number of challenges tackled by the project approach through investigation of morphology and interfaces, combined with optimization and characterization.
- The novel aspect of catalyst development in this project is the tuning of the particle structure and internal porosity for higher activity of a zeolitic imidazolate framework (ZIF)-derived catalyst from the University at Buffalo. The process is yielding very high activity. The performance with alternative metals (Mn, Cr, Ce) is very encouraging. Although the electrode integration, particularly the ionomer–catalyst interface, is outlined as a key aspect of the project, there does not seem to be an approach or tasks specifically addressing that interface or the ionomer integration. IUPUI's coating system is preparing catalyst-coated-membrane MEAs, producing uniform dense electrodes without large aggregates and with high activity. These results are likely due to the high concentration of well-dispersed catalysts at the membrane interface. It is not clear why relative humidity (RH) between 50% and 100% has no impact on performance and performance is higher at 50% RH, which raises some concern in regard to the testing, as the results are inconsistent with the expected effects of RH with thin and thick electrodes.
- Improving catalyst accessibility to improve activity is a great approach. Hierarchical porosities should result in better performance. It would be good to see more in-depth study of the correlation between pore structure and activity by using the Electrocatalysis Consortium's (ElectroCat's) capabilities existing in the national laboratories.
- The approach is to design interfaces and catalysts and is quite comprehensive, which also means it might be tough to understand and optimize. The use of rational design and the inherent built-in feedbacks are good, although it is not clear how things will evolve with time, as most of the focus appears to be more on initial performance. The ZnO template is a good idea, but it is not clear whether these structures are maintained during testing and operating conditions. There is not a clear pathway to surface area and loadings that are high enough that mass transport does not become limiting at higher current densities, as shown in the flooding at high RH. Modeling could help. The ink studies are not comprehensive and just seemingly correlative.

Question 2: Accomplishments and progress

This project was rated **3.8** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Progress is very impressive. Even at 65% completion, the project has met or exceeded most of the end-of-project goals, especially those related to both catalyst activity and MEA performance. The fuel cell hydrogen–oxygen performance of >50 mA/cm^2 (current density) at 0.9 V iR-free is indeed very impressive. The 500 mW/cm^2 power density at 0.67 V in hydrogen–air at 150 kPa is also very impressive.
- The project met or exceeded all targets and milestones with respect to project goals and DOE metrics. Impressive hydrogen–air performance was conducted at 150 kPa. There is a good comparison to current PGM-based cells and their typical operating conditions in the laboratory. Independent hydrogen–air performance is very impressive with regard to RH ranging from 50% to 100%. This seems to indicate that the approach works. The work is combined with advanced microscopy and other diagnostic methods, as it should be, to allow better insight into the reasons this approach works.
- The project is making excellent progress in terms of catalyst activity and PGM-free cathode fuel cell performance.
- The accomplishments in catalyst activity appear good in terms of meeting project goals, with the exception of stability. It is not clear whether CO_2 has evolved from the amorphous carbon that results at end of life or just from the graphitized carbon. It is not clear how pore volume is measured and whether that measurement is accurate for the operating conditions. It is not clear what the estimated surface area of the catalysts is or what pores are active, especially under different RH. The overall porosity and thickness of the different catalyst layers were also not presented. This is especially important with respect to the cracks,

as those may dominate the response. It is not clear why cracks would increase with the ionomer–carbon (I:C) ratio if the ionomer is acting more like a binder. More analysis is required. The optimization is relatively trivial and simple, especially compared to the catalyst design, and so the objectives and focus must be realistic.

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 core project consists of a combination of university and industry partners that complement each other. The project seems to make good use of ElectroCat.
- There appears to be good collaboration between the University at Buffalo and IUPUI, and ElectroCat laboratory support is being used.
- Collaboration between IUPUI and the University at Buffalo is excellent. However, the project could engage ElectroCat better to develop a better fundamental understanding of how catalyst porosity affects catalyst activity and how MEA porosity affects MEA performance.
- Coordination within ElectroCat appears good, although publications seem to be mainly from Gang Wu and collaborators, with none from the principal investigator.

Question 4: Relevance/potential impact

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

- High-potential-impact, PGM-free, oxygen reduction reaction catalysts are needed to significantly reduce the cost of fuel cells. For these systems, competing with Pt is difficult; therefore, higher loadings and thicker catalyst layers are required. Moving away from Fe-based systems is difficult, but since iron can contaminate the membrane, the PGM-free system could be of significant benefit with regard to degradation processes as well.
- The development of new PGM-free catalysts with high activity is well-aligned with the DOE goals and objectives.
- The project is meeting beginning-of-life technical targets for non-PGM systems. Stability needs to be improved, but initial results hold promise of impact; however, for heavy-duty, efficiency needs make it crucial to get higher performance at higher voltages.
- Potential impact is high. It would be good to see the project move to larger-area cells (perhaps 25 or 50 cm^2) and operate them closer to 2.0 stoichiometry of air.

Question 5: Proposed future work

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

- Future work is good and fits well into the continuation of the project.
- The focus on durability is great. However, the project should engage ElectroCat better to improve the understanding of both performance and durability. Understanding mechanisms and processes that correlate porosity to performance and durability will be very valuable for the community.
- A greater future emphasis on stabilizing the PGM-free catalyst's performance would be good. The project is lacking studies focused on degradation, which is becoming the leading challenge for these PGM-free catalysts.
- The project is wrapping up. Focusing more on understanding than on fabrication of new materials is recommended. Stability, in particular, should be a focus. Delta-V analysis should be conducted to understand the nature of the losses.

Project strengths:

- Performance is very impressive. Preliminary durability data are also impressive. This is a great team making excellent progress.
- The project has strong achievements. Operating at relevant hydrogen–air operating conditions allows easy comparison of results and demonstrates the accomplishments well.
- The project has a good design for initial catalyst activity, including multiple approaches. Overall progress and results in terms of activity are good.
- A notable strength of the project is the high catalyst and electrode beginning-of-life activity.

Project weaknesses:

- Durability aspects of the system need to be improved. The future work addresses these aspects.
- Rational design for the ink does not really exist. The project focuses only on changing the I:C ratio and seeing the impact on performance. It is entirely correlative, with not much understanding. The work on durability is nascent in terms of understanding the mechanism loss. Additional accelerated stress tests, such as open circuit voltage hold tests or some RH cycling, could be informative.
- The key weakness is the limited emphasis on durability, beyond applying accelerated stress test characterization.
- There are not enough systematic or fundamental studies that correlate catalyst and MEA properties to performance.

Recommendations for additions/deletions to project scope:

- The project team should consider the following:
    - MEA performance increases with decreasing I:C ratio (down to 0.5), whereas mesopores are optimized at I:C = 0.6. The project needs to add better MEA characterization to figure out mass transport resistance and try to correlate it to MEA pore structure.
    - The project needs to add correlating hierarchical porosity to accessibility, i.e., characterizing the post-treatment effect on the micro/meso porosity and correlate that to accessibility.
    - It is important to make 100% sure that there is no Pt contamination on the cathode (x-ray fluorescence [XRF] can be used). Maybe the project can use two 211 membranes instead of one 212, to make analysis easier, and check for Pt on the cathode side of end-of-test MEAs.
- For even better comparison to PGM systems, it would be necessary also to operate with stoichiometric flow rates during the experiment. It would also be important to clearly identify what role the hierarchical structure plays in achieving the measured performances.
- Carbon corrosion and CO_2 coming out of the system should be measured. Modeling is recommended to understand the losses in the system.

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

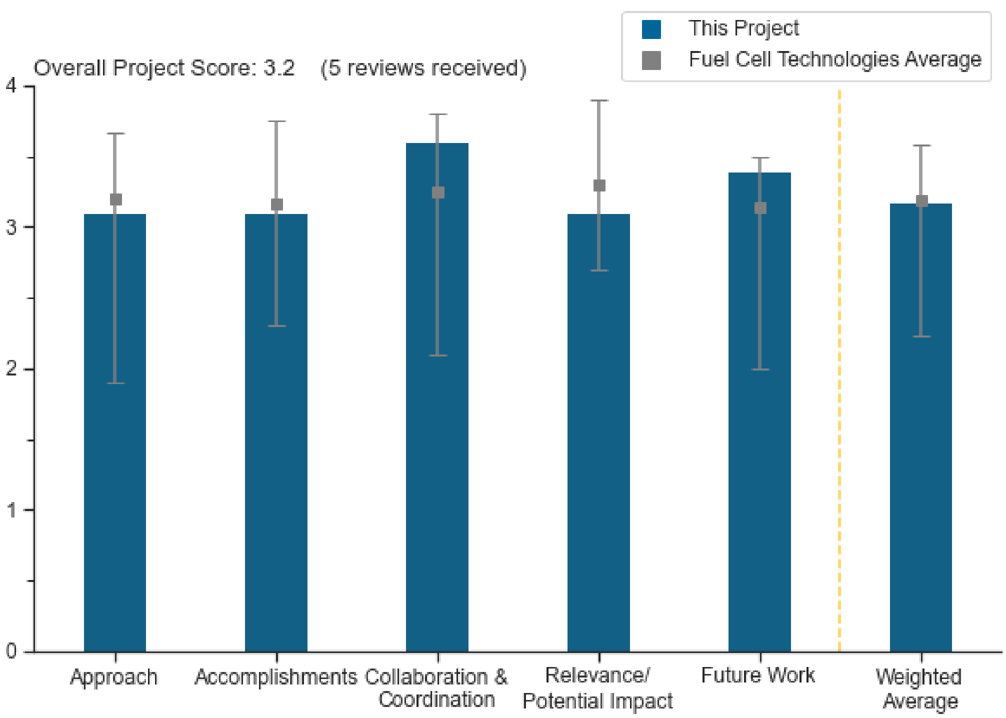
Alexey Serov, Pajarito Powder

DOE Contract #	DE-EE0008419
Start and End Dates	1/2/2019 to 12/31/2021
Partners/Collaborators	IRD Fuel Cells, University of Hawaii, Natural Energy Institute
Barriers Addressed	• Increase activity of platinum-group-metal-free (PGM-free) oxygen reduction reaction (ORR) catalysts • Decrease cost of PGM-free catalyst manufacturing • Increase durability of PGM-free catalysts

Project Goal and Brief Summary

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 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



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

Question 1: Approach to performing the work

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

- The focus of this project is the development of a scalable method for synthesis and manufacturing of PGM-free electrocatalysts (Fe-N-C types) for ORR for polymer electrolyte membrane fuel cell (PEMFC) applications. PGM-free ORR catalysts are integrated into industrial-quality MEAs through cathodic catalyst layer design (optimization of ionomer type and loading, deposition method, and type and concentration of additives). The team studied the electrode thickness to increase performance.
- The approach for this project was more on the applied end (versus the fundamental end) of the spectrum. This makes the project unique compared to other Electrocatalysis Consortium (ElectroCat) projects, which is positive, because the project puts more effort into getting these materials to function in an MEA. The MEA development approach taken was logical. The main criticism of the project approach is that there is a high risk that the catalyst activity and durability trails PGM catalysts by so much that the materials and our fundamental understanding may need to drastically improve before useful MEAs can be produced.
- The team has taken a systematic approach, starting with catalysts and proceeding to MEAs, then to using the electrochemical method to characterize the MEA performance, and then to elucidating the other important parameters, such as mass activity, current density, and catalyst stability. It was unclear whether the go/no-go decision point was for Year 1 or Year 2. The use of pore formers to increase the catalysts' surface area is a good approach.
- The team uses a method of preparing PGM-free ORR catalysts that is somewhat different from the most popular methods currently used in this space, such as metal–organic frameworks. The nature of the approach is essentially the same as others, i.e., using Fe- and N-based precursors plus pore formers. The pore former selected by the team appears large compared to others, which could contribute to performance on par with them. Silicon was used in the precursor, which would trigger the undesirable use of hydrogen fluoride for removal. The authors could be more specific about this. Ultimately, durability is the key challenge in PGM-free ORR catalysts in the acidic electrolyte. There was no clear path or hypothesis from the presentation to articulate how to address this challenge.
- The approach is focused on achieving activity and durability targets for PGM-free electrocatalysts.

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 team has conducted a sufficient number of experiments, from catalyst preparation to testing. The result is also good, on par with other state-of-the-art catalysts in this field. The team also successfully met the go/no-go decision criteria. The durability, though improved, appears to remain the Achilles' heel for this type of PGM-free ORR catalyst.
- The progress was slow because of the pandemic. The team achieved the project targets and the go/no-go ElectroCat target: demonstrate 0.044 A/cm^2 at 0.9 V (iR-free, H_2/O_2 configuration, 1 bar O_2, 80ºC, 100% RH). The team scaled up manufacturing capabilities of the catalyst.
- The biggest accomplishment from the work appears to be the demonstration of an MEA with a PGM-free catalyst that survives 30,000 cycles reasonably well, although still below DOE targets. The project also developed an interesting machine learning tool to determine the importance of MEA factors on resistance. Such a tool would definitely be useful to guide development of electrodes and MEAs with PGM-free catalysts. The downside with the project accomplishments is that the materials developed and the MEAs have very low performance relative to DOE targets. Very little was provided on characterization of the PGM-free materials or on the details of how they were made. The fundamental insights provided by other ElectroCat projects were lacking in this project.
- Significant progress has been made toward project goals and optimization of the catalyst layer design. Although the trend in durability improvement looks very promising, performance at 0.8 A/cm^2 is low.

- The surface area of catalysts using the PF-1 and PF-2 pore formers did not provide either Brunauer–Emmett–Teller (BET) or mercury intrusion porosimetry (MIP) data; it is hard to judge whether these pore formers work as designed, particularly for mesopores. An understanding of the porosimetry of the catalyst and MEA is important, as the mesopore plays a critical role in catalyst and MEA performance. The electrode layer is too thick to have good performance and is the root cause of the low performance. MIP is needed to analyze pore size distribution and pore volume to correlate performance and structure properly (slide 10). Proton conductivity was improved by the use of low-equivalent-weight (EW=720) ionomer and additives, and it is not apparent which one played the major roles here (slide 10). Clarity is needed on how the H+ conductivity of the catalyst layer was measured and whether it was measured by high-frequency resistance. It is strongly suggested that MEA durability, the current loss at 0.80 V, and voltage loss at 0.80 A/cm^2 be provided. It seems losses between 60 and 119 mV are quite significant. There is some interesting progress on ORR mechanisms, but the team should make a clearer and better summary.

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 key personnel are fully qualified to manage this project, as demonstrated by their publications on work in this and related fields, and the proposal clearly and completely defines the roles and contributions of each team member, including the financial support of partners.
- The project team is diverse and led by a respected, world-class scientist. There were no significant weaknesses in the team.
- The project had a very well-coordinated effort between Pajarito Powder, University of Hawaii, IRD Fuel Cells, and ElectroCat.
- The team is working closely with ElectroCat.
- The team showed good collaboration between themselves and with key partners.

Question 4: Relevance/potential impact

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

- Eliminating the PGM while maintaining the current level of specific power, power density, peak energy efficiency, and durability will significantly accelerate the deployment of fuel cell systems.
- The project is critical to the Hydrogen and Fuel Cell Technologies Office and has potential to significantly advance PEMFC commercialization toward DOE research, development, and demonstration goals and objectives. The industrial company that participated in this project is critically needed to accelerate the progress of the PEMFC commercialization.
- The biggest potential impact from the work was the demonstration that PGM-free MEAs with reasonably good durability can be produced, providing hope for further PGM-free MEA development. The modeling tools could also prove impactful for the industry. The biggest concern with potential impact is that DOE has switched focus to heavy-duty vehicle applications, and the materials and MEAs developed on this project are not competitive with the new DOE targets and objectives. The project did not yet investigate the mechanisms for degradation that may make the project more impactful.
- The fuel cell application is now shifting toward use in heavy-duty vehicles, in which the durability requirement becomes more stringent. The PGM-free ORR catalyst becomes less relevant unless some major breakthrough can be achieved.
- The project is relevant to DOE goals on commercialization of PGM-free fuel cells. However, performance of PGM-free catalysts is not sufficient to make a transformational impact in the fuel cell community.

Question 5: Proposed future work

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

- The project team proposes logical next steps to meet the proposed project objectives. The post-accelerated-stress-test (post-AST) characterization could provide interesting insight to degradation of PGM-free catalysts in an MEA.
- Proposed future work is well-organized and feasible. Machine learning is a good approach for rational design of the catalysts and MEA structure.
- Proposed future work is focused on achieving DOE targets in activity and durability.
- The proposed work is well-presented and will lead to meeting the proposed milestones.
- Durability was identified as one of the future work areas, which is important. From a scientific point of view, it is not clear why MEA configuration can significantly improve durability.

Project strengths:

- The strength of this project is that it takes a systematic approach, from the catalyst to the MEA, with the goal to develop a scalable method for synthesis and manufacturing of PGM-free electrocatalysts (Fe-N-C types) for ORR for PEMFC application. This is particularly important for the PEMFC commercialization since the early involvement of the industrial partners will greatly promote the advancement of the PEMFC applications in transportation and other applications, such as renewable energy utilization and hydrogen at scale. The team has good collaboration with ElectroCat and other institutes.
- The project strength is in the combination of strong catalyst synthetic skills with thorough validation of the catalyst performance, use of novel theoretical tools such as machine learning, and modeling of full polarization curves to understand and expedite the design of new materials and catalyst layers.
- The team focuses on the commercialization of the project catalysts and has the venue and capability to do so. It will be exciting to discover fuel cell applications that can serve as early adopters of this type of catalyst. The team also works diligently to address not only the catalyst but also the MEA aspect of the development, which helps to validate the approach. The team demonstrates the capability to produce larger quantities of catalyst.
- This project is unique in comparison with other ElectroCat projects because it puts more effort into getting PGM-free materials to function in an MEA. The durability data to 30,000 cycles was nice to see. The machine learning model was a strong point as well. The project had a strong principal investigator and team.
- There is a clear focus on scale-up and catalyst manufacturing.

Project weaknesses:

- The project does not contribute greatly to the fundamental understanding of how to improve PGM-free catalysts to the point where they could be competitive with PGM catalysts.
- The team has difficulty in controlling the particle size; the porosity of the particles will have a negative impact on long-term stability.
- The project team made good progress but needs to accelerate the effort to meet the project goals and DOE targets. The project status shows that the team met the project milestone but still needs to make great effort to meet the end goals.
- More focus should be given to catalyst durability improvement.
- Fe-N-C electrocatalysts are not new.

Recommendations for additions/deletions to project scope:

- The planned post-AST testing could provide interesting insight as to why the MEA degrades. If the techniques planned do not fully explain the degradation, the team should consider expanding the array of techniques to include electrochemical diagnostics, energy dispersive x-ray spectroscopy (EDX) mapping, pore measurements, or more advanced techniques.

- The proposed team is doing a good job.
- As the project ends in six months, there is not enough time to change its scope.

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

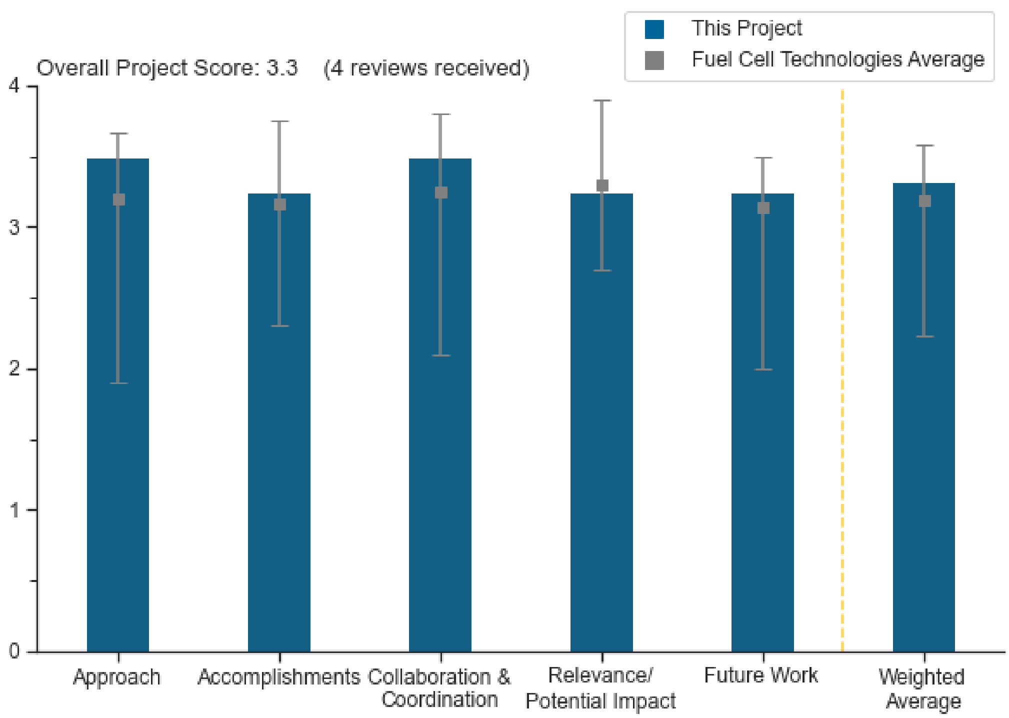
Chulsung Bae, Rensselaer Polytechnic Institute

DOE Contract #	DE-EE0008432
Start and End Dates	4/1/2019 to 4/30/22
Partners/Collaborators	Los Alamos National Laboratory
Barriers Addressed	• Performance • Cost • Durability

Project Goal and Brief Summary

In this project, Rensselaer Polytechnic Institute (RPI) will develop a series of innovative cyclic olefin copolymer (COC)-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 COCs with tunable backbone rigidity, (2) incorporate alkyl chain-tethered quaternary ammoniums of different structures into 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 platinum-based and platinum-group-metal-free (PGM-free) catalysts. The reinforcement of AEM will allow RPI to produce thinner (e.g., 10–25 µm) membranes, affording lower area-specific resistance and better water management in MEAs, particularly with a PGM-free catalyst.

Project Scoring



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

Question 1: Approach to performing the work

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

- The approach of combining ethylene and norbornene has the potential to access low-cost, durable AEMs. The project strategy is a very clever idea: incorporating a non-functional phenyl ring onto the norbornene monomer to enable metallocene catalyst polymerization, followed by functionalization of the phenyl group. The team is focused on the important parameters necessary to advance AEM technology.
- The project is making saturated hydrocarbon and reinforced membranes for high-performance AEMs by using low-cost monomers and polymers, no heteroatom (oxygen or nitrogen) in the backbone, high alkaline chemical stability, tunable rigidity (by varying the ratio of ethylene and norbornene/cyclic olefins in the backbone), and pore-filling reinforcement for enhancing durability and extending lifetime of MEAs.
- The team is making good progress. The work is feasible and will likely be successful.
- This is good work, but the U.S. Department of Energy is now funding at least three aliphatic AEM projects, and the clear advantages or differences between the projects are indistinguishable.

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 progress made by the principal investigator's group is good. The researchers were able to make some really nice progress in developing very nice reinforcement of their AEM. This was also highlighted by the impressive improvement in mechanical properties. Fuel cell performance is good, but it is unclear whether the fuel cell performance is tied to the specific humidification of the cathode and anode.
- The RPI team has shown the feasibility of the synthetic approach, as well as made prototype fuel cell membranes. The researchers are target-driven while still developing the necessary system understanding. Focused performance, mechanical strength, and chemical durability show a well-rounded approach.
- The project has prepared membranes, used reinforcement composite, characterized membranes, and studied hydrogen oxidation reaction catalyst electrodes.
- Some of the targets could be more suited to future applications. For example, peak power density is nice, but it is not really an important practical value. A much better metric would be voltage at 1.0 A/cm^2. The current DOE target is 0.65 V at 1.0 A/cm^2 at steady state (it should be at least a one-hour test). The researchers already seem to meet that target in the pol curves, but it is not clear how stable the performance is, especially given that the membrane is drying out, given the increase in high-frequency resistance shown on slide 12. Additionally, some of the performance variables could be moved closer to reality. For example, the anode flow rate is 1.4 L/min, which is huge, because it is likely to avoid anode flooding. It is also worth noting here that the overall performance with these materials is below the state of the art, and that should be addressed.

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 good collaboration between RPI and Los Alamos National Laboratory (LANL), with good division of labor; the synthesis is mostly at RPI, and the characterization is mostly at LANL.
- The collaboration at this stage of this effort is perfect.
- Collaborations between RPI and LANL are complementary, and the team structure is productive.
- The collaboration seems fine.

Question 4: Relevance/potential impact

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

- The operating environments for AEM fuel cells or electrolyzers are very challenging; however, this team has the potential to address many of these issues. If successful, the RPI technology has the potential to meet many, if not all, of the DOE targets.
- There have been great improvements in AEM technology, but more is needed in terms of CO_2 tolerance and highly active non-PGM catalysts with good stability, and this project is targeting these needs.
- This is good work, but it is in a crowded area. The current results here are similar to the other aliphatic projects, and it is difficult to declare which project is better than the other at this point.

Question 5: Proposed future work

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

- The most relevant future work should be the following:
  - Mechanical property improvement by pore-filling reinforcement and use of higher-molecular-weight cyclic olefin copolymer polymers
  - Evaluation of the AEM performance in a fuel cell long-term test with minimal recoverable and unrecoverable loss
  - Pore fillers for strength and water in membrane optimization.
- Durability will be the next focus, as it should be, to give an idea of the stability of this material. It is interesting that this material was not utilized as the ionomer since it may be a good candidate; perhaps this is something that should be evaluated.
- The proposed work is fine.
- The future work is clear and on point with DOE targets. However, there are very ambitious goals, such as a catalyst layer and AEM compatibility, that will likely exceed the time allotted in this project. The optimization work is not clearly defined. There are multiple layers of polymer composition, crosslinking, support, and process conditions that will be challenging to complete.

Project strengths:

- A key strength of this approach is the ability to incorporate a true commodity monomer (ethylene) into a highly engineered ionomer polymer. Not only does this lower ionomer cost, but ethylene units are thought to be among the most chemically stable in an alkaline environment. The RPI–LANL team comprises some of the most recognized experts in the AEM field. Their understanding of the critical barriers and strategies to address these barriers is of the highest caliber.
- The current strength of this project is the very good reinforced membrane development. The chemistry work is good but is not unique, as many researchers are developing these types of polymers.
- The project has a good team with complementary skills. The project builds well from what is known in the literature. The team is capable of executing all of the aspects of the project.
- There is good synthesis at RPI and characterization at LANL.

Project weaknesses:

- While a very clever approach, optimizing membrane copolymer ratios, crosslink density, support type, membrane thickness, and other properties will be challenging. There are many very good AEM candidates in the literature. More work is needed to demonstrate that this approach has the potential for superior performance, durability, and cost.
- More investigation into this material as an ionomer should be pursed since the material has little aryl groups that have been found to adsorb onto the catalyst with time, affecting durability.

- The team says mechanical properties are being improved, but the characterization of membrane strength is not clear. The relative ranking of membrane strength at 50% relative humidity by stress–strain is useful but does not apply to strength in fuel cell operating conditions, which have much higher water levels.

Recommendations for additions/deletions to project scope:

- This team should focus on membrane optimization, crosslinking, and hydration issues by using a standard anode and cathode catalyst for fuel cell performance comparisons. Any other catalyst work seems a diversion.
- The project should make targets to align better with those laid out in Simon T. Thompson et al., *Journal of The Electrochemical Society* 167 084514 (2020). The project should push the team to be more ambitious with its performance and durability goals, as others have already achieved much better than the final project goals here.
- Ionomer investigation would be beneficial.

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

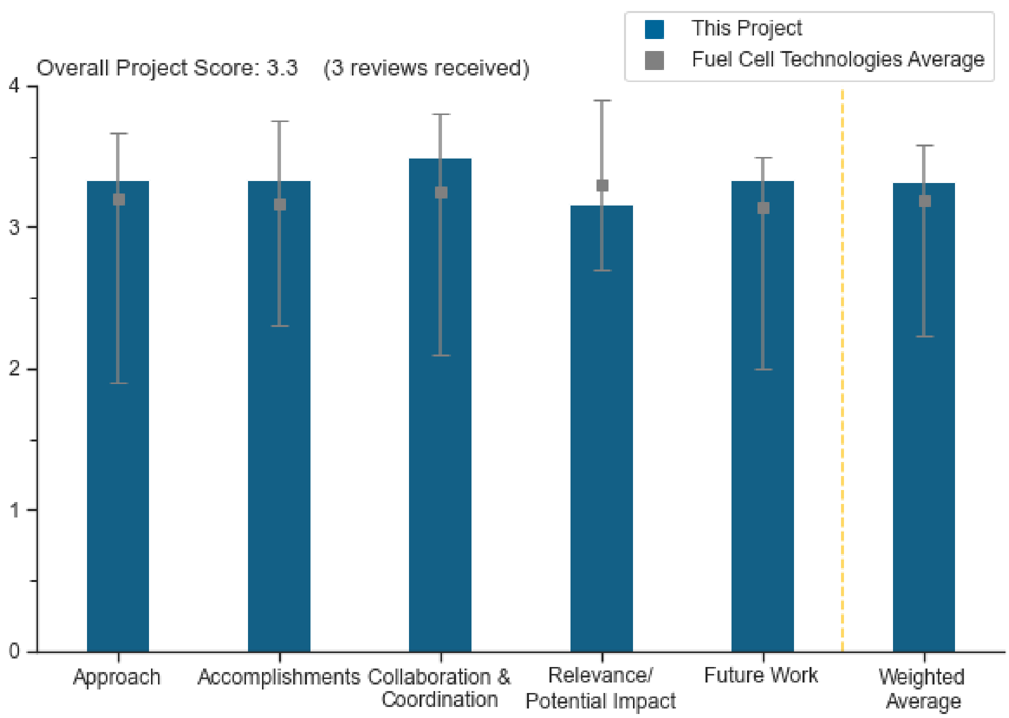
Michael Hickner, The Pennsylvania State University

DOE Contract #	DE-EE0008433
Start and End Dates	10/1/2018 to 3/31/2022
Partners/Collaborators	Pennsylvania State University, University of South Carolina, National Renewable Energy Laboratory, 3M Company
Barriers Addressed	• Durability • Cost • Performance

Project Goal and Brief Summary

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 exceptional 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



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

Question 1: Approach to performing the work

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

- Membrane-making using stable saturated hydrocarbons, using a catalyst specific to AEM, improving cell performance, and using neutron diffraction to study and optimize polymer and water management are approaches that cover all targets.
- The project's focus on large-scale batches and repeatability is important, and it is a valuable part of the approach. The work to focus on such issues should be commended.
- This is good work, but there are now at least three aliphatic AEM projects that the U.S. Department of Energy is funding, and the clear advantages or differences between the projects are indistinguishable.

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 performance results are excellent and above the targets in the milestones. The durability testing, which is what the milestones focus on, are written for 60°C, while this team is showing testing at 80°C, a more challenging temperature. The team is showing 0.69%/100 hours, which translates to 13.8%/2000 hours, which is above the goal of 10%—but again, the work is at a higher temperature. Also, this rate is projected from the first few hundred hours, not actually shown. Overall, this project appears to be on track to hit the final project milestones.
- All targets have been met except the neutron studies, which are behind because the pandemic shut down the National Institute of Standards and Technology (NIST) facility.
- The performance and durability shown were impressive and a good step forward. However, the performance does look similar to Paul Kohl's (Georgia Tech) materials.

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.

- This appears to be a well-integrated team with appropriate partnering.
- All team members share different tasks, such as synthesis at The Pennsylvania State University, catalysts at 3M Company, and characterization at the National Renewable Energy Laboratory and NIST.
- The partners right now are sufficient.

Question 4: Relevance/potential impact

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

- This project is demonstrating steady progress on making the changes needed to advance AEMs: stable saturated hydrocarbon membranes, a catalyst specific to AEM fuel cells, stable cell performance, and water management. The effects of carbon dioxide (CO_2) and high-performance stable non-PGM catalysts still need work.
- This is good work but in a crowded area. The current results here are like those of the other aliphatic projects, and it cannot be declared which project is better than the other at this point.
- The AEM performance is good, but there is still PGM in the electrodes, and then there is the CO_2 issue to address. The likelihood of AEMFCs ever beating polymer electrolyte membrane fuel cells remains in pretty serious question. This is not a negative for this project.

Question 5: Proposed future work

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

- The future work suggested by the team is relevant.
    - The project will synthesize larger-scale batches of polymer to facilitate membrane testing; this is mostly complete, but more material will be filled in as needed.
    - The project will measure water transport in membranes using pulse field gradient–nuclear magnetic resonance (PFG-NMR) and connect to cell water transport observations using water balance measurements.
    - The project will fabricate expanded polytetrafluoroethylene (ePTFE)-supported membranes for cell testing.
    - The project will finalize cell testing electrodes and conditions for demonstration of 2000-hour lifetime end-of-project goals.
    - Someone has to step up on effects of CO_2.
- The future work is reasonable and focused on proving out the final milestone.
- The progress made in measuring the water diffusion and relating it back to fuel cell performance was interesting. In addition, investigation of this material as an ionomer should be pursued since the material has little aryl groups that have been found to adsorb onto the catalyst with time, affecting durability. Perhaps looking at polyethylene instead of PTFE support could reveal better ionomer impregnation.

Project strengths:

- This project has delivered some very nice fuel cell performance and durability, which is very promising.
- This project has good teaming and good progress, except for the NIST water studies.

Project weaknesses:

- There are none, but someone must address CO_2—maybe examine a CO_2-rejecting membrane backbone.
- One weakness is that the material is only being used as a membrane; it should also be looked at as an ionomer. It would be interesting to see if the performance is primarily due to the GT ionomer or not.

Recommendations for additions/deletions to project scope:

- Reviewers did not provide comments in response to this question.

Project #FC-309: Polymerized Ionic Liquid Block Copolymer/Ionic Liquid Composite Ionomers for High-Current-Density Performance

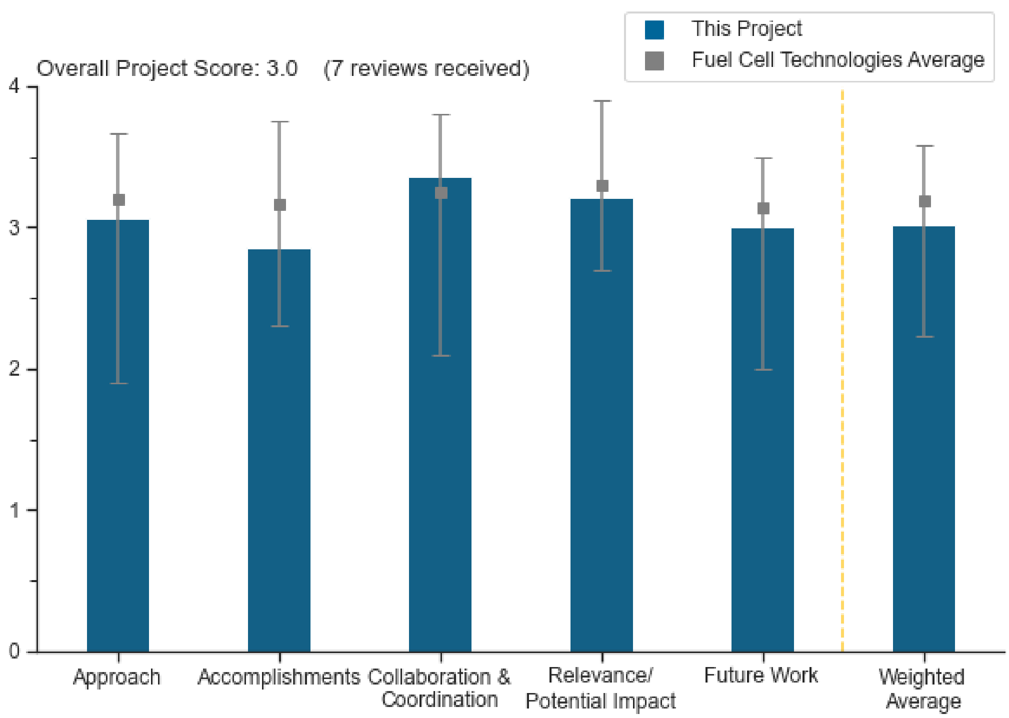
Joshua Snyder, Drexel University

DOE Contract #	DE-EE0008434
Start and End Dates	10/1/2018 to 12/31/2021
Partners/Collaborators	Drexel, Texas A&M, General Motors, National Renewable Energy Laboratory
Barriers Addressed	• O_2 transport through ionomer films • Ionomer adsorption on catalyst • Inaccessible catalyst in porous carbon • Distribution and retention of IL in catalyst layer • Humidity tolerance at HCD

Project Goal and Brief Summary

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 the 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



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

Question 1: Approach to performing the work

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

- The objectives of this project are to improve the performance of fuel cell cathode catalyst layers through reduced sulfonate interactions and lower oxygen transport resistance. The project appears well-suited to prove its hypotheses, and replacing incumbent PFSA ionomers is an interesting concept.
- The approach of using ILs and PILBCPs to enhance electrode performance is innovative and promising. The strong focus on PILBCP synthesis and MEA integration and testing, coupled with fundamental electrochemical studies, makes this a durable approach.
- The approach of using ILs to try to improve Pt utilization and decrease ionomer–catalyst-poisoning-type interactions has potential to improve performance and durability of oxygen reduction reaction catalysts. Tethering ILs to a polymer should help retain the IL in the catalyst layer. The approach of using a block copolymer that also contains a proton-conducting block should alleviate dilution effects and maintain high proton conductivity in the catalyst layer. Slide 14 suggests that the IL can penetrate into the pores of porous carbon supports to provide proton transport to catalyst particles within the pores; however, once the IL is immobilized by tethering to a polymeric backbone, it is unclear whether it will be able to enter these pores. The results shown appear to be for Pt/Vulcan. The project does not appear to be investigating different types of carbons to determine the impacts of carbon support porosity on the tethered IL's benefits or comparing the tethered IL's benefits to use of porous carbon supports to decrease catalyst–Nafion interactions.
- The development alternative ionomer to Nafion is meaningful, and the PILBCP/IL composite ionomer is novel.
- The project has an interesting and novel approach to improving cathode performance.
- While the concept seems applicable, the insolubility in normal solvents and use of Pt/Vulcan are major drawbacks to the approach.
- This presentation was hard to follow. It is clear that the goal is to develop a replacement for PFSA ionomers, but then there is a fair amount about PILBCP–Nafion mixed ionomers. One can see how this could be interesting to look at from a fundamental point of view, but considering the goal is to move away from PFSA, the mixed ionomers seemed to be a big focus of the presentation.

Question 2: Accomplishments and progress

This project was rated **2.9** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The main accomplishment over the last year appears to be in the free IL MEA tests, where IL additives to fuel cell cathodes have demonstrated enhanced performance over the Pt dissolution accelerated stress test (AST). Though degradation rates are maintained, this should help with mechanistic understanding of catalyst degradation. The hypothesis that the polymerized IL does not disperse well in the catalyst ink is supported with good evidence, and the approach of mixing with Nafion to demonstrate progress was a good idea. It is unfortunate that more progress in the polymer work did not meet with any success. There is certainly more to do in durability testing, as indicated.
- The project has met its 2020 go/no-go milestone, with lower Pt loadings than the target, and appears to be on track toward meeting the 2021 target for performance. The project has shown improvements in performance of mass activity, specific activity, and performance at rated power for Pt/Vulcan carbon electrodes using the IL and tethered IL.
- The project has accomplished the following: (1) PILBCP synthesis has been well-progressed, (2) results of specific adsorption in PILBCP–Nafion mixed ionomers are very informative, (3) the microkinetic model is interesting, (4) the new ionomer clearly demonstrated improved performance, but whether IL can be toxic to the catalyst has not been thoroughly studied, and (5) interaction of catalyst with new ionomer in the electrode can be better understood using high-resolution transmission electron microscopy (TEM).
- The higher Pt utilization with high-surface-area carbon at low relative humidity in the presence of free IL is a good result.

- Several questions remain in terms of the combinations of IL, polymerized ionic liquid (PIL), and Nafion. Studies on oxygen transport, pressure, and poisoning of catalysts are needed.
- There does not appear to be substantial progress in the synthesis of PILBCPs in the past year. Most of the work involved mixing PILBCPs with Nafion and demonstrating that the PILBCP can reduce Nafion adsorption and thereby enhance kinetics and transport. The performance enhancement achieved by the mixing of Nafion with PILBCP looks impressive, but the performance of the baseline MEA based on Nafion without PILBCP looks too low. It is not clear whether these materials are truly enabling improved performance over the state of the art.
- This presentation was somewhat hard to follow. It seems like the initial year's milestone was hit, but it is unclear whether the researchers are expecting to meet the final milestone or think this material platform is a no-go.

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 in this project, with strong contributions in ILs from Drexel University and great MEA capabilities at the National Renewable Energy Laboratory (NREL) and General Motors (GM). The roles of NREL vs. GM in MEA analysis should be clarified.
- Collaborations within the project appear to be working effectively. Plans to engage with the Million Mile Fuel Cell Truck (M2FCT) consortium will provide more collaboration and access to additional analytical techniques for characterization. Collaboration with Lawrence Berkeley National Laboratory in the area of thin-film polymer morphology and interactions of the tethered IL with the catalyst could be beneficial.
- The collaboration with NREL is a key enabler of the project, although it is somewhat unclear whether GM has participated in the project yet. The principal investigator (PI) did indicate GM's effort was focused at the end, which makes sense.
- The PI has great collaboration with GM, NREL, and Texas A&M University.
- A capable team has contributed to the progress to date.
- The collaboration appears good.

Question 4: Relevance/potential impact

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

- This project is highly relevant to the goals of the Hydrogen and Fuel Cell Technologies Office (HFTO). This innovative approach to improving electrode performance through development and incorporation of novel materials is a great fit for HFTO and is likely to have substantial impact on the field.
- This work is relevant and has a high potential impact. From preliminary studies, addition of IL can enhance catalyst activity and improve durability. Tethering may limit these impacts in terms of the type of carbon support used. These studies also provide insight into tailoring catalyst surface interactions to improve performance.
- The project is relevant to HFTO in that the ionomer developed can improve power density of polymer electrolyte membrane fuel cells. The long-term durability should be better studied for a higher impact. However, on the slide relating to relevance (slide 5), the table was left empty.
- The work in this project is innovative and potentially high-impact to M2FCT goals.
- A cathode that meets DOE performance targets is needed.
- While the work is focused on PFSA-free ionomers, the testing is all done with Nafion membranes. Also, the slide relating to relevance was mostly left blank.

Question 5: Proposed future work

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

- The project appears to be in no-cost-extension territory to meet its final goals. The focus on durability and 50 cm^2 diagnostics is appropriate.
- Durability testing is very important, especially given the possible mobility of the IL.
- Clearly defined future work has been shown.
- The proposed future work looks reasonable but could have been described in more detail. It seems like all of the future work is on MEA testing. Further work on the PILBCP chemistry would seem worthwhile.
- Durability testing and MEA performance are clearly priorities, but it would have been helpful to have more clarity and language here around what has been learned and how that informs plans going forward.
- Future work should be expanded to look at different carbons. It may be possible to trap free IL in the carbon pores by capping the pores with Nafion or the tethered IL. More durability studies and studies looking for leaching of the IL and decomposition of the tethered IL polymer are needed.
- While durability is important right now, performance is too low to be meaningful to fuel cell performance goals. Durability testing of the IL, PIL, and combinations must be done separately to establish the degradation mechanism.

Project strengths:

- This project provides a highly innovative approach to advancing fuel cell performance and decreasing cost. The dual focus on materials development and MEA testing is a key strength.
- The combination of synthetic capabilities of Drexel University and Texas A&M University with the diagnostic capabilities at NREL and GM is a strength.
- The project has innovative ideas of developing a PILBCP/IL ionomer, a great team has been assembled, and great progress has been achieved.
- The project's innovative approach and excellent collaboration are effective for investigating the concept in relevant systems.
- Novel approach to cathode improvement.
- This project has interesting results.

Project weaknesses:

- Project weaknesses include the following: (1) long-term impact of IL on the catalyst and electrode structures has not been studied, (2) electrode optimization using the new ionomer has not been thoroughly investigated, and (3) PILBCP ionomer–catalyst interaction in the electrode is not shown.
- There are plans to focus on Vulcan carbon in the future work, which is a weakness. The combination of free IL with block copolymer IL and Nafion has potential with porous carbons, as the free IL can access the pores and Nafion and block copolymers may cap the pores, limiting or preventing loss of the free IL.
- There does not seem to have been much recent progress in PILBCP development; most of the progress has been in MEA studies.
- Polymerization work has not appeared promising, and dispersion of the ionomer in catalyst inks has met with initial poor results (though this is a challenging topic).
- There is an insufficient consistency in understanding the effects of IL and PIL mixtures on degradation and performance.

Recommendations for additions/deletions to project scope:

- Recommendations include the following: (1) gas permeability and water uptake of the ionomer-cast thin film can be characterized, and (2) this new ionomer can be used for heavy-duty vehicle application, increasing Pt loading and extensive AST (e.g., 90,000 hours).

- The project should monitor changes to the microstructure of the composite layer of Nafion and PILBCP during, before, and/or after AST.
- The project should expand leaching studies and investigation of IL composites with new advanced porous carbon supports.
- The PI should engage NREL rheology/catalyst ink experts to understand and explore the PIL behavior in inks.
- The project should attempt to quantify coverage of catalysts and the catalyst–electrolyte interface, as well as capacitance. Durability testing of the IL, PIL, and combinations must be done separately to establish the degradation mechanism.

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

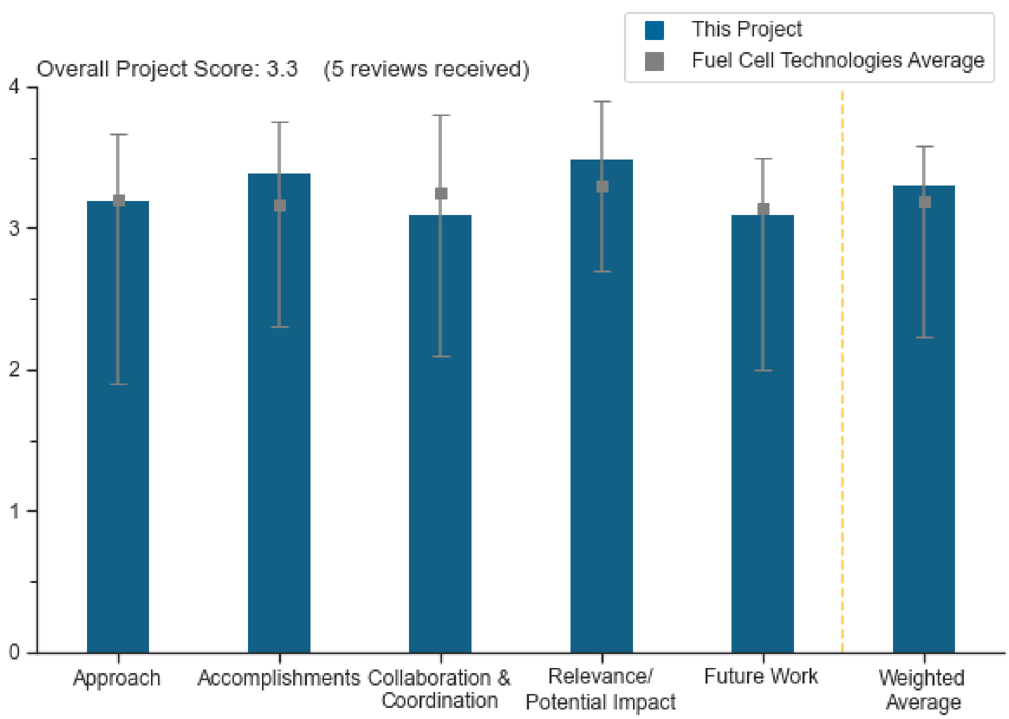
Ryszard Wycisk, Vanderbilt University

DOE Contract #	DE-EE0008435
Start and End Dates	1/9/2019 to 12/31/2021
Partners/Collaborators	Vanderbilt University
Barriers Addressed	• High cost of PFSA membranes • Low proton conductivity at reduced humidity (water partial pressure) • Performance drop above 80°C

Project Goal and Brief Summary

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 Hydrogen and Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan (MYRD&D Plan). The project approach is to develop and 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



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

Question 1: Approach to performing the work

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

- Looking at novel polymers and new methods to make ionomer composites is definitely important. The polyphylene ionomer and electrospinning ideas are worth investigating. The presenter himself correctly identified the core issues with the approach: (1) reproducible synthesis, (2) electrospinning with minimal use of carriers and loss of conductivity, and (3) ionomer integration to create robust membranes. He is fundamentally right. This means the project team has many variables with which to work, and therefore, the research will be complex, which in the end creates a risk of not achieving its goals.
- The data focused on the application of electrospun materials for the fabrication of fuel cell membranes. Preparation of precursor materials is a critical step in this process and clearly identified in Tasks 1–3. The next three tasks address electrospinning. The last two tasks address membrane optimization and fuel cell testing. The systematic organization of the project and progress depend on the success of the previous group of tasks. It is not clear what plans the organization had if any of the first three tasks should reach their expected objectives.
- This is a good approach to improving the mechanical properties of rigid rod sulfonated polyphenylene (SPP), which is brittle and soluble in water.
- The Vanderbilt team has a clear, target-driven approach. Building on the success of the SPP is a credible candidate for realistic hydrocarbon membranes. Electrospinning reinforced fibers has the potential to result in high-quality, commercially viable membranes. However, it seems electrospinning is relied on to the exclusion of other approaches. The new technology in this project is the technique to crosslink the SPP.
- The justification for the PPSA was missing. It is not clear why this poly(aromatic) hydrocarbon polymer electrolyte membrane (PEM) would be more advantageous than sulfated polyether ether ether ketone (PEEK) or PPSU. It is clear that high ion exchange capacity (IEC) values can be attained with PPSA, as two sulfonic acid groups can be installed per phenyl or biphenyl, allowing for high ionic conductivity under low relative humidity (RH) conditions, but the rigid rod nature of the polymer will inevitably lead to poor mechanical properties. Reinforcement is needed to make a mechanically robust hydrocarbon PEM. A clearer picture stating that high IEC is needed for low-RH operation and that the rigid rod poly(aromatic) backbone would prevent water dissolution would help the presentation. It is not clear whether the poly(phenylene) offers greater oxidative stability over PEEK or PPSU backbones because it does not contain ketone, sulfone, or ether functional groups.

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.

- There was good progress in developing membrane mats incorporating SPP.
- A good deal of progress has been made in preparing two reinforced cPPSA PEMs. The pore-filled cPPSA has very high ionic conductivity (450 mS/cm) and conducts protons near 100 mS/cm at 40% RH. The pore-filled approach gives PEMs with better conductivity and mechanical properties. The new pore-filled cPPSA products should be good candidates for 100°C–120°C temperature operation at low RH for heavy-duty vehicles. However, fuel cell performance and chemical stability studies (accelerated stress tests [ASTs]) are needed. These experiments should be done in the no-cost extension period, as the project has been partially delayed because of the COVID-19 pandemic. The following items are missing from the technical target goals: H_2 and O_2 crossover, area-specific resistance, AST, and cost for manufacturing membranes.
- Tasks 1 and 2 appear have achieved success. Task 3 demonstrated two approaches to composite cPPSA–PPSU membranes: dual electrospun and pore-filled PPSU. The data demonstrated that the pore-filled membrane has superior proton conductivity and tensile strength. A problem was reported: mechanical strength characteristics were worse than those obtained in budget period 1. The researchers did not offer a technical path to resolving this issue.
- The proton conductivity reported by the team at 80°C and 40% RH is very impressive. This is perhaps some of the best low-RH conductivity reported by a hydrocarbon membrane. Mechanical properties may

need more improvements. Strain at break of 15% or less is likely to be brittle and may suffer mechanical failure in handling or operation.

- There was good progress in developing membrane mats incorporating SPP.
- The research team synthesized the polymer and made some good mats. The project achieved decent proton conductivity and tensile properties for the items produced. All the accomplishments were very impressive. For this technology or approach to compete, it also has to meet cost, durability, and even higher performance metrics. None of those data were provided in the slides.

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 team leveraged knowledge from Professor Litt at Case Western Reserve University. There was a collaboration with Professor Litt prior to his death. There has been some discussion of testing the PEMs with Dr. Mukundan at Los Alamos National Laboratory (LANL). No interaction with industry or other national laboratory partners is clearly defined.
- The team consists entirely of Vanderbilt University researchers. Given the size of the award, this does not seem to be a fundamental problem. However, the teaming score can be no higher than "good" in this circumstance.
- Collaboration focused on academic researchers. No industrial partners were identified. The researchers reported they would seek an industrial partner/laboratory when optimization is concluded. This should not be a "throwing over the wall" approach to collaboration. The earlier an industrial partner or laboratory participates in a project, the more productive the research and development should be.
- An end user that currently buys membranes would be a great addition to the team, as the end user can provide feedback on the progress and commercial readiness of the technology.
- The team should collaborate with another entity to perform third-party analysis to measure conductivity and measure gas crossover.

Question 4: Relevance/potential impact

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

- The researchers have a good grasp of the importance of their successful demonstration of the technology. Reduced cost and better low-RH performance at higher temperatures would greatly benefit most aspects of fuel cell applications.
- New PEM architectures that conduct protons with low resistance over a wide humidity range, as well as being mechanically robust, are important to the MYRD&D Plan. The project accomplishments and remaining activities are aligned with this plan.
- The SPP approach has the potential to advance the state of the art of hydrocarbon PEMs. For this reason, the project has a high potential impact. Material properties and accelerated durability are necessary for this approach to demonstrate its full relevance and impact.
- If an alternate to Nafion™ PFSA composite materials (currently in the market) is developed, it would be a significant contribution.
- The conductivity of the rigid SPP is one of the highest of all hydrocarbons, and this project improves the mechanical properties. However, there are some concerns that these mats will have issues with gas crossover since the film is really not totally dense (it appears to have some voids). Moreover, the mats are made of fibers that are at the opposite ends of hydrophobic/hydrophilic extremes. With such large differences in a material not covalently bonded, there is a large concern about mechanical stability in RH cycling, which is an AST. Also, these types of structures are typically stable toward highly oxidative environments, which leads to durability concerns. These potential issues will limit the impact of this material until proven otherwise.

Question 5: Proposed future work

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

- The researchers identify three challenges and barriers. The first was a surprise, as it was missed in discussion of Tasks 1–3 and could be a showstopper. The second challenge was identified, and the researchers proposed an approach that appears reasonable. The third challenge states the problem but does not suggest a path to improvement. The future work addresses membrane electrode assemblies and their testing. If membrane challenges are resolved, this is a good approach, but it does not appear the challenges are part of future work. It is recommended the researchers take a step back and solve challenges or at least explain why they are going forward with fuel cell membrane electrode fabrication and testing to resolve the challenges identified.
- Regarding fuel cell testing and AST protocols, perhaps the researchers should look at the water uptake of this material at various temperatures to understand how high temperature and humid environments change the dimensions of the membranes. It would be beneficial to see the impact of this material in Fenton's reagent to determine whether the mat improves chemical stability as well.
- Running fuel cell performance and durability testing—proposed future work—are clearly important next steps; however, the plans for competing this work are unclear. The project does not state whether work will be done at Vanderbilt University, LANL, or another institution.
- The proposed future work primarily focuses on testing the cPPSA in fuel cell devices. The reinforced cPPSA will be the PEM, but it is not clear what electrode binders will be used. Additionally, AST needs to be done. This seems like a good deal of work to complete in six months with limited resources.
- There is much to do. This is all good basic research but has many thresholds to pass before being commercially ready. Some effort should be expended to consider specific tasks relevant to commercialization, such as cost analysis and durability testing.

Project strengths:

- Reinforced cPPSA PEMs have been prepared using two different strategies, co-dual fiber electrospinning and pore-filling. The team has demonstrated remarkably high proton conductivity (450 mS/cm at 90% RH and 100 mS/cm at 40% RH). The pore-filled membrane has good mechanical properties over 18 MPa and 15 strain at break at 50% RH. The new PEMs based upon reinforced cPPSA will be good candidates for membrane electrode assemblies that will operate at higher temperatures that have lower partial pressures of water (important for heavy-duty vehicles).
- Building on one of the most promising hydrocarbon membrane technologies (SPP) is a core strength of this project. The team is highly regarded, has many years of membrane development experience, and clearly understands the important targets for this technology. The proton conductivity presented by the team is quite impressive.
- The research is a good, sound alternative approach, addressing a critical issue in current PEM systems. The strong, highly qualified team has proven competence in this field.
- This is a very creative and technically strong research collaboration. It is good to see electrospinning technology moving forward.
- These materials show one of the highest proton conductivities at low RH of all materials, and this project has shown improvement in the mechanical properties of these materials.

Project weaknesses:

- Mechanical properties to date may be inadequate for truly competitive membranes. Electrospinning fabrication methods make sense for mechanical support but seem to be a "force-fit" in the dual-fiber case. There is no obvious advantage to making an ionomer fiber only to destroy it (i.e., melt or dissolve) later. Additional partners or Million Mile Fuel Cell Truck (M2FCT) laboratories should be enlisted for performance and durability evaluations.
- The largest weakness right now is understanding how humidity and temperature affect the mechanical properties. One of the automotive tests looks at repetitive low/high RH in a membrane electrode assembly

and monitor open circuit voltage loss; this would be a good demonstration of good mechanical–chemical behavior. Also, Fenton's test will give a basic understanding of the membrane's oxidative resistance.

- The project can benefit from a stronger rationale for the PPSA chemistry, especially since it is a non-trivial synthesis and has batch-to-batch consistency issues. Additional work is needed to substantiate the potential of reinforced cPPSA PEMs for fuel cell devices. This work includes gas crossover, fuel cell performance, and durability. The electron micrographs show some porous areas in the membrane, and these pores could cause large gas crossover rates.
- The project needs input from an industrial partner or testing laboratory. The project needs to have a discussion about moving electrospinning to high-rate production. Technically, the project needs to resolve the thermal crosslinking of PPSA ionomers as quickly as possible.
- The project is a long way from commercialization.

Recommendations for additions/deletions to project scope:

- The Vanderbilt team has done a nice job on this project, and clearly, electrospinning is a core strength of the team. However, expanding the potential supports to include expanded polytetrafluoroethylene (ePTFE) is suggested. If nothing else, it would be a good comparison.
- If the polymer system shows promise, there will be a need for much more work involving optimization, electrode attachment, and then preparation for commercial applications.
- The three challenges identified on Chart 14 of the presentation should be added to future work.

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

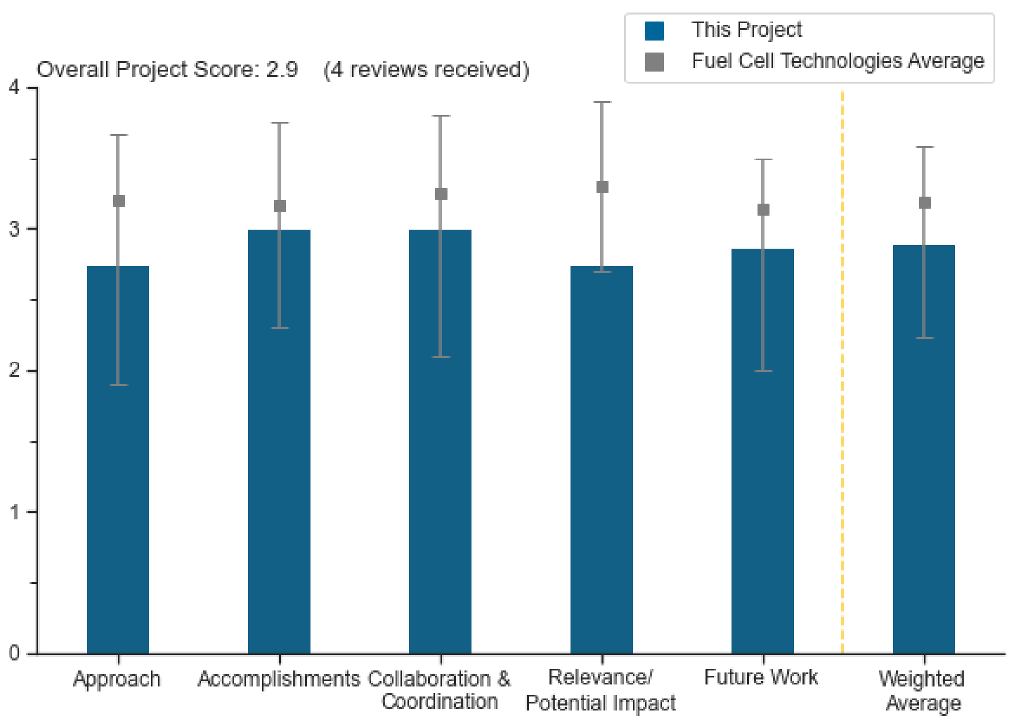
Nem Danilovic, Lawrence Berkeley National Laboratory

DOE Contract #	1.3.0.209
Start and End Dates	7/1/2019
Partners/Collaborators	Nel Hydrogen, Washington University in St. Louis, Ballard Power Systems, Pajarito Powder
Barriers Addressed	• No barriers specific to regenerative fuel cells • Optimization between fuel cell and electrolyzer barriers • Fuel cells (durability, cost, performance) • Hydrogen production (capital cost, system efficiency, and electricity cost)

Project Goal and Brief Summary

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 (RTE) 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



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

Question 1: Approach to performing the work

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

- The approach to move toward more fuel-cell-type materials should lead to increases in RTE. The use of a durability test cycling between fuel cell at ~0.55 V and electrolyzer at 1.55 V should provide an accelerated durability test and advance the field. The addition of work on the porous transport layer (PTL) should help improve performance, as modeling suggests mass transport losses at 1 A/cm^2 are due more to the PTL than to the catalyst layer. There is some question about the durability of the sulfonated poly[ether ether ketone] (SPEEK)-based membranes in electrolyzer operation, and it is not clear what steps are being investigated to improve chemical stability (whether the team is just relying on the base polymer, using current commercial additive packages such as CeO_2, or looking at other additives). Also, it appears membrane degradation testing has looked mainly at discrete mode. Longer durability testing of the membrane in electrolysis mode and cycling between electrolyzer mode and fuel cell mode should be a priority. The targeted current density of 1 A/cm^2 for both fuel cell and electrolyzer mode for the target efficiency seems arbitrary.
- The methods by which the team is investigating URFCs are reasonable; however, the approach is fairly broad, taking on essentially every component of the UFRC, from membranes to catalysts to PTLs to electrodes. This seems too broad a scope for such a limited budget, and it would have likely been more successful to focus on a smaller subset of components with the highest priority, for example, PTL and electrode strategies.
- This project primarily involves testing materials that are already developed, including commercially available materials, to find which combinations of materials give the best URFC performance and durability. The main materials/component development work is in the SPEEK membrane development. Catalyst support development is also mentioned, but no results were shown. Overall, there does not seem to be much novelty in the work being performed.
- The main weakness here lies in the way that the team has been designed, which naturally leads to mostly discrete testing of a fuel cell and then an electrolyzer. There is not much true URFC work here, and that is somewhat of an issue. Making the necessary sacrifices to enable both should be the root of a URFC project, and that is nowhere to be found here.

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 demonstrated improvements in performance with supported Pt/Ir-TiO_2 and has achieved >50% RTE at 1 A/cm^2 (in oxygen). The project has met its milestones for 2021 and is on track to meet end-of-project targets. The project appears to have shifted some from catalyst support development to optimizing systems with commercial materials. It is not clear whether this is due to COVID-19 restrictions or whether doped TiO_2 materials being developed at Washington University in St. Louis showed poor promise. Development of an accelerated stress test (AST) matrix is an important advancement.
- The team's efforts with PTLs and electrodes have yielded good, useful results, but catalyst and membrane work has not shown any specific advances or progress. The investigation of constant gas versus constant electrode approaches are interesting and have shown some ability to increase RTE. The fuel cell conditions presented are a modest window of conditions that could be investigated and seem like they were run at very high stoichiometry, but it is not fully clear. A discussion of the fuel cell operating conditions presented and perhaps presenting how polarization changes as a function of conditions (stoichiometry and back pressure, in particular) would be insightful for overall operating considerations.
- The individual components seem to be performing well. Discrete systems are performing pretty well. Electrolyzer performance is greater than fuel cell performance, but both are fine. Sometimes, it is difficult to judge the accomplishments, as the cell set-up and conditions are often not given in the slides, making it hard to compare to what has been done before or to know whether the experiment done is practically relevant. It is not clear what the team means by "liquid water"—whether it is deionized water or a salt solution, as both would be "liquid water." Durability data are of very limited duration. Practical durability data should be at least 100 hours long.

- Some progress on SPEEK membrane development was reported, but this does not seem to enable a significant benefit for URFC technology. The down-selected membrane is a commercially available perfluorosulfonic acid membrane. Some work was performed on development of novel catalysts for URFCs, but no new results were shown this year. Instead, the catalyst work in the project seems to be focusing on comparing performance of commercially available catalysts. AST development was mentioned, but data are needed to demonstrate that the developed ASTs mimic real-world degradation modes.

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.

- Collaborations within the project appear to be working well.
- The different collaborators all have well-defined roles and have experience in their support areas. The primary concern would be how broad the effort is and how effective the collaborations are. For the membrane synthesis and catalyst development tasks, there seems to be limited synergy with the more general UFRC fabrication and testing tasks.
- The team makes sense and is capable. It seems somewhat less desirable to have one institution on fuel cells (Ballard) and the other on electrolyzers (Nel Hydrogen). It would seem that this would lead to a bifurcated design for the electrodes, which cannot be present in a real URFC. This arrangement also leads to discrete testing dominating, which it obviously has. There is also not much catalyst work mentioned here. It is unclear what Pajarito Powder has done in this particular update.
- The team consists of several of the key players in URFCs. It is a capable team, but in some cases, it is hard to see how the team members are contributing to research and development (R&D). For instance, Nel Hydrogen and Ballard seem to be mainly testing commercially available membranes and catalysts to select the best materials. DOE-funded work should focus more on developing the science or developing new materials and less on screening commercial materials.

Question 4: Relevance/potential impact

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

- The project is in alignment with Hydrogen and Fuel Cell Technologies Office goals and objectives. Improving URFC performance and efficiency will result in increased utility of hydrogen storage as an energy storage option.
- The project seems to be addressing relevant objectives and needs.
- A more science-based approach would make this project more relevant and increase the potential for significant impact.
- Reversible fuel cells do not have well-defined targets, and oftentimes the primary competition for UFRCs would be independent fuel cell and electrolysis stacks. It is far from clear, and perhaps doubtful, that UFRCs will have application in anything beyond a few niche applications, although this is much less a comment on this specific project rather than the area of UFRCs in general. However, science outcomes still have potential value in the fuel cell and/or the electrolysis areas.

Question 5: Proposed future work

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

- Proposed future work addresses most of the challenges for URFCs. Concerns about the durability of the SPEEK-based membranes in electrolyzer operation and how it may handle the humidity changes in reversible mode need to be answered. Longer durability testing of the membrane in electrolysis mode and cycling between electrolyzer mode and fuel cell mode should be a priority.

- As the project is in its late stages, the proposed work is reasonable, but some areas, such as the further development or implementation of membranes and catalysts, seem unlikely to have much chance of meaningful outcomes.
- The proposed future work seems well-planned, but it involves too much demonstration and not enough R&D.
- It seems like the project would really benefit from switching to operating most of the time as a URFC, not discrete systems.

Project strengths:

- There is a strong team, a good principal investigator, and interesting science. The project addresses some gaps in the scientific community. The highest value is in the area of PTLs and in some of the electrode areas, particularly with constant electrode and constant gas comparisons of operating strategies.
- The team's experience in fuel cells and electrolyzers, with Nel Hydrogen and Ballard as partners, is a major strength.
- The team includes many of the key players in URFCs.
- There is a good team. It is making progress toward goals.

Project weaknesses:

- While hydrocarbon implementation in electrolyzers is an interesting area for research and would be great if achieved, the amount of effort in this project seems subcritical and a distraction from other topic areas. The late addition of catalyst development also seems like it serves to distract from the other, more fruitful, areas of investigation in the project. The project really is focused only on performance and not on durability at this point in time, and while this makes sense based on resources, it leaves significant gaps in what is likely the key area for devices.
- The work reported in the past year and the planned future work involve too much screening of existing materials and not enough novel R&D.
- The project is too focused on discrete tests. Experiments are pretty short-term. AST might work fine, but it is unclear whether it is really the correct one.
- Membrane durability aspects do not appear to be receiving the focus they require.

Recommendations for additions/deletions to project scope:

- Extensive work has already been performed on development of polymer electrolyte membranes for fuel cell applications. While there could be room for improvement in membranes, specifically for URFCs, the opportunities for improvements in catalysts and diffusion media are far greater, so it would be better to focus more resources on catalysts and diffusion media and fewer resources on membranes. The work scope associated with screening commercial materials should be reduced.
- The project should be more URFC-focused. It would be good to actually show that the AST is giving relevant information or information that is indicative of how a URFC might respond.
- It is recommended that the project increase work on membrane durability issues.
- With the project only months away from completion, there is not much time to coordinate large changes in scope across the team, but membranes, in particular reinforced membranes, seem to be an area that does not merit further investigation. Catalyst efforts also seem tangential at this point.

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

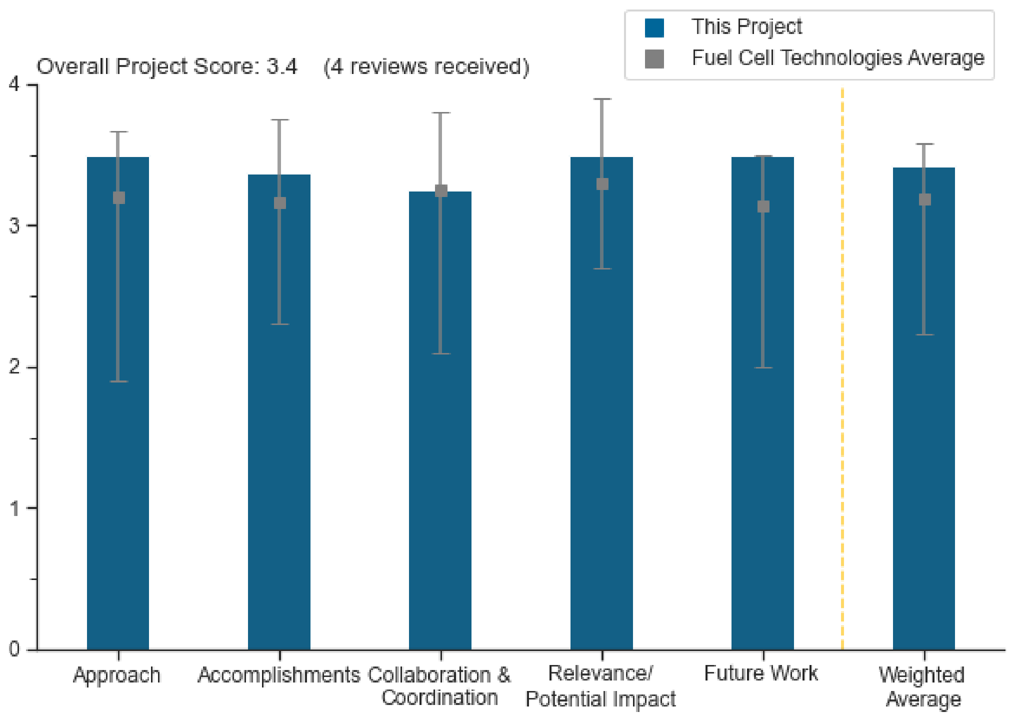
Scott Barnett, Northwestern University

DOE Contract #	DE-EE0008437
Start and End Dates	10/01/18 to 09/31/21
Partners/Collaborators	Colorado School of Mines
Barriers Addressed	• Durability • Performance

Project Goal and Brief Summary

This project will develop and test reversible solid oxide fuel cells (RSOFCs) for electrical energy storage applications, including system concepts for high efficiency. The RSOFCs 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 RSOFCs 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) will inform designs that can achieve cost and efficiency targets for renewable electricity storage.

Project Scoring



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

Question 1: Approach to performing the work

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

- The project is set to develop RSOFCs with high current density, good long-term stability, and high round-trip efficiency. The project will also integrate experiments with system modeling and technoeconomic analysis to provide early feasibility evaluation. These approaches are unique and are critical to ensure that the project goals can be met.
- The pressure effect study was performed on the oxygen electrode side. In practice, pressurizing the hydrogen electrode side is equally important, considering the hydrogen pressurization for downstream storage. Therefore, this portion of the study can be considered in the future. Additionally, the positive effect of gadolinium-doped ceria (GDC) infiltration into the hydrogen electrode microstructure is interesting. A further mechanistic study is deserved. Finally, the need for thermal energy storage can be further discussed for the case of above-thermal-neutral operation.
- The project approach addresses the following key technical barriers: (1) development of RSOFCs allowing high round-trip efficiency, (2) ASR <0.15 Ωcm^2 (achieved in fiscal year 2020), (3) life testing of reversible operation to establish long-term stability, and (4) a reduced degradation rate of ≤6%/kh at 0.75 A cm^{-2}.
- This is a very sound approach.

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.

- This project has made excellent progress; reversible cells were fabricated and tested to show reduced degradation, and a large-area-cell test facility was set up and successfully used to test solid oxide cells with 80 cm^2 active areas.
- The project has good results. However, a solid oxide electrolyzer (SOE) typically operates at around 1.3 V, which is the thermo-neutral point. The V-I (voltage–current density) curve in electrolysis mode should be extended at least up to this point. Likewise, the degradation data at 0.5 A/cm^2 should be increased to higher currents to operate at 1.25–1.3 V. Cell performance is very impressive.
- The project and milestones have progressed well, and go/no-go decision points have been met.
- The progress toward milestones is on track.

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 is led by Northwestern University (NU), with Colorado School of Mines (CSM) as the subcontractor. NU is conducting the experimental work, while CSM is leading the stack and system modeling and technoeconomic analysis. NU provides experimentally measured cell characteristics as input to CSM stack and system models, allowing for accurate prediction of expected system characteristics, while CSM provides input to NU regarding desired cell characteristics and operating parameters, ensuring that test results are relevant.
- NU provides experimentally measured cell characteristics as inputs to CSM stack and system models, allowing for accurate prediction of expected system characteristics. CSM provides input to NU regarding desired cell characteristics and operating parameters, ensuring that test results are relevant.
- The project is a collaboration between NU and CSM, with the former working on experimental and the latter working on theoretical analysis. Both are making good progress toward milestones.
- It would be better to have some industrial partner(s) collaborate on an advisory level.

Question 4: Relevance/potential impact

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

- Developing RSOFCs with high round-trip efficiency and long-term stability will provide an enabling technology for electrical energy storage. GDC infiltration into nickel–yttria-stabilized zirconia (Ni-YSZ) fuel electrodes has been shown to improve cell stability. Oxygen electrode performance under pressurization has been characterized and fully modeled, a large-area-cell test set-up has been assembled and fully vetted, and it will provide a unique testbed for observing effects of RSOFC operation.
- Developing RSOFCs for electrical energy storage with high (60%–90%) round-trip efficiency at ~ 1 A cm^{-2} is a very important part of hydrogen production and storage.
- Some good progress has been achieved on small-cell characterization, while long-term stability needs to be further demonstrated, especially on large-area cells.
- This project is very relevant to improving performance and reducing cost of SOE technology.

Question 5: Proposed future work

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

- The proposed future work is on target, as it aims to further its studies of RSOFC degradation rates and mechanisms, develop procedures for electrode infiltration of large-area cells yielding uniformly low ASR, and continue to refine electrochemical model calibration as additional pressurized cell and 80 cm^2 cell experimental data become available.
- Further work is aimed at identifying main degradation mechanisms, developing preliminary models for both button cell and large-area single-cell tests, and developing procedures for electrode infiltration of large-area cells to yield uniformly low ASR. These studies are essential to developing efficient and durable RSOFCs for energy conversion and storage applications.
- The principal investigators clearly recognize the challenges of the project and have planned well to address them in the coming year.
- Further cell studies at even higher current densities and a stack demonstration are suggested.

Project strengths:

- This is a well-coordinated project with an integration of experimental and modeling efforts to address the critical barriers in RSOFCs for energy conversion and storage. Solid progress has been made, and the project milestones have been met.
- This project combines experimental and theoretical strengths of the two institutions to address the issues of RSOFCs. The two approaches could potentially converge to provide key engineering information to implement in solid oxide cell systems.
- The project is addressing the following key Hydrogen Program technical barriers: (1) development of solid oxide cells allowing high round-trip efficiency, (2) ASR $<0.15\ \Omega cm^2$, (3) life testing of reversible operation to establish long-term stability, and (4) a reduced degradation rate of ≤6%/kh at 0.75 A cm^{-2}.
- There is a great focus on cell performance and degradation. This cell can potentially reduce overall SOE system costs if it can operate at 1–1.5 A/cm^2.

Project weaknesses:

- GDC infiltration into Ni-YSZ has been shown to improve cell performance and durability; however, how to implement this approach for the stack fabrication should be identified and validated.
- The hydrogen storage tank model that is included with additional compressors and intercoolers does not seem well-defined. There is also the question of whether the high pressure will cause any potential problems for the cells during operation.

- The project needs more work on the DOE target current densities of 1–1.5 A/cm^2, with operations at 1.3 V.
- The long-term stability of cells needs to be further demonstrated to meet the cost goal.

Recommendations for additions/deletions to project scope:

- The project scope is well-defined, and no changes are necessary.
- It would be good to see a degradation/performance of at least 1.0 A/cm^2 operating point. The maximum round-trip efficiency is at 0.78 A/cm^2, and the total cost of ownership will also improve with higher operating currents.
- It is recommended that the hydrogen storage tank model be included with additional compressors and intercoolers. Information is needed about the desired properties, such as the volume and the pressure for hydrogen storage; the compressors and intercoolers; and the properties required for the compressors.
- Studying degradation mechanisms should be considered in the remainder of the project.

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

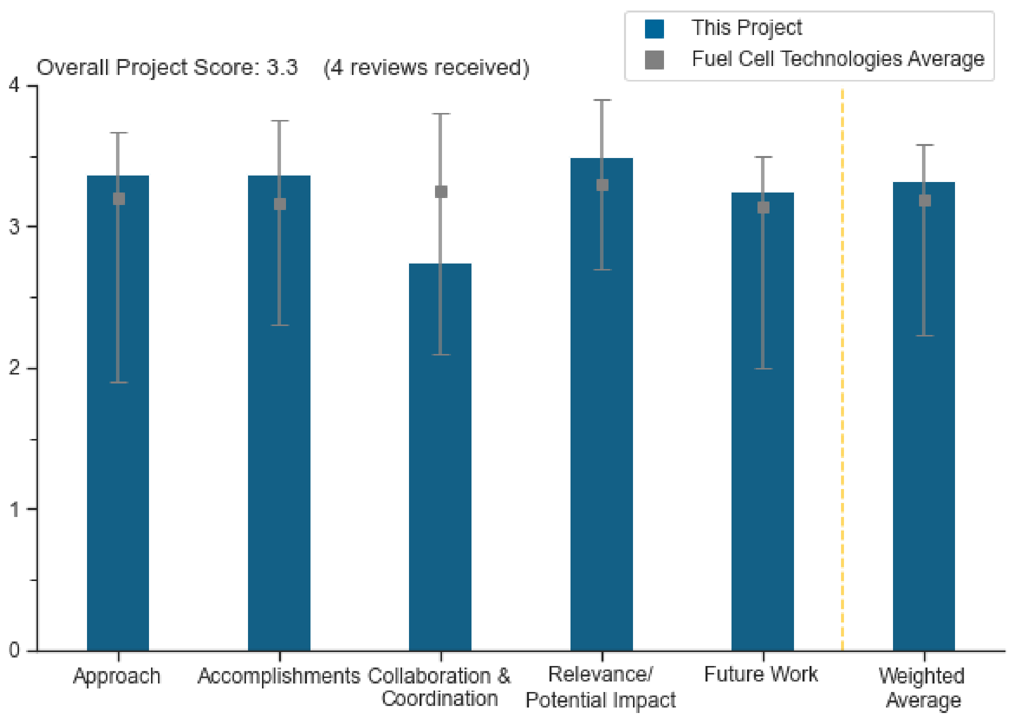
Meilin Liu, Georgia Institute of Technology

DOE Contract #	DE-EE0008439
Start and End Dates	1/1/2019 to 12/31/2021
Partners/Collaborators	N/A
Barriers Addressed	• Capital cost • System efficiency and electricity cost • Operations and maintenance

Project Goal and Brief Summary

This project is developing a robust, highly efficient, and economically viable high-temperature unitized reversible fuel cell 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% round-trip efficiency at 1 A/cm^2 in both operating modes.

Project Scoring



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

Question 1: Approach to performing the work

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

- The approach is quite logical and orderly: (1) synthesize the electrode and electrolyte materials with desired particle size and morphology, (2) fabricate symmetrical cells and single cells, and (3) integrate nanostructured components into the cell design, and modify the electrodes with active bifunctional catalysts and protection coatings in single cells to achieve >70% round-trip efficiency.
- The project studies proton-conducting perovskites as key materials for intermediate-temperature solid-oxide-cell-based water electrolysis. Perovskite proton conductors allow the use of highly active perovskite oxygen electrodes, which ultimately elevate the overall performance and reduce the operating temperature. The demonstrated area-specific resistance (ASR) is very low for the temperature range studied, which is promising for achieving the cost and efficiency goals.
- Very good materials science is being performed in this project. Improved materials and a novel barrier layer approach have been developed to improve resistance to steam, while improving solid oxide electrolyzer cell performance, efficiency, and durability. Significant improvements have been demonstrated as materials formulations have been adapted. The work has led to performance levels that are at or exceed the state of the art. That said, it is a matter of question as to whether it is meaningful to measure round-trip efficiencies via testing at low reaction utilizations and whether button cell testing is the best way to assess durability, given that reactant utilization is extremely low and a number of important degradation mechanisms can occur at higher utilizations.
- Some aspects of stability and durability of this concept have been discussed in the slides and presentation (e.g., see slide 11). However, it is not very clear how this project addresses other aspects, such as mechanical durability related to operating condition changes (such as relative humidity changes after changing the operation mode) and chemical durability, focusing in particular on the many interfaces present. Perhaps the team could elaborate on how these durability aspects are considered.

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 principal investigator (PI) is making good progress toward achieving round-trip efficiency and degradation rate on small button cells. Some density functional theory calculations have also been done to facilitate the understanding of the enhanced catalytic activity.
- The Georgia Institute of Technology (Georgia Tech) had significant accomplishments. The team completed a baseline study of air electrodes (ASR=0.02–0.03 Ω cm^2 under a bias of +0.2 V at 700°C with a durability test of 200 hours), developed fuel electrodes/catalysts with electrode polarization <0.07 V at 1 A cm^{-2}, and demonstrated 200-hour continuous operation of single cells at ≤700°C with >60% round-trip efficiency at 1 A cm^{-2} in both modes with a degradation rate of <2% per 500 hours.
- Very high-performance levels have been demonstrated, with reasonable stability at operating temperatures. Performance, durability, and faradaic efficiency levels still need to be improved at lower operating temperatures that are being targeted, but there is reason to believe that these improvements are possible. The characterization and analysis techniques being employed provide significant insights that will be leveraged in the future development of protonic ceramic electrolytes.
- It would be helpful to understand why the last milestone (>500 hours for a 1-inch button cell with <2% degradation, etc.) was chosen. Perhaps this will help in gaining fundamental understanding of the degradation mechanisms for this concept. If so, information about the type of characterization tools and methods used is also requested.

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.

- This is a collaborative project between Georgia Tech, Idaho National Laboratory (INL), and Phillips 66. The role of Georgia Tech is to develop materials sets, INL is to validate the metrics, and Phillips 66 is to develop a fabrication process and prototype.
- This project's partners for implementation include INL for validation of efficiency measurements and Phillips 66 for development of fabrication processes and prototype.
- This is a single-organization project. Collaborations with INL and Phillips 66 were mentioned, but no evidence (data) of any collaboration was presented. For example, if INL is validating performance levels that have been achieved at Georgia Tech, then it would have been useful to present comparative data. If Phillips 66 has been collaborating to scale up the fabrication to larger cell areas, then photographs of large cells could have been presented.
- It is not clear if and when the partners mentioned in slide 17 have been or will be involved in the implementation.

Question 4: Relevance/potential impact

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

- Proton-conducting solid oxide cells (SOCs) have the potential to operate at a 500°C–600°C range and, thus, to reduce cost and improve durability of SOC systems. However, some critical challenges need to be addressed before scaled-up demonstrations. The project addresses some of the challenges, such as oxygen electrode performance, which will contribute to the overall development of the technology. Other engineering challenges, such as electronic leakage, mechanical strength, and chemical reactivity of proton-conducting perovskites, will have to be addressed in the future.
- Development of reversible SOCs to achieve >70% round-trip efficiency at 1 A cm^{-2} in both solid oxide fuel cell mode and solid oxide electrolysis cell mode is a key element of DOE's energy production and storage targets.
- Excellent materials science is being performed on this project, and the performance and durability of protonic ceramic fuel cells and electrolyzers have been advanced. The work needs to be extended to larger cell areas and higher reactant utilizations before the potential impact of these new materials can be fully assessed.

Question 5: Proposed future work

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

- The proposed future work is targeted to a successful completion. Georgia Tech will make efforts to understand electronic conducting behavior of $BaZr_{0.1}Ce_{0.7}Y_{0.1}Yb_{0.1}O_{3-\delta}$ (BZCYYb)-based electrolytes through conductivity and transference number measurements. The project will also develop new strategies to improve faradic efficiency, including optimization of operating conditions and development of new electrolyte materials with low electronic conductivity. The team will fabricate single cells with desired electrolyte, electrode, and catalyst coatings and run long-term stability tests.
- The PI has identified a key issue of electron leakage through the Ce-containing, proton-conducting perovskite electrolytes used by the project, and addressing this issue is proposed for future work. It is important work to be studied. It will be interesting to see whether the proposed barrier layer will lower the performance.
- The proposed future work does not clearly show how the remaining milestones, related to lower-temperature performance and durability, will be achieved. Larger-area single-cell testing with higher reactant utilizations will help assess the true status of the technology being developed and could provide a decent capstone to the project in its final project year.

Project strengths:

- Georgia Tech had the following significant accomplishments: (1) completed a baseline study of air electrodes (ASR=0.02–0.03 Ω cm^2 under a bias of +0.2 V at 700°C with a durability test of 200 hours), (2) developed fuel electrodes/catalysts with electrode polarization <0.07 V at 1 A cm^{-2}, and (3) demonstrated 200-hour continuous operation of single cells at ≤700°C with >60% round-trip efficiency at 1 A cm^{-2} in both modes, with a degradation rate of <2% per 500 hours.
- There are several characterization methodologies, leading to good understanding of the enhancement mechanisms. From a pure electrocatalytic activity perspective, the achieved ASR is a promising indicator for the materials studied.
- This project has excellent materials science with new materials and approaches, as well as state-of-the-art characterization and analysis techniques.
- The project focuses on a concept that has very high potential for advancement toward the DOE Hydrogen Program goals.

Project weaknesses:

- No progress is reported on developing large cells at Phillips 66. The reported results were all from small button cells. The thermal stability of the catalyst $BaCoO_3$ (BCO) is unknown. In general, BCO is not stable at elevated temperatures because of the loss of oxygen, forming less active brownmillerite or other BaCo-related oxides. The calculation of faradaic efficiency needs further examination, as it requires the analysis of hydrogen produced. The presentation did not elaborate on whether this is the case.
- The approach chooses to demonstrate certain durability targets (the milestones in slide 6) but could focus more on gaining fundamental understanding of possible failure modes and limitations associated with the materials selected/developed.
- There is an over-reliance on button cell testing (low reactant utilizations) for performance and durability demonstrations.

Recommendations for additions/deletions to project scope:

- There may not be sufficient resources available, but larger-area single-cell testing with meaningful reactant utilizations would provide a better assessment of where the technology stands.
- The project’s future work should cover the most pressing work.
- The team should continue the planned project effort.

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

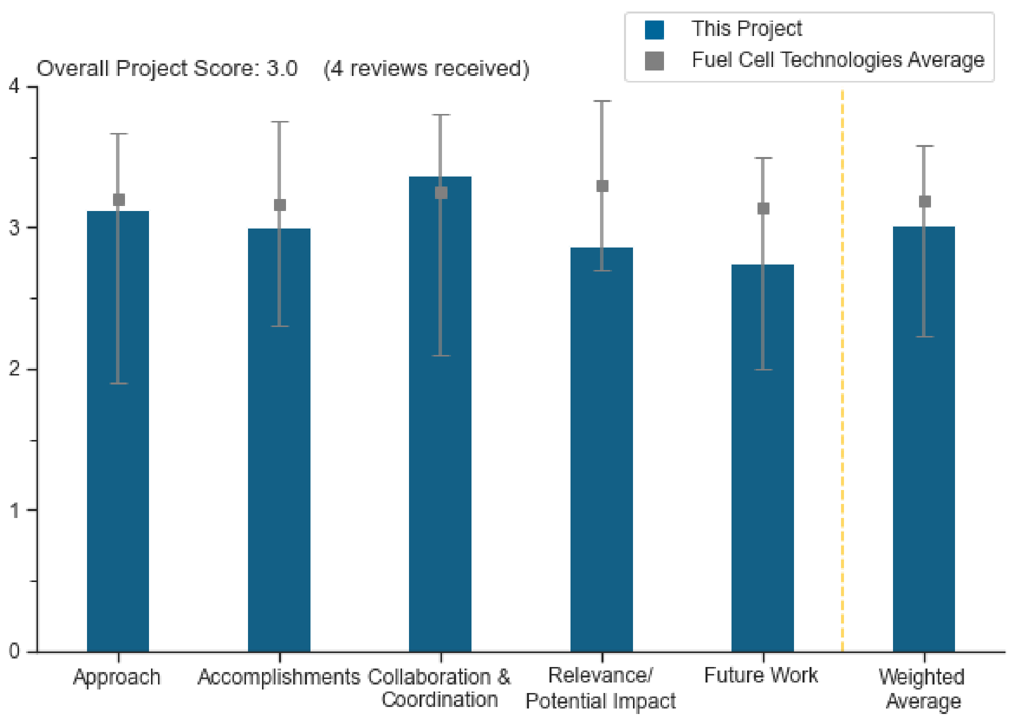
Xianglin Li, University of Kansas

DOE Contract #	DE-EE0008440
Start and End Dates	10/1/2018 to 3/31/2022
Partners/Collaborators	Kansas State University, University of Buffalo, Carnegie Mellon University
Barriers Addressed	• High platinum-group-metal catalyst loading • Catalyst poisoning by methanol • High fuel crossover

Project Goal and Brief Summary

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



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

Question 1: Approach to performing the work

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

- The approach systematically builds on the expertise of four universities, with each university addressing a specific component. This type of project and this type of approach are usually difficult to complete because of the separation of the four research laboratories. The milestone status on chart 5 demonstrates that these four universities were able to coordinate their efforts, contribute their expertise, and successfully complete the milestones in the fifth through eighth quarters.
- Several advances in DMFCs are highlighted in this project. The non-PGM catalyst from the University of Buffalo appears to be an extension of the university's existing catalyst work into a new field, which is reasonable. MEA data is presented for this part of the project and appear to meet targets. It is less clear what the targets for the "ultra-low PtRu anode catalyst" are, and there is no discussion of what scale is necessary for MEA integration or the schedule for it. The electrode work at Carnegie Mellon comprises state-of-the-art techniques, but the MEA performance is one-third that of other partners, and it is unclear whether learnings will be meaningful. Modeling appears to be effective for understanding future materials development needs.
- Overall, it seems to be a good approach in terms of developing active anode catalysts and methanol-tolerant cathode catalysts. There has been no result demonstrating mitigation of methanol crossover, as such. The authors have proposed the idea of fabricating a graphene-based methanol crossover barrier layer.
- The project is broad, taking on many aspects of DMFC performance improvement, including anode and cathode catalyst development, electrode fabrication, and mass transfer considerations. The project is focused primarily on performance metrics but does not quantify all loss metrics, such as methanol crossover or parasitic power/water requirements, and does not address durability at all, which is certainly the most difficult hurdle in terms of making DMFCs relevant.

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 exceeded the target performance while maintaining the PGM at the targeted level. Chart 6 shows the project surpassed literature-reported DMFCs with PGM-free cathodes. The chart was difficult to read because of the overlap of the performance and the target. The authors report that the air flow rate and the pressure are higher than the proposed operating conditions. The proposed operating conditions were not shown in the first 14 charts. The results indicate DMFC lifecycle cost approached the polymer electrolyte membrane fuel cell (PEMFC) target for Class III forklifts, and for Classes I, II, and III, the fuel cost is less. The research team claims DMFC has lower infrastructure costs; however, this does not appear to be consistent with chart 7; this should be explained. The researchers report Pt/Ru/TiO_2/O-NCNTs (nitrogen-doped carbon nanotubes with open-ended channels) have greater current density than commercial PtRu/C catalysts, but the experimental conditions were not reported. It is unclear at what voltage these data were collected. In chart 8, the Johnson Matthey (JM) PtRu/C has a greater mass activity than the Pt/Ru/TiO_2/O-NCNTs. Why the Pt/Ru/TiO_2/O-NCNTs have a greater current density, as claimed in Chart 9, could be better explained. It is unclear whether the sensitivity of the MEA in chart 10 to applied back pressure is unique to the MEA or cathode. If a platinum or platinum alloy catalyst on carbon replaced the PGM-free catalyst, it is unclear whether the same dependence on back pressure would be observed. It would be helpful to know if the back pressure dependence is a product of the MEA design. In chart 11, the researchers report that "additional spray in [gas diffusion electrode] offsets the ohmic loss." This raises questions as to whether:
  - The additional spray changed the catalyst loading
  - The porosity, particle size distribution, and pore size distribution of the catalyst layers fabricated by different methods were considered when comparing spray versus blade coating
  - Spray or blade coating produces the same wettability of the MEA components.

- It is unclear what methods were used to measure the capillary pressure. For two-phase flow, the contact angle is unknown. The conclusion seems correct. The project needs to explain the shape factor from chart 14.
- The go/no-go milestone has been achieved for power density at a given PGM loading. Synthesized anode durability targets in milestone M6 have not been reached or do not appear to have even been started/ discussed.
- Within any given area investigated, the progress is reasonable, although not notable in terms of achieving commercial relevance. The cell performance is good but not remarkable and requires very high temperatures that would lead to significant methanol evaporation losses at the cathode and environmental concerns. The anode catalyst development is struggling to meet quantities for 50 cm^2 testing when the target should be how to synthesize kilogram quantities or hundreds of square meters of MEAs.
- The project demonstrated the achievement of ~250 mW/cm^2 peak power density. However, it is not clear which material, process, or MEA testing operating condition developed as part of the project led to this improvement. Both the PtRu-based anode catalyst and non-PGM-based cathode catalyst are known in the literature. No specific improvement to the catalyst structure that would lead to activity improvement is demonstrated. Therefore, it is not clear why these two known catalysts would lead to higher power densities (approximately five times better than those reported in the literature). Methanol–air polarization curves demonstrated in this project still show a big drop in cell voltage (~150 mV at < 25 mA/cm^2), indicating that the anode kinetics and methanol crossover are still the major limiting factors in performance. Therefore, it is not clear what tangible benefits these new anode and cathode catalysts provided. Using peak current density and peak power density could be misleading in judging catalyst behavior. Also, the beginning-of-life performance is shown at high temperatures of 99.8°C and 300 kPa. Sustaining performance at these conditions is highly unlikely because of durability concerns. Better understanding of the catalyst kinetics and methanol crossover is required.

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 structure of this project was based on collaboration and coordination, with three team members providing outstanding results. The research collaboration was similarly outstanding. It would be great if more collaborative projects worked this well.
- The team is good, with diverse capabilities to focus on individual aspects of methanol fuel cell development.
- The demonstrated collaboration in this project is excellent. Bringing in a diverse set of collaborators magnifies the impact, and the collaborators chosen are preeminent scientists in the field. While there is reported contribution from all parties, it is not clear that the collaborators are intimately integrated in terms of materials-sharing.
- The team includes four universities, each with defined roles, but there are no industrial entities to move the project forward or a system designer that could commercialize DMFCs. This is likely due to the limited interest/viability of DMFCs in different markets.

Question 4: Relevance/potential impact

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

- The research reduced the noble catalyst cost for DMFCs and enhanced the cathode tolerance of methanol poisoning (already known for non-PGM catalysts, but the project results were very good). The researchers claim a decrease of the methanol crossover, but it is unclear what they did (on purpose) to achieve this. The results are good enough to move on to durability testing and stack testing.
- The technoeconomic analysis is questionable. It appears to assume significant cost reductions in the DMFC technology while allocating no similar optimism to reducing the cost of hydrogen in PEMFC technology, which is clearly one of DOE's primary focuses. Given that DOE's focus for fuel cell applications has

moved to the heavy-duty truck, this forklift-/drone-focused project may not fit in the mainstream of efforts. However, small efforts over a broader range of applications seem worthwhile, and the demonstrated expertise is valuable.

- This research is likely to have only indirect impacts on any DOE Hydrogen Program objectives or in the marketplace.

Question 5: Proposed future work

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

- Perhaps one of the team members could attempt to do a voltage loss breakdown analysis to understand the contribution of losses from anode kinetics, cathode kinetics, methanol crossover, transport losses, etc. This would better guide the team to put priorities on challenges to solve. The project should focus on mitigating methanol crossover, fuel and water management, durability, and better understanding of the anode kinetics needed.
- The reviewer agrees with the planned resolution on chart 17. What is missing is the stack testing; durability and decay testing of a 10-cell stack with 50 cm^2 active area per cell is necessary, along with start/stop testing. If this system is proposed for forklift trucks, operation at sub-zero temperatures will be required.
- Graphene-coated membranes have been attempted in a European project (DOLPHIN) and have not produced promising results. Blocking water essentially blocks proton transport, as the proton exists as a hydronium ion in fuel cell transport. It is not clear that there is any proposed work to utilize the synthesized anode catalyst in an MEA, as all MEA data appear to use JM PtRu and no indication of scale-up/utilization is present. Integration of all efforts should be explicitly incorporated in Year 3.
- The proposed future work is limited and not particularly compelling. The membrane work with graphene and polymer coatings is completely disconnected from the current project and serves only to further broaden an already overly broad approach to addressing meaningful advances in the DMFC space.

Project strengths:

- The overall strength is the assembly of very competent researchers who have the ability to collaborate and coordinate a very complex fuel cell research and development task. The performance has emphasized research in this effort.
- This is a broad, collaborative approach that comprises relevant experts to drive the DMFC field forward. Most milestones have been achieved, even with the tough working environment of 2020. Integration of modeling efforts informs future materials development outside the scope of the project.
- This is a good team with diverse capabilities to focus on individual aspects of methanol fuel cell development. There is good progress on the development of methanol-tolerant cathode non-PGM catalysts.
- DMFCs have been largely ignored in the research community, and it is good to have some presence in this space. Broad academic engagement may be a cost-efficient way to maintain some footprint.

Project weaknesses:

- It appears that the team members have focused on meeting the peak power density targets, which is good, but that has been achieved largely by operating the MEA at very high temperatures of ~100°C and high back pressures of 300 kPa. Such extreme conditions might momentarily help achieve the beginning-of-life performance targets. However, the relevance of these conditions for real-life operation is questionable. Also, no quantitative information is available on the major limiting factors of beginning-of-life performance to better guide research priorities. No progress has been reported toward decreasing methanol crossover or toward demonstrating durability.
- The future work is focused on making the MEA and MEA components better, which is good; however, the results are good enough for addressing the next level of complexity—stack testing and real-life operation. The reviewer agrees with the proposed future work and attacking the identified barriers.
- No emphasis on durability is a weakness, and the lack of component integration makes the good collaboration appear more siloed than it should be.

- The project is overly broad and not deep enough to have significant advances arise. The team lacks any sort of commercialization presence or a compelling case of commercial viability or interest.

Recommendations for additions/deletions to project scope:

- The researchers should identify a DMFC manufacturer or a test facility experienced in durability and stress testing of stacks as a testing collaborator.
- An assessment of durability should be present in Year 3, even if the stated focus is on beginning-of-life power.
- Rescoping will be difficult, but not expanding the project to include the membrane parts would make sense.

Project #FC-319: Low-Cost Gas Diffusion Layer Materials and Treatments for Durable High-Performance Polymer Electrolyte Membrane Fuel Cells

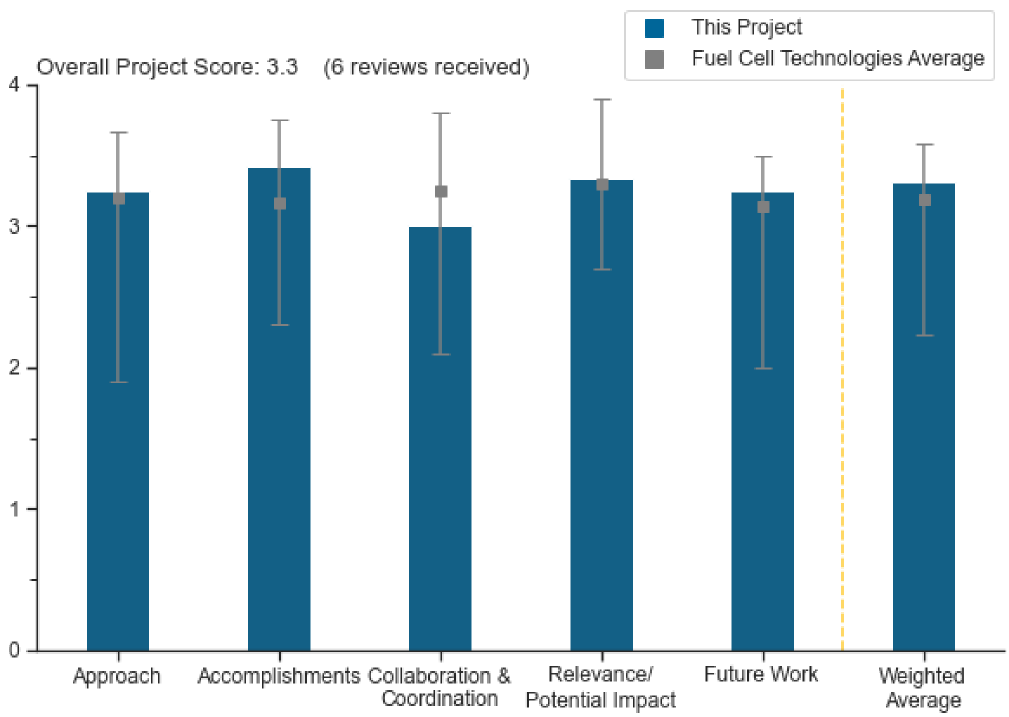
Rod Borup, Los Alamos National Laboratory

DOE Contract #	1.3.0.400
Start and End Dates	10/1/2018
Partners/Collaborators	Oak Ridge National Laboratory, National Renewable Energy Laboratory
Barriers Addressed	• Cost: $14/kW$_{net}$ membrane electrode assembly • Cost: Use of low-cost materials and reduced processing costs • Performance: Mitigation of transport losses through improved water management

Project Goal and Brief Summary

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



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

Question 1: Approach to performing the work

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

- Gas diffusion media have been overlooked by the research community, yet they are a very crucial part of fuel cell systems and need to be looked at more deeply. This is a good approach to looking at an alternative way to make low-cost gas diffusion media. It has merit. The project is looking at reducing cost and improving performance, i.e., adding value to a component that is already in the market. This is a good, sound strategy.
- The project aims at developing new materials and manufacturing methods based on existing fabrics. This is an innovative approach, and the consortium has shown itself to be capable of investigating this approach to the required degree. The approach to researching the suitability of different fabrics for use as GDLs is screening. This is an adequate approach. The consortium is composed of purely academic entities. It is a pity that it was not possible to have a GDL manufacturer participate, as this would have been of great benefit to the project. It is important to notice, though, that the consortium has made efforts to include GDL suppliers.
- The concept of using paper or cloth as a GDL precursor is a good one—so good that it is surprising that microporous layer (MPL) vendors have not already given it a try. Nonetheless, this is a strong effort for a national laboratory team. The potential for elimination of binders and additional pyrolysis is compelling. It would have been good to see a more metric-driven approach, with a spider diagram comparing natural material MPLs to the state of the art. There were metrics distributed throughout but no big-picture summation metrics. Durability is likely to be important, and accelerated testing is recommended to generate faster results with fewer problems. Carbon, hydrogen, and nitrogen (CHN) analysis, or some other structural assay, may help explain variations in hydrophilicity.
- It is good to see a focus on utilizing raw materials that have lower carbon footprints and make the fuel cell technology more cost-competitive.
- The proposed approach—considering materials and processing concurrently to significantly reduce GDL cost while raising performance—appears relevant. Durability aspects should not be forgotten as potential environmental impacts in the process change.
- The project is interesting in its use of so many different natural fiber types. It is not clear whether there are any concerns about the presence of environmental contaminants (trace pesticides, heavy metals, etc.) in the GDLs or whether there is confidence that the processing steps will eliminate the residual trace elements. There is an elemental composition on slide 36, but it is difficult to read. It is unclear whether the team is looking into possible variation in composition due to raw materials from different locations.

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 work is definitely within the scope of DOE's goals for the Hydrogen Program; reducing cost and improving performance of this crucial component in fuel cell systems is critically important. The three-pronged approach of utilizing lower-cost raw materials (fibers), developing hydrophobic surface treatments to replace Teflon, and lowering processing costs is exactly in line with DOE goals. By using cellulosic starting materials and simplifying the production process, the project has reduced costs by over 50%. The team has achieved a high degree of hydrophobicity without agglomeration. This is all very promising, but there is also much more to do, such as looking at the water saturation and looking at overall durability in service.
- The developed process of pyrolysis of cellulosic fibers has proven to be capable of producing materials with, as far as was shown, adequate properties. The process appears to have significant cost-decreasing potential—which is important, as cost is the critical barrier receiving the most attention. For other parameters, such as performance, lifetime, tortuosity, resistivity and conductivity (electric and thermal), and strength, the aim appears to be parity.

- The team has made great progress in identifying top candidate materials and surface treatments. Muslin appears to function poorly on the cathode side without hydrophobic treatment, but with the treatment, muslin is functional. Similar data for Teflonized jute paper would be helpful.
- Very interesting results have been obtained. A large range of raw materials and manufacturing processes led to potential new ways to reduce GDL costs. Among the different characterization tests, more detailed and quantitative mechanical analysis might be carried out to ensure no long-term fatigue issues. Some information on the reproducibility/repeatability of the obtained GDL could be provided. While considering new manufacturing processes in terms of time and energy cost, environmental and safety aspects (the use of silane, for example) should also be considered.
- This project has a very good set of results and a clear presentation. The results of the gas phase treatments look very promising. It is not clear how the cost of chemical vapor deposition treatment with silane precursors compares to the cost of Teflon treatment. There are also questions about slide 18: it is not clear whether the muslin GDL was used at both the anode and the cathode, nor is it clear why the MPL appears to add little to no value at 100% relative humidity (RH).
- The wide range of raw materials that were evaluated showed great diversity in the resulting fibrous structures and properties. Just as discussed for peroxyacetyl nitrate (PAN) materials, more work on looking at the effect of stepwise heat treatment protocols was expected. The hydrophobic post-treatment seems to have the greatest effect on performance, which begs for a closer look at the detailed internal water management.

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 team is composed of well-recognized experts in GDL, imaging, testing, and synthesis capacities. Contacts with GDL industry suppliers will be useful for investigating the upscaling of the most promising solution(s), as well as with Strategic Analysis, Inc., to evaluate the projected GDL costs and the impact on fuel cell system costs for light-, medium-, and heavy-duty vehicles.
- This is a multi-laboratory project, and that form of collaboration is excellent across national laboratories. It would be great, however, if the team had a commercial/industry partner on board that could provide feedback and testing validation and establish a pathway to commercialization.
- There is no doubt about the collaboration between the partners, though the collaboration with external parties could be stronger. The fact that no GDL manufacturer has joined is out of the hands of the consortium, but others, such as commercial users of GDLs in fuel cells, could have been involved.
- The consortium has a strong knowledge base, and collaboration has proven successful, but some business involvement would make it better.
- More industry collaboration would be useful for this project, especially as this shows some promising cost reduction.
- No collaborations were mentioned.

Question 4: Relevance/potential impact

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

- This project is of high relevance to achieving the Hydrogen and Fuel Cell Technologies Office cost, performance, and durability targets, in particular with the current trend to produce membrane electrode assemblies (MEAs) from gas diffusion electrodes.
- This is an important component for fuel cell systems. It has the potential to have a big impact, especially if it leads to significant cost reduction and improvement in performance.
- A main focus of the Multi-Year Research, Development, and Demonstration plan is the reduction of costs, which the project tackles. While other aspects, such as lifetime, thermal conductivity, strength, and water

management, are still subject to research, the results are outstanding. However, other GDL-related goals, such as the avoidance of protruding fibers, are not part of the scope.

- This project has very high potential for impact if all metrics can be met. The team should consider a holistic approach to reporting metrics.
- This project's performance could potentially be improved by the use of high-surface-area fibers, beyond the cost and environmental benefits.
- The story is clear, with a promise of cost reduction, but it remains to be seen how many MEA manufacturers will choose a GDL that has a higher risk of quality issues or year-to-year variability. The financial implications of delivering a defective product to customers could far outweigh the short-term benefits.

Question 5: Proposed future work

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

- The remaining activities are related to lifetime assessment, water management, strength, and thermal conductivity. The project should, however, consider including parameters that are critical for the usability of a GDL in a fuel cell in the remaining characterization schedule.
- Understanding durability and water transport through these layers (with MPL and hydrophobicity) is crucial to realizing the potential of these materials.
- The proposed work is well in line with the project targets. Mechanical characterizations and reproducibility testing of the developed GDL may be explicitly added. Durability testing might be investigated for different applications (i.e., light-, medium-, and heavy-duty vehicles).
- The work proposed could well enhance the performance further, implementing combinations of what has worked before. This reviewer questions the potential success using Nafion™ pillars as hydrophilic pillars, while the porous oxidized organic fibers are among the best water-wicking materials (designed by the plant). The expected effect in the fuel cell should be forecasted by comparing two materials from the SGL Carbon (SGL) range, the second of which also exhibits water-wicking fibers (this is commercially available).
- It would be good to see some study of the GDL thickness. It would be helpful to know how the prepared GDLs compare to SGL 29BC. It is not clear whether compressibility of the GDLs can be measured or how thickness changes with compression. It would also be good to know whether there are plans to test at <50% RH, as the difference between prepared and commercial GDLs is greater at lower RH.
- The team is filing patents and moving the technology forward. It is time to bring in some potential industry partners.

Project strengths:

- The ability to convert raw biological fibers into a fully functional component is impressive—and likely also applicable to other electrochemical fields, such as batteries, flow batteries, electrolysis, and Power-2-X stacks.
- The concept is excellent, with strong potential for impact. A good baseline performance has been established, with a range of design choices. There is a good plan to address stated barriers.
- The project appears well-structured, with a very good level of knowledge for investigating this topic. The partners have strong synthesis, diagnostic, and testing capabilities.
- This is an excellent development effort, with great outcomes and promise. It has strong analytical capability and demonstrated accomplishments as a result.
- An innovative route to the production of GDLs for fuel cells has been taken, and promising results have been obtained, so far. The cost reduction has been the key driver behind the efforts.
- Demonstrating viability of natural fibers is very interesting and sounds promising.

Project weaknesses:

- The project needs industrial partnership to drive this forward. The question is who the partner would be. There are not too many players in this field, nor are there players who would invest the money to scale up this process and optimize it for market applications. It would be worth looking at other potential applications beyond fuel cells and hydrogen devices, such as filtration, to see if there is a potential commercial fit in that area.
- The outcome was anticlimactic because—despite all the variation in the input materials, especially compared to the standard SGL materials—the performance in the fuel cell was pretty much similar. It begs the question of how relevant the anode GDL choice is.
- The team could take a more holistic approach to metrics, including structural aspects and durability, to alleviate the concern that there is some reason this has not been done before.
- The project would have benefited from manufacturer and/or commercial user participation. Further, the parameter relevant to usability could have been given a higher priority.
- Understanding practical considerations such as thickness, compressibility, and performance at low RH would make the study more useful for the industry.
- The reproducibility/repeatability of the obtained GDL could be provided.

Recommendations for additions/deletions to project scope:

- The completion of the lifetime assessment is critical for the overall success of the efforts. Furthermore, it is recommended that the project look into the quality/smoothness of the interface toward the catalyst layer, especially if there is no MPL used in the final iteration. This is one of the highest potential risks remaining. The thickness and compression rate under defined forces are critical for usability and commonly measured. Data should be included.
- While considering new ways of manufacturing process in terms of time and energy cost, environmental (including recycling) and safety aspects (for example, the use of silane) should also be considered.
- This project should add a study on GDL compression and measurements below 50% RH.
- The inherent message in the presentation is that there is no performance sacrifice in comparison to standard materials, but it would be helpful to see if the same goes for durability when lifetimes >10,000 hours are expected.
- The team should start seriously thinking out of the box about how this might move forward into the commercial arena and other potential applications. Then, the project should do enough work in characterizing the materials for other applications to be able to draw new players into this field.

Project #FC-320: Electrode Ionomers for High-Temperature Fuel Cells

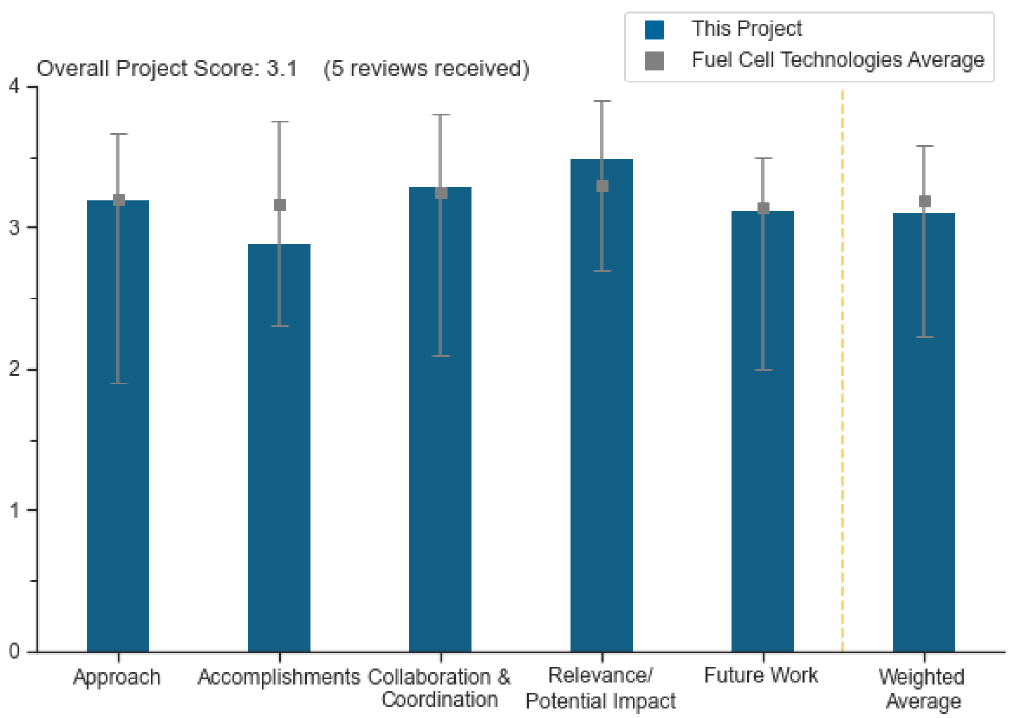
Michael Hibbs, Sandia National Laboratories

DOE Contract #	WBS 1.1.0.804
Start and End Dates	10/1/2018
Partners/Collaborators	Los Alamos National Laboratory
Barriers Addressed	• Cost • Electrode performance • Durability

Project Goal and Brief Summary

Sandia National Laboratories (SNL) seeks 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 Technologies projects.

Question 1: Approach to performing the work

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

- Higher-temperature polymer electrolyte membrane fuel cells (PEMFCs) can be game changers and have large implications for fuel cell system power density, thanks to the potential for decreased thermal management, water management, and increased efficiency. The ion pair approach to high-temperature conductivity in membranes has been one of the most effective approaches for high-temperature polymer electrolyte membranes (HT-PEMs) to date. The use of electron-withdrawing F groups to reduce/eliminate anhydride formation in the phenyl phosphonic acid side chain is a big step forward. Phosphoric acid poisoning of the catalysts can limit catalyst activity. Using a tethered phosphonic acid ionomer may reduce catalyst poisoning compared to an ionomer with phosphoric acid. The project is still using phosphoric-acid-doped membranes. It is not clear if any phosphoric acid is migrating to the catalyst layer during operation and affecting performance.

- The team is well-qualified to perform the work. Investigating different phosphonated ionomer binders over phosphoric acid imbibed polycations is the right approach for reducing kinetic and mass transfer overpotential values in electrode layers. As with most research, new discoveries are made during the project. Here, the team encountered difficulties with anhydride formation with phosphonic acid end groups under dry conditions. This discovery was made after several new ionomer binder chemistries were synthesized and characterized. The anhydride formation precludes their use in fuel cell electrodes, as they do not conduct protons. Borrowing knowledge from poly(tetrafluoro styrene phosphonic acid), it is clear that electron-withdrawing groups in close vicinity to phosphonic acid are needed to increase the functional group's acidity and prevent anhydride formation. The fluorinated styrene moiety has been incorporated into poly(terphenyl) and the SNL polyphenyl backbone to realize new high-temperature phosphonated ionomer materials. There is only one weakness with the approach: the use of ionomers with a lot of phenyl groups that can adsorb to PGM surfaces. Phenyl adsorption can hamper electrode kinetics. Poly(fluorene)-type backbones with phosphonic acid would have been better candidates than the poly(terphenyl).
- The approach to the work appears to be firmly based in rational design of ionomers for the application. More membrane electrode assembly (MEA) diagnostics would benefit the work, as would the discussion of thermal stability and sources of poor performance. Much of this appears to be proprietary.
- Operating at an elevated temperature has many benefits, including easier thermal management and the elimination of humidifiers.
- While the approach in general seems to be supporting higher temperatures for fuel cells, there seems to be some major underlying questions. The chemistry used is very bulky, so it is not clear that it is even possible to achieve theoretical needs for conductivity. Also, it is unclear why the device-level implementations are falling short. Open-circuit voltages (OCVs) are low, while catalyst loadings are very high, suggesting poisoning.

Question 2: Accomplishments and progress

This project was rated **2.9** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- It is clear that the project team encountered the anhydride formation problem during the synthesis of new ionomer materials. Because of this problem, the researchers had to adjust their synthetic strategy to include fluorinated styrene in their problems. The adjustment of the strategy and pivot speaks to the excellent ability of the team. Overall, the team was able to demonstrate excellent fuel cell power density with TPPA [poly(terphenyl-co-tetrafluoro styrene) phosphonic acid] and PA-DAPP [poly(phenylene) with tetrafluoro styrene phosphonic acid side chains] as the ionomer electrode binders. Peak power density values as high as 1.35 W/cm^2 were realized with H_2/O_2, and values of 700 mW/cm^2 were demonstrated with hydrogen–air. These latter values were with the PA-DAPP ionomer; however, a large PGM loading was used: 1.35 mgPGM/cm^2. The goal was to show >500 mW/cm^2 with hydrogen–air, but with a lower PGM loading (<0.125 mgPGM/cm^2). Low-PGM loading research activities will continue in the L'Innovator Pilot project via the Hydrogen and Fuel Cell Technologies Office (HFTO). Still, the team showed 0.5 W/cm^2 with H_2/O_2, with 0.1 mgPt/cm^2 in the cathode with PWN70 [poly(tetrafluoro styrene phosphonic acid-co-pentafluoro styrene)]. This result demonstrates that low-PGM loadings are possible. From a historical perspective, high-temperature polymer electrolyte membrane fuel cells (HT-PEMFCs) typically require high PGM loadings. The team has exceeded state-of-the-art performance and has shown a pathway to low PGM loadings. The team also showed that the PWN70 ionomer with the ion pair HT-PEM is stable for 550 hours at 160°C and 100 hours at 200°C. (The findings were published in the *Nature Materials* journal in late 2020.) This falls short of the stability goal of 1000 hours at 200°C, but it is progress.

- Some of the project milestones have been met, and good progress toward the remaining milestones has been presented. There has been very good improvement in the maximum power density at 200°C over the course of the project.
- The development of fluor-phenyl phosphonic acid side chains to reduce/eliminate anhydride formation in the phenyl phosphonic acid ionomer is a big step forward. Still, it is far from the target of >500 mW/cm^2 peak power density under hydrogen/air conditions with a total PGM loading of <0.125 mgPGM/cm^2, as the performance shown has been with a very high catalyst loading of 1.35 mg/cm^2 platinum, 10 times the target loading.
- The performance gain for the second ionomer discussed appears to be promising. However, limited polymer characterization, no discussion of MEA diagnostic methods, no discussion of platinum content reduction strategies, and hiding durability in the backup slides are all major weaknesses. The project is predicated on higher-efficiency catalyst for lower loadings, but the MEA data being of poorer efficiency, with higher catalyst, requires discussion and justification.
- Materials and testing are taking place, but there is not as systematic of a device for testing, making progress harder to track. While higher-temperature operations are needed, they must also be able to operate at ambient for start-up. There should be testing of start-up and shutdown to confirm if this is possible.

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.

- SNL and Los Alamos National Laboratory (LANL) make a good team. SNL provides expertise in polymer synthesis and characterization, while LANL focuses on fuel cell testing. It is clear the two institutions are sharing materials and results in a timely manner and are moving forward to hit the ambitious project milestones. The project includes industrial partners, such as Advent Technologies.
- This project comprises two national laboratories, where SNL makes the ionomer and LANL tests it in MEAs. This arrangement has appeared to work and has attracted a lab innovator project with Advent to continue. This L'Innovator should focus on the fundamental understanding of MEA performance.
- There is clearly very close collaboration with LANL, which is doing the device construction and testing. Better communication on needs may be helpful.
- There is good collaboration between SNL and LANL.
- The work between LANL and SNL appears to be fairly well-coordinated. Other collaborations, such as with the Fuel Cell Consortium for Performance and Durability (FC-PAD), would be beneficial. FC-PAD labs could perform cell diagnostics and determine voltage loss breakdown, as well as examine durability, phosphoric/phosphonic acid adsorption on catalysts, and effects of decreased catalyst loading. Also of benefit would be collaboration with commercial groups working in the HT-PEM space.

Question 4: Relevance/potential impact

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

- Although HT-PEMFCs have not been a priority for HFTO in the past years, the advent of the ion pair HT-PEMs by LANL has rekindled interest in the technology. In the past, the main problem with HT-PEMFCs has been the loss of phosphoric acid from polybenzimidazole (PBI) at elevated temperatures and humidified conditions and the role of phosphate poisoning in the electrode layers. Because the new ion pair HT-PEM can immobilize the phosphate anions in the membrane matrix via electrostatic interactions, these problems are no longer a serious concern. Furthermore, pairing the ion pair HT-PEM with phosphonated ionomer binders has led to remarkable breakthroughs in peak power density values of HT-PEMFCs that exceed state-of-the-art values (e.g., BASF's Celtec). The HT-PEMFCs with these new materials can operate across a wide temperature range (100°C to 220°C), depending on the application, with low humidity, and can be important for heavy-duty vehicle applications, an important area now for the HFTO. In a very short period of time, the project team has demonstrated HT-PEMFC power density and durability across a wide temperature range, and good performance is starting to be realized with lower

PGM loadings. The high-temperature operation can eliminate the humidifier for the fuel cell stack and reduce radiator size. At elevated temperatures, the fuel cell can run on hydrogen containing CO (i.e., low-cost hydrogen from steam methane reforming). With further maturation of this platform, future priorities should consider potential cycling for drive cycles and start-up/shutdown testing.

- Intermediate-temperature phosphonic acid fuel cells are a potentially high-impact technology that is currently at a low technology readiness level. DOE is well-positioned to support this early work, and ionomer development is a smart area on which to focus.
- Developing lower-cost polymer materials while simplifying the system has excellent potential to advance progress toward DOE research, development, and demonstration goals.
- The approach, if it works, is very much needed. Start-up–shutdown testing is needed to confirm viability, as is the reduction of PGM and the demonstration of needed mechanical properties and electrical conductivity.
- The goal for higher-temperature operation aligns well with the HFTO goals and attempts to improve efficiency and reduce fuel cell balance of plant by operating at higher temperatures. The very high catalyst PGM loadings used decrease the relevance and potential impact. The high loadings can mask durability issues and issues associated with anion adsorption on the catalyst, leading to poisoning and low activity. If poisoning is an issue and these high loadings are required, MEAs of this type will not be economically competitive with traditional low-temperature PEMs, even with the advantages of operating at higher temperature. To be relevant, MEAs with lower PGM loadings need to be demonstrated.

Question 5: Proposed future work

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

- The team has clearly articulated future research directions and has worked on some of them. It is not clear why high ion exchange capacity PA-DAPP is needed. It probably relates to enhanced conductivity in the electrode layers, but it is not clear that greater electrode conductivity is needed. Although not all project milestones were satisfied, many were accomplished, and much progress has been made. The future directions on low PGM loadings and stability with various phosphonic acid ionomers will accelerate HT-PEMFC technology further.
- The plans to work with Advent on future development should be beneficial. The proposed testing with low PGM loadings (or at least normal for low-temperature PEMs) is critical and should be near the highest priority for future work.
- Focusing on durability, platinum loading reduction, and MEA performance are the right places to look for the future.
- Demonstrating long-term durability is a critical next step.
- The PGM loading needs to be reduced, not explored.

Project strengths:

- The major strengths of the project are the availability of three different phosphonic acid ionomers with various backbones, two of which give over 1 W/cm^2 with hydrogen and oxygen. Progress has been made in the project with low PGM loadings, and a peak power density of >0.5 W/cm^2 with hydrogen–air has been demonstrated. The team members' strength is synergistic and contributes to the project goals.
- The project team's approach to HT-PEMs has been one of the few successful approaches in this temperature range.
- The strengths of this project are that it comprises the early leaders and innovators in phosphonic-acid-type intermediate-temperature fuel cell MEAs and that the progress in polymers is promising.
- This is a novel approach to 150°C–250°C fuel cells.
- This is a needed approach, and the project has good device-level testing.

Project weaknesses:

- This project needs more methodical device-level testing of the membrane chemistries to determine whether the ionomers are effective and why OCVs are so low while PGM loading is so high. The principal investigator seems unaware of the device-level testing needs.
- The only weakness was the lack of consideration of phenyl adsorption on PGM catalysts, as the DAPP and TPPA backbones have lots of aryl groups. The poly(fluorene) backbone should be considered.
- MEA diagnostics and durability tests are underdeveloped, and discussion of these activities is not presented sufficiently.
- The use of very high Pt loadings is considered a weakness. The project needs to demonstrate it can get performance with more reasonable Pt content.
- More time would have been needed to complete all project milestones.

Recommendations for additions/deletions to project scope:

- The future work should investigate the potential for replacing the phosphoric acid in the phosphoric-acid-doped quaternary ammonium poly(phenylene) (PA-QAPOH) membrane with the newly developed fluorophenyl phosphonic acid copolymer.
- Understanding sources of kinetic losses would be immensely beneficial to this upstart MEA technology. Discussion of incorporating the ionomers as membrane polymers would be of interest.
- A legend is requested for the comparison plot to commercial PBI on slide 25.
- The project should use start-up/shutdown testing with the use of different catalysts.
- The project funds have been exhausted, and the project is at the end of its timeline, so no recommendations are provided.

Project #FC-323: Durable Fuel Cell Membrane Electrode Assembly through Immobilization of Catalyst Particle and Membrane Chemical Stabilizer

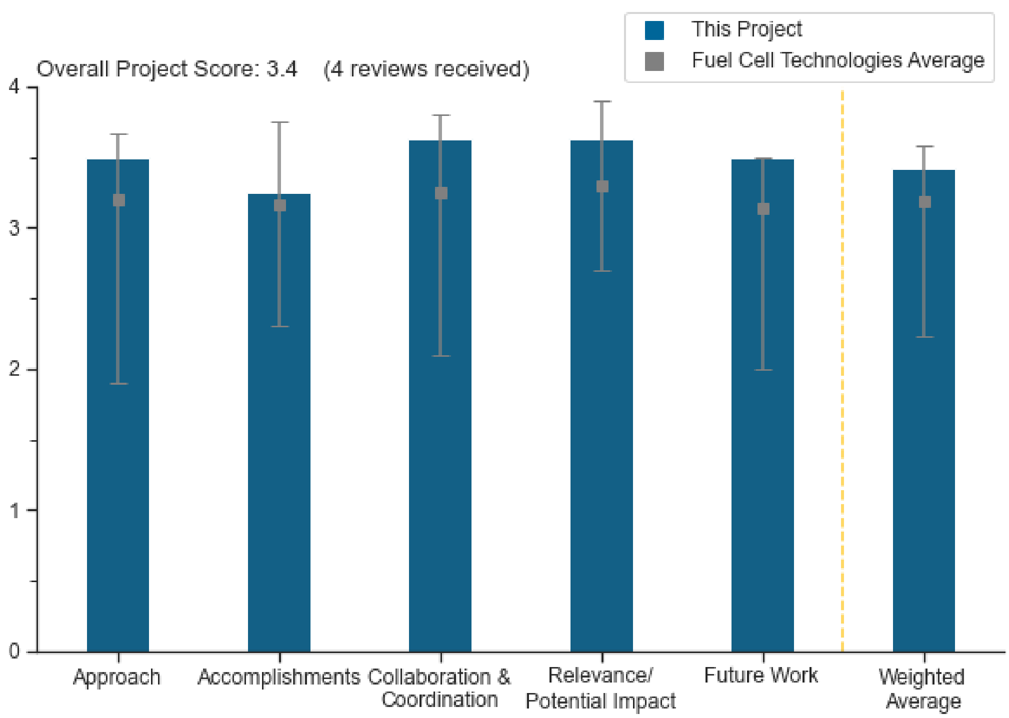
Nagappan Ramaswamy, General Motors LLC

DOE Contract #	DE-EE0008821
Start and End Dates	10/1/2019 to 2/28/2023
Partners/Collaborators	3M Company, Pajarito Powder, LLC, Colorado School of Mines, Cornell University, Million Mile Fuel Cell Truck (M2FCT) Consortium
Barriers Addressed	• Durability: <10% power degradation after 30,000 hours • Cost ≤0.2 mgPt/cm^2 cathode Pt metal loading • Efficiency >65% efficiency to decrease fuel cost

Project Goal and Brief Summary

This project aims to develop highly stable catalysts and membrane materials for use in direct hydrogen-fed polymer electrolyte membrane fuel cell (PEMFC) membrane electrode assemblies (MEAs) in medium-duty (MD) and heavy-duty (HD) truck applications. The materials will feature low platinum-group-metal (PGM) loading, fuel efficiency of greater than 65%, and a lifetime of one million miles. If successful, this project will deliver highly durable MEAs for PEMFC applications to enable use in HD trucks. It will also elucidate the fundamental degradation mechanisms of MEAs.

Project Scoring



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

Question 1: Approach to performing the work

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

- The project aims to develop stable catalysts and membrane materials for MD/HD fuel cell trucks with loading, efficiency, and durability targets. The approach for durable catalyst development includes modification of the Pt-C and Pt-ionomer interfaces using polymer additives and graphitized mesoporous carbon. Then the project proposes a stable membrane development using covalently tethered heteropoly acids (HPAs) and dispersed $Ce_{0.85}Zr_{0.15}O_2$ (CZO) additives. These approaches rely on promising paths for improving component durability, with a well-designed path toward the components' integration into a more durable MEA.
- This project is pursuing multiple promising approaches to address two major PEMFC decay mechanisms: catalyst losses and membrane degradation. The concepts are good, and the plans to execute them are well-designed. However, not all major decay mechanisms will be addressed by this project. For example, it is not focused on carbon corrosion. However, the carbons being used here are either standard materials or are the new engineered catalyst support and can be modified to improve stability if needed (e.g., via additional graphitization).
- The goals and approach to achieving the goals are clearly defined. The project seems well-defined and integrates with other efforts being funded by the U.S. Department of Energy.
- This project is focused on critical barriers, but it seems like two completely separate work streams.

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.

- A range of catalysts has been developed, evaluated, and supported by characterization data that will likely help with material design and down-selection for future tasks. The project reports catalyst accelerated stress tests with end-of-life (EOL) versus beginning-of-life (BOL) comparison that shows possible paths for exploration, such as improved durability for the annealed Pt/high-surface-area carbon catalyst.
- Timely progress toward the objectives has been made (despite Covid-19) and has been demonstrated through measurable performance indicators. In terms of measuring catalyst performance, the focus on electrochemical surface area as an indicator of performance is questionable when it does not correlate well with EOL and BOL performance (slides 9 and 12).
- This project is taking on high-risk research, and it is great that DOE is supporting it. These concepts are very difficult to characterize and validate without a large amount of durability data. The project team seems a bit aggressive with its timeline, given the status presented. MEA integration and interpretation of fuel cell results will be difficult without a fundamental understanding of these prototype materials and the critical formulation parameters.
- The progress the team presented is appropriate, relative to the status of the budget (approximately 25% complete). However, there are no results yet that strongly indicate that critical barriers will be overcome. The results to date do show that the team can make some of the proposed new materials, but further work is required to show that these are truly effective.

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.

- There is a strong collaboration team, from the material (catalyst and ionomer) development side to various characterization efforts. The project aims to coordinate a wide range of tasks, from material design to diagnostics, with other entities, which will help with the progress.
- The project has many great team members, and they appear to be working well with each other. There is good evidence of strong collaboration here.

- There is clearly extensive collaboration with industrial and institutional partners and relevant experts in the field.
- The team is ideal for this work, and the project is leveraging expertise exceptionally.

Question 4: Relevance/potential impact

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

- The project aims to improve MEA efficiency and durability through ionomer and catalyst development, which supports the Fuel Cell Technologies subprogram goals and objectives for achieving the HD targets.
- Anchoring the catalyst particles is a most critical challenge for HD applications and could simplify system operation.
- Catalyst lifetime and membrane lifetime are critical to enabling success of fuel cells in HD trucking. The membrane work might have a greater impact (this failure mode is more critical). In addition, it is interesting that the proposed catalyst additives have yet to show a level of benefit similar to simply annealing the Pt catalyst, so the potential impact of the catalyst additives portion should be downgraded.
- This project addresses a major challenge for HD vehicles, which is much longer durability than light-duty vehicles. However, it is not clear that the project will contribute directly to the other major HD vehicle goal, which is higher efficiency. The project may do so indirectly by enabling higher operating temperatures with more durable components. However, carbon corrosion may be more of a concern at higher average cell potentials, which does not appear to be addressed here.

Question 5: Proposed future work

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

- Future work builds upon the current findings in a logical manner and provides a detailed list of challenges and reasonable methods to overcome them. The proposed future work accounts for some of the possible risks, given that the project has multiple approaches. The other assumptions and risks are also listed explicitly.
- Proposed future work is logical, barriers have been considered, and paths to mitigate risk have been suggested. Given that annealed Pt is identified as a promising path on slide 21, this should be addressed in the scope of future work.
- Future plans are clear and are very appropriate.
- It seems like there is much to do before the first go/no-go milestone, but the plan is appropriate.

Project strengths:

- The project explores both ionomer and catalyst development with novel approaches that have potential to improve MEA durability. The extent of collaboration and the detailed tasks covering design, characterization, modeling, and integration are project strengths.
- There are excellent concepts that show promise. There is a great team with the correct skills and capabilities. There is a good plan for future work.
- Clear goals and barriers have been identified. The project has an interesting approach and technology. The analysis is excellent.
- This project has a strong team that is focused on critical technologies for next-generation fuel cells.

Project weaknesses:

- This is not a major weakness, but some additional clarification on the scope would be helpful, as noted in the recommendations section.
- There is no focus on carbon corrosion. It is not clear if General Motors (GM) is really committed to HD vehicle applications since this project is ultimately the same things needed for light-duty vehicles. (Of course, it is not clear when GM may ultimately introduce any commercial fuel cell electric vehicles.)

- Significant work will be required to understand whether the intended mechanism/structure is achieved.
- The project is not structured to follow up on interesting benchmarking results (the annealed Pt).

Recommendations for additions/deletions to project scope:

- Some information on whether approaches are synergistic or complementary in nature would be helpful. For example, it would be helpful to know whether CZO is being added to the electrodes with a Ce-free membrane or to the MEA membrane with some Ce salts or HPA. The selection methods slide could be edited or expanded to demonstrate the integration of these strategies in the future and the decision-making process.
- The team should consider trying to combine multiple approaches at the end of the project, if possible. For example, if both Pt–carbon and Pt–ionomer interface modifications are successful, then the team should see whether they can be combined in the same catalyst layer. It would be useful to know whether the benefits are additive. Additionally, the project should make an ultimate MEA with all of the successful approaches combined and compare the durability results to each of the individual approaches.
- It would be interesting to see more work understanding the benefit of annealed Pt, including the improved oxygen transport mentioned on slide 26.
- This project would benefit from expanded parametric studies.

Project #FC-324: Reversible Fuel Cell Stacks with Integrated Water Management

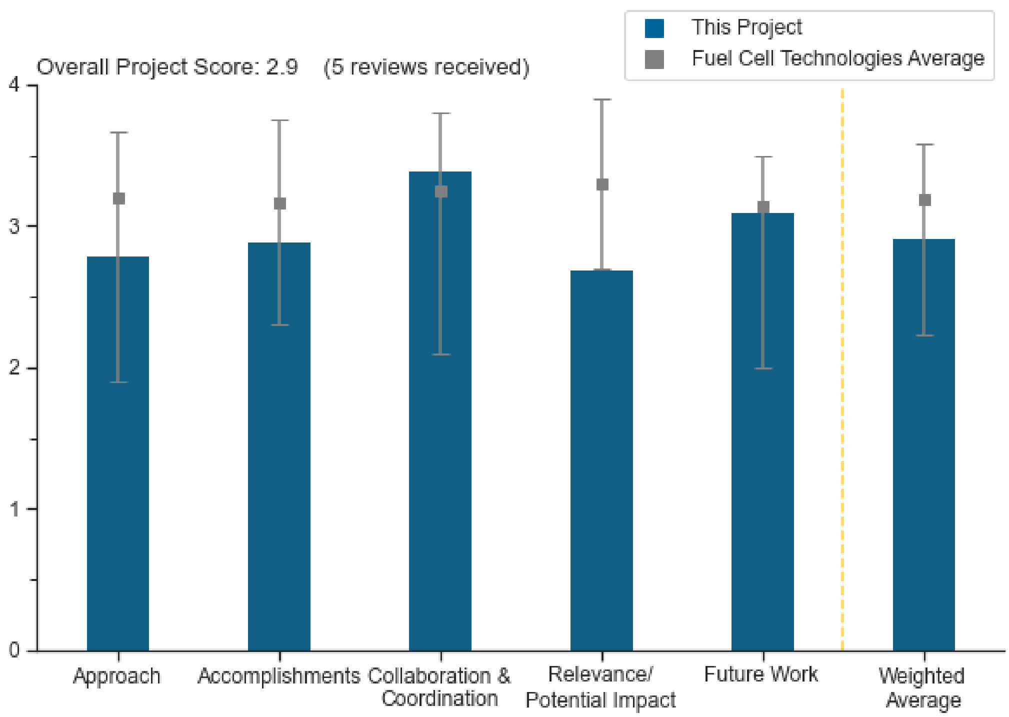
Teddy Wang, Plug Power Inc.

DOE Contract #	DE-EE0008901
Start and End Dates	3/18/2020 to 3/31/2022
Partners/Collaborators	University of Connecticut, Los Alamos National Laboratory
Barriers Addressed	• High capital cost for separate traditional hydrogen production and fuel cell systems • Complicated reversible fuel cell system balance of plant, leading to poor system durability and performance • U.S. Department of Energy reversible fuel cell technical target: 55% voltage round trip efficiency (RTE_V) @ 0.5 A/cm^2 fuel cell and 1 A/cm^2 electrolyzer (2025 target)

Project Goal and Brief Summary

This project aims to develop unitized reversible fuel cell (URFC) stacks and systems by combining Plug Power Inc.'s (Plug Power's) non-flow-through fuel cell design with optimized water management. If successful, this project could dramatically reduce the capital cost of hydrogen fuel cell systems.

Project Scoring



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

Question 1: Approach to performing the work

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

- This is an outstanding approach starting from a strong platform and addressing key deficiencies—namely, catalytic activity and water management—through materials studies.

- There is a good understanding of the operational targets for both modes and an experimental matrix to support it, but it is not clear whether consistent operation is needed (in a tight window for efficiency) or if it is sufficient to measure a fast I-V curve (current–voltage characteristic curve).
- Slide 4 states that the gas compressor and saturators are no longer necessary with Plug Power's design, which simplifies the system layout. An explanation is not given. Diagnostics are not used to identify sources of performance losses more clearly. A simple approach using air and a 79% helium and 21% oxygen mixture would readily provide an estimate of the mass transfer overpotential associated with oxygen transport in the gas phase (porous transport layer [PTL], microporous layer [MPL], and catalyst layer), which is dependent on the level of liquid water present in pores. More elaborate diagnostics could also be used to gain additional information such as the measurement and separation of mass transfer coefficients into fundamental contributions (molecular diffusion, Knudsen diffusion, and ionomer permeability) and neutron imaging. Results obtained with these diagnostics would accelerate design tasks and help determine whether ultimate targets can be met. The University of Connecticut (UConn) is responsible for electrode characterization (slide 17). However, it is unclear which diagnostics will be used to improve the catalyst layer design. The relevance of these test (PTL/MPL) combinations with or without a hydrophobic treatment) is questionable. The planned Los Alamos National Laboratory (LANL) MPL (slide 27) has a different configuration with a non-uniform in-plane distribution of two different hydrophobicity sites. In contrast, the design depicted in slides 11−14 has a through-plane distribution of two different hydrophobicity sites. Therefore, results are not necessarily applicable to the LANL MPL. This situation raises doubts about a successful integration and application of the results shown on slides 11−14 to the design of LANL MPLs.
- The approach consists of leveraging Plug Power’s static feed fuel cell design and adapting it to electrolyzer/URFC performance. The design appears to be limited in capability relative to a forced feed fuel cell design, but ultimately, the work done in the project will show whether the design is effective enough. Some of the discrete testing should be done under more static conditions to mimic the lack of forced flow. It is unclear what benefits the MPL or amphiphilic coatings will have when capillary forces are required to remove water from fuel cells and switch between modes.
- The combination of different electrochemical devices into a single one would imminently result in underperforming on each side, even with the feasibility of the approach proven. The economic analysis, detailing cost benefits of the approach with sensitivities to underperformance and downtime for switching between two modes, should have been conducted prior to starting the experimental work.

Question 2: Accomplishments and progress

This project was rated **2.9** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The UConn team has demonstrated significant progress on developing a materials solution and verifying electrochemical feasibility.
- The presentation showed clear results on screening various components with progress in the preferred selection. It is expected that a large set of variations will be tested going forward.
- Good progress can be made across most tasks; milestones are on track. Results so far indicate the high likelihood of meeting targets and DOE targets. Work was delayed due to COVID-19 and company acquisition.
- Progress appears to have been limited because the project was transferred from Giner, Inc., to Plug Power. For example, baseline performance should probably have been established at this point. It is also important to engage project partners, especially LANL, as soon as possible. It is not clear how the pseudo-MPL (Ti sinter) fits into the overall approach in terms of water management. It is unclear how this meshes with the amphiphilic MPLs.
- Materials shown on slides 6 to 8 indicate a platinum-metal-group loading of 1 mg/cm^2 for the cathode. Materials on slides 9 to 14 show a total metal loading of 2.1 mg/cm^2. It is unclear how the target of 1 mg/cm^2 will be met while concurrently increasing the roundtrip electrical efficiency to 55%. On slide 13, the graph containing the 150% fuel cell polarization curves is seemingly missing data (the sinter + felt -

270 + 250 µm case). On slide 23, results were obtained with a 5 cm^2 active area cell rather than the planned 50 cm^2 active area cell (slide 24).

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 project is structured very well, sharing equally the scope of engagement between industry, university, and national laboratories in the final deliverable.
- There is a large and diverse team of researchers. The project should continue to try to leverage user facilities such as the National Institute of Standards and Technology (via LANL).
- The consortium partners are chosen well, and the takeover by Plug Power does not seems to have affected collaboration.
- There appears to be some good initial coordination, but the contributions from partners could be better emphasized.
- There are indications that coordination could be improved, as the platinum-group-metal loading of the UConn samples does not meet the target (includes only one electrode), and LANL samples have not yet been provided.

Question 4: Relevance/potential impact

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

- The approach of making an operational electrolyzer reversible is a good one because it can take advantage of existing materials properties in the harsher electrolyzer regime. The team must make more progress on water management to realize the largest impact.
- Years of community experience with reversible fuel cells or reversible electrolyzers have always highlighted compromised performances and efficiencies at stack level. The application of this concept is often limited to specific situations.
- The project partially supports the DOE Hydrogen Program and DOE research, development, and demonstration objectives.
- The project is aligned with DOE URFC goals, yet it is unclear what the performance of the stack hardware is, so what its ultimate performance characteristics might be is hard to say.
- Cost and durability aspects will not be addressed. For instance, the added complexity of the cell and stack design with added cell layers (bubble point membrane and water chamber) is expected to increase cost. Therefore, the impact of this project on the DOE Hydrogen Program is limited.

Question 5: Proposed future work

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

- Considering that only three months of work have been reported, the proposed future work, containing the majority of the proposed project, makes sense.
- Future work is outlined well on the materials development side; however, more design work is desirable on the switching mode and optimization of the non-flow plate to avoid starvations.
- The project should drive more out of the reactive spray deposition technology (RSDT) part of the project, if this is on the critical path for membrane electrode assembly. Test stand upgrades to enable automated testing are great.
- The anticipated step to a larger stack configuration will show how scalable this stack technology is while being subjected to natural flow patterns in the active area.
- The fabrication of the two core components, the catalyst layer and MPL, is tunable with multiple parameters, which reduces risks.

Project strengths:

- This is a good team with a proven track record and good previously demonstrated technology from amphiphilic coating, RSDT, and all that Giner, Inc., and Plug Power have done in this space.
- The concept is interesting; it leverages the UTC Power porous bipolar plate water management concept. The fabrication of the two core components, the catalyst layer and MPL, is tunable with multiple parameters, which reduces risks.
- The project shows there is sufficient experience and expertise on board to further develop the existing reversible fuel cell architecture. There is no shortage of conceivable solutions to evaluate.
- The project starts from an excellent working electrolyzer platform. The proposed materials solutions appear promising.
- The project strength is in partnership and material development on UConn side.

Project weaknesses:

- The project weakness is insufficient design work on compatibility between amphiphilic MPLs and the electrolyzer media. The approach to switching modes between fuel cell and electrolyzer is not sufficiently worked out. The non-flow-through plate could contribute to starvation zones, which is necessary to consider if extended lifetime is planned. Durability considerations are not presented, and shorter stack life may negate the entire purpose of integration into the same hardware.
- The approach justification is insufficient. The absence of cell diagnostics is notable, which will slow technology development. The relevance of several completed tests is debatable. There is a high level of risk in meeting electrical efficiency and platinum-group-metal loading targets concurrently, considering the development plan is unclear. Team coordination could be improved. Cost and durability aspects will not be addressed.
- The development in this project is leaning heavily on practical material selections going in one cell platform (up to now), which is then tested to measure its characteristics. Without fundamental understanding or modeling on what issue we are trying to resolve (first), the project is inherently limited to practical compromises.
- There is much recent literature in this space that is being omitted or presumably not used for comparison. RSDT sprayed in layers not mixed together; this might be a problem functionally.
- There was a limited period of operation in 2020. Significant contributions of partners are yet to be realized.

Recommendations for additions/deletions to project scope:

- The project should conduct economical sensitivity analysis accounting for the stack lifetime in the fuel cell mode, time for switching between the modes, and loss of performance on both modes of operation.
- Operational durability and stability need to be taken into account to justify the underlying goal to make this fuel cell/evaporatively cooled architecture more attractive than two dedicated systems. The overall cost assessment needs to consider the lifetime and over-dimensioning catalyst layers to design robustness against off-spec conditions and dynamic cycling.
- The option 2 bifunctional oxygen electrode appears to be too compact (slide 8) and perhaps should not be used. Regarding slide 15, the use of precious metals is not ideal. Titanium is an earth-abundant element, and therefore, it is preferred in comparison to Ru-based materials.

Project #FC-325: Fiscal Year 2019 Small Business Innovation Research Phase II: Controlled Porosity and Surface Coatings for Advanced Gas Diffusion Layers

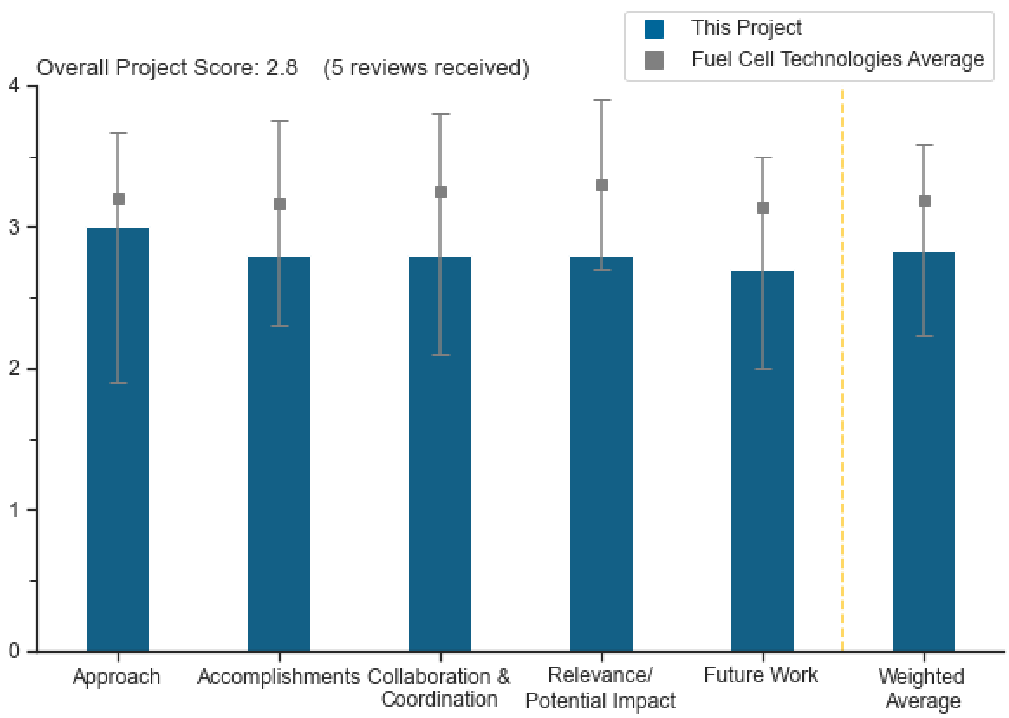
Kristina Bennett, Physical Sciences, Inc.

DOE Contract #	DE-SC0018606
Start and End Dates	5/28/2019 to 5/27/2021
Partners/Collaborators	University of Tennessee, Knoxville, University of Connecticut
Barriers Addressed	• Cost

Project Goal and Brief Summary

This project aims to demonstrate the use of an ice-templating method to tailor the properties of gas diffusion layers (GDLs). This method is scalable and can manufacture GDLs at an estimated 20% lower cost than current methods. If successful, this project will support the adoption of cost-competitive polymer electrolyte membrane fuel cells (PEMFCs) through the production of high-performance GDLs. Physical Sciences, Inc. (PSI) is collaborating with the University of Tennessee, Knoxville (UTK), and the University of Connecticut (UConn) on this project.

Project Scoring



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

Question 1: Approach to performing the work

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

- The project focuses on developing a new approach of ice templating to reduce GDL production costs by 20%. Such a method can also potentially improve performance and durability by tailoring transport properties and using electrochemically stable materials, respectively.

- This is a very interesting approach to fabricating a GDL structure. Overall, the elimination of high-energy heat treatment processing steps and a relatively simple process flow makes this an interesting GDL development process.
- Ice templating should result in a significant reduction in process complexity, leading to cost savings. The challenge will be to demonstrate equivalent performance with ice-templated GDLs across all performance metrics.
- The ice templating method is a seemingly novel technique to make GDLs. The enhanced durability of the GDLs is seemingly due to the electrochemically stable materials; however, no information was given about the materials. It is unclear whether these are standard peroxyacetyl nitrate fibers, carbon, Teflon™, etc. In the cost analysis, an identical raw material cost is used; thus, it appears the materials must be the same; thus, no enhanced durability should be expected. As the material cost is the same, the advantage of this technique would seem to be a reduced cost and an ability to tune the GDL porosity, yet the project does not provide any characterization detail related to the difference in porosity or the desired porosity.
- The idea to use ice as a template is novel and practically explored in reference to a commercial material, but true benefit was not evaluated in a cutting-edge window of operation.

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.

- Overall, good progress was made on this project, with the production of a GDL candidate material that was comparable to a commercial reference. The voltage and current density decay look promising, although the overall performance appears to be much lower than would be expected, as most commercial GDLs operate around 0.7 V at 1 A/cm^2 under hot, wet conditions. It is also unclear from this effort how much material was made and how much variation in typical GDL properties (thickness, basis weight, compressibility, etc.) was seen from batch to batch. Regardless of the drawbacks, the success of making a GDL material with this methodology is impressive.
- The project has demonstrated GDLs with potentially 20% lower cost and identical durability. The project is unclear as to whether the performance of the GDL is identical to commercial GDLs; it is clear only that the cell voltage differential, during a durability test, is identical to that of commercial GDLs over 200 hours and 360 hours. A key to GDL performance is the ability to operate at a range of conditions (e.g., temperature and relative humidity [RH]). The project did not present the operating conditions used, nor did it show a range of operating conditions, so the overall operating ability of these GDLs is unclear.
- A cost reduction of 20% was demonstrated by modeling, with the main savings from reduced energy consumption, less labor, and less equipment and maintenance. The project demonstrated performance and durability competitive with a commercial product (SGL 39AA). However, reported durability and performance were quite far from DOE 2020 targets.
- The presentation showed clear outcomes from the experimental matrix and could show a higher performance in the graphs presented.
- Additional performance metrics, such as resistance, porosity, and hydrophobicity, would provide more confidence of success.

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 collaboration on this project appears to be very good, with each partner having a key role to play in either the generation or the evaluation of the GDL material.
- PSI, UTK, and UConn have good testing capabilities for this project to be successful.
- The project has good partners that could be better integrated. Additional data from UTK could be reported. Stack testing in slide 9 may be from UConn, but if so, it should be attributed to the collaborator.

- PSI collaborates with UTK on GDL performance characterization and with UConn on PEMFC testing. It is unclear how effectively the results from collaborators might have contributed to improvement of GDL production at PSI because very limited data are reported.
- The wish was expressed to work with a GDL manufacturer and reduce the energy spent on drying and firing the GDL processed in the regular manner, but the presentation left the impression that such potential customers were not contacted in prior discussions sufficiently to understand the relevance of this issue.

Question 4: Relevance/potential impact

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

- Lower-cost materials with expanded durability fit into Program goals and are needed for both light-duty and heavy-duty vehicles.
- This is an interesting project, and the initial success is interesting to see. It is difficult to judge the overall impact without more data on piece-to-piece and lot-to-lot variability. The initial cell performance is also somewhat underwhelming, but it will be interesting to see how the cell performs in stack testing. The results achieved so far are interesting, but the potential impact of this project is relatively low right now—but could change as the process is refined and performance improves.
- The impact of a functional, low-cost GDL could be significant. The impact could be better demonstrated here. The membrane electrode assembly (MEA) data shown are not compelling. It is unclear whether the comparison MEA is state of the art, but if it is, it is best to show absolute polarization rather than relative polarization. It is best to compare MEAs under the same conditions. On slide 9, the PSI GDL at 0.6 A/cm^2 is compared to SGL Carbon at 0.7 A/cm^2.
- In the current project period, the goal has been set to reduce production cost by 20%, while demonstrating the same level of performance and durability as commercial products. The project could have a bigger impact if effort were dedicated more to performance and durability improvement instead, considering the GDL is not a top cost contributor to fuel cell stacks, but performance and durability improvements can lead to overall stack cost reduction.
- The functionality of the resulting GDL was not sufficient to evaluate this in the higher fuel cell performance window expected by DOE. It is possible that the party executing the comparison testing had insufficient testing resources.

Question 5: Proposed future work

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

- The proposed future work covers the main concerns associated with the material by continuing to evaluate the performance over longer periods of time, looking at stack testing, and examining the viability of scale-up, which should include challenges related to piece-to-piece and lot-to-lot variability.
- The future work seems reasonable, but the information from the project could be expanded by additional reporting, testing, and characterization.
- The proposed future work should consider including more GDL material property characterization and make an effort to address the big performance gap toward DOE 2020 targets.
- There is limited scope in the project to explore further benefits of using ice particles to template the porosity. The work is independent of the slow production conditions. Making enough material will show process stability and reproducibility.
- The pathway to 10 m^2 is not clear. There should be some effort to identify roadblocks for these materials and mitigation strategies. Process throughput should be considered. For example, the team should consider how hold time to control porosity effects square meters per day.

Project strengths:

- This is an interesting project with a very novel approach to manufacturing a GDL material. There has been some significant success in making a material that performs relatively well compared to a commercially available GDL in one set of conditions.
- The project has a strong starting point with a low-cost process. Good progress has been demonstrated toward scaled-up manufacturing, and project partners are in a position to contribute significant performance metrics.
- This project has a novel design of the ice-templating process that can reduce the cost of production and potentially improve durability and performance.
- A clear effort was made to assemble process instruments to fabricate the targeted structures.
- The project is showing a reduction in cost and equivalent durability.

Project weaknesses:

- The overall weakness in this project is the generally lower-than-expected performance when compared to single-cell testing that has been seen elsewhere (e.g., ~0.7 V at 1 A/cm^2). This may be due to the size of the active area in the cell or poor MEA components. Further data looking at both within-part and part-to-part variability would also be helpful to understanding the manufacturing process. The issues related to scalability will be addressed in the proposed future work and may improve upon this current weakness.
- The project could use more characterization related to the GDL properties and material properties. More information about the overall performance over a range of conditions would be good. Since the strength of the ice templating technique is the ability to control the manufacturing, the team should demonstrate a range of GDL properties and porosity.
- The weakness is the lack of material characterization that is critical to validating the proposed control of GDL physical properties and to helping corelate to MEA performance and durability results. This also makes it difficult to assess how the team can propose effective countermeasures to narrow the gaps between current status and DOE 2020 targets.
- The performance metrics are underreported, external collaborators seem under-engaged, and comparison to commercial GDLs is not compelling. The project must be able to assess the state of the art.
- Without the application of a microporous layer on the GDL, this component is not complete and is unable to demonstrate its true potential.

Recommendations for additions/deletions to project scope:

- The project should expand its characterization and information related to the GDLs and materials. This should include a discussion about porosity, tailoring the porosity, what the technique can do, and what porosity is wanted. Other information should include the material hydrophobicity as a function of operating time. Testing should include showing direct beginning-of-life performance and voltage–current–resistance performance comparisons to commercial materials over a range of temperatures and RH.
- The capability to template a structure, rather than presenting only a reduction in drying costs, seems more attractive from a business point of view. The project is asked to verify the commercial interest in the presented "Unique Selling Points" slide.
- It would be great if the project team could consider adding or enhancing the following studies into the scope:
    - Measure and compare the new GDLs' physical properties (such as electrical resistivity, thermal conductivity, hydrophobicity, porosity, density, and compressibility) to those of commercial products.
    - Include gas diffusion analysis in the fuel cell performance testing to compare with commercial products.
    - Demonstrate how the new process may control key parameters, such as pore structure, that can lead to the change of gas permeability and MEA performance improvement.

- The proposed future work accounts for most of the concerns, although adding in variability analysis of critical GDL values (thickness, basis weight, etc.) would be beneficial for this project.
- The project might consider the effect of adding a microporous layer for water management.

Project #FC-326: Durable Membrane Electrode Assemblies for Heavy-Duty Fuel Cell Electric Trucks

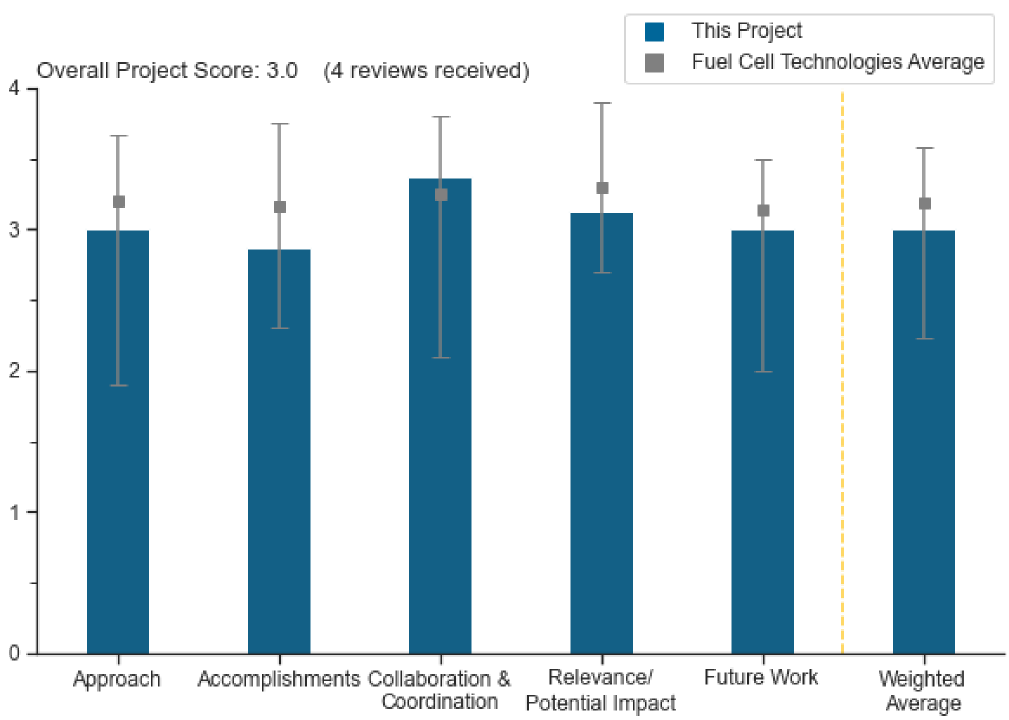
Vivek Murthi, Nikola Motor Company

DOE Contract #	DE-EE0008820
Start and End Dates	Q3 2020 to Q3 2023
Partners/Collaborators	Georgia Institute of Technology, Northeastern University, Carnegie Mellon University, Los Alamos National Laboratory, National Renewable Energy Laboratory, Oak Ridge National Laboratory
Barriers Addressed	• Durability: Improve stability of membrane electrode assemblies for operating conditions relevant to heavy-duty trucks • Performance: Increase catalyst while reducing ionomer poisoning effects to achieve high power density and higher efficiency • Cost: Enable reduction in platinum-group-metal catalyst loading and improve ionomer utilization

Project Goal and Brief Summary

This project will fabricate, characterize, and evaluate a membrane electrode assembly (MEA) with a novel catalyst layer incorporating a "nanocapsule" electrode structure that separates ionomer and platinum to maximize activity while allowing ionic transport. If successful, this project will allow for better use of highly active and/or highly durable catalysts and the bridging of the activity gap between rotating disk electrodes and MEAs.

Project Scoring



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

Question 1: Approach to performing the work

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

- This project is focused on all key improvements sought for heavy-duty (HD) vehicle applications, namely, improved durability, efficiency (performance), and cost. This project is trying some promising concepts, including a highly innovative catalyst layer design. This is an area where more work is justified, since the polymer electrolyte membrane fuel cell community is still essentially using a 30+-year-old catalyst layer architecture that was first developed by Los Alamos National Laboratory circa 1990. However, new catalyst layer architectures are very risky, the failure of 3M's nanostructured thin film (NSTF) being a good example.
- The project approach of developing and optimizing nanocapsule-based electrodes appears to have many significant challenges. While conceptually the approach of minimizing ionomer contact with Pt to minimize activation losses is appropriate, the structure itself may have significant challenges. One concern is associated with having sufficient ionic conductivity within the nanocapsule with reduced or no ionomer within the nanocapsule; maintaining sufficient ionic conductivity at low relative humidity (RH) is a challenge with more traditional electrode structures, and it would appear that the nanocapsule approach may have even more challenges. A second concern is associated with oxygen and water mass transport through the ionomer film around the nanocapsule. A third concern is flooding within the pore structure within the nanocapsule, especially with the ion beam assisted deposition (IBAD)-type catalysts (the pore walls within the nanopores will be lined with Pt, which is very hydrophilic). A final concern is the stability of the nanocapsules in the event of carbon corrosion. It appears (from the reviewer-only section) that the team has considered many of these concerns with modeling, but having more experimental feasibility data would be very useful.
- The concept is a novel approach that is feasible, but it is far from certain that it will work. A number of parameters must be optimized to result in the overall improvement expected. Even if ideal structures can be created, the modeling does not appear sufficient to prove the approach will work. Creating the ideal structures will also be difficult. There are a large number of variables to work with and so many levers to push, but there may be too many. Some early success on achieving an indication that the approach is working will be required before descending into numerous parameter studies. The reviewer-only slides provide a bit more explanation of the model data and parameter sensitivities. Some good physical characterization techniques are shown, but revealing the ionomer in the structure may be difficult. It is unclear how long the HD drive cycle test will be operated. RH sensitivities may be important but were not discussed (the design may suffer from flooding in ionomer-rich regions and drying in ionomer-poor regions).
- It does not seem that the key milestones (slide 7) lead to the go/no-go decisions (slide 8).

Question 2: Accomplishments and progress

This project was rated **2.9** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project has made satisfactory progress, considering the early phase of the project and challenges associated with the pandemic. Initial modeling results appear to show that the nanocapsule approach could provide a large benefit at high humidity conditions but needs to be validated experimentally.
- It is very early in the project in terms of money spent to see much progress.
- It is early in this project, but the reported results are sparse for six months of work. It seems like this group may have missed the first milestones.
- Not much has actually been done yet. This is understandable, though.

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 team is good and has the appropriate skills and capabilities required. There is not much evidence of real collaboration yet, but that is probably because not much has been completed yet. Fabricating and testing MEAs with these new materials will require collaboration.
- An excellent team has been gathered.
- Collaboration appears to be good based on initial results in the early phases of the project.
- Research organizations and project workflow are appropriate.

Question 4: Relevance/potential impact

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

- The project is well-aligned with addressing the key barriers of performance and activity, but there does not appear to be an overt effort toward improving durability other than the stated intention of using catalysts with good durability (e.g., ordered PtCo).
- The project is well-aligned with DOE's objectives and has much promise. Whether it will actually be successful, though, is still to be determined.
- If the approach works, the project may have high impact. There is concern about eventual manufacturability of the designs, but nevertheless, significant learning might be accomplished.
- This work is likely to encounter the same barriers as previous nanostructured catalyst concepts. Water management, catalyst utilization, and sheet resistance are critical issues. It is unclear how these are being addressed differently from issues with 3M's NSTF catalyst layers.

Question 5: Proposed future work

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

- The questions posed on slide 13 seem like an appropriate roadmap for this work. In addition, MEA fabrication studies are going to be important. Catalyst layer thickness, coating method, ionomer distribution, porosity, and other parameters will need to be studied to meet performance and durability project goals.
- The proposed future work of experimental development and testing physical electrodes using the nanocapsule approach is appropriate.
- The plan is okay but presents a significant number of questions to answer. It is not clear if all can be addressed.
- The proposed future plans seem fine, but the details are sparse. The timeline is vague, except for the annual milestones listed on slide 8. These milestones are not very challenging, although these modest performance goals are probably appropriate for a brand new architecture.

Project strengths:

- The project is focused on the development of an innovative catalyst layer architecture. This is the kind of high-risk, high-reward work that DOE should invest in more frequently. The concept is good, and it builds on one of the truly valuable things that was demonstrated by NSTF, namely, that one does not need any ionomer for proton transport over very short distances. The team has the skills required to execute the plan. The new principal investigator (PI) has good communication skills.
- The project aims to use a unique approach for developing a new engineered electrode structure with the potential to significantly improve activity by reducing sulfonate poisoning. The team comprises a set of highly capable researchers, with an appropriate balance of synthesis, modeling, and characterization.

- The project has a strong team, excellent candidate materials (catalyst), and a novel approach, with preliminary indications of possible significant improvements.
- It is too early to tell, but the subcontractors have a strong record of success on other projects.

Project weaknesses:

- This is a high-risk project, and DOE therefore needs to track it carefully to determine whether continued work is justified. However, it is certainly worthwhile to devote three years to it. It is not clear why the project PI is changing. The new PI has minimal project leadership experience.
- The project seems off-balance. Much of the characterization for modeling and fundamental understanding is fantastic, but to meet the go/no-go decision points, the project should consider more process optimization and development studies. It was not clear in the question-and-answer session if the prime organization had a strong background in prior NSFT work. Understanding the successes and challenges documented by these researchers will certainly benefit this project.
- The project does not appear to have any experimental validation that the nanocapsule approach will increase activity, a core tenet of the approach.
- There are too many parameters to consider, and the level of modeling is not clear.

Recommendations for additions/deletions to project scope:

- DOE should continue to fund projects that explore innovative catalyst layers, such as this one and Vanderbilt University’s fiber-based layers. As long as these projects continue to make good technical progress, they should continue to be funded. However, DOE also needs to be diligent about looking for possible fatal flaws in these new architectures to avoid repeating the NSTF experience. In that case, the new architecture was not operationally robust (i.e., it could perform well under only a very narrow range of conditions). The innovative catalyst layer being proposed here has some similarities to NSTF, namely, minimal or no ionomer in some parts. Therefore, the project should be asked to demonstrate operational robustness near the end of the project (i.e., measure performance curves under various RH, temperature, and pressure values and compare to conventional MEAs).
- It is recommended that additional criteria around the first budget period go/no-go decision point be considered beyond the current hydrogen–air kinetic performance at 0.8 V. Some aspect of hydrogen–air-rated power performance and durability should be included in the event that some of the concerns around the nanocapsule approach are larger than anticipated.
- The project should understand feasibility of manufacturing. A 50 cm^2 MEA may be sufficient to prove this concept. The value of a full-size MEA test is not clear.
- This project should consider more process optimization and development studies related to MEA integration.

Project #FC-327: Durable High-Power-Density Fuel Cell Cathodes for Heavy-Duty Vehicles

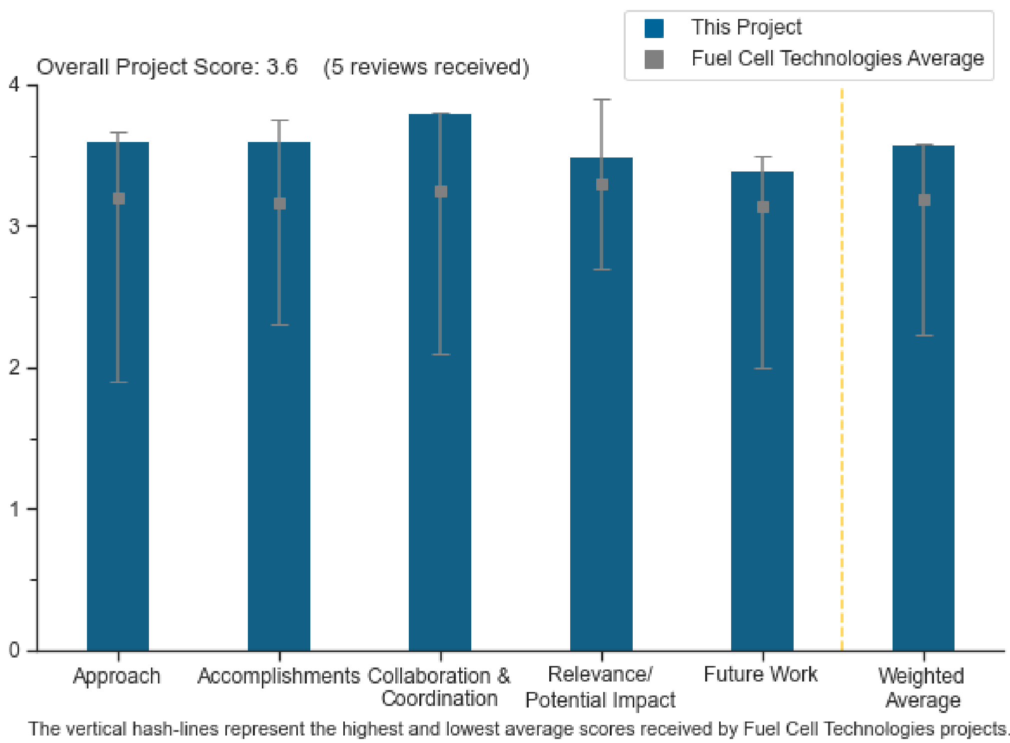
Shawn Litster, Carnegie Mellon University

DOE Contract #	DE-EE0008822
Start and End Dates	10/1/2019 to 1/31/2023
Partners/Collaborators	The Chemours Company, Ballard Power Systems, Inc., Million Mile Fuel Cell Truck (M2FCT) Consortium
Barriers Addressed	• Cost • Performance • Durability

Project Goal and Brief Summary

This project aims to (1) synthesize and implement a custom-designed ionomer that permits enhanced oxygen transport to the platinum surface for improved performance and durability, (2) demonstrate that the ionomer will reduce oxygen transport resistance in a membrane electrode assembly (MEA), and (3) optimize the design of the ionomer for commercialization. If successful, the project will facilitate low platinum loadings in an advanced MEA cathode catalyst layer for heavy-duty vehicles.

Project Scoring



Question 1: Approach to performing the work

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

- Incorporating a high-oxygen-solubility and -transport polymer instead of the traditional ionomers in the cathode catalyst layer is a new novel approach that could significantly improve the performance of the catalyst layers.
- Addressing the long-standing challenge of oxygen transport to the platinum surface has the potential to improve performance and durability for all polymer electrolyte membrane fuel cells (PEMFCs). Shorter side chain perfluorosulfonic acid (PFSA) chemistry is a good choice to implement this concept.
- The project is well-formulated and focuses on the use of high oxygen permeability ionomers (HOPIs) to improve the long-term performance of heavy-duty (HD) fuel cells. The team lays out the rationale well based on the importance of local transport resistance and the loss of electrochemical surface area (ECSA). The primary unknowns of the approach are how central catalyst degradation will be in HD systems and how chemically robust the HOPIs are in use.
- The goals, as well as the progression of work to reach those goals, are well-defined, with reasonable steps and achievable milestones.
- It is unclear whether HOPIs are needed or helpful at 0.2 $mgPt/cm^2$. A significant impact would be expected at lower loadings (<0.1 $mgPt/cm^2$). Modeling at lower loadings should also be considered. The project should focus on high-surface-area-carbon-type (HSC-type) supports, as these work the best at a low loading.

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 project met all Year 1 go/no-go milestones. The project had outstanding progress with polymer synthesis and modeling, as well as electrode fabrication and fuel cell characterization. Impressive voltage gains in fuel cell tests at high current density were accomplished. The project achieved a large reduction in the MEA performance degradation rate in accelerated stress test (AST) cycles.
- The progress to date has demonstrated significant success. The developed ionomer yields significant performance enhancements, and the team has provided insight into the source of the performance enhancements. The developed ionomer also yields a greater degree of MEA performance retention following an AST protocol.
- The results are quite good at such an early stage of the project. The team is obviously leveraging previous work by The Chemours Company (Chemours) in developing HOPIs. The fuel cell results clearly show the ability of HOPIs to improve both specific activity and mass transport losses, although the specific activity improvements seem to be dependent on the use of non-porous carbons. The modeling also shows good initial results.
- Although the modeling work and the ex situ characterization of the new HOPI polymer confirm that it should improve the performance of the cathode catalyst layer, the proof at the end of the day for the high-roughness-factor MEAs was just not there. Ballard tests show only minute improvements over the D2020 baseline when the roughness factor is >100. This begs the question of whether some other factor is dominating the performance of the catalyst layer, such as the catalyst layer resistance being higher for the HOPI, or the optimal ionomer-to-catalyst ratio being different for the HOPI, as compared to D2020, possibly because the density of D2020 is higher. The excellent team approach and well-guided optimization of the materials and structure should lead to further improvements.
- Good progress has been made for meeting go/no-go milestone 1. Perhaps a loading study could replicate the roughness factor vs. performance trend observed in the degradation studies.

Question 3: Collaboration and coordination

This project was rated **3.8** for its engagement with and coordination of project partners and interaction with other entities.

- This project has a great team. Original equipment manufacturers, suppliers, and a well-established university comprise a recipe for fundamental understanding while constraining cost and manufacturability. The principal investigator (PI) has a strong record of collaborative work with successful outcomes.
- This project has excellent teaming and coordinated work. This approach of modeling, ex situ material characterization, and good fuel cell testing work will properly guide the team to further optimize the formulation so that the full benefits of the HOPI can be realized.
- The team, including M2FCT laboratories, is strong, and all team members seem to have a clear role in the project. Almost all are at the very leading edge of what they are doing within the project, with Ballard having a great fit and Chemours bringing unique specific materials and capabilities to the project. The PI is at the top level of researchers at a similar stage of their careers. The modeling effort seems to have all the appropriate skills, but there is less background strength in this area compared to the rest of this extremely strong team.
- The team and the degree of collaboration are good and complementary to the work.
- This is an outstanding team with outstanding collaboration.

Question 4: Relevance/potential impact

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

- This work demonstrates the significant impact of ionomer chemistry on MEA performance. This work is critical for HD PEMFCs, as it will help to remove some of the performance burden from the catalyst.
- Since HD applications typically use higher Pt loadings, degradation studies are critical. For this application, the implementation of HOPI will be highly dependent on cost. Perhaps the investigators can discuss cost impacts in future disclosures.
- Increasing fuel cell performance and durability are key to lifecycle cost reduction.
- It looks as if more work will result in further improvements of the approach.
- Pursuing HOPIs as a pathway to achieve improved durability is interesting. It is essentially trying to take a catalyst degradation and addressing it through improving transport processes. The threefold improvement in oxygen permeability is meaningful, but it is not clear how well it will be maintained over time or whether there are other degradation phenomena that may be worse in HOPI than in other PFSAs. The HOPI approach also requires that Pt ECSA loss be treated as a significant degradation concern because it cannot be addressed either by novel catalysis approaches or by systems controls. This is a worthwhile project, but it is not enough by itself to fully cover durability concerns.

Question 5: Proposed future work

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

- This project has an outstanding plan to build on the results to date to address the remaining challenges.
- The proposed future work is good.
- The topics for future work all make sense and should lead to advances. The work needs clearer priorities of focus, as the list seems more like a laundry list that is not as focused on key issues as would be preferred. Additional studies investigating the HOPI membrane for bulk properties such as water uptake, permeability, conductivity, and durability to radical attack would be good inclusions.
- Future work should be focused on demonstrating a performance improvement in line with what the modeling predicts. A fundamental study of the catalyst layer structures needs to be done so that the high-roughness-factor catalyst layers show a similar level of performance improvement over D2020, as compared to the sub-optimal low-roughness factors.

- The project has a good plan. The team should consider adding cost and HSC support studies.

Project strengths:

- This is a great team that shows strong progress toward fundamental understanding and development of HOPIs. HOPIs are critical technology for next-generation fuel cells, and the team seems to have a well-developed approach.
- The project has well-defined goals and a logical approach. It is an accomplished and well-integrated team that has demonstrated significant performance enhancements with synthesized HOPIs and provided detailed insight into the source of performance enhancement for the HOPIs.
- This project has an outstanding concept, plan, and execution by a well-equipped team. The fabrication processes are scalable to enable faster rollout of the technology once its development is successfully completed.
- This is a novel approach that should give significant catalyst performance gains. The project has an excellent team, and each member's strength is well-utilized.
- Overall, this is a great team and a solid topic with good initial results.

Project weaknesses:

- The HOPI ionomer chemistry adds additional ether linkages in the passive repeating unit of the polymer. Ether linkages have been identified as a weak point in other PFSA ionomers. As this project is targeted for HD PEMFCs, the effort should be focused on fully characterizing the stability of this ionomer against chemical/electrochemical degradation.
- The project's weakness is the difficulty in performing all the needed catalyst layer optimizations to properly demonstrate the improvement in the catalyst layer performance using a HOPI.
- The HOPI will likely benefit light-duty applications in the near term. Constraining development to HD targets does not seem like a logical first step.
- There is uncertainty regarding the criticality of Pt ECSA loss in HD applications and uncertainty regarding chemical stability issues of HOPIs.

Recommendations for additions/deletions to project scope:

- Modeling indicates a lower degree of sulfonate-specific adsorption. This could be supported by direct measurements of sulfonate coverage through techniques such as Co displacement. AST results indicate no significant ECSA retention over D2020, yet there is a marked decrease in MEA performance loss for the HOPI. This is an interesting result, but it appears that why this is the case is not fully understood. It would be beneficial to perform dedicated diagnostics testing to determine exactly what is going on, as it could inform future ionomer design. Kinetic enhancement is attributed to increased oxygen concentration at the catalyst surface. While the researchers provide a simple correlation between oxygen solubility and diffusivity in the HOPI, they could provide direct evidence by looking at the MEA performance as a function of oxygen partial pressure in the cathode. This would prove that it is the high oxygen solubility/concentration in the ionomer that is the source of the performance enhancement. This is important, as it will guide further development of this ionomer, as well as others.
- The inclusion of membrane studies on free-standing films of HOPIs would provide valuable insight into some of the key concerns that may arise from their use in HD fuel cells.
- The project should estimate the cost increase relative to D2020, as well as consider HSC support studies and a loading study to compare with the roughness factor vs. performance trend observed in the degradation studies.

Project #FC-328: Fiscal Year 2019 Small Business Innovation Research Phase II: Novel Fluorinated Ionomer for Polymer Electrolyte Membrane Fuel Cells

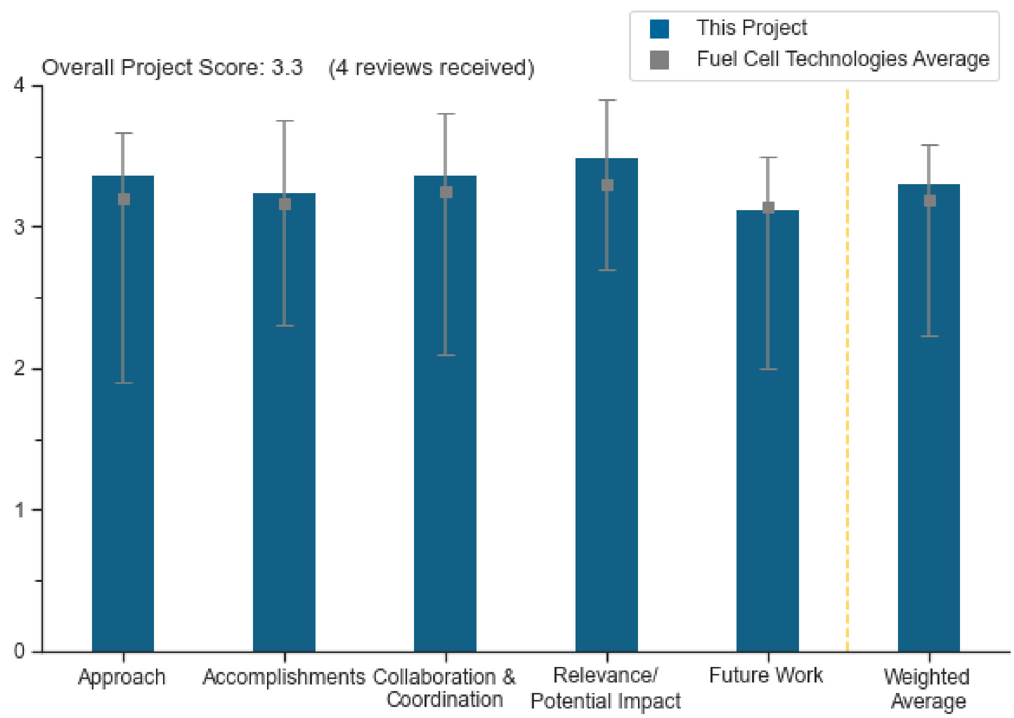
Hui Xu, Giner, Inc.

DOE Contract #	DE-SC0018597
Start and End Dates	5/28/2019 to 5/27/2021
Partners/Collaborators	Compact Membrane Systems, University of Connecticut, University of California
Barriers Addressed	• Polymer electrolyte membrane fuel cell transport loss at low Pt loadings and high power densities

Project Goal and Brief Summary

This project aims to develop a high-oxygen-permeability ionomer that will reduce local oxygen transport loss in polymer electrolyte membrane (PEM) fuel cell cathodes by engineering the polymer backbone to contain molecules with more open space available for transport. If successful, the project will introduce alternative ionomer materials that enable higher power densities compared to state-of-the-art ionomers.

Project Scoring



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

Question 1: Approach to performing the work

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

- This team is investigating perfluorosulfonic acid (PFSA) materials that have substituted tetrafluoroethylene (TFE) with perfluoro-2,2-dimethyl-1,3-dioxole (PDD) for improving oxygen permeability in the cathode layer. The PDD reduces the crystallinity in the material, leading to enhanced oxygen permeability. This is a good strategy for reducing mass transfer resistances and for extending the current density at low cell voltages. The incorporation of non-crystalline fluoroether over TFE into PFSA was demonstrated by Modestino, Weber, and Kusoglu in the *Journal of the American Chemical Society* (2020); however, the fuel cell results and ionomers are much better in this presentation.

- The team has demonstrated the ability to introduce the PDD monomer into the copolymer at different compositions. The PDD is attributed to the increasing free volume and correlates well with the presented oxygen permeability data. The microscopy and other analyses are good tools to help develop the material and processing understanding.
- The copolymerization of the PDD monomer to increase oxygen permeability in ionomers is an interesting idea, as it could improve both PEM fuel cell and electrolysis performance.
- The work is very relevant to the U.S. Department of Energy's focus on improving performance and durability of PEM fuel cell membrane electrode assemblies (MEAs). However, with the recent focus on heavy-duty (HD) applications and the long life requirements for this application, any new material that is being developed should be compared with the best-in-class, state-of-the-art ionomers, rather than just Nafion™-based material. It would be good to see performance and durability comparisons with other PFSA-based materials.

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 team demonstrated several different ionomer variants of PDD PFSA in which the perfluoro(sulfonyl vinyl ether) (PFSVE) or perfluoro(sulfonyl ethyl propyl vinyl ether (PSEPVE) and termonomer were varied. The termonomer is confidential information to Giner, Inc. The oxygen gas permeability improved by a factor of 10 for one variant versus Nafion, but this version did not conduct protons well. The best ionomer variant (PDD4) satisfied proton conductivity requirements at 50%, 70%, and 90% relative humidity (RH) (20 to 90 mS/cm), while showing a threefold to fourfold improvement in oxygen gas permeability over Nafion. PDD4 gave a 150 mV gain at 2.5 A/cm^2 when benchmarked against Nafion. Finally, when electrochemically cycled for HD applications (94°C, 65% RH, 250 kPa), PDD4 also had less fluoride emission rate degradation than Nafion. Overall, the project team has made excellent progress toward the project goals. The researchers have also repeated the synthesis of PDD4 (labeled PDD9).
- The project appears to meet several DOE 2020 targets. The presenter states that other targets may be achieved using W.L. Gore PEMs, but no specifics were provided to support the projection.
- Good catalyst accelerated stress test (AST) durability is shown. However, the ionomer's durability should also be based on other HD operating conditions, such as higher temperature, low-humidity conditions, and durability tests that are similar to membrane AST protocol to assess the open circuit durability tolerance of the ionomer.
- This idea is interesting, but unfortunately, the performance is not great. For example, in slide 7, the authors boast three times or ten times higher oxygen permeation at room temperature, but when the temperature is raised to 60°C, the oxygen permeation drops significantly. The high fractional-free volume of the PDD seems to collapse at a slightly higher temperature. It would be interesting to know what the permeation of oxygen was during fuel cell testing at 80°C. Also, the incorporation of PDD has its tradeoff effect in lower conductivity. PDD7, which had the ten-times-better oxygen permeation, had the worst conductivity. Fuel cell performance was also not very exciting; while there was some improvement over Nafion at high current densities, at the low current densities, there was no improvement. It would have been interesting if the higher concentration of oxygen had led to lower overpotential losses at high voltage, but that was not the case. The authors show some durability improvement over Nafion, but that difference is fairly small—not enough to suggest that this material is better than the state of the art.

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 project includes collaborations with Compact Membrane Systems (CMS), the University of California, Irvine, General Motors (GM), and the University of Connecticut. CMS has contributed with the assistance of PFSA synthesis using PDD. The other collaborators determined the local oxygen mass transfer resistance and ionomer surface coverage for the various new PDD ionomers, in addition to looking at changes in the cathode structure post-mortem cycling (e.g., via transmission electron microscopy [TEM] and nano computed tomography [nano-CT]). These measurements can help optimize the next version of the PDD variant for fuel cell applications.
- The team has successfully collaborated to prepare and evaluate these new ionomers for the cathode with improved oxygen permeation and coauthored a journal publication that has been submitted. Future plans include collaborations with the Fuel Cell Consortium for Performance and Durability (FC-PAD) and original equipment manufacturers (OEMs) toward material validation, which may (ideally) have begun now.
- GM has provided flowfields to test oxygen transport. It would be good to see some MEA testing by industry leaders such as W.L. Gore and The Chemours Company as well.
- The collaborations are sufficient.

Question 4: Relevance/potential impact

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

- The project team has hit most of the 2020 Hydrogen and Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan (MYRD&D Plan) catalyst and HD MEA targets with the new materials. This includes activity and MEA performance requirements at high cell voltages. The durability at high cell voltages was retained. Performance and performance durability at lower cell voltages, where reaction kinetics is more controlling, was not satisfied, but the attained results were close to hitting the targets.
- Considering the relevance and potential impact of the project concept, an investigation to scale up the ionomer polymerization is critical to the success of this approach.
- This is an important area for fuel cell performance. It would have been good to provide the oxygen permeability values from slide 7 at 80°C, as fuel cell tests were performed at 80°C.
- The idea was sound, but unfortunately, the resultant materials did not have high enough oxygen permeation (especially at higher temperatures of 60°C). Moreover, the incorporation of PDD had a large effect on lowering the proton conductivity. The results of both of these issues led to subpar fuel cell performance at fully humidified conditions. It seems likely that at lower RH and higher temperatures, the performance of these materials falls behind the current state of the art.

Question 5: Proposed future work

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

- Future activities focus on material scale-up and providing samples to fuel manufacturer partners. Other activities include adopting a better membrane with free radical scavengers, in addition to testing ionomer durability with a more rigorous stress test (e.g., hydrogen peroxide vapor cell testing). The team is putting together a *Nature Energy* paper based upon the results.
- The presenter stated that there are plans to scale the synthesis of ionomer batches from the current 200 ml to kilogram scale. There were comments related to slight variations between batches (actual percentage of PDD in the product), so it will be important to determine acceptable compositional tolerances and maintain desired performance. No cost analysis projections for the ionomers were noted.
- This project has a good approach to test the MEAs at other locations, including FC-PAD. However, the testing at 50–100 cm^2 is not sufficient. A larger testing platform (200–300 cm^2) should be planned. Most

performance and durability losses due to current distribution and mass transfer losses are more apparent at that scale.

- This project looks to be spent out, and although future work is planned, there is no budget to do the work. The project appears to be done, with a closeout date of May 27, 2021.

Project strengths:

- The new PFSA ionomer binders for the cathode featuring PDD have improved oxygen permeability, while having adequate proton conductivity. Unlike the 2020 report by Modestino and co-workers, this work optimized the PDD binders to ensure cathode kinetics, while also promoting gas mass transfer. The team satisfied several quarterly project goals and has achieved most of the 2020 requirements for the MYRD&D Plan. In addition to the excellent project progress, the team has engaged in good science to understand how these ionomers interact with electrocatalysts and how they degrade differently from Nafion.
- The project objective and approach are clear and appear fairly controlled. The collective team has the skills and resources to achieve the project goals. While these ionomer materials will likely be limited to PFSAs, this approach may be applicable to ionomers for alternative membrane systems.
- The project was based on an interesting concept and, in principle, should result in ionomers with better properties than the current state of the art.
- This project had a good approach to solve for oxygen transport issues in the ionomer and testing the catalyst AST. However, an AST similar to the open circuit test is critical for assessing the ionomer durability under start-up/shutdown conditions. This is good collaborative work to synthesize novel composite ionomers.

Project weaknesses:

- There are no major project weaknesses noted for this project. The presenter stated that some automation within the synthesis may improve composition reproducibility. In general, achieving tight composition control in free radical reactions is challenging.
- Subscale-size MEAs do not shed light on the overall mass transfer and current distribution issues in real-world HD applications. A larger MEA (200 cm^2) should be tested under similar test conditions. Ionomer thickness in the catalyst layer, and its effects on overall performance and durability, is necessary to arrive at an optimal catalyst layer ionomer content, as well as to understand cost and durability tradeoffs.
- Unfortunately, the actual materials did not meet expectations and did not meet DOE Hydrogen and Fuel Cell Technologies Office milestones.
- No major weaknesses were identified with this project.

Recommendations for additions/deletions to project scope:

- This project has ended, but any future funding to this technical approach to improve oxygen transport using PDD-based ionomers should include a scalability study, both for manufacturing ionomers and for testing larger-platform MEAs.
- The collaborations with FC-PAD and OEMs will be important, but delays due to COVID-19 are understood to be a hindrance.
- The project seems to have wrapped up, so there are no recommendations.

Project #FC-330: High-Efficiency Reversible Solid Oxide System

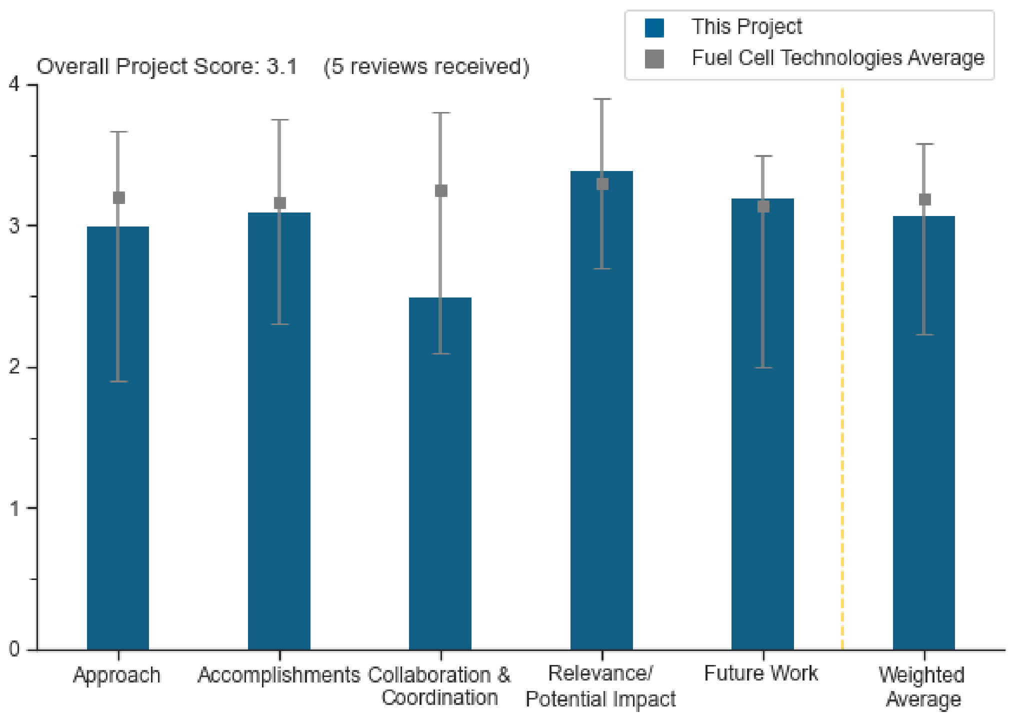
Hossein Ghezel-Ayagh, FuelCell Energy, Inc.

DOE Contract #	DE-EE0008847
Start and End Dates	10/1/2019 to 5/31/2022
Partners/Collaborators	Versa Power Systems
Barriers Addressed	• Capital cost • System efficiency and electricity cost • Renewable electricity generation Integration

Project Goal and Brief Summary

FuelCell Energy, Inc. (FuelCell Energy) will demonstrate a unitized reversible solid oxide fuel cell (RSOFC)-based system, rated at 3 kWe fuel cell power output and 15 kWe electrolyzer power input. The RSOFC system will integrate a novel hot water thermal energy storage system to demonstrate up to 60% system round-trip efficiency (RTE) in testing, with a path to ≥70% RTE. Technoeconomic analysis will validate the projected system costs, which are expected to be at $1,000/kW and $100/kWh. The team will complete a system design and define the required operating conditions for the unitized RSOFC stack, including preferred pressurized operation to achieve the targeted RTE performance. Stack testing will validate the technical approach and operating conditions and will demonstrate less than 5%/1,000 cycles RTE degradation over 100 cycles between fuel cell and electrolysis operating modes.

Project Scoring



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

Question 1: Approach to performing the work

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

- The project objectives and critical barriers have been clearly identified, and tasks are well-designed, feasible, and integrated with other relevant efforts.
- FuelCell Energy is a U.S. leader in solid oxide electrolyzer cells (SOECs) and solid oxide fuel cells (SOFCs). The approach to performing the work is consistent, logical, and aimed at accomplishing the U.S. Department of Energy (DOE) Hydrogen Program goals and milestones. However, FuelCell Energy needs to report the thermal situation of the SOEC, that is, whether it is thermal-neutral. More details need to be given on the thermal storage device and its economic and operational significance.
- The goal of this project is to develop a system for grid-scale storage. The principal investigator shows a depiction of a 1 MW system, which would be the appropriate scale, but the plan for a full-size stack is approximately 10 kW. This means there would be approximately 100 stacks in a 1 MW system (e.g., the process diagram shown in slide 14 with 100 stack modules), which is not viable. Plumbing is required, and half of this plumbing is transporting hydrogen. If DOE sees a need for a small-scale system (e.g., for residential applications), then this approach would be acceptable. However, this technology is not viable for megawatt scale unless the stacks are on the order of 100 kW, which does not seem to be possible for this developer.
- The approach was not clearly described. The objective stated, "Develop storage system design and identify operating conditions that maximize the potential of the RSOFC stack and materials technology in meeting RTE performance and degradation goals; Demonstrate 0.5%/100 cycles RTE degradation," but how exactly RTE could be improved or determined was not explained. It was unclear whether the measured RTE was >70%, how the operating conditions (partial pressure of water [pH_2O], how the voltage–current density curve [V-I], and utilization) would be changed to improve RTE, whether only a pressure effect on RTE would be tested, and what would happen if the pressure does not have a significant effect on RTE.
- It is not clear how the team plans to verify that the energy cost of <$100/kWh is achievable. It would be helpful to clarify how the verification is done—whether only through technoeconomic analysis (TEA), perhaps based on the operating strategy already demonstrated, or through a targeted experiment to verify or prove the technical feasibility.

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 made significant progress, as measured against well-defined performance indicators, and attained accomplishments above the project milestones toward addressing critical barriers to achieving DOE goals.
- An RTE of 75%–80% was achieved; however, the conditions were not mentioned. The stack operating temperature and the gas composition for SOFCs and SOECs should be included. Slide 9 gives 100% H_2 for SOFCs and 22% H_2 for SOECs, but it is unlikely that the gas was completely changed on every cycle. The chosen current densities were low at 0.2–0.35 A/cm^2. These results are quite a bit lower than typically reported by FuelCell Energy; no reason was given. It was also unclear what was causing the main losses on cyclability.
- The results are impressive for this point in the project, with <10% of funding spent; however, the durability results are not adequate. A major issue with solid oxide cells is thermal cycles, and it does not appear that any complete start-and-stop cycles (which will occur in the real world) have been done. Even if the plan is to keep the system hot at all times, it will still need to be put into a standby mode (unless the cells will be kept filled with hydrogen), and the system will be shut down for maintenance. Durability results need to include these stressors, not just continuous cycles.
- There has been interesting and important progress in RTE. The major concern with the project is the acute underspending.

- The project needs to clarify whether there are delays for some of the milestones (milestones 3 to 6 and milestone 9), as appears to be the case on slide 6.

Question 3: Collaboration and coordination

This project was rated **2.5** for its engagement with and coordination of project partners and interaction with other entities.

- A list of potential industry committee members has been identified, and an advisory board is being formed. It is expected that the advisory board will provide valuable guidance for product definition and specifications and oversee the development of the proposed high-efficiency RSOFC system.
- An advisory board is being formed to provide guidance for product definition and specifications and to oversee the development of the energy storage RSOFC systems. Otherwise, there is no apparent technical collaboration.
- Having an advisory board is a good way to identify a demonstration site and receive user feedback; however, having eight very different organizations might be too complicated, as it would be difficult to coordinate, actually collaborate, and discuss the issues. It is suggested that the group be reduced to around three members.
- It is not clear whether the industry committee members (slide 16) have already been engaged and, if so, how. Also, it is not clear how the committee will help with defining the system operating conditions, given the specificity of the system design. Finally, it is not clear what the involvement of Versa Power Systems (VPS) is within the project.
- Only one official collaborator is mentioned—VPS—and VPS's role in this project is never made clear. The researchers claim they are going to form an advisory board, but it is not clear why this has not already been done if the project started in 2019 (as stated on slide 2). The team could definitely use some guidance on storage requirements on the grid.

Question 4: Relevance/potential impact

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

- The project will significantly contribute to the Fuel Cell Technologies subprogram by addressing the barriers of cost and performance and demonstrating RSOFC technology.
- Projects focused on the development of RSOFC systems achieving RTE, cost, efficiency, thermal integration, and operability are key projects for the Fuel Cell Technologies subprogram of the Hydrogen and Fuel Cell Technologies Office.
- The project is well-aligned with the general Hydrogen Program goals.
- There is a question of how easy it would be to translate the data obtained from a 3 kW demo to a 250 kW unit, which would be 80 times larger, and whether there would be additional losses and penalties expected in that scale-up.
- It is unlikely that this project will have any real-world impact, even if it is successful from a technology perspective as it is currently set up. Nonetheless, this project could contribute to the advancement of RSOFCs, which might be useful if the team can find an application that wants a relatively small-scale storage system with ultra-long duration (e.g., >12 hours, which is the point at which the capital expenditures on a dollar-per-kilowatt-hour-stored basis might be somewhat reasonable).

Question 5: Proposed future work

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

- The project has effectively identified the remaining challenges and planned its future in a logical manner by incorporating appropriate decision points and considering barriers to its goals.
- The proposed work is appropriate. Much work remains to come up with an optimized storage system, such as a completing an RSOFC system analysis, completing a process flow diagram, developing steady-state

CHEMCAD models for electrolysis and fuel cell modes, developing a dynamic model for a thermal storage system, running simulations to optimize system configuration and equipment sizes, performing TEA, conducting an RSOFC product configuration, developing stack and system costs at high-volume production, and considering operating and maintenance costs.

- It appears that future tasks are mainly system modeling and TEA. No details on the validation were given. The stack test task is very vague, and no details were provided on how the degradation will be reduced. It was unclear if it is all in operating conditions for parameters such as pressure.
- The Remaining Challenges and Barriers on slide 17 appear to indicate that the current project approach is not viable because improvements in both cyclability and durability are needed (i.e., the successfully completed GN1 milestone is an inadequate assessment of what is actually needed); the system-level RTE is still a challenge, even though the cell RTE is good (this will always be the case in a high-temperature system); the TEA has not been done; and the results could be either poor (and realistic) or decent (with bad assumptions), since it is quite obvious that a 1 MW size system will simply be prohibitively complex.

Project strengths:

- The project has made significant progress successfully building and testing a 50-cell SOEC stack operating up to 410 charge–discharge cycles for >2000 hours, exceeding both RTE and degradation targets.
- This is an important effort to demonstrate a potentially high RTE of SOFCs and SOECs. The calculated data are less optimistic at <45%. Good experimental data are needed.
- This is a well-qualified team. The RTE work is excellent.
- This project is focused on challenging technology that requires substantial improvements.

Project weaknesses:

- The tasks description is very vague throughout. The project needs a clear path on how the losses on cycling and degradation will be addressed and what causes them.
- It is unclear how the project will contribute to identifying limitations related to the materials and cell design used for RSOFCs.
- This project's weakness is underspending. It has a well-qualified team that is drastically underspent.
- The overall system and the viability to meet the DOE objectives is not well-thought-out.
- The team is weak in its collaboration at this juncture.

Recommendations for additions/deletions to project scope:

- The project has proceeded as planned, and there is no need to change the project scope.
- The project should identify possible reasons for the degradation and how it could be reduced or eliminated.
- The team should review the schedule, timeline, and budget for the project and determine whether the work can be completed in a timely manner.
- DOE should consider stopping this project, unless the Department is interested in ultra-long-duration storage for small-scale applications. This project is highly unlikely to address grid-scale needs or the ramping up of intermittent renewables in a significant manner.

Project #FC-331: A Novel Stack Approach to Enable High Round-Trip Efficiencies in Unitized Polymer Electrolyte Membrane Regenerative Fuel Cells

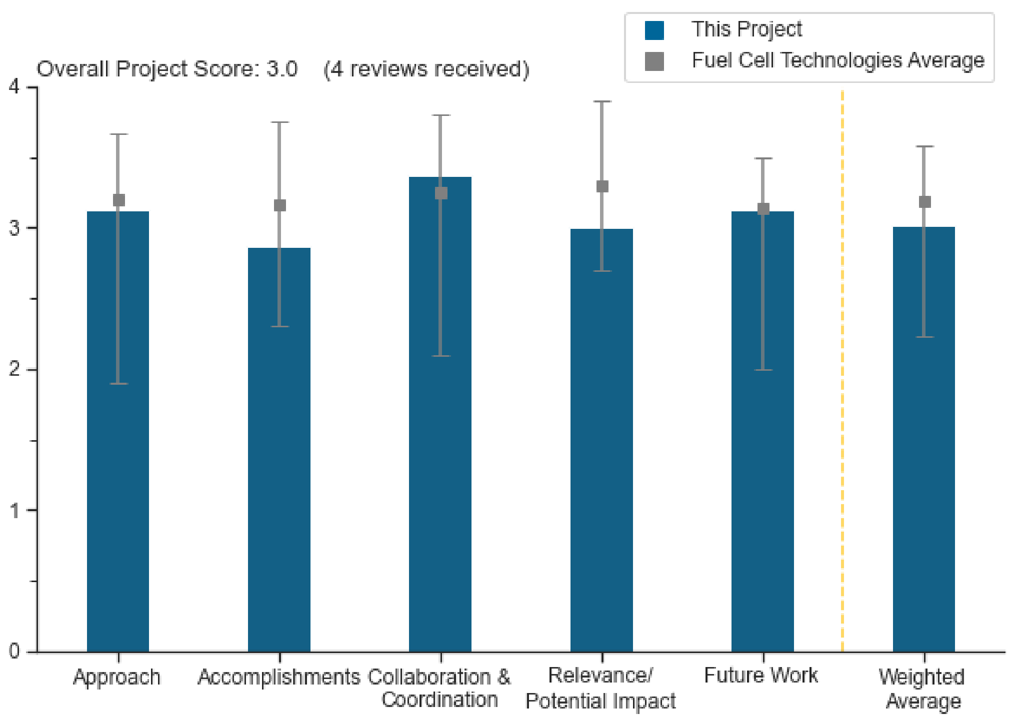
Katherine Ayers, Nel Hydrogen

DOE Contract #	DE-EE0008848
Start and End Dates	5/1/2020 to 12/31/2023
Partners/Collaborators	Electric Power Research Institute, Southern Company, Lawrence Berkeley National Laboratory, Gaia Energy Research Institute
Barriers Addressed	• No regenerative fuel-cell-specific barriers • Optimization between fuel cell and electrolyzer barriers • Fuel cells (durability, cost, performance) • Hydrogen production (capital cost, system efficiency, and electricity cost)

Project Goal and Brief Summary

The overall project goal is to demonstrate a unitized reversible fuel cell (URFC) system based on polymer electrolyte membrane technology that can achieve 50% round trip efficiency (RTE) and reliable performance under relevant duty cycles, with projected costs below $1750/kW. An early focus of this project is to develop a low pressure electrolyzer membrane electrode assembly and stack design which much more closely resembles the fuel cell construction (e.g., thinner membrane), providing a pathway to higher RTEs for URFCs.

Project Scoring



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

Question 1: Approach to performing the work

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

- The project has a well-balanced and well-thought-out approach. Having a trade-off study to vary the extent of stack pressurization vs. mechanical pressurization is a good idea. The idea of a series of tutorial presentations to the power industry is appropriate for improved outreach and has some merit. The key approach to improved systems appears to be a thinner membrane (balanced with an optimized operating pressure) and proper water management.
- This is an experienced team with a significant amount of technology background, integrated with potential end users and those experienced in cost analysis. The approach appears to be sound.
- It is hard for a project to do a great job in addressing U.S. Department of Energy (DOE) objectives, if DOE has not established clear targets. If there are no regenerative fuel-cell-specific barriers, as stated on slide 3, then the principal investigator (PI) should provide some and show how this approach addresses them. There certainly are challenges, which can be more specific than the simple qualitative ones listed on slide 3.
- It is difficult to identify the innovation in this project. The project focuses on stack optimization, system integration, system testing, modeling, and outreach. There is not much focus on scientific development or materials/components development. It seems to be more of a demonstration project than a research and development (R&D) project.

Question 2: Accomplishments and progress

This project was rated **2.9** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Not much work has been done yet because of COVID-19 delays, so most of the work is still future work; however, significant work has been done on down-selection of catalysts and membranes and on modeling and model validation.
- The project is still in the early phases and was delayed by COVID-19 and so simply is not at a point where the team has many technical results to report. What the researchers have so far is good and as expected.
- The team has done a good job at demonstrating electrolyzer performance during the first five quarters, but there is little evidence that much has been done on demonstrating fuel cell performance.
- Little progress is shown on the stack design, although it is listed as a primary accomplishment. The cell optimization graphs show good progress and substantial polarization improvements with thinner membranes and high platinum loadings. The meanings of the open symbols on slide 11 are not defined. Analysis of four different membranes is very good. More description/assessment should be paid to durability. The slight changes in voltage during the short-term-durability testing are not clearly significant, yet durability should be a main factor in cell design selection.

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.

- Collaboration with the Electric Power Research Institute (EPRI), Southern Company, and Lawrence Berkeley National Laboratory (LBNL) grants significant benefits to the project. Gaia Energy Research Institute is qualified to conduct the pressure trade-off study and is very well-suited to conducting the outreach presentations.
- The team is well-composed, and the outreach activities are valuable.
- The project includes some relevant and capable team members, namely LBNL and Gaia Energy Research Institute; however, it is not clear how much these two members have contributed lately. The work with EPRI to educate electric utilities is commendable, as teaching utilities is a major challenge for many reasons.
- Nel Hydrogen's established collaboration with LBNL is a big part of this project. Given that there is a separate project led by LBNL with Nel Hydrogen as a team member, some discussion about how the work

scope is divided between the projects should be included. Two partners are included whose roles seem to relate mainly to outreach and promotion of URFCs. Those partners' contribution to R&D in the project is not clear.

Question 4: Relevance/potential impact

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

- A low-temperature URFC is a viable pathway to using hydrogen for grid-scale energy storage, especially ultra-long duration. Nel Hydrogen has demonstrated the ability to build megawatt-scale electrolyzers, and the approach is credible.
- This project ties into the overall goals of the Hydrogen and Fuel Cells Technologies Office around long-duration storage using hydrogen. It a serious question whether it makes sense to make a URFC, as this team is trying to do, versus two separate systems. The ultimate relevance of this approach should be clearer by the end of the project.
- The relevance and potential impact are significant if the project can meet or beat the targets. There are concerns that the cost goal of $1750/kW is too high for economical implementation.
- The project focuses on screening materials and optimizing stack performance. The project is unlikely to achieve more than incremental improvements in URFC technology. There is not much new science or R&D that could have a substantial impact. The project is about a novel stack approach, but the novelty is not clear.

Question 5: Proposed future work

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

- The future work focuses on analysis, screening, demonstration, and outreach. This work fits within the modest goals of the project but does not seem likely to result in innovation.
- The future work is as expected and is in accordance with the project plan.
- The plan looks good, except there should be more durability testing during this project. It is understood that the primary focus is meeting performance targets, but 100-hour or 200-hour durability tests are anemic. If longer tests are seen as impractical here, then the team should do some accelerated stress tests (ASTs). LBNL has already identified some key durability challenges, such as extreme potential cycles that result in catalyst dissolution and losses.
- The slide for future work does not hold much information, but the slide for key milestones furnishes the needed information.

Project strengths:

- Nel Hydrogen and the PI are very well-qualified to conduct this project. There is a high probability of project success. Collaboration with EPRI, Southern Company, and LBNL brings substantial support and capability to the project.
- The project has a good approach to the development of a viable URFC. There is a great team with the right skills and capabilities.
- Nel Hydrogen is a key developer of electrolyzer and URFC technology and brings substantial background in this area.

Project weaknesses:

- The project is focused on incremental progress through optimization of URFC stacks. There is not much innovation in the approach, nor is there a key idea or hypothesis driving the work. The project would fit better in a demonstration program than in an R&D program.
- There is no explanation of how a thinner membrane can be appropriately supported under the delta pressures contemplated. There is insufficient durability testing and discussion of durability aspects. There is

no discussion of the cost target's being adequate for the overall URFC mission. There is no cost breakdown of the stack or discussion of how the URFC stack differs from dedicated fuel cell or electrolyzer stacks.

- There is not enough emphasis on fuel cell performance to date or enough focus on durability issues.

Recommendations for additions/deletions to project scope:

- More engagement with LBNL should be shown; perhaps it is occurring, but it is not clear. For example, the researchers could explain whether they considered using the Constant Gas vs. Constant Electrode configuration and, if so, share which one was selected and why. Additionally, LBNL could create some key ASTs for URFCs and perform some of these. At least one utility that is actually a leader in grid-scale storage should be added to the team. Working with Southern Company, who is actually doing almost nothing in electrical energy storage, is not ideal (it seems Southern Company is not doing anything new but rather is waiting for the ultimate solution). Even if this works, Southern Company will insist that a gigawatt-hour--scale system is needed to start, which is not viable. The team should find at least one more utility partner.
- The project would be improved with more durability testing early on and confirmation that macro goals are achieved if project cost ($1750/kW) and round-trip efficiency (50%) goals are achieved.
- The focus on outreach and the webcasts to electric utilities belong more in an education or technology acceleration project than in a R&D project.

Project #FC-333: Advanced Membranes for Heavy-Duty Fuel Cell Trucks

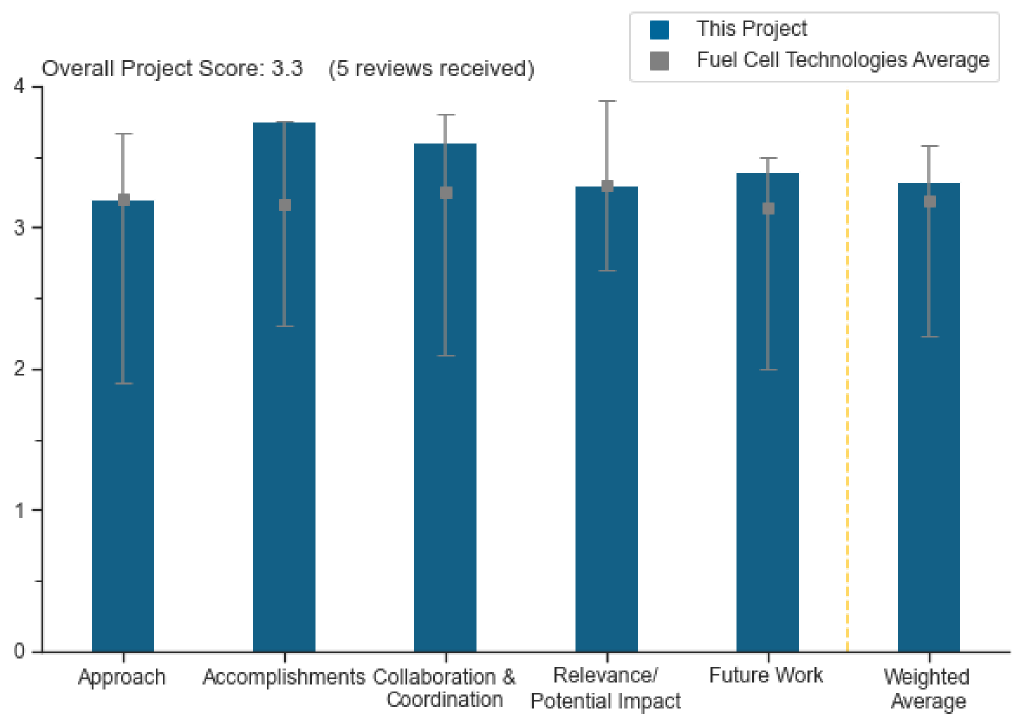
Vivek Murthi, Nikola Motor Company

DOE Contract #	DE-EE0009243
Start and End Dates	Q3 2021 to Q3 2024
Partners/Collaborators	The Chemours Company, Million Mile Fuel Cell Truck (M2FCT) Consortium
Barriers Addressed	• Durability • Performance • Cost

Project Goal and Brief Summary

This project aims to develop membranes with optimized architectures that incorporate thermally stable ionomer chemistries and immobilized radial scavengers. If successful, the project will improve the lifetime efficiencies of membrane electrode assemblies (MEAs) in heavy-duty fuel cell vehicles, reduce the lifetime operational expenses of heavy-duty (HD) fuel cell systems, and improve their commercial viability relative to diesel energy sources. Nikola Motor Company is collaborating with The Chemours (Chemours) Company and the Million Mile Fuel Cell Truck (M2FCT) Consortium on this project.

Project Scoring



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

Note: This is a new project in 2021. 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 objectives and barriers and for project design, feasibility, and integration with other relevant efforts.

- The project has not started; it is anticipated for the third quarter (Q3) of 2021. The project looks at property–function relationships for MEA development for HD truck application. There is new polymer development with various equivalent weights. Radical scavengers in general are an interesting approach to improving fuel cell service life and efficiency. This project looks at radical scavengers with reduced mobility by a factor of two as an alternative approach to cerium systems. The presenter stated that the team could selectively control the area and local concentration of the radical scavengers. It will be interesting to understand more about how this is practically achieved in mass-produced membranes. An organized work plan for the team was presented. It appears to have identified possible challenges and provided initial approaches to mitigate them. Gaining insight into factors that govern dissolution and migration of scavengers, as well as radical scavenger efficacy, would benefit the field. The association of the cerium cation approach was mentioned.
- The project aims to improve the MEA's lifetime and efficiency by incorporating thermally stable ionomer chemistries and immobilized radical scavengers for higher-temperature membranes. This is a feasible approach for realizing this goal, and the approach addresses the challenges and discusses how to overcome them. The project also includes an effort to synthesize new monomers/ionomers.
- The proposed approach to ionomer synthesis, radical scavenger doping, and performance/diagnostic testing is adequate and well-defined. Tailoring analysis to HD drive cycles will do a better job of highlighting early limitations of the developed materials, helping the project to make the needed adjustments.
- Increasing the life and reducing the lifecycle cost of MEAs for fuel cell trucks is critical.
- The approach attempts to modify the polymer chain to increase the stability and also immobilize the ceria radical scavenger. The project identified targets for reduced ceria mobility to one-half of what is the current state of the art. It is not clear that is enough to reach U.S. Department of Energy targets for HD application. Other targets for resistance and hydrogen crossover mechanical and chemical accelerated stress tests (ASTs) are already achieved (aside from a slight improvement targeted for AST) with the state-of-the-art membrane NC-700, so based on the project targets slide, it looks like the project is targeting only a small improvement.

Question 2: Accomplishments and progress

This project was rated **3.8** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- The project is just starting, and no DOE funds have been spent so far. Thus, the progress cannot be evaluated fully.
- The project just started, so it is difficult to make a quantitative analysis of accomplishments.
- The project has not started, so no accomplishments/progress were presented.
- This is not applicable, as the project has not yet started.
- The project is expected to start in Q3 2021.

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.

- There is an excellent team, with an original equipment manufacturer leading the project for measuring lifetime under realistic driving conditions; there is an excellent membrane industrial team member.
- This is a very good team that combines synthesis, characterization, and evaluation expertise. Interaction with M2FCT is planned.

- There are many collaborations, from ionomer suppliers to membrane characterization. A wide range of research activities, from synthesis and cell testing to characterization, will improve the project's likelihood of achieving its objectives.
- A membrane manufacturer is on the team, as well as an academic partner for membrane characterization and AST development.
- The team has been assembled, and the proposed flow of work sets it up for success.

Question 4: Relevance/potential impact

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

- Membrane durability, particularly membrane durability at elevated temperatures, is critical for HD applications. The integration of drive-cycle analysis, organic and inorganic membrane chemistry modifiers for radical scavenging, and detailed analytics position this project to make a significant impact. This work is needed as part of the push to advanced HD polymer electrolyte membrane fuel cells.
- Success in completing this project will be very important to increasing the service life of fuel cells and achieving DOE goals.
- The project goals to improve MEA efficiency and durability through ionomer chemistry and optimization of the radical scavengers and the MEA architecture are in line with DOE Hydrogen Program objectives.
- Developing MEAs that meet the extended lifetimes for fuel cell trucks and expand the operating envelope is on the critical path toward DOE's objectives.
- It is uncertain how much impact this project can make; the team has not yet developed an AST procedure, let alone the target. The go/no-go for Q6 looks like all points already achieved with the NC700, the current state-of-the-art membrane.

Question 5: Proposed future work

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

- Focusing on synthesizing the novel ionomer while in parallel improving membrane reinforcement, ionomer polymerization/dispersion, and refining membrane ASTs is an excellent plan.
- The future work, which is essentially the whole project, is well-defined and has a good chance of accomplishing the proposed goals.
- The proposal includes a plan with tasks involving membrane design and optimization and cell testing with ASTs. Two approaches for immobilization have been proposed, metal-doped ceria and a polymeric route, along with the development of a new ionomer that can help mitigate certain risks associated with the proposed strategies.
- Proposed future work is good; it just needs some work on defining the goals and targets. If the premise is that Ce migration has a strong impact on membrane lifetime (which makes sense), then if the project strongly immobilizes Ce, lifetime (or lifetime AST) should increase dramatically.
- Since the project has not started, the project is considered as future work. The polymer synthesis and structure–property relationship studies are always valuable to the community if the data are shared and published. Only one go/no-go milestone (Q6) was presented.

Project strengths:

- There are well-defined methods to diagnose and characterize the scavenger migration rate and efficacy. They will likely provide useful insights for understanding this phenomenon and for developing immobilization strategies. The team and the collaborations, with well-defined tasks and responsibilities, will likely support the approach and realize the goals.
- There is a good team and good potential to make a fundamental improvement in membrane durability.
- The combined approach of a possible new membrane with Chemours and a possible systematic introduction at the monomer level of tethered radical scavengers is a significant strength.

- The project addresses critical needs for HD fuel cell vehicles in a logical manner with a very good team.
- Risks are mitigated by using both inorganic and organic radical scavengers. Detailed analysis of Ce mobility/transport is included. Detailed drive cycle analysis will help guide other materials development projects. The project has monomer-directed inclusion of radical scavengers.

Project weaknesses:

- No weaknesses were noted in the stated project objectives and approach to addressing the specific goals. The use of perfluorosulfonic-acid-type membranes will continue to limit practical cell operating temperatures. The presenter mentioned the possibility of reducing the radiator size with higher-temperature operations, but it is believed that 80°C–95°C may not be significant enough to warrant that general statement.
- The task lists have too many parameters to choose and optimize from new monomers to membrane parameters and scavenger strategies. It is not clear how they will be prioritized and narrowed down with a clear path forward (work plan).
- Targets and goals should be better defined.

Recommendations for additions/deletions to project scope:

- It is unclear how the team plans to control and optimize local variation with novel monomers or other strategies (slide 15). It is not entirely clear whether the synthesis effort for new monomers is part of the strategy for immobilizing the radicals or whether it will be a separate effort to develop membranes for improved durability. This should be clarified: whether the focus is the optimization of radical scavengers or whether the other membrane properties will also be optimized for HD fuel cell trucks.
- The project should define appropriate targets and ASTs early in the project.
- The project should determine whether scavengers can be regenerated.

Project #FC-334: Extending Perfluorosulfonic Acid Membrane Durability through Enhanced Ionomer Backbone Stability

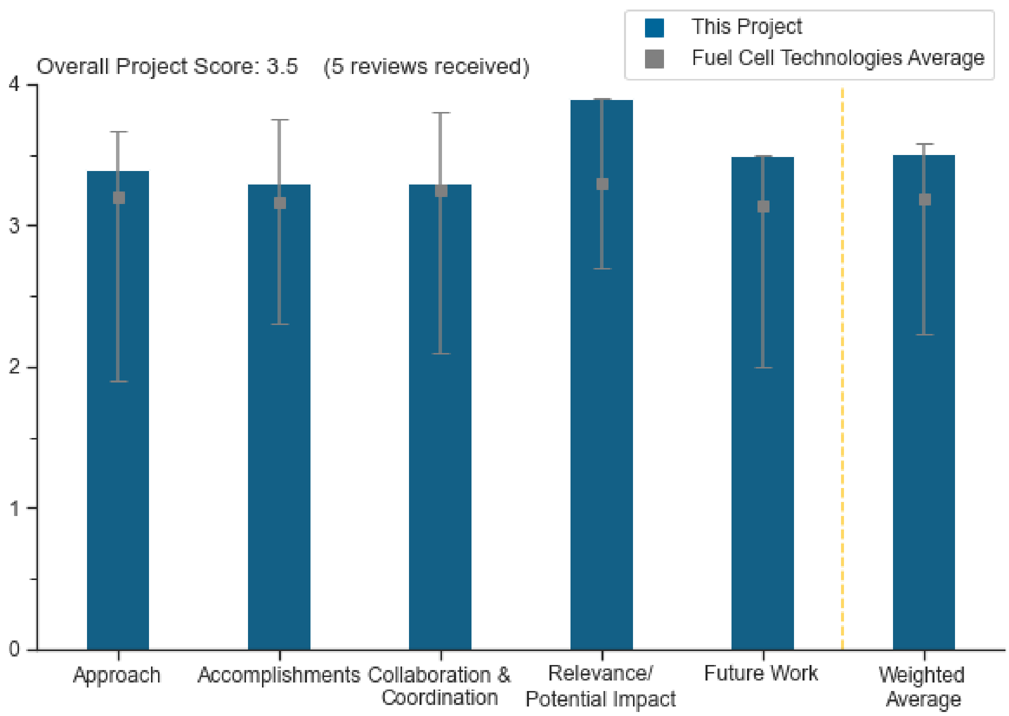
Michael Yandrasits, 3M Company

DOE Contract #	DE-EE0009244
Start and End Dates	1/1/2021 to 12/31/2023
Partners/Collaborators	National Renewable Energy Laboratory, General Motors
Barriers Addressed	• Durability • Performance • Cost

Project Goal and Brief Summary

This project aims to increase membrane lifetimes by improving the inherent chemical stability of perfluorinated membrane ionomers. If successful, the project will increase fuel cell lifetimes and allow fuel cells to meet the U.S. Department of Energy's 2030 heavy-duty transportation target of 25,000 hours of operation.

Project Scoring



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

Note: This is a new project in 2021. 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 objectives and barriers and for project design, feasibility, and integration with other relevant efforts.

- The project approach is excellent. The team first identifies the chemically weak linkage of perfluorosulfonic acid (PFSA) using density functional theory (DFT) calculations and tries to improve the stability by synthesizing novel ionomers. Also, the project approach contains a small molecule study that can verify the DFT results.

- The team has done an excellent job in identifying the critical barriers, setting the project objectives, and developing a very logical approach.
- The team's approach is to get a fundamental understanding of the energy state of degradation initiation atoms and, hence, design a "new" perfluorinated ionomer with a side chain architecture immune to radical scavenging and stable under oxidative attacks under dry fuel cell operational conditions to achieve the desired high-performing/low-degrading membrane. This is the correct approach of balancing the structure–property relationship of new ionomeric membranes. The team's objective of understanding the effect of manipulating C-F and C-O groups to enhance side chain stability is the correct approach. Before spending the resources on synthesizing new ionomer, it is better to conduct the modeling of the proposed side chain structure and understand the oxidative activation barriers of oxidation-susceptive bonds. This will help the team in focusing on an ionomer structure that has a high chance of meeting U.S. Department of Energy's chemical durability targets.
- This is a very interesting approach to improving the durability of PFSA-type materials by replacing various weak links (tertiary fluorine and ether linkage) with perfluoromethyl groups. The fact that this project is looking to stabilize PFSA is great, but the changes will not affect the acid-containing unit, so it is possible to expect higher durability and loss in conductivity. Moreover, fluorochemistry manipulation is not trivial, but the folks at 3M Company (3M) are experts in this area, which gives this project a higher probability of success.
- The project has very detailed and clear tasks. A clear effect of changing the structure on chemical durability would be expected, but it is only about the durability regardless of performance, i.e., ionic conductivity. As mentioned in the slide, the end group working on ionic conducting is out of scope. There should be additional research for the performance improvement.

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 started in January 2021. In six months, the team has accomplished DFT calculation of reaction barrier modeling, DFT solvation, and protonation modeling, as well as establishing the synthetic pathway of a monomer precursor with isopropenyl ether functionality. The team has also established the baseline of 3M standard ionomer degradation under open-circuit voltage (OCV) conditions. Six months into the project, the team has made significant progress into its effort of developing oxidatively stable ionomers for fuel cell use.
- There is not much experimental progress with this project, as the project started on January 1, 2021. The National Renewable Energy Laboratory (NREL) DFT modeling study has made some progress. DFT reaction barrier modeling shows that H radicals attach on ionomer C-F bonds. The 3M ionomer has more stability than a long side chain ionomer. The proposed ionomer that replaces C-F with CF3 in the PFSA backbone may further increase the stability. DFT calculation also shows that introducing CF3 in the polymer backbone would have no impact on the protonation state, which is intuitive. Monomer synthesis seems to be challenging. 3M has started the monomer synthesis. Other accomplishments, such as OCV studies, degradation mechanism studies, and combined chemical–mechanical tests, were presented. Those are relevant to the project, but it is believed that the works were done by other resources. It is acceptable since the project started late.
- The DFT modeling task has progressed well. Good progress has also been made in the effort to establish some baseline performance for the current 3M ionomers. The team seems to have yield issues for the monomer synthesis, but the milestones are not due yet.
- Fluorochemistry is not easy, and monomers are being synthesized, but the optimization will take time, and it should not be expected that optimization occurs in a few months.
- Considering the initial state of the project, the plan and initial study are very logical, with a clear target.

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 constitutes two industry representatives (3M and General Motors [GM]) and one national laboratory (NREL) as partners. The work has been distributed well amongst them based on their core expertise. The six-month progress reported by the team on DFT modeling (NREL), monomer synthesis (3M), and establishing the OCV baseline degradation condition and conducting different accelerated stress test/highly accelerated stress test (HAST) protocols (3M, GM) clearly shows a good collaborative relationship between these three partners. In the past, the prime, 3M, has worked with NREL and GM on several DOE-funded projects, and the three organizations have demonstrated good working relationships. Therefore, it can be expected that the team will demonstrate a similar collaborative environment in this project.
- The team is strong, and the team members have the right expertise for the project. NREL demonstrated modeling work for the project, and GM likely provided valuable durability data using the company's HAST protocol.
- There is a good team of a national laboratory (for modeling), material synthesis, and original equipment manufacturers (OEMs).
- All partners need to be engaged and are very engaged.
- It is still in the initial state, so it is not expected that there is deep collaboration.

Question 4: Relevance/potential impact

This project was rated **3.9** for supporting and advancing progress toward the Hydrogen and Fuel Cell Technologies Office goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- This project is relevant to three out of seven DOE targets established for Class 8 long-haul tractor-trailers. The success of this project will make a significant impact in bringing the fuel cell technology a step closer to commercialization. This project directly addresses the fuel cell system lifetime and consequently addresses system cost and peak efficiency targets. The chemical durability (OCV) target of DOE is >500 hours. The OCV durability target of this project is three times higher (>1500 hours) than DOE's target. In support of the chemical durability of the membrane, over and above DOE's chemical durability target, the team has put in place separate accelerated durability targets, OCV fluoride release rates, and a combined "chemical–mechanical" accelerated durability target of >1,750 hours. These two additional targets will reinforce chemical durability properties of new membrane material and are expected to help push up the DOE chemical durability target.
- This approach to improving the durability of PFSA, while still maintaining its high proton conductivity, is very interesting. If this project is successful in developing the next-generation PFSA, 3M can then readily scale the chemistry, and GM would be a perfect end user. If everything plays out as hoped, this could be a potential game changer.
- Developing chemically stable polymer electrolyte membranes (PEMs) is one of the most urgent challenges for automotive fuel cell applications. The approach to modify the chemical structure of PFSA will have a tremendous impact, if successful, and overall, it is a better approach to enhance chemical stability than using a radical scavenger. The downside of this approach is synthetic uncertainty and cost. Nevertheless, this type of effort should be supported by the U.S. government.
- Developing stable ionomers that are free from those known vulnerable groups is critical to reaching the really long lifetimes required by trucks.
- Using a very popular and fundamental material will have a big impact on other research, once there is a meaningful result.

Question 5: Proposed future work

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

- The future work cited on slide 18 is aligned with the development course of the project. Proposed DFT simulation study of category 1 and category 2 structural linkages on slide 11 is a good way of sorting out the weakest link responsible for degradation initiation under low-relative-humidity (low-RH) fuel cell operational conditions. The team has identified the future work correctly, including the work on (i) degradation studies, (ii) DFT modeling, (iii) monomer synthesis and (iv) durability validation needed for identifying the weakest link of the perfluorinated ionomer and, hence, developing a robust side chain linkage for an oxidatively stable ionomer. There is no obvious gap in the proposed work in terms of executing the proposed project.
- The future work is well-organized, including degradation studies, DFT modeling, monomer synthesis, and durability validation. The project needs to focus more on monomer synthesis and polymerization.
- The proposed future work is reasonable and planned out well.
- It is in the initial state. It is expected that there will be a significant development.
- The approaches for the future are very logical.

Project strengths:

- The strength of the project is 3M’s expertise in PFSA membrane design. Variations of the chemical structure for current PFSA PEMs can provide valuable information, and the tasks should be done. 3M has done much work in this area, and the studies contributed much to advanced PEM design. The project team is strong and complementary. The project’s principal investigator understands the PFSA chemistry well, and the tasks can be accomplished efficiently. GM’s involvement in this project is also advantageous.
- Overall, the project is a very well-thought-out project, focusing on identifying the weakest link in perfluorinated ionomer chains and, hence, developing a tailor-made side chain structure that will be stable under low-RH fuel cell operational conditions and, hence, helping DOE to achieve its desired fuel cell membrane target for Class 8 long-haul tractor–trailers. The project partnered with respective technical experts from reputable organizations, well known for their fuel cell expertise. The technical approach and future work proposed by the team are systematic and logical.
- There are many strengths of this project, which has PFSA manufacturing experts who are partnered with a large OEM at GM working to improve PFSA durability. It will be exciting to see what results they can demonstrate next year.
- This project is based on very fundamental materials, and the target is very clear. If there is a good result, then there will be a big impact on future research and development.
- The project team is very strong. The plans of the project, current and future, have been well-thought-out and are well-executed.

Project weaknesses:

- There are none. The project is wished luck, as it is not easy to work with fluorinated polymers.
- There are no particular project weaknesses, other than the fact that this is high-risk and high-reward type of project.
- The team should have a strategy to determine the possibility of synthesizing DFT simulation-predicted side chain structures. The team also needs to have a strategy to rate and compare DFT simulation-predicted stable side chain structures on the scale of OCV oxidative stability and synthesizability of those side chain structures.
- The reservation for this project is how the chemistry manipulation will affect the final cost of the new PFSA. These new monomers will have different reaction steps, product yields, and reactivity ratios, which makes cost very concerning. Also, even small changes in the polymer structure are sure to affect the mechanical properties and hydration characteristics.

- Research using a very fundamental material has two different views. Many similar chemical structures studied in past decades have not shown better properties. If the results show similar or even worse properties, then there would be no impact on future development.

Recommendations for additions/deletions to project scope:

- More work is suggested on monomer or model polymer synthesis before extensive degradation studies and modeling. Novel PFSA development is limited mostly by polymer synthesis.
- To improve ionic conductivity, there should be additional research on the end groups.

Project #FC-335: Additive Functionalized Polymers for Extended Heavy-Duty Polymer Electrolyte Membrane Lifetimes

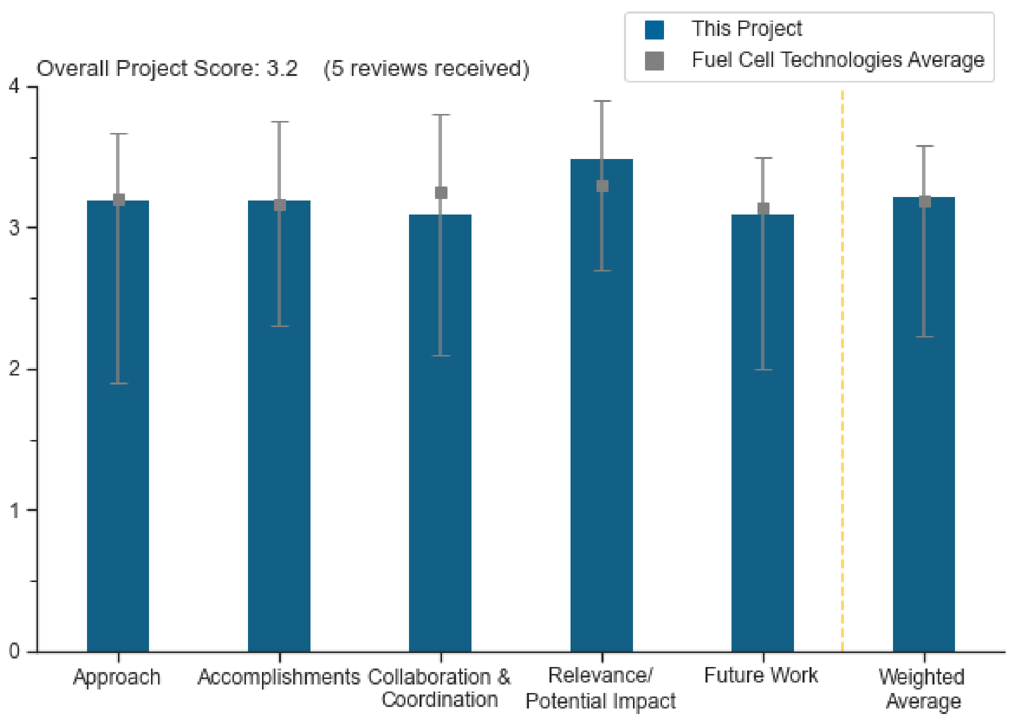
Tom Corrigan, The Lubrizol Corporation

DOE Contract #	DE-EE0009245
Start and End Dates	Q2 2021 to Q3 2023
Partners/Collaborators	National Renewable Energy Laboratory
Barriers Addressed	• Chemical durability of polymer electrolyte membranes • Shortcomings of current radical scavengers • Fuel cell stack lifetime

Project Goal and Brief Summary

The Lubrizol Corporation (Lubrizol) will work with the National Renewable Energy Laboratory (NREL) to develop membranes with enhanced chemical durability, with the goal of improving the lifetimes of polymer electrolyte membrane (PEM) fuel cells for heavy-duty vehicles (HDVs). The research team will identify novel additives to mitigate chemical degradation and find strategies to immobilize these additives, thereby addressing radical scavenger shortcomings. The improved membrane durability could enable PEM fuel cell HDV lifetimes that achieve the U.S. Department of Energy (DOE) target of 25,000 hours (one million miles for long-haul trucks).

Project Scoring



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

Note: This is a new project in 2021. 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 objectives and barriers and for project design, feasibility, and integration with other relevant efforts.

- The development of membranes with enhanced chemical durability for HDVs is important to achieving DOE's and original equipment manufacturers' (OEMs') goals. Lubrizol is a leading manufacturer of additives for various industries, including radical scavengers. The project wants to demonstrate better scavenging performance than that achieved with cerium, including no migration. The physical blending and covalent attachment are approaches for application of scavengers. The team will use open-circuit voltage accelerated testing to qualify the material. The Year 1 go/no-go milestone to demonstrate chemical stability is comparable with or better than Ce-doped perfluorosulfonic acid (PFSA), with three times greater stability in hydrocarbon PEMs.
- The project goals are to improve durability by overcoming the shortcomings of the current radical scavengers by using two covalent functionalization strategies to immobilize the scavengers. The strategies are explained clearly with specific deliverables.
- This project has a good plan for screening, incorporating, and characterizing membranes with the novel additives. An understanding of the underlying mechanisms of the additives was not discussed.
- The project's approach is good; however, there is some doubt about the point of using an additive to improve the durability of a hydrocarbon membrane that will not be stable enough itself.
- The approach is centered on two methodologies to incorporate additives into the membrane to mitigate chemical degradation. A significant amount of information is left out, most likely for proprietary reasons, making it challenging to judge the potential effectiveness of the proposed strategy.

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 is new, with a start date of April 1, 2021. The project has explored two membrane impregnation procedures: swollen membrane and solution casting. The team has demonstrated the preparation of impregnated PFSA membranes via solution casting, with good homogeneity for initial results, and presented initial conductivity data for impregnated PFSA membranes. The preliminary in-plane conductivity shows some effect on conductivity for some additives, but there are no comments on the reason. The preliminary qualitative Fenton test performed shows promising results.
- The team has made excellent progress, given that the project only just started. The team has already demonstrated some durability improvements over non-Ce-doped control samples, which demonstrates that the approach is working. Goals should be set higher than the go/no-go of only equal durability to state-of-the-art PFSA materials. In fact, the team should use a Chemours NC700 as the benchmark, or at least a Nafion™ XL membrane, to determine whether improvement over the state of the art is made.
- The membranes incorporating non-covalent additives have been fabricated using two strategies. The project identified the pros and cons for each strategy and membrane. Casting and fabrication conditions have been optimized to cast membranes that are intact. The bar plot in slide 9 zooms into a narrow range of values and could be misleading since the actual reduction achieved would be less than it appears.
- A number of control samples and samples with additives have already been prepared. The Fenton test results and membrane conductivity may be within the error margin of the measurement.
- The project just started, but it has demonstrated two methods for additive incorporation, and early testing indicates that chemical stability improved with the additive.

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 listed NREL (Million Mile Fuel Cell Truck [M2FCT]) for various types of evaluation and cell testing. Such testing will help with the project's progress and membrane assessment in the cell.

- NREL is a good partner that can verify the accelerated stress test improvements by comparing results to the laboratory's own baseline.
- NREL has very good expertise in membrane electrode assembly fabrication and characterization.
- NREL is the only partner mentioned. The general work plan with NREL was presented.
- The team is sufficient to complete the proposed work.

Question 4: Relevance/potential impact

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

- The approach has excellent potential to improve the state of the art if the project can demonstrate the immobilization of the additive and demonstrate that the additive will not leach out or get destroyed during operation.
- If Lubrizol can identify a low-cost additive membrane system that can be solution-cast and demonstrate between radical scavenging behavior, without the migration observed with Ce, it would have great impact on the DOE goals for fuel cell electric vehicles (FCEVs).
- The goal to improve the durability by altering the radical scavengers or their attachment to the membrane for immobilization is in line with the Fuel Cell Technologies subprogram goals and supports the need for durable PEMs for HDVs.
- The work is very relevant to the HDV funding opportunity announcement. If successful, the project could have a significant impact on membrane longevity.
- The extending membrane lifetime is a critical need for heavy-duty FCEVs.

Question 5: Proposed future work

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

- The listed activities are all relevant and important for the realization of the project goals. The crossover and stability, in addition to the relative humidity conductivity, are all important properties.
- The proposed work is good, and the project should continue to benchmark its own additive-prepared membranes to control membranes even without Ce+ additive. This will demonstrate that the project's approach is working. However, the team should also use commercially available membranes with Ce+ additive in the tests to determine the target over and above the state of the art. There should be an improvement in durability over existing Ce+-based membranes.
- Considering that this is a new project, the presented work plan details are very good for achieving goals with the collaboration with NREL.
- The project just started, but the proposed future work appears adequate at this point.
- Potentially only one additive will be evaluated.

Project strengths:

- Radical scavenging is important for cell durability. Lubrizol has a catalog of additives and experience in radical scavenging in general. The collaboration with NREL should allow for the identification of at least one candidate additive that will be effective for scavenging but not hinder or deteriorate other membrane properties (i.e., conductivity, mechanical). Details and progress on the covalent additive systems are happily anticipated.
- The combination of casting strategies as part of the impregnation and fabrication could be effective and useful for improving membrane properties. The membrane preparation and processing are explained in detail.
- This project's strengths are the use of multiple additive incorporation strategies and the focus on unique additives beyond cerium.

- Studying the effect of additive concentration is a good plan.
- An immobilized additive has potential for large improvements in lifetime.

Project weaknesses:

- There are no significant weakness noted for the project objective. It would be good to collaborate with OEMs sooner rather than later. There is no mention of consideration as to whether the impregnated additives, if liquid or low-melting, will plasticize the membrane, thereby depressing the glass transition of the polymer, which may limit cell operation temperature. This would not be an issue with covalently bonded additives.
- The effectiveness of the proposed immobilization strategies is yet to be proven since the current progress is based on a limited dataset with Fenton tests only.
- While the project has two methods of incorporating additives, to mitigate the risk of any of those additives not working, it would be helpful to have an additional approach to improving membrane durability.
- At this stage, with limited information, it is difficult to assess the risk of the novel additives.
- The project's goals and targets are not set high enough.

Recommendations for additions/deletions to project scope:

- The project reports and compares fluoride (F^-) release rates, which are helpful for evaluating the effectiveness of the additives, but the changes in F emission are different from what was reported in the literature. The procedure and values should be checked against the previously reported data. The project could benefit from some additional membrane characterization efforts, including temperature-dependent properties, maybe with dimethylacetamide (DMA), and some additional transport properties. It would be helpful to know the percentage of loading of the additives relative to SO_3 groups.
- As with similar projects, it is important to determine whether the additive is regenerable to have a long-term impact on membrane lifetime.
- It is suggested that the project use nuclear magnetic resonance analysis to assess chemical degradation of membranes and additive incorporated membranes.
- Multiple samples of each process/composition should be prepared and characterized to ensure repeatable results.
- The project should raise the bar for the performance targets.

Project #FC-336: A Systematic Approach to Developing Durable Conductive Membranes for Operation at 120°C

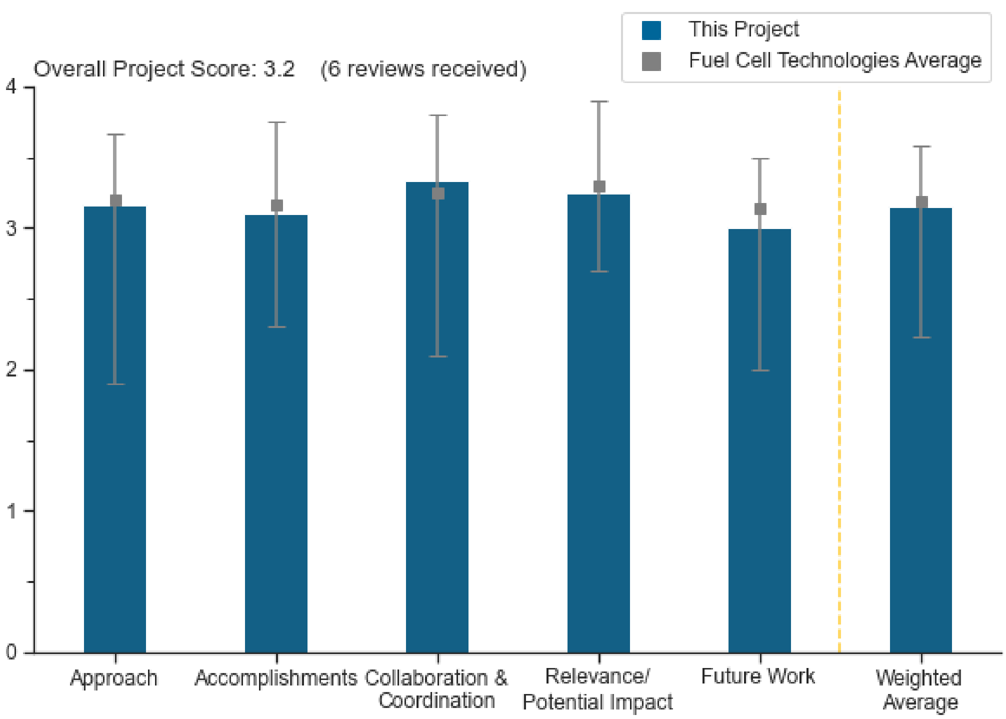
Tom Zawodzinski, University of Tennessee, Knoxville

DOE Contract #	DE-EE0009246
Start and End Dates	10/1/2020 to 3/31/2021
Partners/Collaborators	Oak Ridge National Laboratory, Akron Polymer Systems
Barriers Addressed	• Low area-specific resistance • Low oxygen, hydrogen crossover • 25,000-hour lifetime

Project Goal and Brief Summary

This project aims to develop membranes with sufficient performance and lifetime to meet the requirements of long-term applications of polymer electrolyte membrane fuel cells (PEMFCs) for heavy-duty (HD) vehicles. The research team will use background measurements and literature evaluation to inform paths forward for membrane development to meet cell resistance requirements over ranges of temperature and relative humidity (RH) that reflect operating conditions in HD vehicles. Researchers will then identify and prepare new membrane materials with side chain and polymer chemistry tailored to achieve acceptable conductivity and resistance, with low water uptake and swelling. The University of Tennessee, Knoxville (UTK) is collaborating with Oak Ridge National Laboratory (ORNL) and Akron Polymer Systems (APS) on this project.

Project Scoring



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

Note: This is a new project in 2021. 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 objectives and barriers and for project design, feasibility, and integration with other relevant efforts.

- The team's approach of "mining for protons" is a good way to bring the results from past work together and gather more information on the mode of water transport per mole of sulfonic acid group. This will help the researchers to design their proposed multi-acid exchange site ionomer, as shown in slides 11 and 12. The team's approach of manipulating composite structure to balance proton conductivity and water transport is a good way to determine the balance needed for the 120°C functional fuel cell membrane. Overall, the approach is adequate and aligned with the proposed goal of the project to develop a low-RH membrane.
- The project approach is clearly defined as obtaining acceptable conductivity with minimal water and obtaining high durability. For the acceptable conductivity with minimal water, the team plans to work on preparing composite structures and high ion-exchange capacity (IEC). For durability, the team plans to minimize dimensional changes over RH/temperature, gas crossover. Those approaches that the team is pursuing are not new but seem worth trying further with this project. The team understands the issues with sulfonated polymers for high-temperature (HT) and low-RH fuel cell operations. The technical approach is technically sound.
- The project aims to address membrane performance with a focus on the role of water, specifically retention of water and proton conduction of different ionomer types, structures, and composites at elevated temperatures. Another important aspect of determining a path toward low gas crossover is described but is not part of the Year 1 go/no-go milestones. Both published data and ex situ material properties will be collected through data mining and direct measurements, respectively, to discern potential modeling or predictive parameters leading to enhanced proton conductivity and increased lifetimes at hot and dry conditions relevant to heavy vehicle PEMFC applications. An interesting approach to increasing the acid content of the perfluorinated sulfonic acid (PFSA) using multifunctional phenyl groups, based on previous UTK work, will help probe IEC–structure relationships. The reason to use bis(trifluoromethylsulfonyl)imide (TFSI) as an analogous acidic group was less clear. The gas phase acidity of perfluorinated bis(sulfonyl)imides has been shown to be stronger than that of PFSA for small molecules; however, the added mass (i.e., lower IEC per gram) and the mixed phenyl-trifluoromethyl imide in the hydrocarbon (HC) systems may be less acidic. The bis(sulfonyl)imide structures also lead to the potential for increased gas crossover, and impact to bulk structure may confound performance results; that said, measurements of all proposed material properties on these systems may give direction as to which functional groups to focus on for HT applications.
- The approach of the project is rather unique in that the team does not have its own particular polymer to promote. Instead, the project is using a set of guiding principles to look at all possibilities and try to arrive at a polymer that is good. There is an intrinsic risk to this approach.
- Tom Zawodzinski has made a tremendous impact in PEMFCs over several decades, and it will be interesting to see the progress that his team makes in the next year. However, the team has already seen the use of HC polymer in PEMFCs and additives, as well. The team should, as planned, "mine" the literature to further develop concepts that have shown promise; however, while high IEC, additives, and sulfonamides have shown promise, they have also proven to have issues. High IEC materials have high water content and typically quickly degrade mechanically and/or chemically (radical attack). Additives, although not described in detail, will result in brittle membranes at high contents, and the additives can migrate or degrade. Finally, sulfonamides have higher acidity than sulfonic acids, but the impact in fuel cell performance is small, as was discovered by 3M Company. It will be interesting to see the progress of this project.
- To obtain higher proton conductivity at 120°C, the overall approach looks very reasonable and promising. However, there is no clear approach on how to get high durability. There would likely be a trade-off between high conductivity and high durability, especially at high temperatures, such as above 100°C. The reviewer is a little worried that the current membrane material would have a limitation on durability at high temperatures, even though there would be a significant modification from the current material.

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 team has acquired PFSA resins with a large range of equivalent weights, from which two bis(sulfonyl)imide derivatives have been prepared. Additionally, precursor HC polymers (e.g., PPO-Br) have been acquired, and APS polymers have been prepared for membrane property testing. As this project has just begun, the collection of a suite of materials, clear plans of polymer functionalization and fabrication, and the acquisition of material characterization equipment to date are appropriate. The team has made progress since the slides were submitted, as described during the presentation. The details about the data mining team's progress were lacking, but certain literature information the team will collect was discussed.
- The project started in April 2021, and the subcontractor agreement was completed in May 2021. This gave the team very little time to report any measurable progress for the 2021 DOE Hydrogen Program Annual Merit Review (AMR). Within this short time, the team has conducted "mining for protons"; prepared and tested the membrane materials; and deployed and used new test beds for automated testing over a full range of RH and temperature. Overall, the progress is significant within this short period of time.
- It seems that the project just received its funding not long ago. Its milestones have been pushed back, likely due to the delay of funding. The team has made some progress on the data mining part of the project. Materials have been acquired, and things are beginning to move.
- This project has just started, so it's hard to tell about the results, but there's been very good preparation on background research on how to get high conductivity.
- There wasn't much done in terms of accomplishment, but that is not the fault of the principal investigator (PI), as the funds arrived in mid-March. It seems the time was spent planning future synthesis and partnering with a toll manufacturer.
- The actual start date for the project is March 15, 2021. Because of the late start date, there are limited accomplishments. The chemical structures of the planned polymers were shown, but no supporting spectroscopic data were provided.

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 is a well-balanced research team composed of an academic institute, a national laboratory, and a polymer company. The team is well-posed and has the necessary expertise to conduct the proposed development work. The role of each partner is well-defined, and the tasks are assigned appropriately and are aligned with the partners' core expertise. The PI is a well-known researcher in the fuel cell area and is engaged with both UTK and ORNL. His link with both entities will help the progress of the project due to effective communication between both entities. The team is tightly engaged.
- UTK has clearly established the roles of all partners in the project plan. APS is providing scalable HC polymers, and UTK will handle some polymer modification synthesis, detailed material testing, and, eventually, data mining. Engagement with ORNL for side chain modification was apparent, and the presenter described having an industrial partner with precision coating capability, Kodak, for when scaled-up coatings are needed. The project is in its early stages, so defining the roles of the team partners and establishing partnerships to generate high-quality materials for characterization were critical.
- The team is composed of a university (the prime), a national laboratory, and an industry partner. All three institutions are involved with polymer synthesis and characterizations. Collaboration with Kodak for a large-scale film preparation looks like a good plan. It is possible that institutions for fuel cell testing can be included. The validation of membrane performance in membrane electrode assemblies (MEAs) by fuel cell or automotive companies may be desirable.
- The partnership is fairly small right now, but it makes sense. Initially, ORNL and APS were focusing on making small batches of materials and looking at blending additives. If ORNL comes across a material that

is interesting, it can hand it off to APS to scale. Eventually, another partner will have to be added, such as Los Alamos National Laboratory or General Motors, to run third-party fuel cell testing.

- The project has just started. There seems to be a very good collaboration to get the plan going.
- All partners seem to be engaged.

Question 4: Relevance/potential impact

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

- The project is relevant to the multi-year research, development, and demonstration plan (MYRD&D plan) and DOE's targets for long-range trucks. The success of this project will make a significant impact in the progress of fuel cells to large-vehicle use. This project addresses DOE goals by providing paths to (i) lower greenhouse gas emissions, (ii) create good-paying jobs in the U.S., (iii) build clean energy infrastructure, and (iv) support environmental justice.
- The project supports the Million Mile Fuel Cell Truck (M2FCT) consortium goals, which include material (component-level) targets that are relevant to long-haul trucking (e.g., a wide range of operating humidity and temperature with minimized gas crossover). This project supports high-level goals of DOE, including advancing competitive materials for the market, with a focus on durable PEMFC MEA components, and an emphasis on HD efforts that include enhancing efficiency and durability, which should also benefit medium-duty and stationary applications. Moving toward zero emissions and clean energy infrastructure are outcomes that could be accelerated by the anticipated results of this project. A partial focus on HC membranes targets costs associated with PEMFC components and new strategies to probe structure–performance relationships. These hit directly on the Hydrogen and Fuel Cell Technologies Office's research, development, and demonstration (RD&D) strategies involving innovative membranes and ionomers to achieve lower costs, higher durability, and improved power density.
- Developing polymer electrolyte membranes (PEMs) for HT and low-RH operations is critical for HD fuel cell applications. The project goal is well-aligned with MYRD&D plan goals.
- Once there is a higher-performance and higher-durability membrane for 120°C operation, there will be a great variety of applications in the HD truck industry because heat rejection is one of the biggest challenges.
- The PI is experienced in the type of work proposed, so it is expected that the work will stay focused on the relevant goals.
- It is too early to predict the potential impact of this project since it is still getting off the ground. The PI has a plan, and he does have ideas of improving membrane performance to meet DOE targets. There are not many details of what the PI has planned, but that is understandable, as the field is very competitive and they must be careful not to disclose information at this stage of development.

Question 5: Proposed future work

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

- The project's planned approach is logical, including wide-net literature mining and data generation to complement sparse published data at low RH and high temperatures. The emphasis on formulating and scaling materials to explore avenues (i.e., cross-linking, reinforcements) for swelling control in a chosen system to enhance durability is appropriate. However, it was not clear whether only materials meeting the Year 1 go/no-go would be moved forward in the scaling effort. Synthetic strategies aimed at both HC block copolymer and PFSA polymer modifications, such as the "Ball o' Sulfonates" approach to study structure–performance relationships, could provide important insight into management of water in membrane materials.
- The future work cited on slide 16 is aligned with the development course of the project. Since the project just started and very little progress has been made, it is difficult to comment on the future work and judge the relevance and alignment of the future work to the project goal. Maybe in the next AMR, there will be enough information to judge the relevance of the future work.

- It is a good approach to obtaining high conductivity, but it is still unclear how to get higher durability at 120°C.
- The proposed work is relevant. Once more progress is made, it will be easier to determine whether the future plans are logical.
- The future works are described in general terms. It seems there are no specific structures to get to the performance and durability targets, for example, approaches to making composite membranes that include cross-linking, reinforcing, and fillers. There needs to be more specific methodology for cross-linking and similar types of inclusions. The future work looks more like basic research rather than PEM development for HD fuel cells.
- The material that was presented in the slides was not particularly exciting since what was shown has already been looked at in some shape or form in the past. However, the PI is not showing his cards yet. It will be interesting to see the progress that is made from now until next year's review since that will be a better indicator of the project's direction and will give the reviewers a better idea of the materials–performance synergy that is being made.

Project strengths:

- There is significant potential for the project to generate very useful parameters for determining how a membrane material will perform at low RH and high temperatures, as well as structures that provide consistent performance over the RH range at a given temperature. The team has experience determining critical parameters for predicting performance, such as water and proton mobilities, as well as composite materials that flatten the conductivity curve over an 80% RH range. Collaboration with an HC team that has scaled materials is helpful for determining feasibility of material sets and producing relevant quantities of polymers for this project.
- The personnel in this team and their technical expertise is the strength of the project. The PI and ORNL have immense knowledge and resources to explore the proposed objectives of the project. ORNL has world-class analytical techniques for fuel cell components, especially in the MEA space. The project is expected to get the best possible support needed for its progress.
- The strength of this project is that Tom Zawodzinski is leading it. With his leadership, at the very least, we will gain better fundamental understanding in the area of PFSA/HC/additives and its impact on water diffusion, proton conduction, gas permeation, degradation, and fuel cell performance.
- The project PI is a well-known expert on PEMs and has worked for a long time to develop PEMs for HT and low-RH fuel cell operation. The proposed HC membranes can be tailored easily to accomplish the project approaches. Once desirable materials are prepared, the scale-up of those materials may not be challenging compared to that of PFSAs.
- The target is very clear, and the high performance and durability are very important for the next-generation fuel cell system. This project has a good plan for how to reach the target, and there will be a good chance of solving current practical issues.
- The team is strong, and the approach is unique. The potential is large.

Project weaknesses:

- The project is new, and not enough progress has been made to understand the weaknesses. Overall, the proposed steps of the project seem to be logical, and it is too early to point out any weaknesses of the project.
- The weakness is that the materials that are discussed are materials that have been already evaluated; however, it seems likely that the PI is holding some ideas back to protect them.
- The project is intrinsically a high-risk one, as it does not have a "promising" material with which to begin. However, this has been the uniqueness of the project.
- This project needs more engagement with fuel cell testing. It needs a solid plan with M2FCT and other coordination for the purpose. Fundamental understanding is important, but it should not be the main part of the project. Currently, the project looks like low-technology-readiness-level research. The project should not pursue large-scale production until the materials show high performance in an MEA under HT and low-

RH conditions. The area-specific resistance (ASR) milestone is 0.02 ohm cm^2. However, the project is pursuing acceptable conductivity at high temperatures and low RH. Therefore, the milestone and project target are not aligned. If the team is indeed looking for an HT membrane with such low ASR, this is a very challenging project, and no clear pathway was provided.

- The materials set includes materials known to the investigators as ones that likely will not meet durability requirements (e.g., styrenic materials). Prioritizing HC-based materials with structures that have proven adequate chemical stability is more appropriate when determining parameters for increased performance and water management relative to PFSAs. It is unclear what materials set the investigators will move forward with if, for instance, only high-IEC PFSA materials meet the Year 1 go/no-go targets.
- There is no clear approach on how to get high durability.

Recommendations for additions/deletions to project scope:

- The team should consider going through the work of Professor Alan Hay from McGill University on the dendrimer-based ionomeric polymer. Some of his work has been published in *Macromolecules* 41, 2 (2008): 281–284. The proposed project on preparing densely sulfonate-packed structures (e.g., Ball o' Sulfonates) has some similarity to Professor Hay's work.
- To get the high durability at 120°C, it is necessary to develop a different structure of the polymer, or modification from the current material could be an option. The team is asked to make it clear how to approach that issue.
- The project has an is appropriate scope: to determine the most important material parameters for effective water usage with minimal dimensional changes that first meet DOE performance targets by the Year 1 go/no-go point (material milestones). Milestones include publishing findings from data mining and choosing or developing a relevant durability test with which to test the team's materials solutions. No additions or deletions to the project scope are recommended at this time.
- No particular tasks need to be deleted, but the team may consider narrowing down the composite membrane approach. It is essential for fuel cell testing (single-cell level) to evaluate the membrane performance and durability. Most other MEA component development includes fuel cell testing or demonstration to verify that the proposed approaches are valid.

Project #FC-337: Cummins Polymer Electrolyte Membrane Fuel Cell System for Heavy-Duty Applications

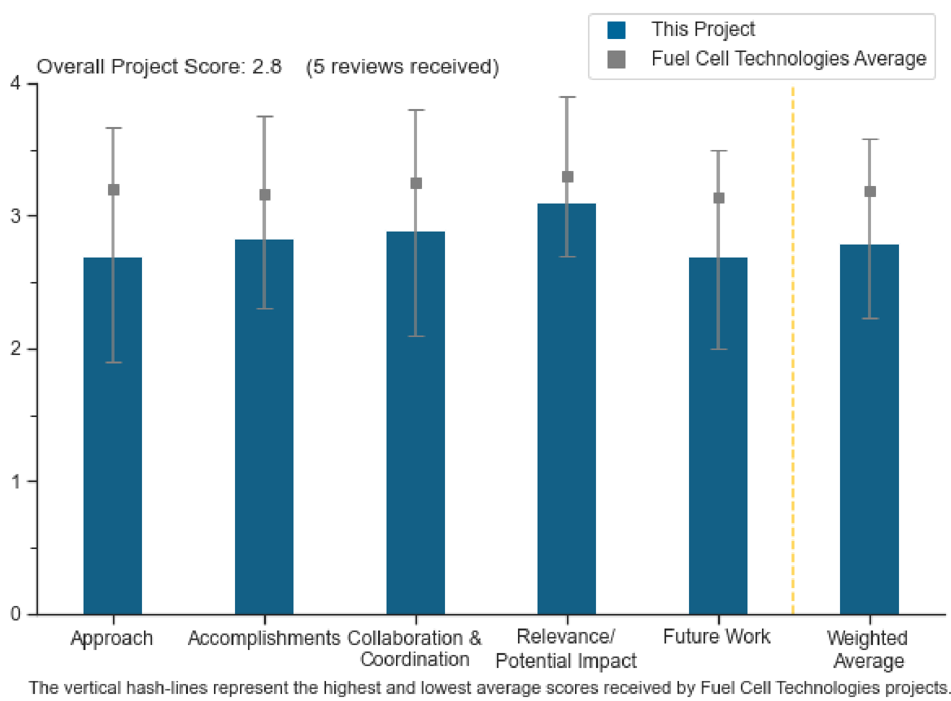
Darren Hickey, Cummins Inc.

DOE Contract #	DE-EE0009247
Start and End Dates	8/1/2021–7/31/2024
Partners/Collaborators	Cummins Hydrogenics, Argonne National Laboratory, W.L. Gore & Associates, Inc., Dana Incorporated, Cummins Turbo Technologies
Barriers Addressed	• Operation ≥100°C • 68% peak efficiency • 25,000-hour life • $80/kW system cost in mass manufacture

Project Goal and Brief Summary

The objective of this project is to develop and demonstrate a new standardized, modular, and scalable 100 kW polymer electrolyte membrane fuel cell (PEMFC) stack that meets performance, efficiency, durability, and affordability requirements for heavy-duty (HD) applications. Membrane electrode assembly (MEA) and bipolar plate (BPP) development efforts will be undertaken and demonstrated in progressively larger stacks. The stack will be designed to run at higher pressure and tolerate high temperatures (≥100°C) during peak power excursions. A key metric is the system cost of $80/kW at 100,000 units per year volume. To achieve this objective. a study on advanced manufacturing methods to reduce production costs will be undertaken. This project is a collaboration between Cummins Inc.; its Fuel Cells and Hydrogen Technologies division (FCHT, comprised in part by Cummins' acquisition of Hydrogenics); Cummins Turbo Technologies (CTT); Argonne National Laboratory (ANL); W.L. Gore & Associates, Inc. (W.L. Gore); and Dana Incorporated (Dana).

Project Scoring



Note: This is a new project in 2021. 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 **2.7** for identifying and addressing objectives and barriers and for project design, feasibility, and integration with other relevant efforts.

- The approach as cited on slides 6–7 of the presentation is a well-thought-out approach for developing a high-pressure PEMFC stack and system for HD applications with high efficiency. The team's top-to-bottom task flow is well-connected and logical and is owned by partners with the appropriate expertise needed to address the criticality of individual tasks. One thing is not clear from slide 6, and that is why the team is selecting the W.L. Gore membrane to make a MEA with >100°C operational capability. The operational temperature has impacts on the membrane dryness and, hence, the performance. Presumably, the team looked into this matter before selecting the W.L. Gore membrane. Another point to be noticed is that the team wants Dana to develop the manufacturing process of graphite BPPs (slide 11). This could become a bottleneck of the project. It is to be hoped that Dana already has a manufacturing plan to provide an adequate amount of graphite BPP for this project.
- The high-temperature membrane is an interesting and potentially exciting portion of this project; more details will be helpful. A more in-depth discussion about the time at high temperatures (e.g., 1% of 25,000 hours at >100°C) will be very valuable.
- The market/industry-requirements-led approach is effective, and the focus on high-temperature capability, combined with a simultaneous focus on system components (e.g., the air compressor), is good and is needed for successful HD applications. However, the focus on membrane capability at high temperature may not be sufficient, as catalyst failure modes are also accelerated at high temperature; this may be the limiting factor in achieving the lifetime requirement.
- The project properly notes the importance of higher maximum operating temperature to meet Class 8 long-haul HD requirements. However, it is unclear how durability will be achieved while simultaneously increasing maximum temperature. The approach to do so is not described. Similarly, how to achieve higher efficiency, other than the reduction in electrode activation at higher temperature, is not described. Further, this implies operating at elevated temperature not just during extreme events but all the time (to realize efficiency and fuel economy increases). There was no description of the approach to doing so while not compromising electrode and membrane life. It would be good to demonstrate that graphite plates can be cost-competitive and meet the necessary power density for the chosen application.
- No technical approach was addressed, even though reviewers asked about one. The project set a goal to achieve 25,000 hours of life and to be able to operate at temperature of 100ºC or more. Usually, higher temperatures accelerate catalyst degradation, so the goal is very challenging. It is very important to address how to technically achieve it. Achieving 68% of peak efficiency is also challenging. The cell voltage should be very high. This also accelerates catalyst degradation.

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.

- The project did not start until April 12, 2021. Therefore, the team did not have much time before the DOE Hydrogen Program Annual Merit Review to progress through the planned work. However, CTT has received external funding and is working with FCHT to define the specifications of the e-turbo. CTT has started working on material selection, turbine wheel design, and e-turbo concept design, which will be used in designing the e-turbo for this project.
- The project has not yet been started, but progress is being made on overcoming the hurdles to start the project.
- This project has not started yet; there are no accomplishments. This should not be rated yet.
- The project is just commencing, so it is impossible to rate, so it was given a middle grade, which should be ignored.
- This is a new project.

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.

- The list of collaborators looks to be excellent and includes Cummins, FCHT (Hydrogenics), ANL, W.L. Gore, Dana, and CTT. It is reasonably clear what each of the collaborators will provide to this project, although it is unclear whether W.L. Gore will provide just the membrane or the full MEA; it seems as though Hydrogenics will do the full MEA using the W.L. Gore membrane. The project should discuss whether and how the national laboratories' Million Mile Fuel Cell Truck (M2FCT) can provide assistance to this project and should leverage the characterization abilities of M2FCT, especially related to durability.
- The team is composed of institutions with appropriate resources and expertise. The project's partners and their roles in this project are shown in slide 11. There is no mention of the feedback process from the team to W.L. Gore and Dana. Preferably, no membrane and graphite BPP development work will be done at W.L. Gore and Dana. The team will just be buying their commercial membrane and BPP, and the researchers will develop/optimize their own MEA and stack at FCHT.
- ANL, W.L. Gore, and Dana are capable partners for modeling, membranes, and BPPs, respectively. However, it is not clear who will lead electrode development, a key element, given the plan to run at elevated temperature and cell voltage to meet the thermal and efficiency targets.
- It is difficult to judge the level and effectiveness of collaboration on a project that has not yet started, but the plans and connections to industrial, government, and commercial partners are there.
- Catalysts could be one of the key enablers, but it is unclear who within the collaboration roster is supposed to develop these. This funding opportunity announcement was the development of the domestically manufactured stack. It is not clear these partners can meet this requirement.

Question 4: Relevance/potential impact

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

- This project is relevant to DOE Class 8 HD targets. Success of this project will make a significant impact in proving the MEA/stack and balance-of-plant design parameters for commercial trucks, bringing fuel cell truck technology close to commercialization. This project plans to address all four critical barriers of DOE Class 8 HD targets, i.e., operation ≥100°C, 68% peak efficiency, 25,000-hour life, and $80/kW system cost in mass manufacture. It would have been nice if the team had disclosed an estimated cost of high-temperature MEAs and graphite BPPs. Both high-temperature membranes and graphite BPPs are expensive. Although the team claims it will meet $80/kW system cost in mass manufacture, it would be nice to know the present cost of membranes and BPPs and what commercial volume has been considered for mass manufacture cost modeling.
- The project clearly aligns with the high-level DOE research, development, and demonstration objectives (durability, efficiency, cost, and higher-temperature operation).
- Industry-led definition of requirements and evaluation of fuel cell technology are critical to advancing DOE Hydrogen Program (Program) goals and objectives.
- A domestically supplied and manufactured stack fits directly into the Program and applies to the emphasis on HD trucks.
- High-temperature operation is a goal, but it is very difficult to achieve the lifetime with this condition. The project needs to qualitatively show how much benefit can be obtained if this goal is achieved.

Question 5: Proposed future work

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

- The proposed work for this project, on slide 13, is logical. The team has thought through all necessary elements of the project and has spelled out the future work appropriately. The team should consider cost analysis as a part of a go/no-go decision in fiscal year (FY) 2022 during short stack testing. Assembly of the short test stack should give the team an understanding of the potential cost of a 100 kW commercially viable stack. Given the cost of the stack components, cost analysis and potential stack cost information will be crucial to ensuring the viability of meeting DOE's cost target during the development phase of the project.

- The catalyst is an important enabler for high-temperature operation and long lifetime, but no task was defined for the catalyst.
- Without a clear approach, it is difficult to comment on the future work. The basic steps are there but lacking in detail, with no mitigation pathways cited.
- As the project has not started yet, it is premature to comment on whether the outlined work builds on past work and will overcome barriers. In general, the plan and decision points make sense.
- The project has not yet started, but the project plan seems solid at this stage.

Project strengths:

- This is a well-crafted project with capable partners with necessary expertise for executing this project. The team's decision to use a commercial membrane and BPP to assemble a 100 kW stack and focus on stack manufacture/optimization is the correct course of action for this project. By involving ANL for high-temperature MEA/stack system development, the team has made a good choice. The decision to have the go/no-go decision in FY 2022 after short stack testing is the correct decision. The short stack testing should give the team ample information to understand the viability of meeting DOE Class 8 HD targets for a 100 kW stack.
- The project collaborators look to be in place for a successful project. The project plan looks to be in place to cover the needed portions to make a stack that can meet DOE HD targets.
- The strength of this project lies in combining industry-led requirements evaluation (real-world requirements and use case) with the technological advancements needed at both the fuel cell and system levels.
- The strengths are a group of capable partners (ANL, W.L. Gore, and Dana).
- It is hard to see any project strengths.

Project weaknesses:

- The project has not started yet, and not much information is available to understand the weaknesses. Overall, proposed steps of the project seem to be logical, and it is too early to point out any weakness of the project.
- Hydrogenics is in Canada, and a barrier was mentioned about that and legal agreements. Clearly, as Hydrogenics is making the MEA from the W.L. Gore membrane, this is a critical issue that has to be solved. The project seems to lack any fundamental aspect related to understanding performance and material degradation. The team should work with the national laboratory M2FCT consortium to fill that gap. The DOE targets are being based on end-of-life targets; the researchers discussed little to no information about how they will meet the durability target of 25,000 hours, which is especially critical with the discussed use of high operating temperatures.
- This project may be overlooking some of the high-temperature failure modes, which might prevent the project from meeting its lifetime objectives.
- The project needs to quantitatively clarify the benefit of this challenging goal (high-temperature operation and long lifetime). Also, the project needs to show the technical approach and identify what the enabler to achieve the goal is. The peak efficiency goal needs a clear technical approach to achieve it. The principal investigator's answers to reviewers' questions were not enough.
- A weakness is the lack of a clear approach to achieve the aggressive requirements that often have non-complementary traditional solutions (high temperature and high cell voltage to meet heat rejection and efficiency versus low temperature and low cell voltage generally used to achieve durability).

Recommendations for additions/deletions to project scope:

- The team should consider adding stack cost analysis as a part of the short stack development work and report to DOE during the go/no-go decision.
- Expanding the collaboration to M2FCT to understand MEA performance and durability should have a positive impact on the project.
- The project will demonstrate a full stack plus turbo-compressor. However, to assess durability, it is recommended that a complete system, including humidification, be designed and tested, as these play integral roles in demonstrating system durability.
- It might be beneficial to add a collaboration with some of the catalyst durability projects or groups to ensure success.
- It is highly recommended that the project re-define the goal and address the technical approach.

Project #FC-338: Domestically Manufactured Fuel Cells for Heavy-Duty Applications

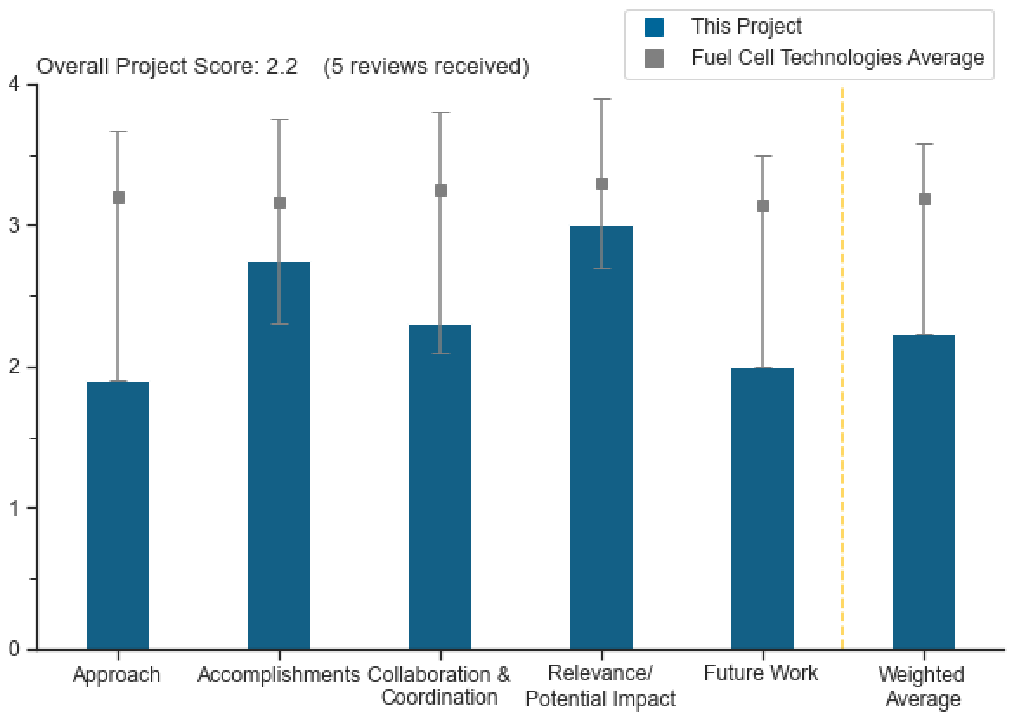
John Lawler, Plug Power Inc.

DOE Contract #	DE-EE0009248
Start and End Dates	5/1/2021 to 5/1/2024
Partners/Collaborators	Argonne National Laboratory
Barriers Addressed	• Durability (25,000-hour life) • Cost ($80/kW) • Performance (limited platinum group metal loading 0.2–0.3 mg PGM/cm^2)

Project Goal and Brief Summary

Plug Power Inc. (Plug Power) is working with Argonne National Laboratory (ANL) to develop a heavy-duty (HD) fuel cell stack that is a suitable drop-in replacement for diesel engine applications. If successful, this project will enable high-volume production of bipolar plates (BPPs) and 100 kW modular stack systems to create a reliable and efficient stack with improved durability, cost-effectiveness, and performance.

Project Scoring



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

Note: This is a new project in 2021. 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 **1.9** for identifying and addressing objectives and barriers and for project design, feasibility, and integration with other relevant efforts.

- The approach of this project is to develop new plate and seal designs for a 5 kW short stack and the U.S. Department of Energy's (DOE's) target 100 kW stack for testing. This stack is a single-power unit intended for use in multi-stack systems with a single balance of plant (BOP). The team is also taking an approach to determine the appropriate seal testing method for a commercial production environment. The material choice for sealing elements and BPPs is appropriate for large-scale manufacturing.
- The approach is logical. Starting with a 5 kW substack for an eventual 100 kW stack is reasonable. The 100 kW stack may still be considered small for a Class 8 truck application but is large enough to be representative. Because of proprietary information issues (and as Plug Power is not under contract yet), there is a lack of detail regarding specifics of the design approach, particularly for the BPP and coating. This is both understandable and frustrating.
- The presentation focuses on two aspects: leak testing and design for manufacture automation. While these are important and possibly overlooked aspects, details of the team's approach to meeting the most difficult and high-level targets (cost, durability, and high-temperature operation) are lacking.
- This pre-start project presentation lacks detail. No information is provided related to the type of materials used. Future presentations should include details related to the membrane electrode assembly (MEA), membrane, catalyst, gas diffusion layer (GDL), BPP material/coating, and/or the expected operating conditions. At present, it is impossible to know whether this design will meet DOE targets or be suitable for HD applications.
- The presentation and the slides do not provide enough information to be able to discern the approach or approaches that will be used to meet the performance targets and metrics. The presentation lacks information about the MEA, membrane, or catalyst improvements that will be investigated to provide the durability needed for the HD application. While BPPs and seals were discussed briefly, information on BPP or coating material strategies and seal materials or strategies was also lacking. Some of the information appears to suggest that by optimizing BPP materials and operating conditions, durability and performance targets can be met with current MEA designs, but that is considered highly unlikely. The principal investigator (PI) appeared to be unaware of previous DOE work looking at seal materials and the accumulated materials compatibility database that contains information on seal materials.

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.

- The project started on May 1, 2021. Therefore, the team did not have much time to work on the project and report notable progress in the June 2021 DOE Hydrogen Program Annual Merit Review (AMR). The team has completed the workplan for the 5 kW stack demonstration. The workplan is sound and covers necessary elements of redesigning the stack based on the new BPP design.
- This project has not started; thus, there are no accomplishments or progress. The score is irrelevant because it is too early to evaluate the project when no funding has been spent.
- The project is just commencing, and rating it is impossible, so it received a middle grade. This section should be ignored.
- The project has not yet officially started, so there are no accomplishments yet.
- The project has not started.

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 constitutes two partners: Plug Power (prime contractor) and ANL (subcontractor). Plug Power is constructing a test bench capable of running the accelerated stress tests proposed by the Million Mile Fuel

Cell Truck (M2FCT) consortium at the 100 kW system power level to generate performance data for modeling the HD fuel cell stack at ANL. The collaboration meets the project objective. Both Plug Power and ANL have the required resources and expertise needed for conducting the tasks proposed in the project.

- Collaboration with ANL for system modeling is prudent and appropriate. There is substantial experience in the community and among vendors regarding design and fabrication of metal plate and MEA systems. Plug Power's desire to create its own proprietary system is understandable; however, additional collaboration with vendors or consultants that have that development experience already would offer substantial risk reduction benefits. The technology transfer associated with the alliances with Renault Group and SK Group appears to be significant to the project, and future presentations should mention any overlap and collaborative efforts where relevant.
- The only collaboration listed is with ANL, on a limited work scope. Most of that modeling is not even related to the actual stack manufacturing, which is the title of the project. The team comments that the project will follow M2FCT-developed accelerated stress tests.
- This project has not yet started. It appears that Plug Power is collaborating with ANL, but it is not clear whether that is through M2FCT or separate from that. Collaboration with the M2FCT consortium would be beneficial.
- No collaborations are listed.

Question 4: Relevance/potential impact

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

- The team's goal is to develop and demonstrate a 100 kW fuel cell stack with domestically sourced components, which will serve as a single-power unit intended for use in multi-stack systems with a single BOP and which has potential to meet the 2030 system targets for HD fuel cells. This project will focus on key technical barriers, such as durability, cost, and performance, that are key to meeting DOE's 2030 interim goal of 25,000 hours of stack/system life for the Class 8 tractor–trailer application. Instead of researching many fuel cell components, the team has taken on the task of using existing material and BOP systems and maximizing the stack performance through BPP flow field and sealing optimization to reduce leakage-related performance loss. The project has direct relevance to DOE's developmental goal, and the project's success will make significant impact in driving the progress of hydrogen fuel cell applications in larger vehicles. The team is also concentrating on a closely coupled stack design and manufacturing system that has the potential to meet production volumes for Class 8 trucks and marine and rail markets.
- The project tracks the Hydrogen and Fuel Cell Technologies Office (HFTO) targets and planning near-verbatim. Thus, the project is well-aligned regarding relevance.
- It appears that the overall goals align with high-level DOE targets and goals, though the lack of information provided makes it difficult to discern whether lower-level targets and goals at the component level are aligned. The project plans to integrate BPP design and manufacturing and move Plug Power's sourcing of BPPs from outside the United States to the facility inside the United States, which aligns with the goals to strengthen the U.S. supply chain.
- A U.S. manufactured fuel cell stack for HD applications aligns with the DOE Hydrogen Program. Not enough information was provided to understand whether the project will be able to meet DOE targets.
- The project listed the primary DOE research, development, and demonstration objectives as stated project targets, but little can be gleaned without any detail or approach to how to achieve these targets (and only some comments on sealing, automation, and optimizing operating conditions, followed by BPP design, to align with these). The project needs to detail the approach and priorities.

Question 5: Proposed future work

This project was rated **2.0** for effective and logical planning.

- The project has not started yet. The team is in final contract negotiations with DOE. The team's immediate goal after the start of the project is to begin developing a 5 kW short stack.

- Since the project has not yet started, everything is considered future work. The slide on future work refers only to developing the 5 kW substack. Additional information should have been described.
- Only plans for the first budget period are described. The plans that were presented left several points unclear: why the flow analysis is delayed until after the 5 kW stack testing, whether there will be initial computational fluid dynamics/finite element analysis modeling and design, and what the target operating conditions are. The presentation is lacking in any other details on the proposed and future work.
- This project has not started yet, so the information is all about future work. The presentation discussed only BPPs (but did not give any information), gave a list of possible gasket materials, and discussed leak testing. Essentially no information is provided about the MEA, which is the critical component in determining whether the project can meet DOE targets.
- The project provided insufficient detail to allow reviewers to determine what the proposed future work is and how it addresses the barriers.

Project strengths:

- The team's knowledge and experience is the strength of the project. Both Plug Power and ANL have performed hydrogen fuel cell research and development for decades, and combined, they have experience to drive such a project to develop understanding in BOP and system levels, which is necessary for developing HD fuel cells for larger vehicles.
- Plug Power has substantial internal capability and is well-suited to developing a U.S. domestic product. Teaming with ANL is a good alliance. The selection of 5 kW and 100 kW demonstration power levels is appropriate.
- Plug Power is a leading manufacturer of MEAs and fuel cells for industrial applications. The proposed work to bring BPP manufacturing in-house could be beneficial to U.S. manufacturing and the fuel cell supply chain.
- Plug Power has a history of focusing on what is needed to enter the market with a saleable product.
- Plug Power has commercially sold fuel cells for applications such as forklifts. That work may translate to making HD fuel cell stacks, but as to whether it actually will, the team did not present enough information to enable that determination.

Project weaknesses:

- The team is working on domestic fuel cell manufacturing. It is not clear whence the MEA will be sourced; the reviewer's understanding is that W.L. Gore's commercial MEAs are manufactured outside the United States. The team did not provide the MEA sourcing information.
- The selection of metal plates with coatings for a 25,000-hour lifetime is not well-supported or -explained. The selection of 0.769 V/cell at end of life, even though specified by DOE, is an aggressive value that may not be consistent with cell degradation and a 25,000-hour lifetime.
- There are no listed collaborations and no details of plans. Having neither gives little confidence that the aggressive targets can be met successfully, and there is no method to gauge whether the project has potential even to advance from the state of the art.
- Details regarding Plug Power's proposed work and approach to improving durability and addressing performance were lacking. It was not possible to determine what the approach is or how the team proposes to address the challenges associated with fuel cells for HD trucking.
- The project did not provide information to clarify what the researchers are going to develop during the project. There was a total lack of information presented in the AMR. It is basically impossible to know the weaknesses of this project because the presentation did not give any information.

Recommendations for additions/deletions to project scope:

- The end-of-life cell voltage is unreasonably high for this project and should be reconsidered. Consideration should be given to the metal plate/coating and MEA experience of the past from other groups. Perhaps this is being done, but as presented, it seems a Plug Power development is in isolation. The involvement and technology transfer to and from partners Renault Group and SK Group should be more clearly defined.

- Plug Power is being funded by the federal government for this project. Part of being funded by the federal government is being reviewed at the AMR. Projects are supposed to be additive to the HFTO portfolio, and this project is currently not. Plug Power presented no information that would help any other project, whether government, academic, or industrial. This project needs to provide a minimum amount of information so that it can be reviewed to enable understanding of whether it can meet future DOE targets.
- The PI needs to provide more information about the approach. The project is receiving public money, and the PI needs to provide more information so we know the researchers are using it wisely and that their plans address the relevant barriers.
- The team should clearly delineate an approach and plan to meet the high-level targets set forth in the DOE Hydrogen Program.

Project #FC-339: M2FCT: Million Mile Fuel Cell Truck Consortium

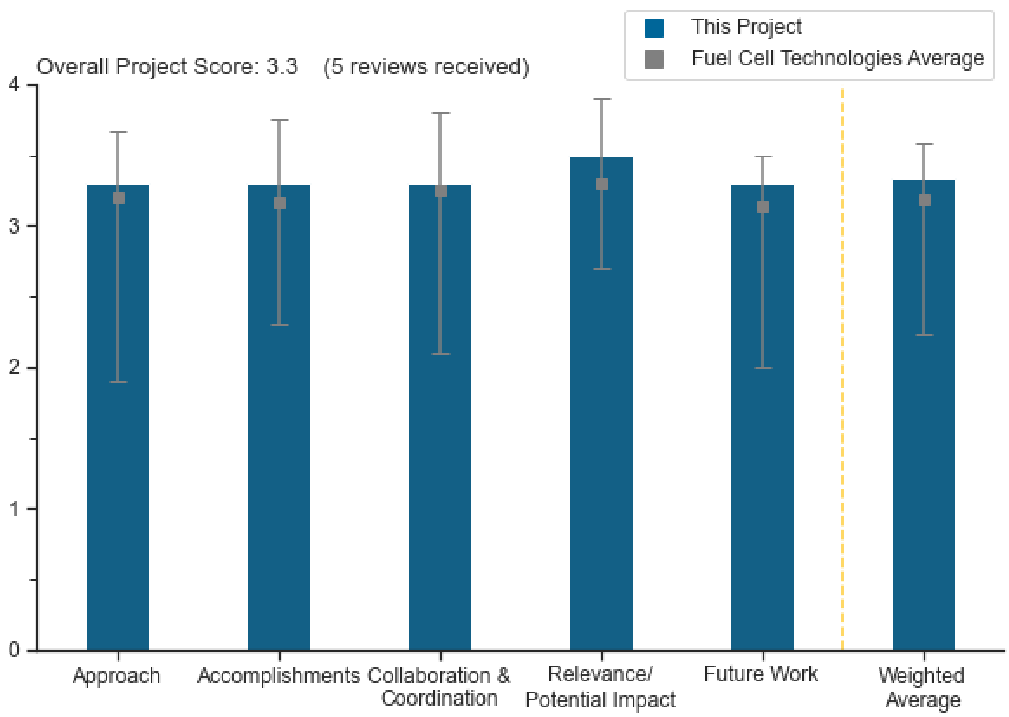
Rod Borup and Adam Weber, M2FCT

DOE Contract #	WBS 1.5.0.402
Start and End Dates	10/1/2020 to 9/30/2025
Partners/Collaborators	Los Alamos National Laboratory, Lawrence Berkeley National Laboratory, Argonne National Laboratory, National Renewable Energy Laboratory, Oak Ridge National Laboratory, Pacific Northwest National Laboratory, Brookhaven National Laboratory
Barriers Addressed	• Durability: 30,000-hour lifetime • 72% peak efficiency • $60/kW fuel cell system cost

Project Goal and Brief Summary

The project team is working to construct fuel cells that provide 2.5 kW of power per gram of platinum group metal (PGM) after a 25,000-hour-equivalent accelerated durability test. The purpose is to create durable and efficient fuel cell designs suitable for adoption by the heavy-duty vehicle (HDV) market.

Project Scoring



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

Note: This is a new project in 2021. 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 objectives and barriers and for project design, feasibility, and integration with other relevant efforts.

- The proposed approach is to combine different activities toward common objectives, including materials development, component integration, component/cell durability, and system analysis. The proposed workflows for each segment appear relevant for achieving the project targets. The organization of the project should ensure good communication between the different members. The "team-of-teams" approach is composed of well-recognized experts in the field and, as it has already demonstrated its relevance in several U.S. Department of Energy-funded projects, appears to be the most effective way to work on these important challenges. Because most of these national laboratories are/were also participating in complementary projects such as the Fuel Cell Consortium for Performance and Durability (FC-PAD) and the Electrocatalysis Consortium (ElectroCat), this consortium's approach will facilitate the capitalization and valorization of the results in profit of this new project. Even if many stack components are mentioned, it is clear that membrane electrode assemblies (MEAs) will be at the heart of the activities. The approach of developing an MEA and then validating it in industrial stacks is interesting if there are regular loops during the project. Concurrently developing MEAs and bipolar plates, however, usually leads to the best performing stacks.
- The shift to heavy duty (HD) increases the relevance of polymer electrolyte membrane fuel cells (PEMFCs). This consortium brings together a broad range of expertise at the national laboratories. They have already announced a few funding opportunity announcements (FOAs) demonstrating their commitment to bringing in outside expertise from industry and academia. The proposed approach is a logical integration of all members and resources.
- There is not much to say here. This looks really great. One thing that the team might want to keep in mind is that there does not appear to be any evidence that the accelerated stress test (AST) is truly indicative of million-mile performance. The team plans to use the same AST but move to 90,000 instead of 30,000 cycles. However, HDVs are often operated much differently from light-duty vehicles (LDVs), with much of the lifetime spent operating at near-constant load (driving on highways and not in cities, towns, etc.). Surely, this can be reflected in the AST.
- The overall approach is sound, with the "team-of-teams" approach providing comprehensive and integrated activities to address the challenges. The project correctly places a high emphasis on durability, benchmarking, and models for Year 1. The high-level overview slides on the approach are all well-thought-out and show a very comprehensive approach, i.e., in building goals and in the interactions, length and time scales, advanced characterization approaches, etc. The advanced computing approach will be very useful if the researchers are able to create the digital twin. The monolayer studies for the membrane seem to transition into the catalyst as well. However, the membrane goals do not appear to be clearly stated. The multivariable parameter sensitivity analysis will be valuable to drive experimental studies, once validated. While the system-related analysis is very useful and should be done, it may be better for it to report out as a separate project. This team excels at activities related to fuel cell MEA development, and that is where the researchers should keep their focus.
- A team of national laboratory scientists is working together to attempt to make long-endurance fuel cells. While the concept is good, the approach is risky. The researchers have no controls engineering, the key to fuel cell management. Rod Borup reminded everyone that U.S. Department of Energy laboratories predicted poor stability of Pt-Co electrocatalysts, but their reverse engineering of a Mirai showed the catalyst intact because of controls. Controls are really cheap compared to materials invention and characterization. The team should really be focused on understanding when materials are robust and when they are at risk, and the project should publish the parameters. Control engineers can then use the information to write controls. The team organization is confusing. For instance, it is unclear why a leading expert in microscopy (Cullen) is being reassigned to a communications job. It would make more sense for researchers to do research and then use a communications specialist to communicate. The group is also going to be soliciting proposals, so the team will be bogged down in contract awards and program reviews. In theory, this might make sense, but the organization chart looks very top-heavy and loaded with scientists in contract and management jobs. The presentation was surprising in terms of the lack of new topics being studied. There are tens to hundreds of papers on sulfate anion adsorption. It is unclear why this is being

studied again. A thorough literature review would be adequate. Likewise, there has been significant work on Pt on oxides. The key issue was that the rotating disk electrode results did not match the MEA results because of hydrophobicity issues. The key to this work is water management, not repeating synthesis and characterization approaches from 10–15 years ago. The only thing somewhat new is the machine learning. This will be very tough because it is hard to model an unknown. Machine learning is very much based on labeling, meaning that you need to know what is most important. Perhaps a neural net would be a better approach. There needs to be more focus on water management and humidification at start-up and shutdown.

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.

- All of the work shown appears to be well-thought-out and focused appropriately on elucidating key mechanisms and/or improving materials and designs. There is so much work shown that it is difficult to go into detail on any one subject to fully explain work being completed. Not all charts have full legends, and the reader can only assume the meaning, e.g., the right-hand plot on slide 33, with three PtCo/C values, showing the lowest change in power density although it is stated to have the highest performance loss. Adding the hydrophobic layer to the gas diffusion layer is an interesting concept to reduce cation migration. There are also data in the additional slides showing some good performance increase. Novel materials work (catalyst, ionic liquid) is showing good promise. Some of this work also appears to be reviewed under separate projects; therefore, it is not clear how much is done under Million Mile Fuel Cell Truck (M2FCT). It seems that this may be just an integration of concepts from other projects.
- The consortium is still in its early stages. However, the team has made progress on identifying critical target metrics, impacts of aging on critical materials, baseline testing, and funded FOAs.
- Many interesting results have been obtained during the first year of the project (analysis of operating conditions, performance and efficiency, durability measurements at projected HD loadings within development of HD-related ASTs, catalyst inks to performance and catalyst layer analysis, and some material developments). It appears that, considering the HDV operation conditions and defining accordingly, the different AST protocols appear as a critical point to which high attention has to be paid, in particular, when adapting AST protocols developed for LDVs. The system model considers that there will be no humidifier anymore. It is unclear whether this has been validated by the HDV manufacturers for all HDVs.
- The project is very new. The team seems to be focused on organizing the project.

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 organization chart presented appears well-adapted to ensuring an efficient and fluid collaboration between the project partners. Special attention has to be paid to the regular exchanges with industrial partners implementing the new materials and components in their stacks. Regarding outreach activities, organization of regular webinars with international institutions is encouraged, in particular on the AST protocols.
- So far, this is extremely laboratory-focused, which is good, given the capabilities at the labs. The industry interaction is clear here. There is limited room for fundamental academic work.
- There is great integration of resources and personnel.
- Significant collaborations and collaboration mechanisms are well-described.
- The DOE team is attempting to work with new organizations and even award some funds to new contributors. Unfortunately, the management team is an old group of colleagues. It would have been very pleasing to see a new name, someone with knowledge of heat transfer or manufacturing or packaging (surely the DOE laboratories have this expertise). With the same group of colleagues, there will be little room for innovation. Industry is poised to have a small voice.

Question 4: Relevance/potential impact

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

- Hydrogen-fueled mobility applications will become more and more important during the decade. In complementarity with batteries, the hydrogen solution is the most relevant for the need of high payload, high autonomy, while keeping the flexibility of today's thermal engines. Therefore, developing specific components adapted from LDV developments for HDVs is of utmost importance. This project will provide results of high impact for the industrial development of HD hydrogen transportation, in line with DOE targets.
- There is great potential for this consortium—not only in bringing together disparate resources at multiple national laboratories but also in highlighting targets, publishing benchmarks, and developing drive cycles and AST protocols for HD. There is great promise here not only to develop new materials but also to aggregate and promote best practices for testing and assessing new materials. One thing that has been missing in materials development is a standardization of how those materials can be integrated and accurately tested in PEMFCs. This consortium has the opportunity to develop the needed standardized synthesis, integration, and testing of new materials.
- The project is highly focused on advancements required to meet HDV and medium-duty truck targets.
- The project is extremely well-aligned.
- At this point, the project is poised to have low impact because the team is a group of long-term colleagues who are doing work that they have been doing for years. Yes, durability is very important, but the focus is not clear. Durability is challenged with start-up/shutdown conditions of stacks, uneven fuel distribution, and poor humidification. So much is systems-level-dependent. It is not clear how the basic research is being poised to affect real-world durability.

Question 5: Proposed future work

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

- The proposed work covers many topics in line with the project objectives. Most of the presented results relate to the electrodes. As durability is key for HDVs, it is expected that membrane stability will also be covered in the coming years. As, up to now, stacks using metallic bipolar plates have not demonstrated durabilities of tens of thousands of hours, the impact of bipolar plate corrosion products either affecting the contact resistance or polluting the MEA should be investigated. Shutdown/start-up testing is foreseen. Freeze start testing may also be considered, even if occurrences should be lower than for LDVs.
- There is no need for comments here. The project is outstanding.
- Proposed work is well-defined and critical for the advance of HD PEMFCs.
- The proposed future work is appropriate. The project should include a manufacturability and cost assessment with delivered MEAs.
- As noted above, the proposed work has largely been done in the past at DOE laboratories and elsewhere.

Project strengths:

- Really, everything is a strength. The team is great. The institutional structure is very good. The project is tackling the correct problems.
- The team and collaborations, baseline testing and standardizations, and incorporation of academia and industry through FOAs are all strengths.
- The project has a very strong team, excellent analytical capabilities, and the latest approaches, e.g., machine learning. There is excellent collaboration.
- The "team-of-teams" approach grouping well-recognized experts is a real strength of this project.
- The goal of the project, understanding how to increase the durability of fuel cell MEAs, is important.

Project weaknesses:

- Even if considering all stack components in the scope of the project, it appears to be very focused (perhaps too much so) on MEA developments for HDVs. Connections with ongoing DOE-funded projects on MEAs, membranes, and stacks seem to be put in place, and it will be important to ensure regular effective exchanges.
- The project may be too large and all-encompassing for effective management.
- The project is not tailored to guide controls designs, and there are few new ideas in the project/presentation.

Recommendations for additions/deletions to project scope:

- The DOE researchers need to do a comprehensive literature review of much of the proposed work and determine whether the proposed experimental work needs to be completed, or whether controls parameters can be recommended on existing publications and patents. The team must include one or two people from outside the DOE fuel cell community who can offer a fresh perspective; otherwise, this appears to be just a jobs program. The researchers should not be in administrative roles. If possible, the researchers should do a review of where their prior projects successfully affected the fuel cell industry (licensed patents), take those lessons learned, and try to replicate in this project.
- The project is so large and includes so many different activities that it feels like a shotgun of work. It may be better to separate out some of the work for separate reviews, rather than review all the work together. There could still be a high-level review of the consortium/approach in one normal-length review session. The team might consider studies to support freeze-start operation, such as material and MEA properties under sub-zero conditions.
- The scope of the project is quite large and ambitious. The challenge will be to cover all the foreseen items and to ensure effective coordination.

Project #FC-341: Advanced Anion Exchange Membrane Fuel Cells through Material Innovation

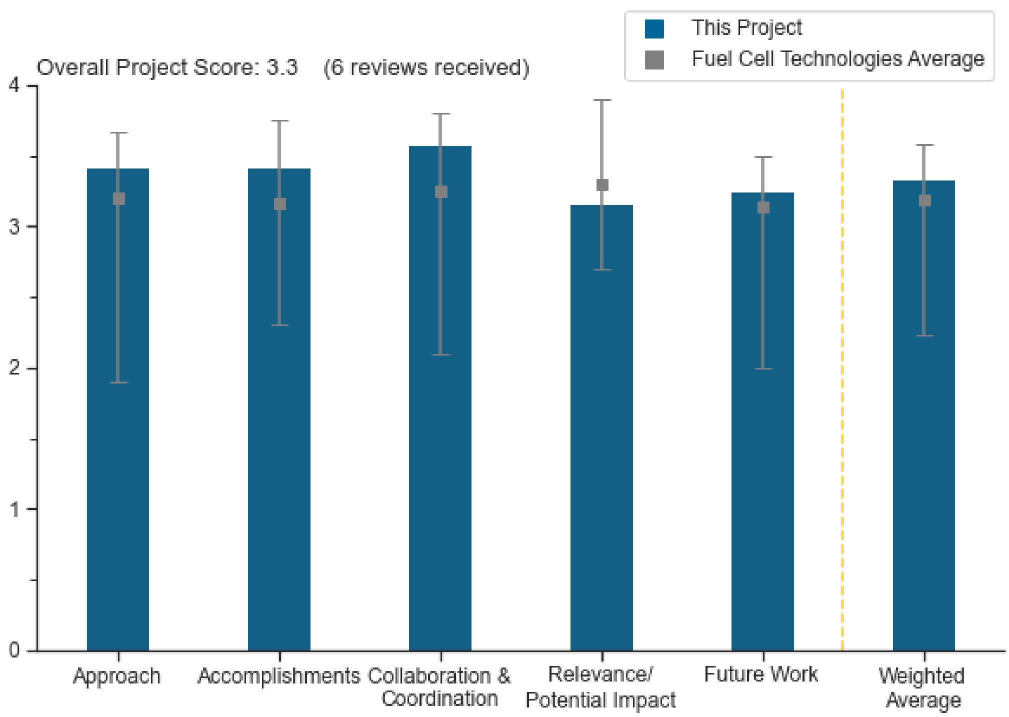
Yu Seung Kim, Los Alamos National Laboratory

DOE Contract #	WBS 1.3.0.440
Start and End Dates	8/1/2020
Partners/Collaborators	Sandia National Laboratories, Oak Ridge National Laboratory
Barriers Addressed	• Cost • Electrode performance • Durability

Project Goal and Brief Summary

The project team is working to develop advanced anion exchange membrane (AEM) fuel cells (AEMFCs) through materials innovation and deliver 50 cm^2 membrane electrode assemblies (MEAs) that meet Hydrogen and Fuel Cell Technologies Office (HFTO) performance and durability milestones. The goal is to improve AEMFC performance and durability while working toward a commercially viable system. The project could also provide scientific knowledge of AEM stability that can be transferred to other AEM-based technologies, such as an AEM electrolyzer. Los Alamos National Laboratory (LANL) is collaborating with Sandia National Laboratories (SNL) and Oak Ridge National Laboratory (ORNL) on this project.

Project Scoring



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

Note: This is a new project in 2021. 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 objectives and barriers and for project design, feasibility, and integration with other relevant efforts.

- The overall approach is sound. This is exactly the kind of work that a national laboratory should be doing – understanding the fundamental principles of how AEMs work and how to improve them. Reading Dr. Kim's papers and listening to his presentations are always enjoyable. They are very informative. The problem is that there are at least a dozen ionomers competing for the AEM crown, and we need a core understanding of these ionomers, their structure, test methods, etc. to be able to (1) understand how they work and (2) develop more advanced polymers and electrochemical systems. This work is therefore extremely valuable.
- The ideal approach would make the ionomer/membrane insensitive to CO_2 and would focus on non-PGM-based MEAs. However, the proposed work is also very valuable, assuming the oxidant fuel cell feed will be CO_2-free air. An aryl ether-free backbone, along with stable fixed cations, appears to be the logical way to go.
- The approach is good and very clear.
- The approach addresses durability and water management, which are major barriers for AEMFCs. The approach to use high-platinum-group-metal (PGM)-loaded catalysts in MEAs may lead to conclusions that are not applicable to PGM-free or low-PGM catalyst layers.
- This project focuses on AEM materials for the AEMFCs. The proposed AEM materials show some novelty. However, the AEMFC has been progressed significantly in the past using PGM catalysts and hydrogen–oxygen. More focus should be on AEMFCs with PGM-free catalysts and hydrogen–air (with trace amounts of CO_2) performance. In addition, LANL AEMFCs have been funded before. This project is the development of new AEMs (from ORNL and SNL). It is not clear why LANL does not use the previous AEM materials but focus on electrode design.

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.

- There has been a systematic analysis of polymer degradation for a variety of structures, and there is new insight gained by the work. We need much more of this type of work for AEMs to emerge as viable options for major electrochemical applications.
- Most strategies have been implemented in the approach, such as saturated hydrocarbon, polyfluorene, polyphenylene, and styrene-based ionomers, typically with thickness of 37 microns, water uptake of 76%, and good ionic content (International Electrotechnical Commission [IEC] 1.7).
- This is a relatively new project, but a significant number of valuable data have already been collected. The team's findings regarding MEAs with asymmetric electrodes are particularly interesting.
- New development of different ionomers for the anode and cathode (asymmetric ionomers) provides another dial to turn to help with water management and should prove useful. Development of a homogeneous method for functionalization of polyphenylene polymers provided significant improvements in membrane durability in ex situ testing. While ex situ membrane stability has been increased substantially, stability of ionomers still appears to be an issue, with significant degradation over hundreds of hours of operation in an MEA.
- The project is progressing well.
    - Durability is still variable (slide 12). This needs to be addressed. Based on the data, there is also a need to refresh the cell with KOH periodically. This is also a limitation, as it is not needed with some other systems and is not generally practical.
    - Loading of 0.6 mg/cm^2 is not really "low." It is suggested that this low-loading target be moved to be in line with Thompson et. al and have a final target of 0.125, or maybe slightly above, such as 0.2.
- This is a new project. Its current progress is fair.

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.

- Collaboration between partners is effective. Yu Seung's collaboration with others in the AEM community is apparent through his publications and work co-leading the membrane working group.
- Dr. Kim has reached out to a wide range of institutions—not just his contractual partners in the project. This reviewer knows from firsthand experience that he keeps them regularly informed of his progress.
- The project work is spread out over LANL, ORNL, and SNL, with well-defined roles and responsibilities to optimize meeting all targets.
- This project has twelve collaborators and two partners; it is an excellent example of teamwork.
- It is good to have three approaches for the polymers, but it is clear that one institution is producing the best results so far.
- This project is just a collaboration with national laboratories. An industrial participant will be helpful.

Question 4: Relevance/potential impact

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

- AEMs are sort of a holy grail. They offer the potential for significant cost reduction in catalysts and membranes for many critically important devices such as fuel cells, electrolyzers, and flow batteries. A fundamental understanding of AEM properties and performance in situ is key to improving these materials for these applications. It is the next big breakthrough as hydrogen technologies and the hydrogen economy emerge commercially. This will make a huge impact.
- The project aligns well with the DOE Hydrogen Program objectives. The work on polyphenylene-based AEMs can be of great value.
- This project is targeted at AEMFC technology, which is a potential alternative to PEMFCs. Over the past decade, the performance of AEMFCs has been greatly improved, but research activity is still needed to improve durability, increase carbonation tolerance, decrease gas flow rate, and implement low-PGM or PGM-free catalysts. This project focuses on AEMFC performance under hydrogen–air (CO_2-free and CO_2-containing) and low RH conditions.
- This project is partially relevant to DOE fuel cell research, development, and demonstration objectives.
    - Provided significant progress in reducing Pt loading has been made in polymer electrolyte membrane fuel cells (PEMFCs), the development of AEMFC gas becomes less meaningful.
    - If AEMFCs will be pursued, the integration of AEM materials with PGM-free catalysts is a must and more significant.
    - Water management of AEMFCs is more complex, making the application less viable.
- The project is clearly targeting the AEMFC goals well, although the "low PGM" in the project is not really low PGM.
- AEM and ionomers are relevant to the HFTO goals, as these technologies have potential to reduce fuel cell and electrolyzer costs by reducing PGM content and enabling use of lower-cost bipolar plate materials. AEMFCs with high PGM loadings are not relevant to the HFTO goals.

Question 5: Proposed future work

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

- There is a clear, systematic approach to the work, as can be seen from the polymer variants studied sequentially. The presentation and data make sense.
- All the proposed materials synthesis work and MEA and fuel cell fabrication and testing are well-aligned with the project goals.
- Noteworthy future work includes the following:

  - AEM and ionomer optimization (ORNL, SNL, and second-year LANL [end-of-year milestone, July 31, 2022]) AEMFC performance: 0.65 V at 1 A cm^2 on hydrogen–oxygen at 80°C
  - Demonstration of target performance and P = 150 kPa durability, (LANL) AEMFC durability: <10% voltage degradation after 1,000 hours, 1 A/cm^2 operation (total PGM loading = 0.6 mg/cm^2)
  - Synthesis and characterization of fluorinated first-year go/no-go milestones (July 31, 2021): poly(fluorene) ionomers (LANL), poly(phenylene) AEMs with technoeconomic analysis and piperidinium cationic groups (SNL) and polyolefinic AEMs via ring-opening metathesis polymerization (ROMP) (ORNL).
- Using fluorinated polymers is not a good idea. Avoiding fluorination would be a huge bonus over polymer electrolyte membranes. It would drive down cost and environmental impact. It should be noted that not all of the performance issues require a new polymer. It is suggested that the team also do work on the electrode composition and MEA methodology.
- Future catalyst layer ionomer work should focus on PGM-free catalysts, as their hydrophobicity and water transport are significantly different from those for high-PGM-loaded catalyst layers.
- The focus of future work is still on AEM materials development. More focus should be on electrode design using PGM-free catalysts.

Project strengths:

- The project has good fundamental data on a variety of different candidate polymer structures. These data are very useful. There is also good systematic testing of candidate polymer performance. There are clues in the test results that suggest new directions for polymer development. The project has very large, wide-ranging direct and indirect partners, which is a strength. The project is designed inherently to share knowledge widely and encourage technology commercialization.
- This is a well-planned and -executed project. Better AEMFC membranes/ionomers are needed, which necessitates a better understanding of the unique features of AEMFCs as compared to PEMFCs. The project team consists of world-class experts, so significantly improved membranes, MEAs, and new knowledge are highly likely to become available through this project.
- The project has a very strong membrane team, the project's AEM materials are innovative, and LANL has good small device integration experience.
- The principal investigator and collaborators have been leaders in the AEMFC field and have determined the main AEM degradation modes.
- This project has smart approaches that target defined, known issues.
- This project demonstrates good teaming.

Project weaknesses:

- This is a very difficult problem. Membrane durability is a critical and complex issue. There are many approaches and many angles that have to be investigated. The problem is simply too large for this one research project. There must be ongoing research and expansion of the overall effort to address this most critical of issues. A breakthrough on durability could literally catapult the hydrogen economy forward.
- It would be good to bring in one or two things that are not related to just the polymer, but this is minor and should not be considered a fundamental project weakness.
- There has been great progress in AEM performance, but AEMs continue to be speculative and necessitate a focus on effects of CO_2 and non-PGM catalysts and catalyst durability issues. Making a great laboratory demonstration on hydrogen and oxygen with high platinum loading will not make a commercial fuel cell or electrolyzer.
- The use of catalysts with high PGM loadings in the MEAs provides electrodes with substantially different water transport characteristics not representative of the desired end product and may lead to conclusions that do not apply when a PGM-free catalyst is finally used.
- The project still focuses on hydrogen–oxygen (no CO_2 impact will be investigated). The project also still uses PGM catalysts (no PGM-free catalyst is planned). AEM mechanical durability needs to be examined.

- The project does not promise CO_2-resistant membranes/ionomers. The cost of the proposed materials is also not addressed.

Recommendations for additions/deletions to project scope:

- The project should expand funding, increase the overall scope of this research, and add more scientists; this effort should be elevated to a Manhattan-Project-style effort. The project is addressing a critical issue, a critical problem that must be solved.
- The project should add PGM-free catalyst integration with membranes and ionomers. Also, the project needs to investigate the impact of CO_2 on the membrane and mitigation strategies.
- The project should have more focus on effects of CO_2 and non-PGM catalysts and catalyst durability issues.
- Testing with PGM-free cathode catalysts should be added.
- The "low loading" issue, noted above, should be addressed.
- The project should include cost estimation of the proposed materials.

Project #FC-342: Advanced Ionomers and Membrane Electrode Assemblies for Alkaline Membrane Fuel Cells

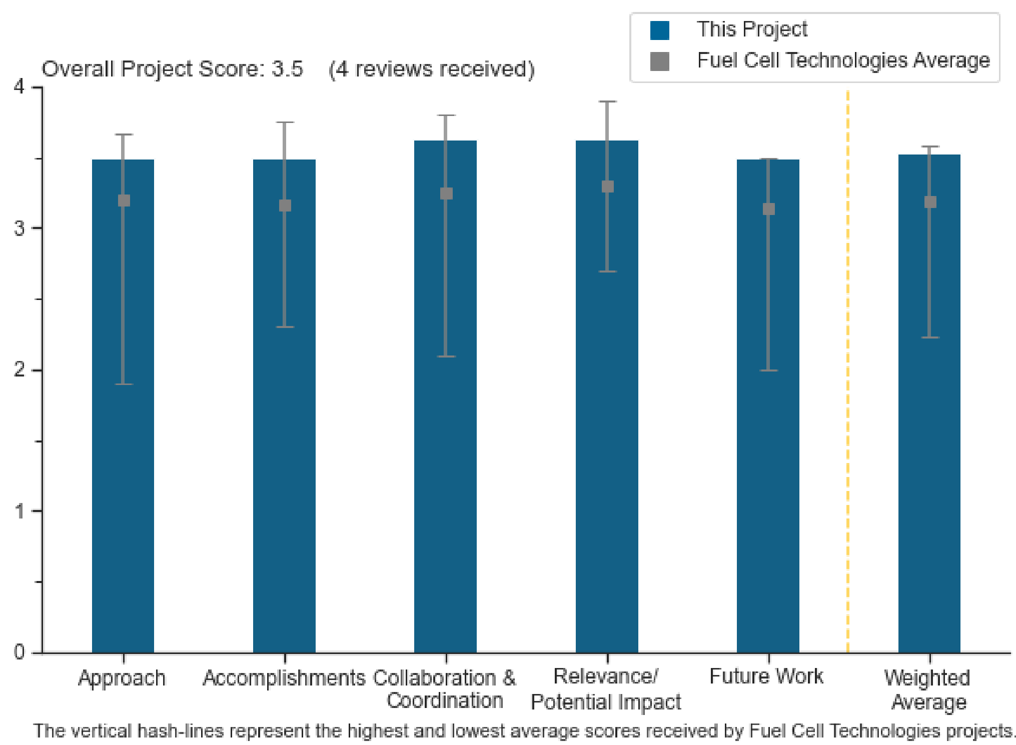
Bryan Pivovar, National Renewable Energy Laboratory

DOE Contract #	WBS 1.3.0.540
Start and End Dates	10/1/2020
Partners/Collaborators	Lawrence Berkeley National Laboratory, Oak Ridge National Laboratory, Colorado School of Mines, 3M
Barriers Addressed	• Durability • Cost • Performance

Project Goal and Brief Summary

This project aims to improve the performance, durability, and operating window of alkaline membrane fuel cells (AMFCs) and enable them to function with electrodes with minimal platinum group metal (PGM) content. If successful, the project will produce AMFCs with durability equal to polymer electrolyte membrane (PEM) fuel cells, addressing greenhouse gas emissions, air quality, and environmental justice concerns while enabling domestic energy competitiveness.

Project Scoring



Note: This is a new project in 2021. 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 objectives and barriers and for project design, feasibility, and integration with other relevant efforts.

- This research looks at a set of critically important problems involving membrane, electrode, and catalyst performance and durability. It is a very broad subject, but it must be investigated and understood in order to support a breakthrough in cost for many different components of the hydrogen technology stream and, therefore, hydrogen economy. The overall approach is sound. The National Renewable Energy Laboratory (NREL) obviously has the core competence and the tools to do this work. The overall project appears to be well-sequenced, and the data presentation is logical and systematic.
- The project has a very nice set of materials and approaches.
- The project focus on performance and durability is appropriate. The project approach of using techniques that have been successful in determining where degradation is happening in PEM systems (such as electrochemical impedance spectroscopy and voltage loss breakdown) should prove effective. There is a disconnect between how alkaline electrolyte membrane fuel cell (AEMFC) systems are marketed as potentially low-cost options that can utilize PGM-free catalysts and how this work is being performed (both this project and the one at Los Alamos National Laboratory [LANL]). It appears a significant effort is being put into optimizing these very high Pt loading electrodes that do not appear to be economically viable for applications of interest. Water management, which is key to AEMFC operation and likely plays a large part in durability, will be quite different for high-Pt-loading and PGM-free catalyst layers (and likely for low-loaded Pt catalysts). PGM-free catalyst hydrophobicity is different from that for PGM-based catalysts, and ionomer interactions with PGM-free catalysts with atomically dispersed active sites are different from those with Pt-based catalysts with Pt particles. It is not clear that learnings from high Pt-loading catalysts will be transferable to fuel cell systems that may be of interest commercially (with PGM-free or low-PGM-loading electrodes).
- The proposed approach involves a combination of alkaline electrolyte membrane polymer development, membrane electrode assembly (MEA) fabrication, and fuel cell testing. It is not clear where the focus is, but all the aspects of the proposed work are of great interest of the AEMFC community and the U.S. Department of Energy Hydrogen Program (the Program).

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 researchers very systematically looked at improving the performance of the electrode(s), then looked at improving the performance and degradation of the membranes, and then looked at improving cell design and engineering. This is precisely the order in which the work should be done, and thus the team was able to demonstrate some clear improvements and, therefore, accomplishments in overall system performance. The researchers moved the needle forward. They can make progress toward a PGM-free system and identify good membrane candidates for cell engineering.
- The project has begun work integrating PGM-free cathode catalysts. It is important to work with PGM-free catalysts rather than the very high-PGM-loading MEAs being studied, as the PGM-free catalysts are integral to achieving the promise of lower costs for AEMFCs. The project has identified cathode catalyst degradation and particle growth as an important degradation mode. Given the timing of the start of this project and COVID-19 restrictions, the researchers have managed to accomplish quite a bit. They have a long way to go to meet the identified target for the fourth quarter of 2021: 100 mA/cm^2 at 0.8 V in hydrogen–air for loading of 0.2 mg PGM/cm^2. Most of the work has been with much higher loading (four times the target or more), and data shown with the PGM-free cathode appears to be a long way from the target.
- So far, the team is hitting its targets. It would be nice to see the project be more aggressive with its cell-level low-PGM target. The team has also improved its demonstrated durability.
- Impressive results of the electrode performance improvements have been reported, mainly involving ink composition and processing conditions. There is interesting work on powder processing versus dispersion processing. Not exactly sure what was “unique” and “advanced” in the reported degradation diagnostics.

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.

- NREL's team of industry partners is impressive. There is widescale coordination and very high-quality information exchange across all stakeholders with interest in the work.
- There is a very impressive portfolio of collaborators.
- Collaborations are apparent in the water transport study. Collaborations could be expanded to include LANL and others, particularly in the PGM-free catalyst area.
- Right now, it seems that most of the work is at NREL. It would be helpful if the Colorado School of Mines work were better highlighted. It is also less clear in the presentation what 3M is doing. However, the role that ORNL can play is clear.

Question 4: Relevance/potential impact

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

- This is the holy grail of the hydrogen economy. If the project is able to move to non-PGM electrodes and low-cost membranes for critical devices, such as fuel cells and electrolyzers, the cost breakthrough (and opportunities) would have a dramatic impact on the future trajectory of this industry and the global energy picture. This is a crucial project that is very relevant and important, with very high potential impact.
- Improvements in performance and durability and the introduction of non-PGM catalysts into AEMFC MEAs are perfectly aligned with the Program goals. The proposed work, if successful, might lead to significant advancement of AEMFC technology.
- The work is very focused on DOE goals.
- AEMFCs have promise to provide lower costs and may help reach cost targets if they can reach performance and durability targets. This work is directed at meeting the performance and durability targets. Whether reduced costs can be achieved is likely dependent on using PGM-free catalysts. The majority of the current work has been with very high-PGM-loading MEAs, which are not relevant to the Fuel Cell Technologies subprogram. Alkaline membrane systems may be more applicable in electrolysis and reversible fuel cells.

Question 5: Proposed future work

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

- The proposed future work addresses the appropriate barriers and is moving toward low-PGM and PGM-free catalysts.
- The only concern is that the work on segmented cells, which is obviously important and novel, should be pushed back, and the project should focus more on the first three components of the research. The first three components are critically important, and those problems are so big, important, mysterious, and not fully understood that all resources in this project and beyond—i.e., a Manhattan-Project-style program with vast resources—should be applied to study them and come up with a real pathway forward.
- Future work is well-defined. It may occur that switching to non-PGM catalysts will make some data obtained with Pt-based catalysts obsolete.
- It seems that the project will move in the right direction, but the listed items are pretty vague.

Project strengths:

- There is an excellent research plan and excellent team; this combination will most probably lead to very important findings. Inclusion of modeling is also a great idea. The future belongs to computers and to the software.

- The overall competency of the research team and access to important testing and analytical tools are impressive. The analytics and diagnostics effort is excellent and very important and informative. Real progress has been demonstrated, which suggests the project is well-managed and systematic.
- The project team's capabilities in fuel cell diagnostics and methods to determine where degradation is occurring are strengths.
- There is a good group of researchers at NREL using their capabilities well.

Project weaknesses:

- The new cell designs, while being very important, should take a back seat to fundamental work on the electrodes, membranes, etc. Really great material options that work and that are durable are needed first, before we look at new cell designs.
- The current focus on high-PGM-loading catalysts is considered a weakness.

Recommendations for additions/deletions to project scope:

- The planned move to PGM-free and low-PGM catalysts will be beneficial.
- The project should drive down the PGM loading for the fuel cell later to be more in line with the work performed by Simon T. Thompson, et al., and reported in the *Journal of the Electrochemical Society* (167 084514, 2020).
- New cell designs, while very important, should take a back seat to fundamental work on the electrodes, membranes, etc. Material options that work and that are durable are needed first, before we look at new cell designs.
- There needs to be more focus on non-PGM catalysts and inexpensive ionomers/membranes, such as the possibility of the elimination/minimization of the effect of CO_2 on AEMFC performance.

Project #FC-343: Fiscal Year 2020 Small Business Innovation Research Phase II: Improved Ionomers and Membranes for Fuel Cells

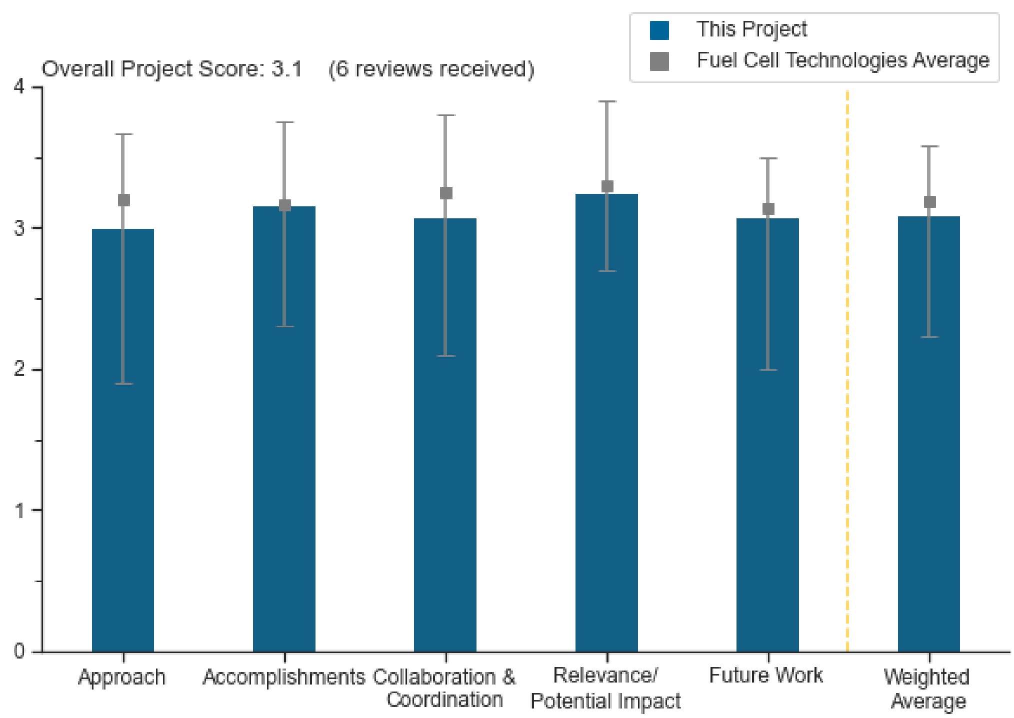
Chris Topping, Tetramer Technologies, LLC

DOE Contract #	DE-SC0019980
Start and End Dates	7/1/2019 to 8/23/2022
Partners/Collaborators	Commercial Fuel Cell Systems Manufacturer, National Renewable Energy Laboratory
Barriers Addressed	• Stable, enhanced performance across a range of temperature and humidity conditions • Reduced gas crossover (hydrogen and oxygen) • Reduced costs

Project Goal and Brief Summary

The project objective is to develop cost-effective, high-performance, durable polymer electrolyte membranes (PEMs) that do not use perfluorosulfonic acid (PFSA) as a base. Tetramer Technologies, LLC (Tetramer) will work with the National Renewable Energy Laboratory (NREL) and a commercial fuel cell systems manufacturer to produce new cost-effective PEMs for integration into existing and future commercial fuel cell devices. These membranes will meet U.S. Department of Energy and industry performance and cost targets that are not attainable with currently available materials.

Project Scoring



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

Note: This is a new project in 2021. 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 objectives and barriers and for project design, feasibility, and integration with other relevant efforts.

- Tetramer makes the polymer and works with customers to do the testing. The approach of using block copolymer with hydrophobic and hydrophilic parts with lower projected cost is great. Unfortunately, the presenter could not reveal the customer and did not provide too much detail on the approach to testing and evaluation. Typically, the team plans to do 500-hour tests (not enough details). If these are accelerated stress tests (ASTs), then it makes sense, but 500 hours of simple fuel cell testing is not enough to evaluate these membranes. It would also be useful to provide more details on what the elected degradation mechanisms for these membranes are and evaluate them in ASTs that accelerate those mechanisms.
- The block copolymer approach employed by the Tetramer team has the potential to advance hydrocarbon PEMs to be competitive with PFSAs. There is an understandable need to provide generalized structures until all the intellectual property is secured. However, it is difficult to evaluate the project merits based on blocks "A" and "B" and to compare against other hydrocarbon systems. The principal investigator addressed this issue during the poster discussion session. Low humidity performance and chemical stability have been traditional barriers to hydrocarbon membrane success, and the Tetramer team should prioritize these aspects if this approach is to be a genuine advance over previous work.
- The overall concept underlying the research project is important and sound. Based on the slides provided, it was hard to tell what polymer development program was being pursued. The test data were helpful and clearly an important part of the work and presentation.
- The team is preparing several new hydrocarbon ionomers based upon block copolymer designs for use as PEMs in fuel cells. Much of the information about the chemistry and block copolymer characteristics (such as morphology, molecular number, and block lengths) was missing because that information is confidential and proprietary. The central premise of the project asserts that the tunability of the different blocks can give rise to desired properties that maximize ionic conductivity while also providing robust mechanical properties. There are no plans to assess chemical durability of the new ionomers. It is unclear whether the team is selecting chemistries that can tolerate reactive oxygen species or whether free radical scavengers will be incorporated to ensure PEM durability.
- The systematic approach to membrane synthesis is valuable and has yielded interesting initial results, but there is not enough focus in the early evaluation stages on durability (the focus seems to be primarily on performance, which is important but not sufficient).
- The approach is very difficult to determine; there were not enough technical details to determine whether the approach is viable.

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.

- All of the new hydrocarbon PEMs achieved <0.02 ohm cm^2 at 80°C. Some, such as membrane Type 2, showed less than 0.01 ohm cm^2. This was a key metric in Year 1 of the project. PEMs Type 1 and Type 2 also showed hydrogen crossover less than 2 mA/cm^2. The new block copolymer ionomer PEMs have been characterized, but these data were not shared. Polarization curves were also generated, but the cell voltage data were not included. Overall, the project team has accomplished the goals for Year 1 of the Phase II Small Business Innovation Research project. No information has been provided yet on whether the team could hit the membrane cost (high volume) of $\$20/m^2$.
- DOE goals are clear and are clearly laid out for this research project. The work is obviously targeted at meeting those goals. It was not clear what polymer was being developed. The fuel cell test data did, however, suggest that the work was moving in the directions laid out by DOE.
- It is difficult to determine the amount of progress; the polarization curves do not even include voltage numbers on the y-axis. The team could have solved this by applying a benchmark such as the NR-211 in the polarization curves, but the presenter showed only the area-specific resistance (ASR) comparison to

NR-211. Nafion™-based catalyst layers married to new non-PFSA membranes have traditionally become an issue.

- The progress and accomplishments to date are reasonable for this stage of the project. Preliminary data at 100% relative humidity (RH) shows good progress; however, low-RH conductivity and fuel cell performance are necessary to be competitive with PFSA incumbents. Ionomer swell and water uptake data would be helpful for comparing to other technologies.
- The project has successfully made four different ionomers, and the progress in the synthesis sections is good. However, it is very difficult to evaluate this when few data were presented on the performance. Polarization curves alone are not very informative for evaluating a membrane. The ASR data were excellent, but they were presented only for 100% RH and 80°C. Moreover, the pol-curves had no correlation to the ASR. Additional characterization, including electrochemical impedance spectroscopy, should be presented to show feasibility. Crossover measurements should be presented under various conditions and directly compared to, say, NR-211. It is unclear under what conditions the <2 mA/cm^2 crossover was measured. A better comparison of these membranes to a baseline NR-211 or a state-of-the-art chemically and mechanically stabilized PFSA membrane would be great.
- Progress to date has yielded interesting results that address project objectives (performance characteristics of block copolymer membrane), but some barriers (durability) will not be adequately addressed by performance alone.

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.

- NREL is a great partner. The "leading" fuel cell developer's name was not disclosed.
- Collaboration with NREL looks to be well-coordinated, and NREL is a good partner in this project. The "commercial fuel cell systems manufacturer" is a curious choice. This reviewer is not aware of another instance in which a partner identification is kept secret or of the business advantages of this secrecy. It is to be hoped that the partner can be named at the next review.
- The quality of the level of coordination is difficult to judge from the poster materials but, at face value, seems to go in the right direction.
- There is a good collaboration between Tetramer and NREL. The commercial fuel cell systems manufacturer was not identified. It was unclear whether NREL or the manufacturer prepared the membrane electrode assemblies. Some of the tasks for this manufacturer and NREL seem to overlap. NREL has generated quality polarization curves with the PEM ionomers. However, the electrode composition is missing. It is not clear whether the team will use the new ionomers as electrode binders.
- The project has industrial partners to evaluate the membrane, and NREL is doing some durability testing. The project could engage the Million Mile Fuel Cell Truck (M2FCT) consortium to evaluate these membranes under the various DOE-specified ASTs, or the team could engage other industrial partners to get more data on these newer materials.
- The presentation did not even disclose the team members.

Question 4: Relevance/potential impact

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

- The impact of developing a lower-cost, higher-performing, higher-durability membrane is excellent.
- The Tetramer team is addressing the key DOE membrane targets. The team deserves credit for taking on the challenging work of exploring hydrocarbon membrane technology from the perspective of molecular architecture to address the traditional shortfalls of hydrocarbon membranes. The plan for systematically studying block copolymer architectures is unclear. While some degree of "trial and error" is necessary to establish feasibility, more systematic approaches need to be employed to optimize the system for maximum performance.

- The relevance of this project is excellent; low-cost membrane alternatives are essential to the success of the Program objectives. However, durability should be given more priority to align better with Program goals.
- The project team has hit the 80°C ASR and hydrogen crossover requirements for the Hydrogen and Fuel Cell Technologies Office multi-year research, development, and demonstration plan. There are not results for 120°C, which is important for heavy-duty vehicles, or for 30°C. Mechanical property and durability information for the new PEMs is also missing.
- It is difficult to determine the real potential here since very little technical information is given on the approach and the results.

Question 5: Proposed future work

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

- The future work plan proposed is described well and is certainly in line with what needs to be done going forward.
- Working on enhanced performance and durability is great. More details need to be provided on the types of tests that are to be run. In addition to open-circuit voltage (OCV) testing, combined RH cycling–OCV testing (similar to the DOE protocol for the combined mechanical–chemical stability or the General Motors highly accelerated stress test) should be performed.
- Future plans include ASR and fuel cell performance assessments at 90°C to 120°C, as well as durability tests and swelling tests. There are plans to supply materials to a fuel cell manufacturer for stack testing and field testing with a system.
- Durability has long been an obstacle for block copolymer structures and should be addressed earlier in the project (it should be part of a go/no-go decision point).
- The Tetramer team has outlined the necessary future steps in broad terms. However, the work around optimization is not well-defined. It is unclear whether this will be iterative or will include design of experiments or other approaches.
- The team did not show any durability data, and this is a typical issue with new materials. More durability data would help in the down-selection process. Building stacks prior to any of these durability and DOE target demonstrations on single cells is premature.

Project strengths:

- Hydrocarbon block copolymers are a good idea for realizing optimized PEMs that provide good proton conductivity and mechanical properties. Using cheap monomers as starting materials could hit the manufacturing cost target of <$20/m^2. The project team has demonstrated that several of the new PEM variants can meet both the conductivity and hydrogen crossover requirements. The team has a good collaboration with NREL and a fuel cell manufacturer to demonstrate actual performance in a few cell devices.
- The block copolymer approach to hydrocarbon PEMs is likely the most productive path to hydrocarbon membranes that are competitive with PFSAs. This project builds on past hydrocarbon technology and is poised to advance this technology. The Tetramer team has the polymer, membrane, and fuel cell expertise to successfully complete this project. Collaborations with NREL and a fuel cell manufacturer provide complementary expertise.
- Polymer synthesis ideas are good, and good materials to test seem to be provided. Testing data are sensible and provide encouraging outcomes.
- Polymer synthesis is the team's strength. It seems like good progress is being made on this front.
- The high-risk approach could yield a breakthrough.
- There is strong initial success with meeting performance objectives.

Project weaknesses:

- This is a difficult project to assess because much of the pertinent information is confidential and proprietary. This is understandable. The team has made good progress on ASR at 80°C and hydrogen crossover and has generated reasonable polarization curves. It was unclear what the electrodes were for the polarization curves (e.g., platinum-group-metal loading). The team needs to make a statement as to whether the new block copolymer chemistries can potentially tolerate reactive oxygen species.

- The exact polymer structure could not be determined, and any "weakness" in the project would be associated with the starting materials and the core technology being developed. It is therefore not possible to comment.
- The true test of hydrocarbon membrane technology is low-RH performance and accelerated durability. Until those are demonstrated, the potential of this project cannot be fully evaluated. Optimization tasks lack clarity. Variable space for di- and tri-block copolymers can get out of hand without a focused approach. More mechanical property data are necessary for full evaluation.
- Characterization of the fuel cell performance and durability of these materials is weak at this point. More emphasis needs to be placed in this area in collaboration with the existing partners, or new partners need to be added to advance this further.
- It is very difficult to determine because of the limited technical details given in the presentation.
- Failure modes should be addressed earlier in the project.

Recommendations for additions/deletions to project scope:

- The project is not clear on a pathway to commercialization and issues related to that, such as monomer cost, scale-up issues, toxicity of intermediates, and potential environmental impact. No real analysis appears to have been done on a pathway forward to the market.
- The project should perform the DOE-recommended AST protocols on these advanced membranes. The team should better understand degradation mechanisms in these membranes with fuel cell testing and develop ASTs to study these materials better.
- The project should include screening criteria at an earlier phase that address other barriers mentioned on slide 18.
- Mechanical property assessment and durability should be added as a project goal.
- More technical details should be given for reviewers to do a proper review of the project.