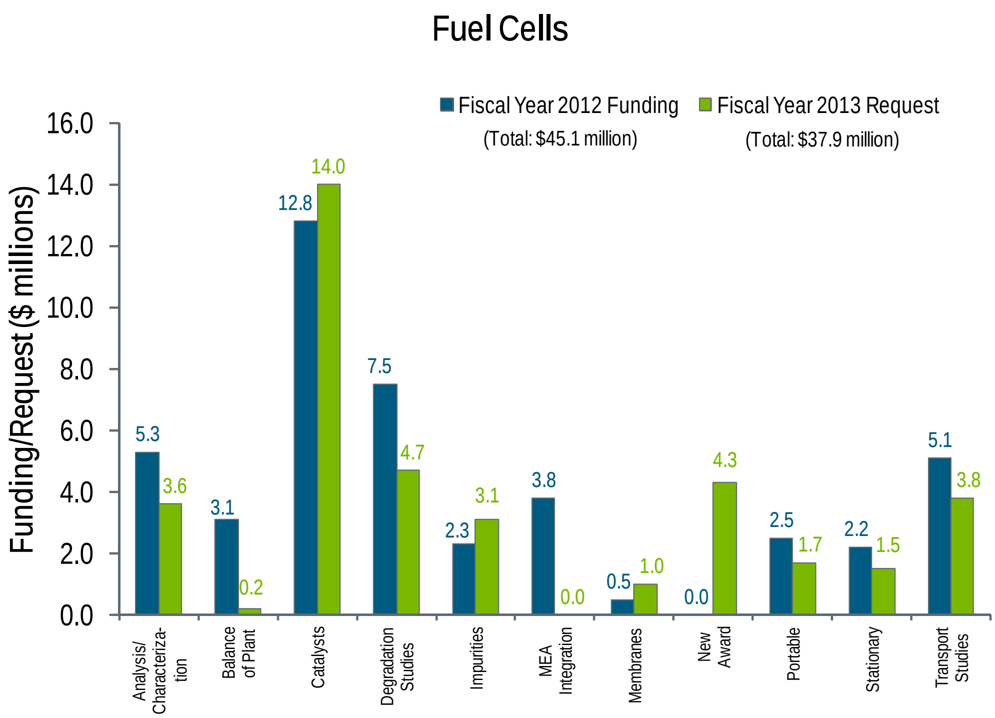
2012 — Fuel Cells
Summary of Annual Merit Review of the Fuel Cells Sub-Program

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

Reviewers commended the Fuel Cells sub-program for being well managed and clearly focused, with annual progress demonstrated. They noted that key challenges were identified, and good plans and a project portfolio were in place to address the key challenges. However, reviewers also commented that certain areas of the portfolio, such as membranes, need to be further strengthened. They also expressed concern about whether current funding levels would be able to support expanding the portfolio.

Fuel Cells Funding by Technology:

The Fuel Cells sub-program received $45 million in fiscal year (FY) 2012 and $38 million is requested for FY 2013. The sub-program continues to focus on reducing costs and improving durability with an emphasis on fuel cell stack components. Differences in the funding profiles between FY 2012 and the FY 2013 request are due in part to a mandate by Congress to fully fund projects in the year they are awarded rather than obligate future funds. In 2012, two new projects were awarded in membrane electrode assembly (MEA) integration and balance of plant (BOP), and the total cost of the projects was assigned to the FY 2012 budget.



Majority of Reviewer Comments and Recommendations:

At this year's review, 66 projects funded by the Fuel Cells sub-program were presented and 45 were reviewed. Projects were reviewed by between four and eight reviewers with an average of seven experts reviewing each project. Reviewer scores for these projects ranged from 1.8 to 3.6, with an average score of 3.0. Both this year's highest score of 3.6 and average score of 3.0 were similar to last year's highest score of 3.7 and average score of 2.9. The lowest score of 1.8 for all projects reviewed in 2012 was lower than 2011's low score of 2.0 for all projects reviewed in 2011. The lowest scoring project in 2012 was different than in 2011.

Analysis/Characterization: Seven projects were reviewed and received scores between 3.3 and 3.4 with an average score of 3.3. The reviewers praised the projects for significant accomplishments and progress in areas of system analysis, imaging, and development of testing protocols. Analysis/characterization projects were noted for accomplishments related to optimal platinum loading, providing unique imaging capabilities, and addressing performance and durability. Suggestions for future improvement included development of in situ liquid transmission electron microscope (TEM) techniques, including more extensive error and uncertainty analysis; description of the connections between imaging and reaching DOE's technical targets; and enhancement of the accuracy of the decay prediction.

Balance of Plant: Three BOP projects were reviewed this year, each receiving a score of 3.0. According to reviewers, the BOP projects may lead to a significant positive impact on the fuel cell system in terms of volume, weight, and maintenance. The reviewers expressed concern about the long-term durability of humidifiers and coolant systems being studied and suggested that projects should increase the understanding of degradation mechanisms.

Catalysts: The scores for the 11 catalyst projects ranged from 2.7 to 3.6 with an average of 3.1. According to the reviewers, catalyst projects have demonstrated significant improvements in catalyst cost, durability, and oxygen reduction reaction (ORR) activity. Reviewers expressed concern about some projects relying on rotating disk electrode (RDE) measurements to predict performance and suggested that performance of catalysts should be tested in MEAs. The reviewers suggested that more emphasis be placed on direct measurements of conductivity after exposure to relevant conditions for some projects.

Cross-Cutting: The four cross-cutting projects received an average score of 2.3 and ranged from 1.8 to 2.8. Reviews were mixed for cross-cutting projects. One project was praised for having a good mix of industry and academic expertise. For the other projects, reviewers expressed concern that some are either not relevant to the sub-program's goals or have a scope that is too broad. Reviewers suggested that the remaining tasks need to be reevaluated and prioritized.

Degradation Studies: The average score for the six degradation projects was 3.1. Scores ranged from 2.3 to 3.5. Reviewers praised the degradation projects for focusing on key parameters that affect fuel cell durability and for collecting and analyzing necessary data to highlight the key factors that impact voltage loss. According to the reviewers, the projects have good collaborations. Reviewers expressed a desire to see model results that more accurately address commercial fuel cell systems and for more evident correlation between model and experimental characterization.

Impurities: The two impurities projects reviewed received scores of 2.9 and 3.5. Reviewers commended the projects for collecting an extensive knowledge base about contaminants, generating a reliable model, and collaborating with stack manufacturers. Reviewers were concerned there is not sufficient data to make the models as reliable as possible. Reviewers suggested developing a better understanding of the causes of the performance losses and that tolerance limits for contaminants be recommended.

Membranes: Two membrane projects were reviewed, receiving scores of 2.6 and 2.9. Reviewers felt the projects had good concepts and approaches. There was some concern about the focus of one project on developing better membrane supports rather than better membranes. Reviewers also had concerns about the likelihood of success of one project.

Portable Power: The three portable power projects reviewed received an average score of 3.0, with scores ranging from 2.6 to 3.4. Reviewers felt the projects were relevant, with strong teams making good progress in water management and durability. Reviewers recommended increasing focus on developing membranes with high conductivity and reduced methanol permeability. The reviewers suggested increased interactions with original equipment manufacturers where relevant.

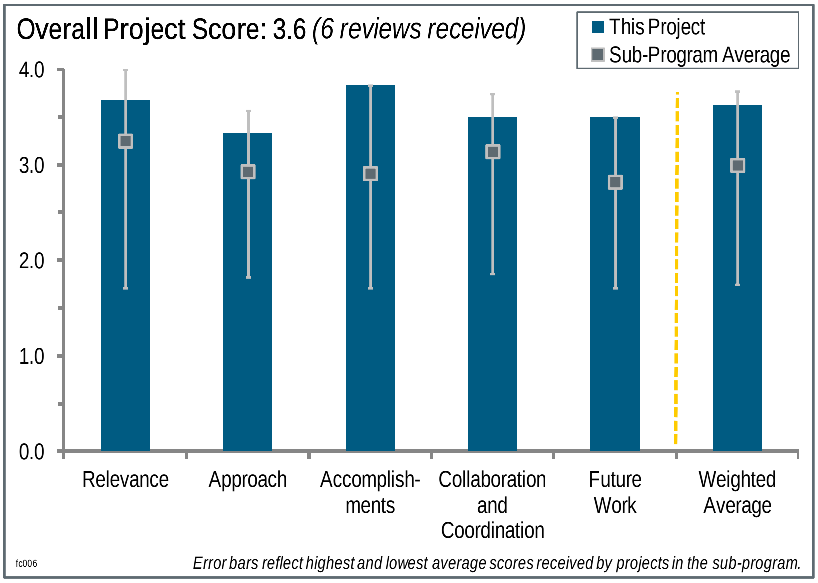
Stationary Power: The three stationary power projects reviewed received an average score of 2.9, with scores ranging from 2.7 to 3.4. According to the reviewers, the stationary projects are relevant to the sub-program's goals and have technically strong teams. Reviewers commended one project for making good progress towards addressing critical barriers of performance, durability, and cost. The two remaining projects received mixed reviews. One project was commended for making good progress, while reviewers were skeptical that the technical milestone could be met. Reviewers commended the final project for having a technically sound team, but were concerned about the lack of progress made.

Transport Studies: The four transport projects reviewed received an average score of 3.0, with scores ranging from 2.7 to 3.4. Some transport projects were praised for being well designed and for making good progress. Reviewers suggest adding studies of a larger variety of MEA configurations and of stack-level freeze cycle transients. For one project, reviewers expressed concern about lack of published results and recommended disclosure of the model in a peer-reviewed forum.

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

Radoslav Atanasoski; 3M

Brief Summary of Project:
The objective of this project is to develop catalysts that will enable polymer electrolyte membrane (PEM) fuel cell systems to weather the damaging conditions in individual fuel cells during transient periods of fuel starvation, thus making it possible to satisfy 2015 U.S. Department of Energy (DOE) targets for catalyst performance, platinum group metal (PGM) loading, and durability. This project will develop a catalyst that favors the oxidation of water over the dissolution of platinum (Pt) and carbon at voltages encountered beyond the range of normal fuel cell operation and beyond the thermodynamic stability of water.



Question 1: Relevance to overall DOE objectives

This project was rated **3.7** for its relevance to DOE objectives.

- By design, the program addresses prescribed targets and therefore is relevant to national goals.
- This project addresses challenges related to catalyst performance, PGM loading, and durability. It is intended to meet DOE 2015 targets. The project is fully aligned with DOE research, development, and demonstration objectives.
- This project is working to use material-based solutions to increase durability (as opposed to engineering-based solutions), which is what a DOE project should do.
- Catalyst durability under start-up, shutdown, and cell reversal is an important issue to address and is relevant to DOE's overall objectives. A more rigorous milestone for PGM and electrochemical surface area (ECSA) loss may be able to be achieved.
- This project addresses durability, one of the key barriers to the adoption of fuel cell technology, using a passive materials-based approach that could reduce system costs and is relevant to the DOE objectives of reducing the cost and increasing the durability of fuel cell systems. This project addresses degradation due to start-up, shutdown, and cell reversal, key degradation modes in automotive fuel cells.

Question 2: Approach to performing the work

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

- The approach is to add "depolarizers" to the catalysts that inherently limit potentials to values that preclude adverse side effects. This idea has considerable history and is a sound and proven concept.
- The approach section showed the employment of go/no-go decisions for specific quantified parameters, which were met and exceeded in the past year. The approach includes stepwise development toward the desired targets.
- The approach to use oxygen evolution reaction (OER) catalysts on cathodes to mitigate dissolution of Pt and carbon during start-up/shutdown and cell reversal is good. The use of these precious metals to prove the concept has been valuable. Exploring less-expensive, lower-loading, and perhaps more-durable OER catalysts would be the next challenge.

- The materials approach includes the use of ruthenium (Ru) and iridium (Ir) on the anode. The project must be careful in the use of these materials. Ru is known to crossover to the cathode and poison the oxygen reduction reaction (ORR). Ir is a relatively rare metal (worse than gold [Au] and Pt). The main approach appears to limit the loading of the Ir/Ru, which likely limits both issues and may also limit the long-term effect of the intended approach. The purpose of the material development is the protection of hydrogen (H_2) starvation and shutdown/start-up. All analysis is based on accelerated stress testing, including gas switching. It is not clear how the project intends to show that the catalysts are effective during H_2 starvation (which is often due to anode water flooding), and whether these materials suffer a degradation in activity from high water content.
- This project addresses degradation due to start-up/shutdown and cell reversal—key degradation modes in automotive fuel cells—and addresses key barriers of durability and cost. The project is designed to address key steps in the carbon corrosion mechanism at both the anode side and the cathode side, and it provides flexibility and an improved chance of success. The work at 3M is focused on addressing the durability of DOE target catalyst loadings.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.8** for its accomplishments and progress.

- This project shows promising results.
- Excellent progress was made toward improving catalyst stability.
- The objectives have been met and the enhanced durability is evident.
- The accomplishments and progress have been tremendous. The progress ranged from developing and improving SU/SD durability protocols for these types of catalysts to many rotating disk electrode and membrane electrode assembly (MEA) tests to show the durability of these catalysts. The scale-up of the catalysts to short stack and the evaluation of many stacks and MEAs by an independent institution are appreciated and impressive. To test Ru stability, the team should use CO oxidation.
- This project has already demonstrated significant improvements in catalyst durability to start-stop cycling protocols on the cathode, showing less than 10% loss in electrochemically active surface area on the cathode after 5,000 cycles simulating start/stop. This project has demonstrated a three orders of magnitude reduction in the ORR current at the anode and 200 cycles of 200 mA/cm^2 simulating cell reversal with anode loadings of 0.045 mg/cm^2 PGM with the upper potential remaining <1.8 V. This project has demonstrated improved tolerance to cell reversal in stack testing using the OER modified nanostructured thin film (NSTF) anode at an original equipment manufacturer (OEM). The OEM considers this anode to be “a promising MEA vehicle component” after evaluations including CO-tolerance and freeze-tolerance testing. 3M has demonstrated the importance of including cycles that bring the cell down to normal operating potentials during stop/start durability cycling.

Question 4: Collaboration and coordination with other institutions

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

- The collaborators are appropriate for the tasks.
- The partners are full participants and the project is considering expanding the role of an automotive OEM from a collaborator to a subcontractor, which will be beneficial in the future stack-level testing.
- This project shows a very good mixture of project participation from national laboratories, academic institutions, and industry. The clear distribution of tasks, regular communication, and data indicates an excellent collaborative effort.
- The collaboration with the Automotive Fuel Cell Cooperation (AFCC) is clear and excellent. It is unclear how much characterization is being done by Oak Ridge National Laboratory. It is entirely unclear what Dalhousie is adding to the project and if Argonne National Laboratory did the OER activity measurements, or what they exactly did for the project.
- The 3M projects tend to confuse collaborators with vendors, which is fully acceptable. One suspects that the real collaborations are within the 3M community, a rather unusually smart group of people who have a long history of invention and progress. The catalyst business is highly proprietary, as fuel cells become “car parts.” It is apparent that some cards need to be held close to the chest.

Question 5: Proposed future work

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

- Future works includes progressing toward meeting DOE 2015 targets combined with fundamental studies.
- It would be nice to include some H_2 starvation due to water-induced flooding experiments to see the durability effect of the anode catalysts and if they are stable over operation.
- Future work plans, including fundamental materials studies, engineering to understand interaction with other MEA components, and lowering PGM loading with enhanced durability, are all appropriate.
- Work evaluating the OER catalysts (especially on the cathode) on a dispersed catalyst, such as Pt on a durable carbon support (graphitized carbon?), would be beneficial.
- The results suggest that major program objectives have already been met. One wonders about the contamination implications with this approach. The PGM additives have seen earlier fuel cell applications (and electrolyzer). There is evidence that some additives tend to migrate away from the catalyst or move from the cathode to anode. Most likely they will end up in the membrane. The effects of these new "contaminants" need to be considered. Others who used similar formulations found that the "additives" tended to dissolve and migrate.

Project strengths:

- This project has a good team, a good approach, and good effort.
- The strength of this project appears to be the 3M and AFCC collaboration.
- The project concept, approach, accomplishments, and collaborators are all strengths.
- This is a comprehensive study leading from fundamental understanding over development to in situ full-size stack testing.
- The concept to attack carbon corrosion by interrupting several steps in the corrosion mechanism is a strength.
- The task successfully addressed key durability issues in PEM stacks. The work was excellent and showed clear direction and progress.

Project weaknesses:

- It is unclear what Dalhoussie is adding to the project. There is no information on other degradation mechanisms, such as H_2O_2 formation results, for these catalysts. The accelerated stress tests that are used to evaluate the catalysts (the DOE accelerated test protocol) have been shown to be useful, but they also need to include lower voltages (e.g., 0.1–0.65 V) to test the stability of the added metals.
- There was lots of information on the slides, making it difficult to follow; the oral presentation was difficult to understand as well. For example, the difference between nm1 and n1 cycles was unclear. There is so much color-coding and information that the key messages and significance of the data shown is lost.
- This project has no weaknesses.
- Nothing appears weak in this project.

Recommendations for additions/deletions to project scope:

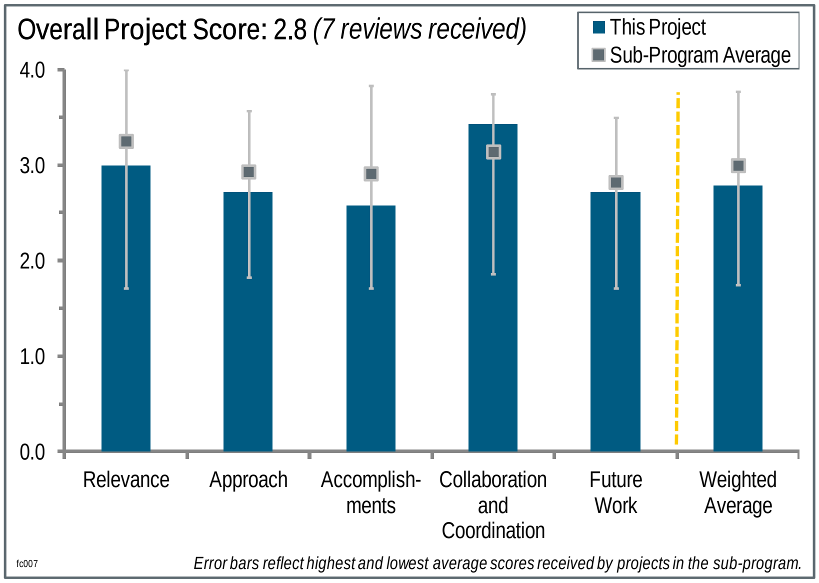
- The researchers should include a test that involves an H_2/air front on the anode, such as start/stop on a single cell. They should also apply this concept to dispersed catalysts on carbon supports.
- Work evaluating the OER catalysts (especially on the cathode) on a dispersed catalyst, such as Pt on a durable arbon support (graphitized carbon?), would be beneficial.
- Possible contamination should be explored. It would be interesting to compare the cost of this modification to the cost of improved stack management (control of anode and cathode gaseous constituents to preclude oxygen), and to control the stack potential using an external power source (battery).

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

Bryan Pivovar; National Renewable Energy Laboratory

Brief Summary of Project:

The objective of this project is to overcome the most critical barriers for fuel cell membrane electrode assembly (MEA) development: durability, cost, and performance. The project works to develop extended thin film electrocatalyst structures for increased activity and durability, and to explore the incorporation of these structures into robust, high-efficiency MEAs. The long-term goal is to incorporate materials with improved mass activity and voltage cycling stability into high-performance electrodes.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.0** for its relevance to DOE objectives.

- This project addresses the challenges of performance and catalyst durability in a novel and exciting way.
- This project addresses durability and precious metal thrift through novel geometries of platinum catalysts.
- This program is consistent with the DOE project aspects of higher oxygen reduction reaction (ORR) performance to minimize metal loadings. Further, it swings widely to look at a breadth of geometries, catalyst formation methods, support types, etc. The approach is good if sufficient iterations can be tested in a timely manner to glean insights.
- This project is directly relevant to DOE's goals of developing high-activity, durable catalysts for polymer electrolyte membrane fuel cells. The thin film catalyst approach has implications for the durability and performance (especially in improving mass activity) of catalysts that are distinctly different from traditional nanoparticle catalyst systems.
- This project is focused on trying to flush out new versions of one of the most successful approaches identified thus far for effective fuel cell cathodes, namely extended surface area catalysts. In that respect, it addresses the activity and platinum group metal (PGM) loading barriers. High-volume manufacturability and associated costs are apparently not being considered as a criterion at this early stage.
- The project objectives are relevant to DOE's overall objectives, but the project has too many unanswered questions. The deposition method is not finalized, surface areas are still too low, quantities are still at the gram level (10 grams is the most that will be generated by completion of the project), and there is no time to evaluate the MEA if it is even fabricated.
- The relevance of this work is to "focus on overcoming the most critical barriers for fuel cell MEA development," but the experiments are all on rotating disk electrodes (RDEs). There seems to be a major disconnect because RDEs have been shown to have a correlation with MEA results for platinum (Pt)-based/carbon materials, but there is no evidence that any of the RDE work for Pt blacks will correlate to MEAs. The flooded electrolyte condition of the RDE cell makes the whole Pt surface available for H+ and mitigates the role of the Nafion® interconnectivity, which most assuredly will play a large role in the Pt electrochemical active area in an MEA.

Question 2: Approach to performing the work

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

- This broad approach using several different methods of fabrication is well thought out. This late in the project, there should be some down-selection of the methods most likely to attract a potential supplier so that this work can be scaled up to stack testing in the near future if all targets are met.
- The approach is well founded and based on a strong team. While some of the more esoteric preparations were pursued in order to gain understanding, it may be time for the program to focus on the most promising spontaneous galvanic displacement (SGD), chemical vapor deposition (CVD) and work on making electrodes, the effort of which may be substantial. Using Pt black as a "surrogate" to understand electrode difficulties was a good approach.
- The efforts of this broad-based team are generating many new results and gaining experience discovering the issues with making effective electrodes with a wide variety of non-traditional catalysts. The wide variety of new catalyst forms the researchers have considered successfully demonstrate multiple ways to generate the high-specific activities of extended surface area catalysts.
- The sole focus on RDE is very concerning when the objective of the project is to understand the role of extended Pt structures (with and without carbon) in MEAs. With one year left, the researchers should move as quickly to MEA work as possible.
- The approach is good, although some of the processes are not realistic for manufacturing, but that is fine for understanding potential directions to pursue. The gains made in making optimized Pt black have been around for a number of years and there is no reason to believe further improvement will happen in that area, but they do make a good base case for comparison. Hopefully, more publications will come out that would be more about the catalytic activities than the process of formation. It would be best to test 3M nanostructured thin film (NSTF) to complete the breadth of geometries and types and plot that versus all the other catalyst types. The 3M NSTF does seem to be highly supported by DOE, so it should be a benchmark by which to compare other materials.
- It is not clear how the barriers will be overcome with respect to: (1) increased surface area; (2) a reduction in thickness using the atomic layer deposition (ALD) method (currently at 6nm, needs to be at 1–2nm); (3) the inability for the ALD method to scale up (continuing with this method does not seem reasonable); and (4) catalyst support not being selected. This project is not utilizing the model effort to make decisions/down-selection of catalyst particles, electrode structure, and performance. Very little information was provided on the modeling effort and its benefits, and data should include error bars when presented.
- Though the specific activities of CVD-grown Pt nanotubes are high, there does not appear to be a possible step forward for large-scale synthesis of these structures at volume. The mass activity of the CVD-grown structures is also low. The transition metal templates used for the SGD reaction still contain some of the template material post Pt deposition, and these are highly mobile cations (silver [Ag], copper [Cu]), which can strongly affect fuel cell performance. Images of ALD-grown films show films composed of small grains or Pt nanoparticles, similar in morphology to published images of the 3M NSTF catalyst system. If the growth mechanism for Pt on the chosen substrates is similar, is it unclear what the implications are for the ultimate required Pt thickness to reach high-specific and mass activities while maintaining aspects of thin-film durability.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- Extremely good progress has been made on all fronts. It would be good to understand more about residual Ag in the SGD method and how this may impact both performance and durability. It should be noted that Ag solutions may be classed as toxic, making process scale-up costly.
- The quality and understanding of the complexity of RDE work is transparent and outstanding, but the overall progress seems stunted by the low throughput of materials tested. The presentation communicated more about the means and types of catalyst than the ORR results.
- The researchers have accomplished much, but at approximately 60% complete, it would be good to see a MEA/polarization curve from the best to-date preparation just to get a real sense of progress. Working on

mixing carbon with the catalysts and exploring the impact of Nafion is good progress in preparation for the electrode-making part (MEAs).
- The researchers have made good progress demonstrating many varieties of extended surface catalysts that do indeed have the higher specific and mass activities they are looking for. They are also discovering many issues with learning how to disperse them effectively for both accurate activity characterization and electrode formation. They are also learning new information about how measured surface areas and mass activities may not be a simple function of particle size, as is conventionally believed. This is good information for the whole electrocatalyst community.
- Durability data is still not available, MEAs are not fabricated and tested to-date, and the quantities generated are still too low for meaningful verification. Yield was not discussed during review and it was unclear what the yield of substrates was. This project has investigated and synthesized many novel catalysts with several geometries and has shown good cyclic durability, but at this stage in the project (60% complete) the project needs to focus and down-select the palladium (Pd) deposition method, fabricate MEA, and conduct durability fuel cell testing.
- Batch-to-batch variability for synthesized materials is of concern. The activities reported for the Pt black work, though useful in comparison to the synthesized structures, are different than accepted values in the literature. This project has made positive progress toward improving specific and mass activities of SGD scaled-up materials. There is low total PGM activity of multistep Pd and Au materials. CVD grown materials have not improved catalyst mass activity.
- Excellent progress was made on looking at different Pt extended structures and finding the best materials. But the materials cannot truly be compared until they are cast into MEAs. It is not clear if having high, specific-activity Pt materials by a modified RDE process should be counted as a success, especially because they show that the materials have poor mass activity. If the DOE target for high-specific activity was eliminated and the only goal was mass activity, they would have few accomplishments. If the only goal was mass activity in MEAs, which really is the most relevant parameter, the project would have no accomplishments. This is not to say that the project has not produced good materials, the researchers just need a better practical measure of their accomplishments, which should probably be Pt mass activity in MEAs.

Question 4: Collaboration and coordination with other institutions

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

- This project has well-coordinated work with other organizations and institutions.
- This project appears to be very well coordinated among the very large team.
- A lot of institutions are involved with good collaboration on this project.
- There has been very good coordination between the teams. It appears some of the lessons from one track are being applied to others.
- The principal investigator (PI) clearly has good command of the activities and the contributions of the various collaborators. Perhaps due to the limited presentation time, it was not clear what the contributions were from three or four of the principal partners.
- An outstanding team has been assembled for this project, with the qualifications/strengths needed to accomplish the goals of the project. It is unclear how much interaction is taking place among the team members. It would be helpful to know how often the team interacts via conference calls, emails, visits, etc.
- There is a large list of collaborators, including national laboratories, industry, and academia, which is very good. It is, however, difficult to see a significant engagement with industry. A catalyst manufacturing company such as TKK or Cabot should become more involved in the scale-up of the materials that show promise, especially in the later stages of the project.

Question 5: Proposed future work

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

- There should be more focus going forward. The modeling work and the ALD work should be cut and the focus on SGD, and CVD with alloys, should be continued.

- Focusing on a much smaller subset of the approaches investigated to date is the right direction to go. Being able to demonstrate that good performing electrodes can be made and useful fuel cell results can be obtained with at least one of the three approaches (CVD, ALD, or SGD) is critical for the next year of the project.
- Continued studies of the role of carbon in RDE electrodes should be eliminated and the researchers should move all effort to optimizing mass activity in MEAs (via optimization of carbon and Nafion® loading) for the last year. There still may not be enough time.
- Given the suitability of Pt black as a surrogate, and the risk of unknowns in scaling up some of the other avenues, it is questionable whether the team should pursue making high(er) surface area Pt black. A now defunct company called E-TEK was commercially selling high-surface area Pt black with B.E.T. (N2) areas of >45 m^2/gram. With the tools and knowledge from some of the solution phase methods here, trying to recreate this material would be worthwhile. Also, a lot of focus should be put on electrode/MEA making as the next step.
- The proposed work is fine. A published paper summarizing the efforts and finding common threads that are not currently being communicated would be helpful. Full fuel cell testing coming at this late stage in the project seems problematic, as there will be a lot of issues in integrating any of them and realizing their full potential in less than a year. Starting earlier with the Pt black would give insights into future issues that may be battled.
- The proposed future work plan is clear for the electrode studies and the modeling effort. The Pt deposition task still continues down the path of evaluating three deposition methods; this task should be down to one method so that optimization can take place during this final year. The ALS method cannot scale up and should be eliminated. The team should down-select between CVD and SGD methods based on the work to date and modeling predictions. The presentation did not address the possibility of catalyst leaching and degrading fuel cell performance and should be evaluated before long-term fuel cell testing.
- With the inclusion of Los Alamos National Laboratory, there is a partner with demonstrated capabilities for incorporating materials into fuel cell MEAs. Though mentioned in last year's comments, there was no discussion of what alternate materials could be used to replace the nanowire templates. The lower-temperature ALD approach seems to be in conflict with the higher-temperature CVD annealing approach with respect to Pt nucleation and growth, as well as surface diffusion of deposited Pt during the formation of catalysts by both methods.

Project strengths:

- Large teams are able to look at a lot of different materials.
- The researchers are highly capable, well directed, and very knowledgeable.
- This is a distinctly different approach to catalyst development through thin-film methods.
- This is a great research team. Valuable work has been accomplished in learning about the synthesis of novel catalysts and their properties.
- This project's strengths are the breadth of constructions and the quality of RDE work, although that can be a "Catch 22" if only a few samples get tested.
- This is a well coordinated and managed effort across a broad front. There is lots of technical innovation and exciting progress has been made and demonstrated.
- The research team has good depth in the area of carbon-free (Pt) materials and a wide assortment of approaches. This project has good approaches to understanding limitations, such as the dispersion of these novel materials and adding carbon.

Project weaknesses:

- Too much emphasis has been put on RDE to predict performance in MEAs.
- There is a lack of focus going forward that will not reach a conclusion that has value to the industry. Also, the engagement of catalyst suppliers could be much stronger.
- Electrode (MEA) making appears late in the program. Earlier attempts, even at a small scale, may have assisted in down-selecting from the numerous approaches in the portfolio.
- Sample testing throughput seems slow, based on the limited data; if this is not true, it should be communicated in next year's review. It is understandable why some reviewers might not like the breadth of catalyst types researched, but the breadth is great as long as there are conclusions that can be drawn across platforms.

- The team must down-select a deposition method, generate MEAs, and test them to learn the benefits of electrodes and fuel cell improvements. The researchers also need to incorporate the modeling effort to make decisions.
- The use of readily leachable transition metal nanowire templates is of concern with respect to fuel cell durability. There is an unexplained difference in the project reported Pt black activities versus literature values.

Recommendations for additions/deletions to project scope:

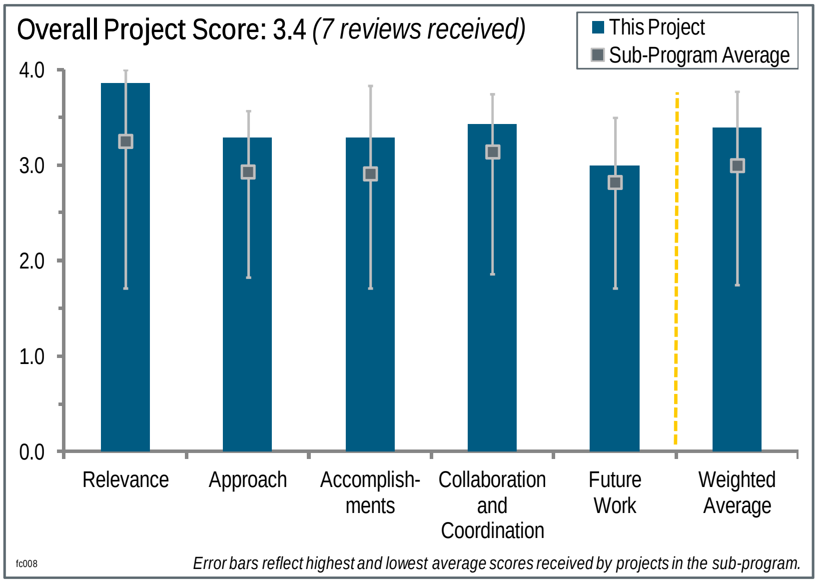
- The researchers should stop doing RDE work.
- The project should focus on one Pt deposition method for future work. The other tasks are reasonable and achievable.
- The researchers should cut out modeling efforts and stop ALD work. They should scale up the SGD method with suppliers and study the effect of residual Ag. The CVD method can be used to try alloys.
- The PI's listed focus on electrode making is endorsed. The project should consider pushing the limits of Pt black. It is questionable whether this group can make a higher surface area version that would be appropriate for the electrode structures they envision.
- The researchers should add 3M NSTF data if at all possible, because it is held as a standard now and is a necessary comparative to draw conclusions.
- A greater emphasis on Pt only or Pt and non-PGM alloy systems to focus improvement on total PGM mass activity would be beneficial.

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

Nenad Markovic; Argonne National Laboratory

Brief Summary of Project:

The main focus of this project is to develop an understanding of the oxygen reduction reaction (ORR) on multi-metallic systems of PtMN-alloys (M=Co, Ni; N=Fe, Mn, Cr, V, Ti, etc.). This knowledge will lead to the development of highly efficient and durable real-world nanosegregated platinum (Pt)-skin catalysts with low Pt content. Argonne National Laboratory's (ANL's) technical targets exceed or match the 2015 U.S. Department of Energy (DOE) targets for specific activity, mass activity, and electrochemical area loss. The target total content of platinum group metals (PGMs) is less than 0.1 g/kW, which is less than the original DOE target of 0.2 g/kW.



Question 1: Relevance to overall DOE objectives

This project was rated **3.9** for its relevance to DOE objectives.

- Simultaneous improved catalyst activity and durability is relevant to the program goal. The stretch target is even better.
- This is a very relevant project, with both cost and durability being considered from a fundamental point of view.
- Higher ORR activity is one of the key targets that need to be addressed for lowering the cost of polymer electrolyte membrane (PEM) fuel cells.
- Developing high-performing and durable catalysts, as well as lowering costs by reducing the PGM total content, is highly relevant to the DOE Hydrogen and Fuel Cells Program. The technical targets that the researchers set for themselves are also high.
- This project is outstanding. This thorough work is providing an important framework to guide others in what are the most important factors in making better, more active ORR catalysts.
- Fundamental studies of optimized activity and durability of nanosegregated Pt-skin catalysts are a promising approach to achieve high utilization of expensive Pt catalyst—one of key technical gaps for PEM fuel cell commercialization.
- The overall approach for making Pt skins on top of Pt alloys is relevant from the perspective of attempts to improve mass activity of these catalysts for oxygen reduction. The results reported are a culmination of nanoparticle synthesis and studies conducted on single crystals.

Question 2: Approach to performing the work

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

- The progress is excellent. It would have been nice to see catalysts in fuel cells sooner in the plan to understand any issues with integration and processing.
- It is good to see preferred crystalline orientation and ternary alloys used to improve material stability in the fuel cell environment.

- This is very careful work, including looking at particles in a disciplined way. Very few places in the world can take such a careful and thorough look at these catalysts.
- It is not clear whether the catalyst preparation is scalable.
- This project has a good approach. It encompasses studies to gain a fundamental understanding of the ORR on the catalyst systems, as well as practical membrane electrode assembly (MEA) fabrication and testing. The techniques used to characterize the catalyst to gain a fundamental understanding are also impressive. In terms of the catalyst synthesis, it would be good to know how easily these catalysts can be synthesized, how much it will cost to synthesize them due to the multiple steps involved, and if the process is scalable. It would also be good to know if the shape and structure of the catalysts are thermodynamically stable.
- The approach is structured and rigorous, uses well-defined model systems, and is based on surface science. This approach is a step above most catalysis work in that it is a forward design process determining the fundamentals of the activity/structure design.
- The stability aspect appears to be addressed adequately. However, there is no indication of other factors, such as mass transport or cost. This work has the appropriate balance of ex situ and modeling compared to fuel cell testing. Although there is a risk that MEA testing has been left too late and new issues will arise, and there has not been a lot of durability work, these efforts should be placed on the catalyst design.
- The approach of this effort is to engender with reproducibility thin films of Pt on top of Pt alloy to effect improvements in ORR activity for PEM fuel cell cathodes. The approach is based on nanoparticle synthesis with capping agents and understanding the increased activity in terms of careful structural studies. This approach though laudatory has to be tempered with realities, such as the challenges of translating these concepts into large-scale synthesis and understanding the issue of a limited potential window where such structures are maintained.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- The accomplishments and progress appears to be on target with the goals and objectives of this effort. There seem to be nice, incremental improvements being made.
- The project showed preferred performance with ternary alloy. It is necessary to investigate the mechanism of catalyst activity improvement. It needs material stability under the fuel cell environment.
- The accomplishments presented are excellent and impressive. More fuel cell performance and durability testing would be good, which seems to be the plan for future work. It is great how the principal investigator (PI) ties the fundamental understanding gained to designing better catalysts.
- Between the 2011 and 2012 presentations, there are many results that are shown again, sometimes just reordered. While the quality of work is excellent, it would be nice to get a sense of the quantity of work and how many catalysts are generated, characterized, and tested. There are references to 3M nanostructured thin film (NSTF) catalysts, but there are no comparisons between that system, the nanoparticle approach, and conventional catalysts to get a true sense of the amount of change in activities seen.
- The activity and durability results are great. The level of understanding this project brings should not be ignored. Some may complain about the cost of this kind of work, and they should not. This is remarkably important and there are few places that can be as productive as this group, which happens to work at a national laboratory, which is expensive.
- The project is achieving results at a suitable pace. The reaction pathways and mechanism have been confirmed, but more understanding is required on the segregation profile.
- Whereas the catalyst has been prepared in an MEA with General Motors (GM), only mass activity was shown, but not the polarization curve. This may indicate problems with mass transport or ohmic losses.
- The practicality of scale-up is not clear, but the direction has been changed to improve this.
- The project was able to build on experience with binary systems to develop a shortcut approach for ternary systems, in order to get the desired structures.
- The stability of the catalyst was assessed at 0.6–0.95 V, with minimal change in catalyst structure after 20,000 cycles. A higher upper potential limit needs to be assessed. While a fundamental approach is very good and needed, an increased effort of understanding MEA scale effects should be introduced at this point in the project.
- ORR activity in rotating disk electrodes (RDEs) has not been correlated to that in MEAs of subscale fuel cells. Even though durability has been reported in MEAs, the ORR activity in MEAs has not been clearly

reported and compared to the extremely high values reported in the RDE half cell setup. It appears that the activity is no different than that of Pt/carbon in MEAs, based on incomplete information of the conditions in the slide on MEA durability studies.
- It seems likely that the activity in MEAs will be lower than those in a fuel cell based on the typical experience of the research community. Milestone 1.2 on ORR mechanisms has apparently been met, but simply obtaining a Tafel plot slope is not evidence for having accomplished this milestone.

Question 4: Collaboration and coordination with other institutions

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

- The project has appropriate partners and they seem to be well coordinated.
- The researchers are working with the best.
- ANL has assembled a good team; however, it is not clear the extent to which some team members are contributing.
- This project has good collaborations with 3M and others; this is a bonus. The work is so valuable that reviewers should give them extra points for reaching out to industry.
- Good collaborations seem to be the case here, though the role of the Jet Propulsion Laboratory (JPL) is not clear in terms of the results presented, and neither are the roles of Brown University and the University of Pittsburgh.
- GM has tested durability but not reported the activity compared to the baseline Pt catalyst typically used in real-world fuel cells.

Question 5: Proposed future work

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

- This project has well-targeted improvements that are not only scientifically important, but relevant.
- The plan forward looks appropriate, with the exception that the practicality of scale-up of the structures is not clear.
- Putting catalysts in fuel cells sooner would be better, because it will take some development to get the catalyst to reach its optimal performance.
- The proposed future work sounds very much like an incremental effort toward expectedly same results. No new ideas or concepts seem to manifest in the future efforts.
- It would be good to see more transmission electron microscopy, RDE, and fuel cell results for the catalysts on a carbon support. It would be good to know how high or low of a catalyst loading on carbon can be prepared. Some evidence that the catalyst shape and structure do not change with time would be valuable.
- ANL should focus on the materials stability of the ternary alloy catalyst, which shows high activity. It is questionable that the PI can take enough time for alternative approaches for fabrication of thin-film nanoscale catalysts with ultralow PGM content.
- The ORR activity of the new catalysts in MEA versus that in RDE is of primary importance. The values reported in RDE are extremely high and unlikely to be reproduced in a fuel cell. Independent laboratories need to evaluate the electrocatalysts in RDE and MEA to ascertain the ORR activity and ECA.

Project strengths:

- The catalyst fundamentals are excellent.
- This project has good material synthesis and analysis capabilities.
- The researchers take a careful and disciplined approach.
- The novel electrocatalysts and preparation method with apparent high activity are strengths of this project.
- This project provides a very structured and fundamental approach to establishing active and durable catalysts using ternary systems.
- The project team is of the highest caliber.
- Project strengths involve the ability to control Pt alloy nanoparticle morphology and surface structure to engender high mass activity for ORR. This is carefully done in the context of studies conducted using single

crystals and nanoparticle synthesis, and advanced tools and methods, such as synchrotron-based X-ray absorption.

Project weaknesses:

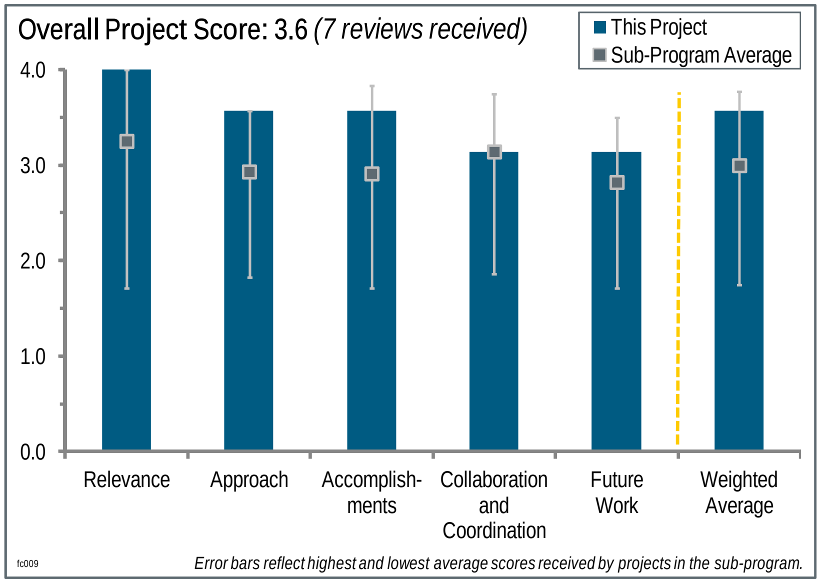
- Reliance on RDE may not map quickly into fuel cells.
- The principal weakness in this project is that this proposed effort is a marginal improvement over decades of similar work and is quickly getting dated.
- Expected costs and manufacturability of the catalyst need to be addressed. Also, MEA testing has been minimal.
- The scalability of manufacturing of the electrocatalysts has not been explored. Support interaction and durability with the electrocatalyst need to be evaluated to obtain a working catalyst.

Recommendations for additions/deletions to project scope:

- A potential manufacturing partner and cost should be addressed.
- ANL should increase MEA testing and include a feasibility assessment on practical catalyst manufacturing.
- ANL needs an independent laboratory to evaluate the ORR activity in both MEAs and RDEs.
- This type of work should be a larger fraction of the portfolio, with this type of electrocatalysis at the center. The science this group produces is a national treasure and we are not supporting it well enough.
- The roles of the partners mentioned in this effort are not clear. For example, the role of JPL, which mentions a combinatorial approach, is at odds with the thought-based approach being shown in this effort.

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

Radoslav Adzic; Brookhaven National Laboratory



Brief Summary of Project:

The objectives of this project are to: (1) develop high-performance fuel cell electrocatalysts for the oxygen reduction reaction (ORR) that are comprised of a contiguous platinum (Pt) monolayer on a stable, inexpensive metal or alloy nanostructure; (2) increase the activity and stability of the Pt monolayer shell and the stability of supporting cores, while reducing the content of noble metals; (3) maximize Pt utilization to use every Pt atom; (4) scale up the conventional synthesis of ultra-thin palladium (Pd) alloys (refractory metals or gold [Au]) nanowires or hollow nanoparticles as supports for a Pt monolayer; and (5) scale up synthesis based entirely on electrodeposition of nanowires or nanorods.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **4.0** for its relevance to DOE objectives.

- This project is critical. It has the best chance to lower Pt loadings to μg/cm^2 quantities.
- This project is in full alignment with DOE research and development objectives. It targets reduced Pt loading and cost while increasing activity and durability.
- The development of low platinum group metal (PGM)-loaded electrodes is absolutely essential to enable the commercialization of light-duty fuel cell electric vehicles.
- The project is very well conceived and is relevant to the DOE Hydrogen and Fuel Cells Program goals, specifically the goals related to enhancing catalytic activity and stability.
- This project does a very nice job of looking at derivatives of Brookhaven National Laboratory (BNL) core shell technology to address DOE activity objectives for Pt.
- This project provides one of the most promising pathways to reduce the use of expensive Pt in fuel cells and thus could allow fuel cells to become cost competitive with other power sources for a range of applications.
- BNL has made considerable strides in reducing total precious metal use, as well as reducing Pt. The activities are finally consistently listed both against Pt and against the total PGM use.
- BNL has succeeded in transferring its technology to a catalyst supplier (albeit not the supplier that is a formal partner within the project) that is making sufficient quantities of catalysts available to enable thorough evaluations of the concepts to be made under actual working conditions of applications.
- The most impactful possible development toward the commercialization of hydrogen (H_2) fuel cell vehicles is the development of high-activity, durable catalysts. BNL is working on exactly that topic.
- BNL understands that high activity must imply the reduction of Pt by expressing the activity in units of amps per mass of Pt. BNL understands that high activity must also imply the reduction of all precious metals, including Pd, ruthenium (Ru), and Au. However, there could be a better focus on the elimination of other precious metals.

Question 2: Approach to performing the work

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

- This project has a clear set of goals to be accomplished by a clear set of pathways. The approach is sound and the results show that it is successful.
- The approach appears to be successful, as seen from the catalyst activity gains. The team seems to leverage basic research funding well to do complementary theory, which helps the project.
- There was no discussion of the approach to be used by Johnson-Matthey Fuel Cells, Inc. (JMFC) for catalyst scale-up.
- BNL's approach to develop core-shell nanoparticles is one of the most promising concepts to date for both maximizing the fraction of Pt available for the ORR reaction and enhancing its stabilization. Refractory-alloy and/or hollow particles could do a lot to reduce total PGM. It would be good to see continued effort on the hollow particle approach that BNL does not appear to be pursuing, because it eliminated the need for Au or Pd. It is nice to see membrane electrode assembly (MEA) testing done on promising concepts. The multiwalled nanotubes (MWNTs) work is a duplication of other efforts.
- The approach is very well conceived. There is a strong emphasis on fundamentals and there is a pathway presented for scale-up of promising systems. There are clear underlying hypotheses presented, and a well-designed research approach to test each hypothesis.
- The barriers addressed are very relevant and the targets set are challenging.
- Multiple pathways are being followed to reduce the total precious metal, so the project is not dependent upon any one approach working.
- The use of refractory metals in the core, alloyed with limited amounts of precious metals, could finally put to rest longstanding concerns about the amounts of precious metals used in the cores.
- Extensive use of Pd in the cores of many of the systems being studied raise concerns of a return to 2000–2001, when Pd was more expensive than Pt on a mass basis; however, the lesser density of Pd made it a bargain versus Pt, even under those unusual and unlikely-to-be-replicated pricing conditions. (There are twice as many Pd atoms as Pt atoms per gram, with a similar volume per atom.)
- The researchers should be wary of the false assumption that (111) crystal faces always give higher ORR activities. While this appears to be true for Pt alloys, it is not true for pure Pt.
- While some examples are included, BNL needs to provide more focus on cheaper core materials, such as those with hollow cores or more refractory cores (e.g., NiW).
- The approach is somewhat compromised by the tendency to use metals that, while capable of stabilizing a monolayer of Pt, may themselves be unstable in a fuel cell cathode environment; examples include Pd and nickel (Ni). To the principal investigator's (PI's) credit, there is an attempt to stabilize a Pd core with Au. However, the use of Au again prompts questions about cost and the usage of other precious metals.
- Modeling efforts to predict stability need to incorporate a more realistic environment that accounts for the presence of oxygen (O_2) and water. While the approaches of other projects may more clearly show a roadmap toward high-specific activity using thin-film structures, the approach of this project is worthwhile due to the need to see if nanoparticles and related morphologies can yield high activity.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.6** for its accomplishments and progress.

- BNL has done an excellent job working with industry (General Motors [GM]) in the practical evaluation of catalysts and the evaluation of catalyst stability. Also, kudos for the transition of the BNL catalyst technology to industry.
- BNL has made very good progress toward the targets of the program. Specifically, the demonstration of high MEA performance with very low loading is impressive and shows applicability.
- BNL's fuel cell data on Pt/Pd/Au look encouraging. The GM data on the Pt/Pd is a decent first step, but significant improvements in activity are still required. The voltage cycling results are also encouraging, although the Pt/C MEA baseline seems surprisingly low performing by comparison. PtNi nanoparticles on MWNTs are showing high activities, but stability is unproven.

- The primary issue here is the noble metal content (Pd, Au) in the core of the catalyst. Minimizing Pt content is of little use if it is supported on an Au core. However, the project team is well aware of this issue and is making significant progress in modifying the core composition. The carbon nanotube approach looks promising.
- The team has made very good progress using multiple approaches. The use of Pt monolayers deposited on stand-alone or alloy cores has shown promising results. Accomplishments have been validated by MEA performance evaluation, yielding very encouraging results.
- The durability of the system has been demonstrated through potential cycling tests.
- The progress on refractory-metal-alloy cores is very encouraging. BNL's demonstration of a process to put down high-quality Pt monolayers without an intermediate copper underpotential deposition step could greatly reduce manufacturing costs and make this catalyst technology more widely accessible. BNL has made good progress on extending the testing of activity and durability in MEAs, which complements the rotating disk electrodes (RDEs) used in the past. This trend toward working with practical MEAs should be continued.
- More attention could be paid to the hollow Pt catalysts that were described several years ago, as these fully avoid residual concerns about the durability of any system containing Pd that are held by catalyst technologists with considerable Pd experience.
- Results for catalysts that include PGMs other than Pt need to have mass activity reported in A/mg PGM. Out of approximately seven distinct families of catalysts reporting, only three show A/mg PGM (Pt/Ru, Pt/Pd, and Pt/PdW). This needs to be done for the rest.
- Performance of the PtML/Pd/NiW catalyst needs to be reported in mass and specific activities to allow comparison with other catalysts. The in situ results shown are iR-free using O_2 at the cathode, and as such, they appear poor.
- Only stability results are shown for a catalyst that likely has poor total precious-metal-basis mass activity (Pt/PdAu). It would be interesting to see stability results for high-mass-activity PtNi supported on MWNTs, as well as anything involving a cheap core (e.g., PtML/Pd/NiW).
- BNL showed high-activity results for hollow core materials in past years, but there has been little follow up. This needs to be done.
- At present, no individual catalyst has been developed that is known to meet both cost and durability targets.

Question 4: Collaboration and coordination with other institutions

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

- This project has a large team of partners and collaborators from industry, academia, and one national laboratory. The team provides the large pool of resources necessary to perform the intended work.
- It was nice to see MEA testing done and at original equipment manufacturer (OEM) (GM). Pt/Pd nanoparticles are being scaled up by a catalyst supplier. Massachusetts Institute of Technology (MIT) has done some work on PtNi nanoparticles on MWNTs. It was unclear from the presentation if there are key contributions from JMFC and UTC Power at this point.
- BNL showed good collaboration with industry. The extent and nature of collaboration with academic partners was not quite as clear. Perhaps the details of this collaboration will be presented on the next occasion.
- Next year (2013) should be a very important year for this project, and collaborations will be critical in areas such as scale-up of synthesis, technology transfer to licensee, and MEA and stack testing. All these activities depend on successful collaborations. If this team cannot successfully transfer and scale its technology, then a very promising approach will have been wasted.
- The roles of UTC Power and JMFC are not clear, and the MEAs appeared to be tested by GM. Because UTC Power is a Cooperative Research and Development Agreement (CRADA) partner, this may not be a significant issue, but hopefully in the last year of the project, the objectives of UTC Power can be met.
- There is excellent coordination of theory and MEA evaluation, especially as it seems to be at no cost to this project.
- There has been no apparent contribution in this year's slides from funded partner JMFC. Technology transfer and scale-up have occurred, but to a manufacturer outside the project that does not have production facilities in the United States.
- The licensing of BNL's monolayer catalyst technology to a catalyst supplier outside the project should make this material generally available to the industry. However, it is not clear that formal protection and licensing of intellectual property (IP) generated by national laboratories, rather than putting the information into the public domain, is the most efficient pathway to attain national technical goals; it has, in this case, inhibited similar scale-up and commercialization efforts by other competent manufacturers.

- MIT has done interesting work under the project, but no coordination between the MIT efforts and the main project at BNL was evident from the presentation.
- Contributions of partner UTC Power were not clear in past work, but they should become important in future MEA testing. There has also been effective collaboration with several organizations outside the formal project.
- The collaborations that involve MEA fabrication and fuel cell testing have resulted in data that generate quite a challenge in comparison to the program targets. The polarization data associated with Pt/PdAu nanorods and PtML/Pd/NiW are shown for very low loadings (0.027 and <0.04 mg Pt/cm^2, respectively), but also with iR-free potentials and O_2 as the cathode oxidant. It would be much better to use a loading, such as 0.075 mg PGM/cm^2 (which would total 0.125 mg PGM/cm^2 if combined with a 0.05 mg Pt/cm^2 anode), and then report both iR-free and cell voltage using air. This kind of experiment could yield data that would be useful for comparison.
- Collaborations on fuel cell testing should also aim to provide mass and specific activities in situ.
- Modeling efforts do not appear to be on track to provide useful information. They also need to require a realistic environment (O_2 and water).

Question 5: Proposed future work

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

- The proposed future work is in line with the project requirements.
- The proposed future work is exactly what it should be. One hopes that the team can stick to this plan and not get sidetracked.
- It is not clear from here what happens to the MIT catalysts.
- The team should focus on MEA development for the last year. The tech transfer of the catalyst IP to industry was well done.
- It would be beneficial to understand how the progress currently made in this project would actually translate into a cost reduction of fuel cells. The project team should give an estimate of the potential cost benefits.
- BNL should emphasize Pt on refractory alloy cores and hollow Pt nanoparticles rather than Au- and Pd-based cores. A lot of the future work seems to focus on the nanowire supports, on which it will likely be more difficult to create very thin Pt films. There are also already other projects (National Renewable Energy Laboratory) focusing on these types of materials. BNL needs to keep stressing MEA tests and work to understand what gives high durability. Also, there should be increased focus on high current density performance.
- BNL needs to focus on scaling up the best precious-metal-basis mass activity materials, provided there is an expectation of stability. These materials should include the hollow cores, which have been reported in years past. Thankfully, returning to the hollow nanoparticles is mentioned in the future work for 2012.
- BNL should look at the stability of the PtNi nanoparticles on MWNTs before scaling up.
- The Pt/PdAu nanowire precious metal mass activity should be reported before there is any decision to scale up the production of this catalyst.
- Refractory core work should continue as noted.
- Refractory-alloy cores are being properly emphasized for future work.
- BNL should apply plans for microemulsion synthesis work on hollow Pd to hollow-Pt catalysts that avoid possible durability issues with Pd.
- It is not clear that the increased emphasis on nanotube and platelet cores is warranted, as the project claims benefits for (111) orientations of pure-Pt systems that have not been borne out by careful single-crystal experiments.
- Increased emphasis should be placed on the detailed characterization of these catalysts after they have been tested in MEAs for activity and durability; it is not clear that the active materials are the same as those characterized before testing.
- Further emphasis should be placed on understanding why the electrochemical surface areas of these materials are in the range of approximately 120 m^2/g Pt rather than the 240 m^2/g Pt that would be expected for a true monolayer catalyst.
- The researchers should be wary of overreliance on electrodeposition as a means of depositing catalysts onto diffusion media, as electrochemical methods are typically harder to scale up than chemical methods.
- The researchers should keep focused on durability testing under a range of conditions (including extended times at the H_2 potential) and further characterize the effects of core materials that may escape into the ionomer and then to other parts of the cell.

Project strengths:

- This project has a good approach and thorough execution.
- The potential for order of magnitude reduction in Pt loading is a strength.
- The culmination of BNL's excellent catalyst work is playing out well in practical MEAs.
- This project has very promising catalyst concepts already scaled up for MEA testing at an OEM. Initial durability results are very encouraging. There was good engagement with both catalyst suppliers and end users. Thus, scalability is being considered.
- This is an excellent team and an outstanding PI. There is a strong focus on fundamentals and there are excellent results attesting to progress. There is an emphasis on stability in addition to activity.
- There is a good commitment to scale-up and interaction with industry partners.
- The creative choice of catalyst systems, synthesis, and pre-testing characterization of the catalysts is a strength of this project. There was effective technology transfer to get Pd-core catalysts scaled up and into the hands of industry in adequate quantities for thorough testing under a wide range of conditions.
- The researchers are making progress on testing materials in MEAs, though this needs continuing emphasis.
- The durability results suggest improved uniformity of Pt monolayers versus earlier efforts.
- BNL is able to leverage the investigators' well-established expertise in generating Pt monolayers on a variety of materials.
- BNL is able to quickly churn through the synthesis of a wide variety of catalyst families.
- RDE techniques and other experimental techniques are well understood and implemented.

Project weaknesses:

- BNL needs to focus on improving materials stability.
- The impact of the modeling work was unclear. The most developed concepts still use PGMs (Au and Pd) in the cores. BNL seems to have abandoned the promising hollow core Pt nanoparticle concept.
- The nature of collaboration with academic partners and their roles was not very clear.
- The durability tests employed should perhaps be revisited.
- Further description of the methods needed to get improved uniformity of Pt monolayers should be given, accompanied by detailed characterization showing these improvements. Surmising increased uniformity merely from durability data is not enough.
- Noble metal (Pd, Au) use in catalyst core is a weakness of this project. There is some reluctance to work aggressively on changing the cores. The Pd or Au used maybe relatively small (mg/cm^2), but if the total noble metal content is similar to conventional catalysts, the benefits of this project are essentially wasted.
- BNL often fails to recognize that the inclusion of lower-cost precious metals will not in fact result in lower costs when catalysts are commercialized. BNL needs to be better focused on thrifting all precious metals.
- The nanoparticle focus puts the project at a disadvantage of usually beginning with materials that are going to be low for specific activity.
- The modeling assumptions were not well defined.
- The fuel cell data have not been generated in a manner that is easy to compare to program targets.

Recommendations for additions/deletions to project scope:

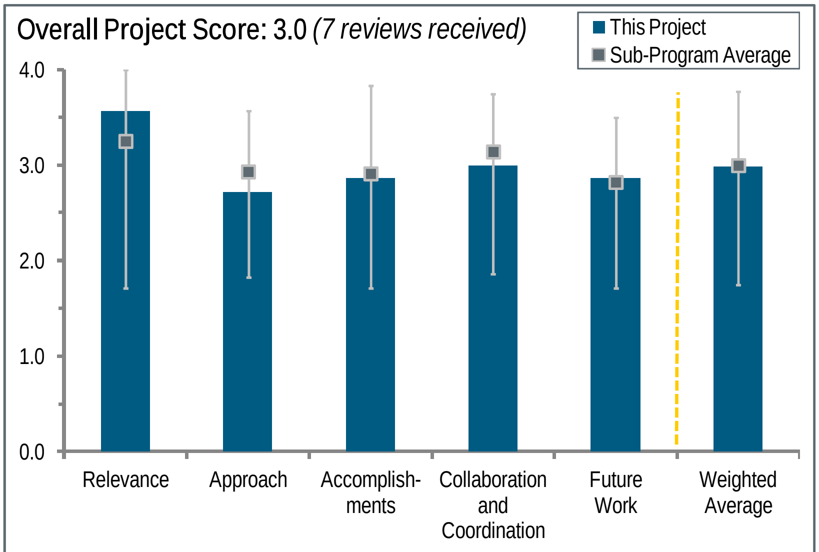
- This is a very nicely structured project and should continue as per the scheduled plan/scope.
- BNL should evaluate the MIT catalysts in MEAs.
- This project should have less of a focus on Au/Pd-containing cores and nanowire supports. More focus should be on hollow and refractory metal cores. BNL should continue MEA testing with an emphasis on durability and high-current density performance.
- BNL should focus exclusively on: (1) cheap cores, hollow and refractory, and (2) other catalysts that have demonstrated high mass activity on a precious metals basis. All other material developments should be deleted.
- The modeling activity should be deleted because the prospects are low that it will provide quality information toward predicting stability.

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

Fernando Garzon; Los Alamos National Laboratory

Brief Summary of Project:

The goals of this project are to: (1) develop durable high-mass-activity platinum group metal (PGM) cathode catalysts to enable lower-cost fuel cells; (2) elucidate the fundamental relationships between PGM catalyst shape, particle size, and activity to help design better catalysts; (3) optimize the cathode electrode layer to maximize the performance of PGM catalysts for improved fuel cell performance and lower cost; (4) understand the performance degradation mechanisms of high-mass-activity cathode catalysts to improve catalyst design; and (5) develop and test fuel cells using ultra-low loading, high-activity PGM catalysts.



Error bars reflect highest and lowest average scores received by projects in the sub-program.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.6** for its relevance to DOE objectives.

- This project supports DOE's objectives to lower costs and increase durability of the cathode catalysts for polymer electrolyte membrane (PEM) fuel cells.
- The stated objectives will deliver a high-performing, durable catalyst that can be made to deliver these attributes in a practical fuel cell. There appears to be no clear path to meet that objective half way through the project.
- This project is directly relevant to the overall DOE objectives. Los Alamos National Laboratory (LANL) is working to generate a stable, lower-cost cell catalyst.
- Membrane electrode assembly (MEA) cathodes typically consume two-thirds of catalysts and are a good target for cost reduction for both quantity and material design.
- Understanding the durability of low-loaded PGM catalysts is essential to enable commercialization of fuel cell vehicles.
- The reduction of total platinum (Pt) loading in the fuel cell stack below 10 g is vital for fuel cell vehicle commercialization. This project is clearly aligned with this target and its development of durable, ultra-low Pt alloy cathode catalysts.

Question 2: Approach to performing the work

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

- The options should be assessed against a metric, prioritized, and down-selected.
- The science within each topic or task within the project is fine, but there is no coordination between these tasks so that the overall effect is the simple dilution of resources and effort.
- The approach appears to be rational and it should be expected that the project will yield interesting and useful results.
- The basic approach of using modeling to complement the experimental work is a very valid one. In-depth analytical support is also a great asset to this project. Some of the catalyst ideas are quite novel and interesting,

such as metal nanotubes and Pt-Ce-carbon. However, others are well understood and are not novel, such as Vulcan v. Norit carbon supports and ethylene glycol studies.
- The approach is well designed, as it combines both theoretical and experimental components. If it is a modeling-driven approach, model validation should occur in parallel to model development to ensure that the modeling assumptions are correct. It would also help to correct modeling assumptions at the early stage.
- This project needs greater emphasis on model validation in parallel with good performance in electrochemical characterization. There may never be good performance, but the models could be validated. The researchers need some tests just to validate the model, even if the results are not good. There is not enough single-cell testing being performed, because not enough materials are getting through the initial screening.
- It is unclear how the current dispersed work streams will improve the understanding of ultralow PGM catalysts. The density functional theory (DFT) studies on nanotubes will only have value if those structures can be made and tested to validate the model. The microstructural modeling needs to be validated for standard electrodes first before using it to investigate ultralow PGM catalysts. It is not clear how the CeO mitigation addresses specific ultralow PGM catalysts issues. It is also not clear how the ethylene glycol method enables ultralow PGM catalysts. The nanoplatelet work is the one promising area of the project. The accelerated stress test (AST) work is being done elsewhere, and developing ASTs for ultralow PGM catalysts will not be fruitful until there is field data to validate failure mechanisms from these ASTs.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- The conclusions are unclear conclusions. Only qualitative statements are given and hard data are missing.
- The accomplishments to date in modeling, nucleation growth studies, and synthesis and characterization are interesting. It is unclear if the use of yttrium (Y) actually reduces the cost of the catalyst.
- Significant progress has been made in advanced modeling, and modest progress has been made in overcoming barriers in catalyst synthesis.
- The modeling work has progressed well, but the experimental reduction to practice has lagged behind. There appears to be little integration of the modeling work into the catalyst work.
- The Pt_3Sc and Pt_3Y work has not really started.
- Overall, a lot of great work has been done on this project, all the way from modeling to analytical characterization to fuel cell testing.
- Perhaps in this particular presentation, too much emphasis was put on the modeling work and not enough was put on the actual experiments.
- The work done on Norit and Vulcan has already been done before, so it is unclear what the principal investigator is bringing that is new or exciting to the community.
- The mass activities for both the polypyrrole (PPY) and TKK benchmark appear too low.
- Pt-ceria-carbon catalysts are very unique with a lot of potential, and the analytical work thus far is quite positive. It will be exciting to see the future results.
- Many structural models are being generated, but not enough are being tested and initial results are limited. The project is 60% complete, but only limited testing has been performed to indicate progress toward objectives. The test results shown do not indicate any significant progress. Very limited cell and no stack testing has been performed.
- There is a smattering of results from the various parts of the project. All of the results seem reasonable, but it is not clear how they will, either independently or as a whole, lead to the development of an ultralow PGM catalysts with the exception of the PtY and PtSc alloy catalysts, which show some clear activity benefits. The PPY-Pt materials seem unlikely to meet activity targets.

Question 4: Collaboration and coordination with other institutions

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

- This project has good collaboration among national laboratories, universities, and industry.
- The collaboration and coordination with other institutions appear appropriate for this type of project.
- There seems to be little coordination between the groups.

- All of the subcontractors appear to be making contributions to the project. It is not clear that there is a strong interaction among all of the partners.
- An exceptional team has been brought together with universities, national laboratories, and stack original equipment manufacturers. Collaboration between all of the partners is well described and coordination/interaction appears to be very high.
- This project is dominated by national laboratories and universities. It would be good to see more involvement with the catalyst industry if there is any chance of taking the success of this project beyond the academic curiosity.
- Collaboration within the project team appears to be on track, but only limited coordination is shown with non-team members. Catalyst and MEA manufacturers should be included somehow, despite proprietary information restrictions (maybe as peer reviewers).

Question 5: Proposed future work

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

- The proposed future work appears logical.
- So far this seems to be a bunch of independent projects. The future plans do not include tying the work streams (i.e., materials and modeling work) together or down-selecting promising concepts.
- The future is focused on three areas of high potential: Pt/Ce/C, Pt/PPY, and Pt/Y, Sc with modeling support. More experimental work and less modeling is great. Additional fuel cell testing will be a great addition to the work thus far.
- Overall, the plans are built on achieved progress to date.
- The plans need to be more focused on model validation. Prior to extending the DFT model to catalyst coated nanotubes and nanowires, model predictions should be validated on uncoated nanotubes and nanowires. Prior to Pt/PPY fuel cell testing, the catalyst's activity needs to be properly evaluated by rotating disk electrode (RDE). Prior to moving to Pt/Y, Sc nanoplatelet research, the activity of Pt/Y, Sc nanoparticles needs to be properly evaluated by RDE. This project needs to be more focused on DOE targets with respect to mass activities.
- The PPY/Pt work is irrelevant and should be stopped. There should be more focus on tasks with the project so that the resources are not diluted too much.
- Decision points versus a schedule or budget are not addressed in the presentation. The greatest weakness of work to date is the minimal level of cell and stack testing. This is addressed at an increased level in the proposed future work. Verification of models needs further attention in future plans. The relation between DOE and Ballard Power Systems AST protocols should be related to actual test data, and one or the other selected for screening.

Project strengths:

- The strengths of this project are the knowledge and dedication of the project members.
- This project has a strong consortium in principle.
- LANL has assembled and managed a great team.
- Some of the catalysts investigated are really interesting and the complementary modeling work is very useful.
- Future fuel cell testing will be a great addition.
- Using a theoretical approach as guidance for the synthesis of new catalysts and for building more efficient catalyst layers is a strength of this project.
- A strong team of national laboratories and universities has been assembled.
- This project's greatest strength is the development and verification of electrochemical and structural models with test data. Hopefully, there will be progress toward performance and durability targets.
- This project has highly competent researchers with outstanding facilities. The preliminary results on Pt/Y, Sc nanoplatelet catalysts are promising.

Project weaknesses:

- This project has little coordination, and conclusions and down-selection seem to be missing.

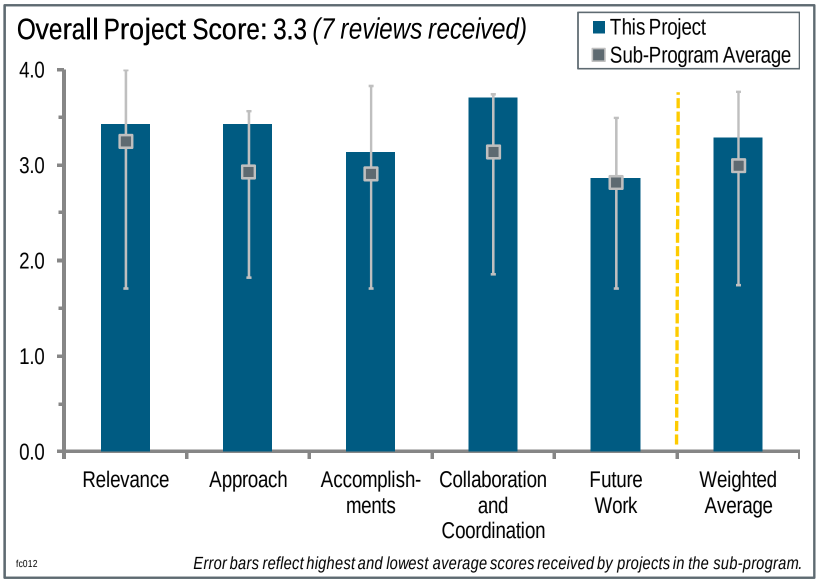
- The Vulcan/Norit work is not the best use of time. There is perhaps too much emphasis on modeling work.
- RDE activities are too low and need to be brought in line with literature values.
- There is a disconnect between modeling efforts and experimental work.
- There is limited involvement with industry. This means the project will have a limited impact on the industry if it was successful.
- The individual tasks are in silos, with no integration or down-selection planned.
- LANL has yet to show that any of the materials/structural designs can even meet the performance and durability levels of currently used cathode catalysts at costs comparable to current products, never mind at reduced costs. There is no discussion or analysis of costs for the tested materials/structural designs.
- There is a lack of clear indication of how the isolated projects tie together. Several tasks, such as ASTs, CeO_2 studies, and the Ballard electrode model, do not seem directly relevant to the development of ultralow PGM catalysts. The model validation plans are unclear.
- Model validation follows model development. It is unclear if the authors are planning to validate the model predictions that nanotubes are more susceptible to electrochemical dissolution than Pt (111). It is unclear if the researchers are planning to perform experiments on single crystals.
- Electron-beam techniques are not scalable for the synthesis of small-diameter nanotubes. It is unclear if there is a scalable approach for synthesis of small-diameter Pt nanotubes.
- With respect to RDE characterization of Pt_3Y and Pt_3Sc and bulk catalysts, the advantage of Pt_3Y and Pt_3Sc over Pt is not obvious, since the Pt standard has poor quality. Mass activities of PtY/C and PtSc/C catalysts are not shown, probably because they are low.
- With respect to RDE characterization of PPY Pt catalysts, the thin-film limit is not fulfilled, meaning that activities cannot be properly evaluated.

Recommendations for additions/deletions to project scope:

- The researchers should continue the good work.
- The presentation slides were far too busy.
- This project is doing well. The only real comment is to continue to focus in on the new, novel high-potential catalysts shown on slide 26 along with more experimental work and more complementary modeling work.
- The project needs to be more focused with respect to catalysts research. The researchers should focus on three areas: nanotubes and nanoplatelets, alloys, and pyrolized PPY Pt catalysts. This will leave behind Pt-ceria catalysts and studies of Pt nucleation and growth on carbons.
- The researchers should stop work on PPY/Pt and seek to focus efforts on the most favorable approaches. The researchers should also integrate the modeling into the catalyst design. If no Pt nanotubes are being made, there seems to be no reason to do this work. It seems like this should be part of the work at the National Renewable Energy Laboratory (FC-007).
- There needs to be greater emphasis on testing at the cell and stack levels and feedback to the modelers and theoreticians. The involvement of cathode material and MEA manufacturers would help to keep the focus on cost and durability.

Project # FC-012: Polymer Electrolyte Fuel Cell Lifetime Limitations: The Role of Electrocatalyst Degradation

Deborah Myers; Argonne National Laboratory



Brief Summary of Project:

This project's objectives include: (1) understanding the role of cathode electrocatalyst degradation in the long-term loss of polymer electrolyte membrane (PEM) fuel cell performance; (2) establishing dominant catalyst and electrode degradation mechanisms; (3) identifying key properties of catalysts and catalyst supports that influence and determine their degradation rates; (4) quantifying the effect of cell operating conditions, load profiles, and the type of electrocatalyst on the performance degradation; and (5) determining operating conditions and catalyst types and structures that will mitigate performance loss and allow PEM fuel cell systems to achieve the U.S. Department of Energy (DOE) lifetime targets.

Question 1: Relevance to overall DOE objectives

This project was rated **3.4** for its relevance to DOE objectives.

- The topic and program are highly relevant to the DOE Hydrogen and Fuel Cells Program's objectives.
- This project is a good fundamental study on basic degradation mechanisms. The relevance is high, but it is limited to the catalyst.
- This project is very relevant to DOE's objectives. In order to meet DOE's durability targets, all decay mechanisms and rates from each of the components must be known.
- Understanding the effect of platinum (Pt) and Pt alloy properties on voltage decay is a critical issue that must be addressed to enable fuel cell electric vehicle commercialization.
- Argonne National Laboratory (ANL) seeks to understand in depth the nature of catalyst dissolution. It repeats in a very thorough and methodical way work that has been done already. The dependence of crystallite size is fairly well known. The pulse shape dependence has been published already.
- Degradation of catalyst activity is clearly a very relevant issue for DOE. This project contains a lot of work on different ways to degrade catalysts. However, it is not clear that all of them are relevant to real fuel cell conditions. For example, it is unclear why someone would use perchloric or sulfuric acid to examine acid leaching and dissolution. Triflic acid would be more relevant.
- ANL is making progress toward understanding the factors that govern the durability of oxygen reduction catalysts in fuel cells. Such knowledge is critical to the development and relevant testing of advanced catalysts to meet all of DOE's performance targets simultaneously.
- ANL is attempting to generate data to allow testing of a model of catalyst degradation that one would hope would have predictive value. This could be very valuable if it can be reliably and verifiably done.
- The comparisons of pure-Pt and Pt-alloy catalysts should be important in guiding how much emphasis should be placed on the development of alloy systems. It would be helpful to compare alloys prepared using different methods (e.g., codeposition versus sequential deposition) to check the generality of conclusions drawn from materials made by one preparative method, as appears to be the case here.

Question 2: Approach to performing the work

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

- The approach is a little too limited.
- This is a good approach. It would have been good to see PtNi results on the non-carbon support done earlier.
- This project is a “tour de force” of organizing a large group of collaborators to produce a single, coherent view of predominant catalyst degradation mechanisms.
- ANL has done an outstanding job collecting and analyzing an abundance of data to highlight the key factors that impact voltage loss in dispersed platinum group metal (PGM) catalysts and membrane electrode assemblies (MEAs). The approach could benefit from a more thorough statistical analysis to identify better second and higher-order effects. There is even more that can be gained from the vast amount of data collected by taking advantage of the design of the experiments analysis. It is not clear how the modeling is fitting in.
- ANL accomplishes a lot of work and provides lots of data. There is an attempt to correlate with models, but it is not established very well, at least as it appears in this presentation. One is left with the impression that there is not a strong theoretical basis for the design of the experiments.
- The lead institution and partners have a very comprehensive approach and well-defined milestones. The project is well structured. There is a good balance between theory and experiment.
- Good work has been done to check correlations between electrochemical behavior in aqueous solutions and performance in real fuel cells.
- Nice efforts were made to separate Pt versus alloy effects from particle-size effects. The presentation could be a bit more clear on exactly how the different initial particle sizes were achieved, either by thermal annealing of a given small-particle precursor or by synthesis to differing initial particle sizes, as the results from these two sample-preparation strategies could be quite different.
- ANL has made good efforts to try to separate out the relative contributions of multiple effects: Pt dissolution/reprecipitation, particle coalescence, loss of alloying-element atoms, and corrosion of the support.
- A greater diversity of preparation methods should be employed for any given alloy composition. It is not clear if the conclusions on the effects of acid leaching, either before or after MEA preparation, would be the same for materials generated by different synthetic approaches.
- More detailed structural evaluations following durability testing could be informative.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- ANL has achieved an excellent level of accomplishment so far.
- ANL has generated a tremendous amount of data, but the progress is not so clear. After the presentation, it was still unclear whether this project is meeting DOE’s goals. The presenter went long and failed to make this point clear.
- This project has done very well on the first goal of elucidating predominant degradation mechanisms. It is not as strong in the second stated goal of mitigation strategies. The reviewer did not see a recommendation of running boundary conditions or whether there is a way to carry out the recommendations on operating limits based on this work.
- The key results involve the role of particle size and electrochemical surface area on durability and the fact that the relationship holds for both Pt and Pt alloy nanoparticles. It is not clear that the findings are universally true, as conflicting data sets exist, but the conclusions are merited, based on ANL’s data. It is not clear how the modeling work is driving the understanding, but that may be due to the lack of time during the talk to present the modeling work.
- The project goals are to understand the degradation of the cathode catalyst and its impact on performance. The project has done that from two points of view in a very in-depth way: at the catalyst level and at the voltage point of view, which are fairly well documented. What the researchers have not done is look at this project from the point of view of the acidity of ionomer, the type of ionomer, the temperature of operation, and the local current density.
- The team has made great progress toward the stated goals and has taken a nicely structured approach that has yielded many fundamental insights. The volume of work performed is truly impressive. The stated milestones

have been substantially met. The clarification of the relationship between ECA loss, mass activity, and particle size was particularly useful.
- Good progress has been made in measuring and modeling changes in particle size distributions for several different catalysts, and the process has been modeled.
- The output of the modeling effort should include not only the predicted evolution of the particle size distributions, but also the numerical values of the diffusion coefficients used for alloy systems and a description of how they were derived and the extent to which these coefficients agree with previous experimental data.
- The experimental work with the novel Pt_3Y and Pt_3Sc systems was very useful to fuel cell development in terms of showing the limited level of trust that can be placed in current calculational estimates of both activity and durability of oxygen reduction reaction (ORR) catalysts (and in experimental results obtained entirely through rotating disk electrodes).

Question 4: Collaboration and coordination with other institutions

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

- This project has very good collaboration.
- The project has an excellent range of collaborators, but it is not very clear if they are all collaborating.
- There is phenomenal coordination and collaboration between the partners and their specialty/contribution. It is great to see Johnson-Matthey Fuel Cells Inc., make a catalyst control at the diameter needed to compare to alloys.
- This project has a solid coordination of organization within the project and clear interaction with other durability and catalyst projects through working groups. It is good to see the inclusion of ANL's PtNi alloys in future work, although the project is almost over. It would be interesting to know what impact Jeremy Meyers leaving University of Texas-Austin would have on his part of the project.
- The team is well structured and has demonstrated effective collaboration between national laboratories, industry, and university partners.
- The coordination between the team members appears to be quite good, including correlations drawn between experimental work and calculations. The project team appears to interact productively with other DOE ORR catalyst durability project teams. The project serves the fuel cell community in general through the principal investigator's chairing of the Durability Working Group.

Question 5: Proposed future work

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

- The proposed future work is logical and in line with expectations.
- Mitigation strategies appear to be focused on carbon supports, which are covered with other DOE-sponsored programs. Most of the focus is on cathodes. It was unclear if ANL is doing anything on anodes.
- Future work on PtNi alloys, temperature dependence studies, and post-test diagnostics of the PtCo MEAs is justified. More statistical analysis of existing data should be added.
- There were no specific slides calling out future work. The reviewer is looking forward to the work on chlorides and 3M's NSTF and hopefully some core shell materials, as that work has not been published much.
- There were not many suggestions for where this project would go in the future besides responding to last year's reviewer comments.
- This project is approximately 83% done, so the amount of future work that can be done is limited. Plans for studying temperature effects on oxides, new work on PtNi, and completion of a cathode performance degradation model seem very ambitious.
- A bit more attention to possible changes in area-specific activity during analysis of the data might lead to insights useful for the design of new catalysts.
- It would be nice if ANL could make more progress with in operando XAS studies of catalyst degradation in working fuel cells to show scientists outside national laboratories whether such experiments are likely to be worth the trouble of doing.

Project strengths:

- This team is very industrious with lots of data generated.
- This is a strong team with great analytical skills and equipment. The leadership is excellent.
- This project has a strong team. There is a good balance between theory and experiment. A large volume of work was performed and data was duly analyzed to provide key insights.
- This project carried out a degradation analysis through systematic and sophisticated analytical instrumentation. There was good selection of controls. The team is strong, as are the team's contributions.
- ANL has executed a solid plan and collected and analyzed a lot of useful data. It is a solid approach to address a critical issue. There is strong collaboration both within the project and with the fuel cell community.
- The quality of work is extremely high in documenting catalyst degradation at the catalyst level. It provides thorough documentation of what others have observed and forms a basis of prediction for modeling efforts in this narrow set of constructions.
- A very large amount of systematic work has been accomplished under this project, and several factors have been confirmed as major drivers of catalyst degradation.
- The level of relevance of studies concerning aqueous electrolytes to the operation of PEM fuel cells has been successfully tested.
- Initial particle size distributions were shown to exercise a strong control over subsequent changes in both surface area and mass activity.

Project weaknesses:

- The project's mitigation strategies should be communicated as part of recommendations or areas for further research.
- There is a lack of statistical analysis and links between modeling and experimental work.
- The work is not held together by a strong hypothesis and is very empirical.
- The modeling component was not well explained. While a number of different types of catalysts have been examined, the question about how the observed results can be generalized will always remain and the team should try to address this issue.
- The dependence of many mechanisms on particle size and distribution is apparent, but other effects such as contaminants in the carbon or other material making up the ink solution are not explored. It is unclear why scandium was chosen as the other metal to evaluate; the results are underwhelming.
- Most of the work on alloy catalysts was done from one type of preparation of PtCo catalysts, and the results may not be fully representative of alloy catalysts in general.
- A bit more attention to possible changes in area-specific ORR activities might give a fuller picture of catalyst degradation, particularly with a more diverse set of starting materials.
- The project documents catalyst dissolution with outstanding science and powerful tools, but there is no new science here giving direction for what to do to moderate this. The researchers may be filing intellectual property, but there is no indication that they are. It would be desirable to see what deleterious effects come from alloy catalyst dissolution besides the loss of activity, namely performance losses at high current densities due to concentration gradients.
- The project focuses on one-cell temperature. An understanding of the activation energies of the process would be most helpful in determining the trade-offs between durability, performance, and balance of plant.

Recommendations for additions/deletions to project scope:

- This project needs stronger theoretical justification.
- There is not much time left, but doing more thorough statistical analysis to look for second-order effects is recommended.
- The impact of dissolved metal ions from the catalyst in the catalyst layer ionomer, at high current densities, would be very interesting to study because it might be more important than the loss of activity in the catalyst in determining the overall MEA performance.
- This project should expand to look beyond chlorine, even though this is the most likely real-world contaminant. Other contaminants will be there such as sulfur and CO.

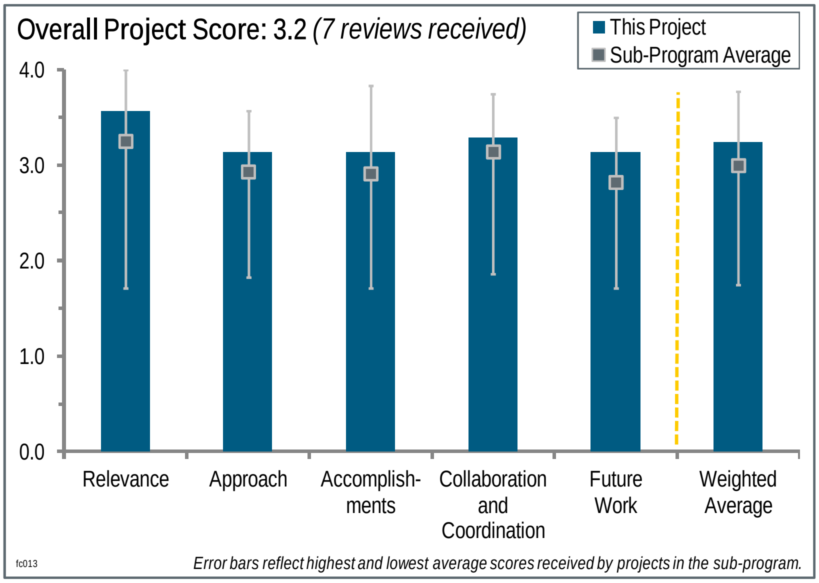
- Because the strength here is the analysis, ANL should collaborate with other DOE project teams that are “best in class” for stable support/alloy and apply the methodology developed here.
- In addition to using waveforms that comply with DOE durability protocols (cycling above or below approximately 1 V), perhaps upsets that typically occur in the stack, such as current reversal or the actual time spent at >1 V during start-up/stop of an automotive stack, can be employed as part of the degradation studies.
- The stated objective is cathodes, and clearly this is where most of the degradation action occurs, but perhaps anodes should also be examined. Even some small efforts to confirm there are no concerns would be useful.

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

Rod Borup; Los Alamos National Laboratory

Brief Summary of Project:

The objectives of this project are to: (1) identify and quantify degradation mechanisms in fuel cells by developing advanced in situ and ex situ characterization techniques to evaluate individual fuel cell components, component interfaces, and component interactions; (2) understand the impact of the electrode structure on durability and performance; (3) develop models that relate components and operation to fuel cell durability, and make an integrated comprehensive model of fuel cell durability available to the public; and (4) develop methods to mitigate the degradation of components.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.6** for its relevance to DOE objectives.

- This project is very relevant to the goals and objectives in the DOE Fuel Cell Technologies Program Multi-Year Research, Development, and Demonstration Plan.
- This project is fairly well aligned with DOE Hydrogen and Fuel Cells Program objectives, particularly in the area of durability.
- The project is relevant to DOE's objectives. Fuel cell component durability is a key area of interest where insights are needed.
- This is a fine beginning for the base case model of durability with a particular construct. The model might not have as much instructive value as hoped for, given the limited material set.
- This project is addressing the most relevant issues facing the fuel cell technical community. If this group is able to contribute to the durability solutions, this project will have been successful and will absolutely contribute to the overall DOE objectives.
- Durability is one of the key catalysts' properties. Improving durability is a very important segment of catalyst development. Knowing the degradation mechanism is the first step in the rational approach to improve durability.
- This is a well-executed project with a focus on the key parameters and interactions needed for high fuel cell durability without added costs. The project has great visibility across all of the DOE projects and Los Alamos National Laboratory (LANL) has used accelerated stress tests (ASTs) that are relevant to the key decay mechanisms.

Question 2: Approach to performing the work

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

- The approach taken is comprehensive, though it appears that there are a large number of parameters/materials being studied and it is not very clear how all of it will be integrated.
- Basically, the approach is good. However, final changes in the material properties by AST are to be correlated with overpotentials and finally I-V performance using a MEA/CL model, which can predict I-V performance.

- The approach involving in situ and ex situ studies, varying catalysts and ionomers, determining components degradation, and looking at structural effects to identify the mechanism of degradation is quite appropriate for the durability problem.
- The approach taken by this team is comprehensive, coordinated, and well organized. All the major barriers are being addressed with well-defined milestones. Although all the major themes are being addressed very well, in the presentation it would have been helpful if a flowchart-like overview of the entire test plan had been shown. This would have been helpful in assessing which tests are being carried out time-wise and would allow comment on how findings from one set of tests might impact the test program in this comprehensive undertaking. Overall, the approach taken by this team in addressing the interrelationships of many variables is long overdue.
- This approach seems unnecessarily broad and because of that many slides have observations or theories that are left out there without further testing.
- The approach to carbon-supported catalysts is good; LANL should have been more focused on that and the overall impact on performance. Looking at platinum (Pt) blacks, 3M nanostructured thin films (NSTF), core shell, and alloy catalysts would be very interesting to see how they affect durability/performance as these are leached into the material. Investigating the impact of additives on performance, durability, and high current operation would have made for a more comprehensive model of durability. Changes in mechanical properties of the PEM MEA with relative humidity cycling seem neglected here.
- The approach overall is very good, with a couple areas for potential improvement:
    - Catalyst degradation: The anode loadings in MEAs are somewhat high. It would be good to see evidence that shows that this is not a factor in the overall performance loss measurements. The low cathode loading data are really interesting. It would be good to see more information on the microstructural changes in the electrode preparation. The lack of decay in the low-Pt loading samples seems anomalous with respect to the industry, so a deeper dive on this would be helpful.
    - Membrane degradation: LANL shows a very interesting effect of the cathode preparation on membrane durability and should consider the effects of this on Pt dissolution, with subsequent effects on membrane durability.
    - Carbon corrosion: It would be good to see the results of electrode degradation combining carbon corrosion and load cycling as seen in practical power plants.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- This project has made good progress against its milestones and appears to be on track.
- The study on “low Pt-loaded MEA” is good. The study and modeling of the individual phenomena is going well. The quantification of material properties decayed by testing is necessary, and finally correlation with overpotentials/I-V should be revealed.
- This project has achieved great progress and insight on electrodes and electrode effects on membrane degradation. There should be more carbon corrosion mitigation combinations tested (i.e., combinations of stable carbon in gas diffusion layers [GDLs] and catalyst layers, PTFE content).
- The accomplishments to date are very interesting and there has been a large volume of work performed. Most of the milestones/targets have been achieved. One thing that did not come through was the work on the integrated modeling of all the degradation processes.
- The authors reported several interesting results that elucidate catalysts’ degradation, including catalyst agglomeration as a function of loading and localized structure changes. It is not clear why no structural changes were observed in the microporous layer (MPL), even though CO_2 originates from it.
- Degradation of the ionomer interacting with Pt is an important observation.
- Some of the results confirmed previous findings, such as the role of increased hydrophilicity with oxidation and the role of dissolved Pt on hydrogen (H_2) and oxygen (O_2) crossover.
- Improved stability has been achieved with some catalysts, but no description of the procedure to obtain it was given.
- LANL has made excellent progress. New findings are leading to new durability perspectives and insight into this multidimensional problem. The team appears to be making progress on all fronts. The only serious weakness is the dependence on students to perform the work. As reported, one milestone was not accomplished

because of the loss of a student. It is also difficult to assess how other pertinent electrode elements are being addressed, if at all. For example, ionomer content and influence in the electrode layer, carbon purity (of the MPL and GDL and electrode support), and pre-treatment of the membrane can and have been shown to impact performance.
- There appears to be a tremendous amount of work being carried out in this project, but it is a mixture of studies that range from very relevant to an exceedingly thorough work of previously well-documented results. This reviewer will be going through the papers to understand the degree to which the review results were snapshots of scattered results or statistically significant results. LANL showed data for areas such as XPS shifts for fluorine binding energy without stating the hypothesis being tested until the backup slides. The open circuit voltage (OCV) data slides are ambiguous and perplexing. LANL stated that, for the aged MEA, unequal anode and cathode pressure data are unreliable due to H_2 outboard leaks, and the analysis is based on these data. Further, the observation that there was no change in OCV going from 10 to 35 psig with the fresh sample is a surprise.
- LANL should carry out pore size geometry measurements on tested catalyst-coated membranes to see how the electrode morphology is changing. It was unclear if this was a porosimetry measurement or BET.

Question 4: Collaboration and coordination with other institutions

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

- Collaboration with partners appears to be going well.
- The project has shown good collaboration between partners.
- Very good collaboration is evident from the contributions of each collaborator.
- This is a well-coordinated collaboration with credible organizations and researchers.
- It seems like all of the groups are contributing proportional to their budget.
- The list of partners and collaborators is extensive. There is good coordination between the partners/collaborators, and this year's additional partners (Nuvera, Argonne National Laboratory [ANL], the University of Nancy, etc.) make sense.
- Great modeling progress has been made with Lawrence Berkeley National Laboratory; good characterization work was done with Oak Ridge National Laboratory, and nice start-up/shutdown (SU/SD) cell development was done with the University of Nancy. LANL should identify the ongoing gap of high-power durability issues at low loadings.

Question 5: Proposed future work

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

- The proposed work is a logical extension of the project's accomplishments.
- The proposed work is clearly derived from open questions that remained unanswered in the reported studies.
- The proposed future work is in order. However, not much was mentioned about the integrated modeling component.
- Much more work is needed to complete the metal and carbon bipolar plate durability studies and the correct areas seem to be captured in the slides.
- This project's plan is OK. The temperature dependence of various degradation phenomena should be investigated to allow people to predict the MEA/cell lifetime for individual operating conditions by interpolation or extrapolation of the Arrhenius plot.
- The future plans presented are appropriate for this effort. It is concerning that there are still many tests that need to be completed within the next 18 months, and there is not much time considering that many tests may have a significant impact on the conclusions of the entire project.
- One significant outcome of this project is the integrated model for fuel cell durability; it will be a baseline to compare against.
- The studies comparing SU/SD cycling with potential cycling will be most interesting and hopefully show high correlation, which would greatly simplify SU/SD testing.

Project strengths:

- The carbon-supported catalyst work is good.
- The theme, network, and collaboration are all strengths of this project.
- The breadth of the team and its experience bring a vast amount of expertise to the project.
- A strong team and strong collaboration are the strengths of this project. The treatment of the durability problem is quite comprehensive.
- This is an excellent team that is addressing key variables in a difficult system (MEA). The available testing resources are very good, as are the communication and coordination.
- LANL effectively uses ASTs to separate and identify the effects of different mechanisms. This project is a great use of MEA variants to show very interesting mitigations for durability issues (electrode and membrane), and a good use of characterization and modeling to register results.
- This is a very good team, which is addressing an important problem. LANL is using a sound approach and proper management with respect to meeting milestones. The researchers are attempting to have modeling in conjunction with experiments. The insights on localized structural changes in carbon were quite interesting.

Project weaknesses:

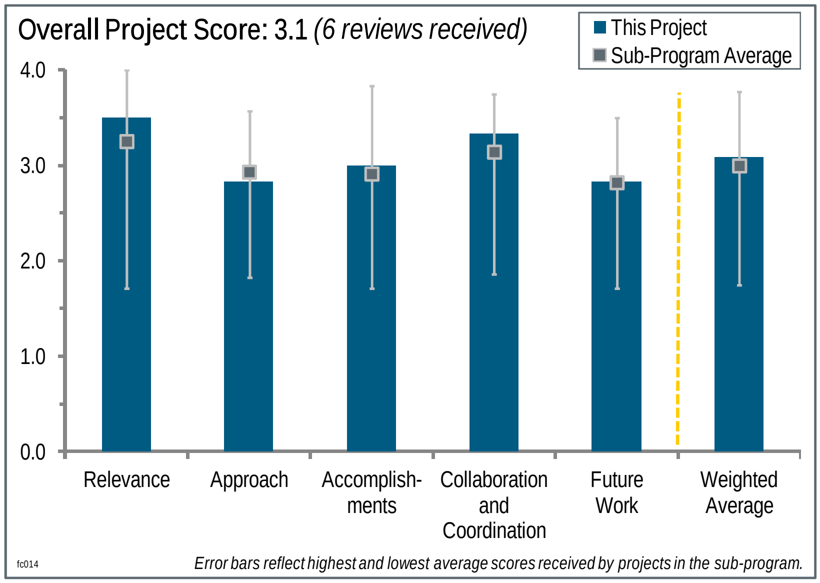
- The quantification of material properties and the prediction of I-V by modeling should be improved.
- A large number of MEA variants have been chosen; however, it is unclear how all of the results can be integrated and generalized.
- It looks like there is some overlap with the ANL durability project; these redundancies should be removed where possible.
- This is a massive (technical) undertaking and researcher continuity is critical. The use of students is clearly understood; however, selected tests carried out by the students may not be completed on time.
- It appears that test result reproducibility was not reported.
- The integrated model LANL will produce does not appear to represent technology in current practice. All ionomers in use have fluorinated end groups and all manufacturers are using some peroxide mitigation strategies. The integrated model may be limited to a particular set of materials.

Recommendations for additions/deletions to project scope:

- The researchers should further explore ways to preclude Pt dissolution.
- At this stage, it is not advisable to add additional elements to the scope of the work plan. This project is already complicated enough.
- LANL should predict I-V performance based on the properties of fresh/decayed materials.
- No further studies on fluorinated end groups are necessary. Original equipment manufacturers use peroxide mitigating additives, and studies looking at the balance of these versus performance and durability might be nice.

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

Scott Blanchet; Nuvera Fuel Cells



Brief Summary of Project:

The objective of this project is to identify and model polymer electrolyte membrane (PEM) fuel cell durability factors associated with low-platinum (Pt) membrane electrode assemblies (MEAs) operating at high (>1 W/cm^2) power densities. The key deliverable of this project is a durability model that is experimentally validated over a range of stack technologies operating at high power.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.5** for its relevance to DOE objectives.

- The project is very relevant, but component development is more critical than overall stack development because stack development does not contribute to companies using different stack configurations and systems.
- This program is addressing critical issues related to fuel cell viability. If successful, this group will provide additional evidence related to the durability questions plaguing the progress of attaining lifetime and performance targets. The objectives and activities in this effort align with the goals of the DOE Hydrogen and Fuel Cells Program (the Program).
- Maintaining high performance at high power densities using reduced platinum group metal content is a key enabler for PEM fuel cell automotive applications. The Nuvera design seems capable of very high power operation, so it is a good system to do this work.
- This project is well aligned with theProgram. The key to developing low-cost, durable fuel cell stacks is to be able to use models and accelerated stress tests (ASTs) to predict the voltage degradation under different operating regimes. This allows industry to conduct the relevant trade-offs to understand technical feasibility and the cost of each of the different system components (including stack).
- Reducing electrocatalyst is a good objective. The data reported indicates changes in the durability with changes in the catalyst loading. The data suggest reduced availability of oxygen (O_2) for the thinner catalyst layers; this is to be expected and should not be considered breakthrough technology, but rather a confirmation of the results from previous studies in the literature. The increased diffusion losses in the thicker cathode catalyst layer are consistent with previous works. The carbon corrosion with unmitigated start stops is expected. It was surprising to see the high reverse-current decay (RCD) not accelerating the degradation of high-loaded Pt electrodes. The objectives of the study were worthwhile; but they only provide confirmation of previous research and development.
- This project addresses DOE's cost and durability goals, and fully supports the Program objectives. Nuvera's approach to improve power density by operating at higher current densities offers cost advantages, but others have seen durability issues when operating in this regime with low-loaded catalysts. This work seeks to address the durability issues associated with operating at the higher current densities. This strategy is applicable to fuel cells outside the automotive arena as well. Automotive original equipment manufacturers (OEMs) have stressed heat rejection as an issue when operating at high current density; however, not all fuel cell applications have as stringent heat rejection requirements (Q/iTD) as autos. In addition, there are advanced thermal management techniques, which could allow for easing of the Q/iTD requirement that could make operating at higher current density more favorable.

Question 2: Approach to performing the work

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

- The durability of catalysts has been evaluated by several groups using various drive cycles, and it has not been considered and utilized, or incorporated in this project.
- The approach taken is appropriate and addresses the durability and performance targets. The key durability issues are being investigated and, if successful, should provide guidance to resolving the lifetime limitations. It is not clear as to how the model will incorporate the different key variables being studied.
- The approach was very good, but there is nothing outstanding about it. If there was something new in the approach, it was not clear. The approach is sufficient to answer the objectives of the work.
- The program has done a good job in comparing performance across stacks, single cells, and subscale cells. There needs to be more about how the project is concluding that the main source of degradation in the low-loaded MEA's effects ionomer degradation. The use of electrochemical impedance spectroscopy to make quantitative determinations of losses within the electrodes can be highly suspect, and it needs to be confirmed with other measurements. The presentation needs to better highlight the way these conclusions are reached. The same must be said for the determination of the O_2 pressure at the electrode level. Also, the tests need to be run at the proper temperature in order to reject the heat at such high power densities; this factor needs to be included to see if increased degradation results.
- The project team has a balanced approach to the development of understanding of failure mechanisms in operating fuel cell stacks. The project team has done a good job of trying to link the impact of ASTs on voltage degradation performed at the single cell and multiple cell stack level. There appears to be good agreement between the different architectures investigated and between the different groups using those architectures (not a trivial task). The approach could be more relevant if the project team were to consider in more detail the impact of off-spec operating conditions on the fuel cell durability of low-cost MEA options. As an example, in an air-air start scenario, the voltages expected to be seen by the cathode far exceed 1.0 V (i.e., they are outside the stated AST protocol range). The team appears to have touched on this when trying to understand the impact of start-stop operation in NST3 (slide 15). However, it appears that the researchers have not been able to measure half-cell potentials to indicate the degree of stress in terms of voltage that differentiate mitigated and unmitigated. Also, the data shown is only for the high-loaded MEA case. One would hope that they have similar data for the low-loaded case.
- The approach is focused on using modeling and accelerated aging tests and drive-cycle tests to validate models and determine the effects of high current density operation on fuel cell durability. Testing and modeling both Nuvera’s open flow-field design and land-channel architecture makes the work applicable to the whole fuel cell community. It is not clear that the ASTs being used will determine how operating at high current density affects aging, particularly for current densities of 2.5 A/cm^2 and higher. The discussion of aging or of ASTs operating above 2 A/cm^2 is limited. Tests where the cell is operated at the highest current densities for extended times (e.g., a soak at 2.5 or 3 A/cm^2 for 100 hours or more) are not described. The results from some cycling to high current density (lower V of 0.4 V) are shown, but it appears most of the testing has been limited to current densities below 2 A/cm^2. The testing to date appears to be more focused on how aging under normal conditions affects performance at high current density, rather than how operation at high current density affects aging (e.g., NST3 appears to only go to 2 A/cm^2 [from the performance curve on slide 10], and its effect is evaluated on performance at different current densities up to 3 A/cm^2).

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- Some of the studies, such as on Pt loading, have been reported many times for the last decade and the results are not new or interesting.
- It is surprising that the main, salient result is the continued degradation at low loadings during the drive-cycle testing, yet no real attention is paid to how that will be mitigated in the future. This is the main focus of the program, so much more focus on this point is needed. Given the concerns about the assigning of electrode losses earlier, it is hard to see that fundamental progress on the low loading durability will be made.

- These researchers systematically answered questions regarding the stability and durability of the catalyst. It was surprising that the RCD did not accelerate degradation of high-loaded MEAs; this would suggest that much of the higher loading is not participating in the electrochemistry and that it may be unnecessary to have such a high catalyst loading.
- The progress to date has been good, and Nuvera has demonstrated consistency between single-cell open flow field and stack beginning of life performance, temperature and pressure gradients, and oxygen pressure at the catalyst. This project has benchmarked open flow field and land-channel architectures, and found similar aging and limitations for comparisons above 1 A/cm^2 due to the pressure differences in land-channel versus open flow field. The researchers should focus more on how normal aging affects operation at high current density.
- The low-cost MEA design appears to be on the path toward meeting DOE cost targets based on initial performance. However, the voltage degradation observed appears to indicate that there is still a significant hurdle in terms of achieving realistic lifetime expectations (i.e., 5,000 hours). This in itself is an important result. The key deliverable from this project is a validated model and the team appears to be on track. The low- and higher-cost MEA technologies studied have provided good data to which voltage degradation models can be developed from. At present, the correlations between model data and AST data (slide 6) are in good agreement for only a couple of cases. This would generally seem to suggest that there is still some more refinement required.
- There results presented are very good. It would have been helpful to understand what thermodynamic studies were carried out and what they yielded in relation to the evaluation. It also appears the data on slide 6 regarding Pt dissolution was force fit. The data looks quite linear but with a Y-axis offset. Many other questions are raised based on the waveform used, but they were not discussed in the presentation. Furthermore, it is not clear whether the statements on the progress slides are observations or conclusions. The reproducibility of the data should have been reported. More data on all that is known on the MEAs would have been beneficial. If the MEAs are proprietary, it is unclear how one will know what to correlate with what on a fundamental basis.

Question 4: Collaboration and coordination with other institutions

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

- Sufficient collaboration exists in this project.
- The partners seem to be engaged based on the data presented.
- The collaborative efforts are well balanced and appropriate.
- The program has established a highly qualified collaboration team.
- Collaborations within the project are well coordinated and appear to be working smoothly. Integration of the modeling, characterization, and testing efforts is excellent.
- It seems that the benchmarking done with Los Alamos National Laboratory and the post-test collaboration with Oak Ridge National Laboratory have been well executed, although these are the easiest. It seems that the modeling work with Argonne National Laboratory has only been applied to Pt dissolution, which is not the key phenomenon for the degradation. With 70% of the project complete, the model needs to have progressed to the point where quantitative reconciliation with the high-power, low-loading decay is occurring.

Question 5: Proposed future work

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

- This work is consistent with the scope and project objectives.
- The future work does not clearly highlight the need for the model development to include the right mechanisms for the key decay phenomena, nor does it outline a credible plan for experimental validation of the model.
- How the future work will increase durability of fuel cell systems is not addressed. The proposed future work appears to be more basic science than direct benefit to commercial fuel cell development.
- There is some question as to whether there is satisfactory data available to begin building the Pt dissolution model. The researchers should decide what additional experiments need to be performed and what other electrode factors need to be understood, such as percent Pt on the carbon, ionomer content and influence, and micro-layer characteristics (i.e., pore size distribution and d_{10} data).

Project strengths:

- This project has a strong team and good systematic analysis of the data.
- The high-power-density stacks are operated with realistic loadings. There is good benchmarking across experimental platforms.
- This project has good collaboration partners, consistent experimental data sets, and a balanced approach of in situ and ex situ measurement correlations.
- This project has an excellent and very experienced team. The team is not resource limited, nor is it without the knowledge of degradation issues.
- This project has been very responsive to DOE and Fuel Cell Technical Team concerns about heat rejection and Q/iTD requirements, and it has begun testing at higher temperatures.

Project weaknesses:

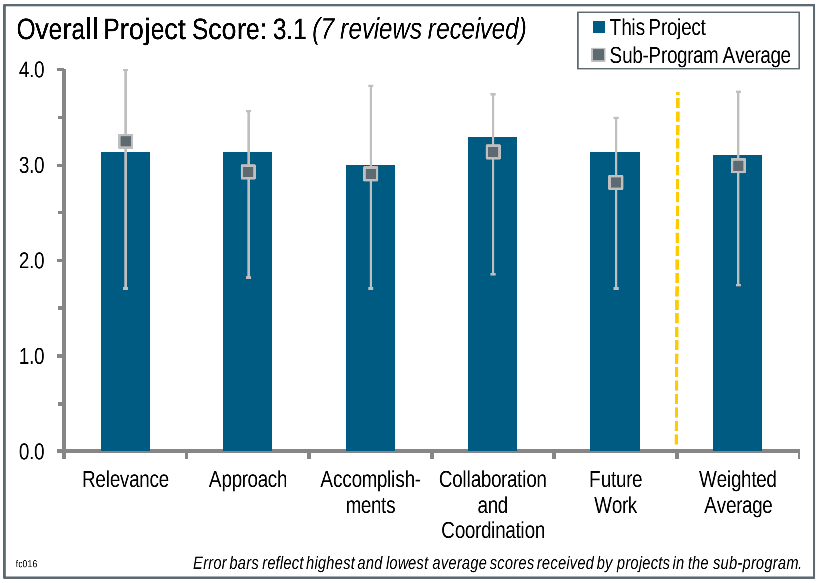
- There is a lack of focus on reducing high power decay with low electrocatalyst loadings, which is the stated purpose of the program. There is a lack of progress on model development and proper diagnostics for model validation.
- This project is repeating the results of many others. It would appear the researchers thought the increased catalyst loading would provide different results than other studies with higher loadings, and that the low-loaded results would yield different data than previous studies on low-loaded results. It is unclear why the researchers and DOE thought this.
- This project does not have enough half-cell voltage mappings of duty cycles. This would help to develop understandings of effective stress load on MEAs and would help develop more effective ASTs. There was no focus on recovery techniques for degraded MEAs or other modes of operation that would help maximize performance from degraded MEAs.
- It is hard to understand the level of detail that will be in the model results. It is unclear how the system characteristics (e.g., thermal gradients in the cell, for one) will be taken into account. It would be good to know about the specifics of the MEA. The potential weakness is embedded in the possible lack of detail.
- It is not clear how much of the testing to date has been at high current conditions versus aging under normal conditions to determine how it affects performance at high current density (e.g., NST3 appears to only go to 2 A/cm^2, and its effect is evaluated on performance at different current densities up to 3 A/cm^2).

Recommendations for additions/deletions to project scope:

- If it makes sense, incorporate operational conditions that the MEA will encounter in a stack.
- The researchers should have the models more accurately address commercial fuel cell systems.
- The researchers should look more closely at linking off-spec operating conditions from duty cycle to voltage degradation.
- More focus on testing at high-current-density conditions would be beneficial, even if OEMs think this is beyond the current densities they could use. Determining the degradation modes under higher-current-density operation would be beneficial to the fuel cell community at large.

Project # FC-016: Accelerated Testing Validation

Rangachary Mukundan; Los Alamos National Laboratory



Brief Summary of Project:

Accelerated stress tests (ASTs) are important because they allow for faster evaluation of new materials and provide standardized tests to benchmark existing materials. The objectives of this project are to: (1) correlate the fuel cell component lifetimes measured in an AST to the real-world behavior of that component; (2) validate the existing ASTs for catalyst layers and membranes; and (3) develop new ASTs for gas diffusion layers (GDLs), bipolar plates, and interfaces.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.1** for its relevance to DOE objectives.

- The correlation of ASTs to real-world experience is valuable information for fuel cell developers.
- This project is relevant to the DOE Hydrogen and Fuel Cells Program. Validating ASTs is important to the industry.
- The project goal is well aligned with the DOE goal of improving the durability of polymer electrolyte membrane (PEM) fuel cells. Newly developed ASTs for GDLs, bipolar plates, and interfaces will provide researchers with a very useful standardized instrument to evaluate new and existing fuel cell materials.
- This is a very important project with good, solid work. Despite only evaluating post-mortem data from bus stacks, the general principles learned may be validly applied elsewhere. It is relevant to the key issue of improving durability.
- Overall, this project is relevant to meeting DOE's durability barriers. However, a significant portion of the project depends on data from buses operated by Ballard using older membrane electrode assembly (MEA) technology for field correlation. The level of mitigation for start-stop within the Ballard program is unclear, and a small number of unmitigated starts can have a significant impact on the catalyst and support durability, which could render the field comparison irrelevant. Further, the ASTs developed for the GDLs seem irrelevant because this failure mode is uncommon.
- This project makes an early attempt at correlating changes occurring in MEAs in a field application (buses) with changes induced by laboratory ASTs. As such, it provides a start toward a critical need in the development and validation of fuel cells for transportation applications.
- The total database from the field was limited to a few used stacks from buses, which apparently used platinum (Pt) loadings higher than those that could be considered for any application in larger numbers. While buses are a good early market application for fuel cells, the modest number of field samples obtained and the limited level of control over the duty cycles in the field have made this project appear a bit premature. For example, it does not appear that there were any tests of different carbon supports in the buses, so all of the support-variation data from the laboratory could not be checked with field data.
- The importance of understanding the durability and life of fuel cell systems makes this one of the most relevant programs supported by DOE. This is an extremely ambitious undertaking.
- The 40,000 hour stationary durability target may be difficult to meet with PEM. The 20,000 hour status was obtained under either laboratory or commercial conditions. If under laboratory conditions, it is not a reasonable durability claim.

- The AST's lack of correlation to "real-world" data needs better definition. It is unclear what "real world" means. Maybe Los Alamos National Laboratory (LANL) should take on the task of defining "real-world" conditions for the multiple fuel cell applications.

Question 2: Approach to performing the work

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

- The approach is well designed and focused on establishing correlations between real-world fuel cell data and the results of accelerated tests to develop reasonable accelerated tests.
- The approach to correlate field ASTs and material property changes cannot be improved upon. The principal investigator has met the milestones required.
- LANL is taking on catalyst, membrane, and GDL issues for a well-rounded approach. It would be useful to see a correlation to more than the limited set of bus data. Light-duty vehicle (LDV) data would be more relevant due to the larger potential market and the very different drive cycle experienced by a vehicle rather than a bus.
- The approach seems sound. It might be useful to understand the sensitivity of the degradation mechanisms to the operating parameters rather than trying to find a single AST.
- The approach is well organized. There was no discussion on the operating history of the bus or a correlation with specific events. The impact of an air/air shutdown was not explained. The operating temperature spikes and the length of time that the fuel cells were at a peak temperature were not explained. The current approach gives an average temperature dependence, but in many cases this unusual event has a greater impact on durability. It is unclear if the anodes or cathodes were ever reactant-starved. All of these performance conditions exist and need to be documented for correlation with ASTs.
- The approach does not include pore size distribution, porosity, or mean pore size of GDL or microporous layer. It is unclear if the researchers think these values are unimportant.
- While it is a very good idea to try to validate ASTs by comparing the results with data from actual applications, the quality of the field data available to this project and the level of information that can be shared about the materials used in the field tests were inadequate for this project to really deliver what its title promises.
- Two different MEA types were used in the bus stacks. While the P5 stacks actually ran in buses, the HD6 stack was a bench test run under a presumed bus operation schedule. One therefore cannot really know whether the observed differences between P5 and HD6 behavior were due to the different materials used or the different protocols. For the P5 stacks, only averaged values of stressors are available, compromising the utility of comparing P5 bus results to results from ASTs. Because the differences between the materials for P5 and HD6 are not fully shared, the fuel cell community cannot draw useful information from this comparison.
- LANL plans to bridge the significant gap between the duty cycles of buses and those of the larger market of LDVs through small-scale laboratory fuel cell tests incorporating protocols intended to mimic light-duty behavior. While this work will be of some interest, it does not permit the "validation" of protocols listed in the project title. Until automotive original equipment manufacturers (OEMs) release samples and service histories from their vehicle fleets, no one will be able to do better on validation under a DOE project.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- Significant progress has been made toward the evaluation of ASTs for catalysts, carbon, and membrane degradation. There is significant progress toward establishing correlations between field and laboratory tests.
- A significant amount of work has been done on catalysts and membrane ASTs. The rate of progress on automotive-related drive cycles and the timing for related failure analysis is unclear.
- LANL has developed good correlations between field data and catalyst ASTs, but it lacks good correlations for membranes and GDLs. Most fuel cell failures occur due to the mechanical breakdown of the membrane (crossover). It would be valuable to spend more time correlating membrane breakdown in the field to an AST. This would help screen new membrane materials more rapidly in the laboratory and avoid the cost of putting faulty materials in the field.
- The accomplishments/progress have produced some notable results that may not have been anticipated, and so they are even more valuable. Quantifying the Pt in the membrane ratios for the field versus AST is one such

result. The second is that there can be significant Pt particle growth as well as carbon corrosion during just the high-voltage hold (maybe without cycling). It is unclear if the mechanism for this sintering is understood. Because this is more of a diagnostic-type project and not a solution-finding project, it is difficult to rate this a 4, but it is a very good, solid project.
- The accomplishments are very helpful and will increase the knowledge base for fuel cell durability.
- It is unclear why high-surface-area carbon is being tested. It is unclear if there was something new in this carbon. Over the past 10 years, high-surface-area carbon has been evaluated and reported in the literature. It is unclear if a new AST will repeat experiments that compare high and low SA carbons.
- In slide 8, the researchers made no comment on the order of magnitude decrease in the number of air/air starts per hour.
- Results on electrochemical surface area (ECSA) and Pt-particle size appear to be a repeat of the last 10 years' work; it was unclear what was new.
- The AST underestimation of Pt in the membrane may mean that the test is invalid or that field tests were operated at harsher conditions.
- For the graphitization data, the initial size of the Pt particles on graphitized carbon compared to Pt particle size on higher-surface-area carbon was not given; it would be interesting to know if this is important.
- Position-in-cell-resolved characterization of bus MEAs is a useful contribution, but the presentation of the data on the "AST/Field correlation-electro-catalyst" slide was confusing, in that increases in ECSA are discussed but increases in crystallite size are what were shown in charts (i.e., inverted polarity of the effect).
- In terms of carbon corrosion, as measured by electrode layer thinning, a correlation of bus and 1.2 V hold data is encouraging, but it is likely to be highly dependent on the exact protocols used in the buses.
- It appears that post mortem characterization (failure analysis) of the bus-stack MEAs is still not complete, even though the researchers are 70% into the project. It would seem that this analysis should have been completed quite early in the project to provide a baseline for characterization of the post-AST MEAs.
- For the Pt in the membrane, it should have come as no surprise that the redeposited Pt lined up near the cathode for the hydrogen/nitrogen cycling AST, while for the bus it is more centered in the membrane; these contrasts have been previously observed and published.

Question 4: Collaboration and coordination with other institutions

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

- A good team has been put together.
- There has been good coordination and collaboration between the partners.
- The collaboration within the project appears to be working well.
- Efforts should be taken to reach out to automotive OEMs to gather any relevant field data.
- This project has well-coordinated collaboration between the national laboratories and industry.
- The collaboration seems reasonable, but it would be good to see a broader range of field data for the correlation. The team should focus on making partnerships with organizations that can provide that data.
- The coordination with partners in this project is apparently limited by the willingness of the partners to provide materials and information on testing for evaluation. It could always be improved upon.

Question 5: Proposed future work

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

- Plans are based on the past progress and are focused on filling the gaps between the results of field and laboratory tests.
- The proposed work builds on past work and expands to meet some of the critiques from last year.
- Completion of the automotive drive-cycle durability test and any related failure analysis is critical.
- The future work should yield data for a correlation between bus data and laboratory data. Failure analysis of bus stacks should quantify performance and durability losses with operation conditions and operation events. This did not appear to be mentioned in the presentation.

- The team is focused on improving the understanding of the relationship between ASTs and field data for GDLs and membranes. This is very important work to focus on.
- The researchers need to finally complete the failure analysis on the old bus stacks. Auto drive-cycle testing will be done with small-scale cells and serves as a patch to the project to make up for the limited field samples available. Much of the total impact of the project will ride on what can be accomplished in such tests.
- It is not clear that enough different membranes are being tested to allow a meaningful recalibration of the combined mechanical/chemical cycling AST, nor is it clear that this project is the right place for such a recalibration to be done.

Project strengths:

- The researchers have significant experience at LANL with studying durability issues.
- The ability to run tests and correlate the observed losses using models is a strength of this project.
- This project has a strong team and good knowledge of laboratory accelerated testing procedures.
- LANL is a taking a holistic approach to the problem, including the carbon support, catalyst, membrane, and GDL.
- This project combines extensive experimental effort with modeling. LANL has carried out very careful analysis of different components to make a valid comparison of field and laboratory samples.
- This is one of the few public-sector analyses used in vehicular MEAs, even if they are limited. This project combines thoughtful experimentation and data analysis.

Project weaknesses:

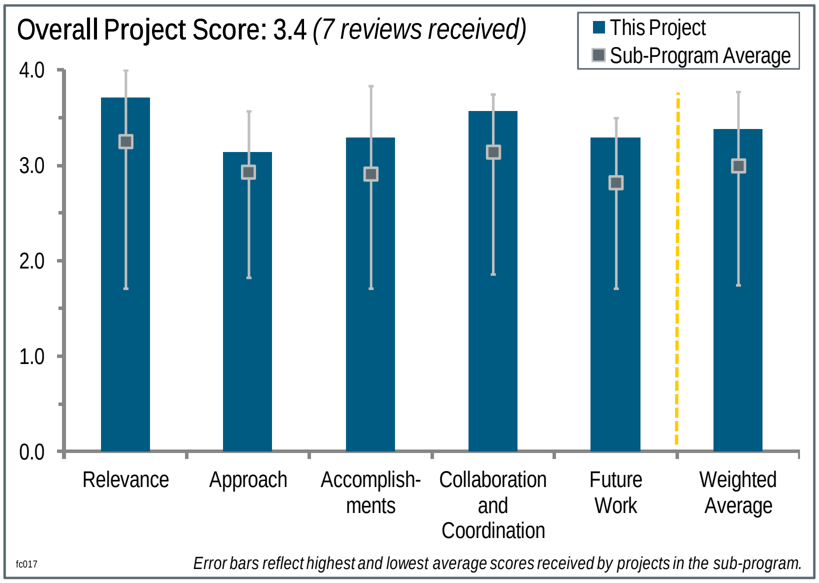
- This project lacks other field data, such as from LDVs.
- This project lacks field data with relevant mitigation strategies for start-stop and chemically mitigated membranes.
- The durability tests at different catalyst loadings overlap with Dr. Borup's project. It is unclear how analysis of the particle size distribution after durability tests was carried out and if catalyst-coated membrane (CCM) cross-sections were analyzed.
- This project lacks full (100%) collaboration with the partners supplying the materials and conditions. This project may be providing data on MEAs that are now old technology. As such, it is important to try to wring-out generally valid principles.
- This project has inadequate field databases with high-loaded electrodes, which will not be carried further into fuel cell development projects, and have only one type of MEA actually used in buses (with another on a bench running a different bus-operation-like protocol). Without a more extensive publicly released field database, true validation of ASTs is still impossible.
- LANL is also unable to disclose publicly the nature of the materials used in the field tests.
- This project is limited in its attempt to correlate laboratory results and commercial performance results (i.e., the ability to catalog all operational events in commercial operations and to assign performance or durability losses in a quantitative manner to operational events.) The overall scope of the objectives may be greater than the available funding.

Recommendations for additions/deletions to project scope:

- This project should add LDV field data.
- The researchers should check if there are any relevant data from the National Renewable Energy Laboratory from the Learning Demonstration project to look at the number of voltage, relative humidity, and start-stop cycles.
- It does not appear that the team is focusing on stationary fuel cells. This should be removed from the scope of the project.
- More careful HRTEM analysis on CCM cross-sections, both for field and laboratory samples, would be helpful. More careful analysis of GDL microstructure, including analysis of hydrophilic and hydrophobic pores, pore size distribution, etc. would help to better validate AST protocols for GDLs.
- It seems like the researchers are collecting the data that seemed to be missing from the presentation, and that they are reporting what they believe are the critical parameters. When an AST is developed, it is unclear whether it will be generalized to all fuel cell systems or specific to the ones (Ballard?) included in this study.

Project # FC-017: Fuel Cells Systems Analysis

Rajesh Ahluwalia; Argonne National Laboratory



Brief Summary of Project:

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

Question 1: Relevance to overall DOE objectives

This project was rated **3.7** for its relevance to DOE objectives.

- This project is very relevant and is one of the most important studies to drive the identification of targets for fuel cell applications.
- This project is highly relevant because it forms the technical basis of DOE's cost model work, and as such it is fundamental to managing the cost aspects of the program.
- This project supports DOE R&D with an independent analysis to assess design-point, part-load, and dynamic performance of automotive fuel cell systems. Stationary systems are not integrated, or were not presented.
- This project provides essential intelligence that informs the research, development, and demonstration plans by providing a comprehensive, fact-based, and data-rich analysis of the actual performance of fuel cell materials and components and how they relate to the overall performance of a fuel cell system.
- The project objectives do support the DOE Hydrogen and Fuel Cells Program (the Program) in a critical manner, but the focus of the principal investigator (PI) recently is too narrow, both with respect to the system options considered and studies undertaken.
- This project is important to DOE for target setting and as input into the high-volume cost estimates by other contractors. The project continues to evolve as more information is learned from industry about simplified designs (such as removing components, etc.).

Question 2: Approach to performing the work

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

- The approach is solid and has remained the same for many years.
- The general approach to this project is sound. More extensive validation would be good, considering the use the model is put to.
- The approach followed is effective and contributes to overcoming barriers. Argonne National Laboratory (ANL) aims to provide a validated versatile system design and analysis tools adapted to handle issues of current interest. Validation on real systems should be investigated in the future.
- The approach is to validate the model against data collected through collaborations with industry and academia. The tests run on a 50 cm^2 cell to simulate stack data should be replicated with a more realistic hardware to

really prove performances. It is strongly recommended that other stack manufacturers are engaged to validate stack data and accuracy.

- The approach to the work is superlative. While this project, at its core, involves the creation of a model system (slide 6) and modeling the characteristics and performance of the materials and subsystems of which this model is composed (passim), the project is exemplary in its integration of actual performance data from a wide range of industry sources to validate and exercise the model in useful ways.
- The critical barriers are cost and durability. However, the PI is presenting minor system improvements (e.g., hydrogen [H_2] purge schedule) instead of studies that can have a major impact on cost or durability. The PI needs to include decay to help the Program understand how this impacts system and vehicle performance, to better understand what different types of decay mean to system design and cost. It would be interesting to know how adding complexity to the membrane electrode assembly (MEA) to mitigate start/stop decay with oxygen evolution reaction catalysts/eliminating carbon supports (such as 3M is exploring in multiple other DOE-supported projects) compares to system-level mitigations (such as those UTC Power has developed and utilized to obtain 12,000 hour durability on fuel cell-powered buses with conventional MEAs).

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- The work is good, but the focus is not on the high-level, broadly applicable studies that would have the most impact.
- ANL is highly productive and provides outstanding value to the Program. The technical accomplishments (analytical results) summarized on slide 6 do not do justice to the myriad analytical results found on slides 7–20 and in the Technical Backup slides.
- The artificial neural network and physical models are a good way to capture the complexities of real systems without a model that is too complicated for the purposes of these studies.
- The calculated optimum purge schedule should be validated against existing fuel cell electric vehicle (FCEV) systems, because the optimal system may not always be able to be implemented in a real-world system for a variety of reasons.
- The model has been exercised to identify trade-offs. Studies suggest that low platinum (Pt) loading has an optimal value, but the data are biased by collaboration with only one stack supplier. More validation of the model on real systems is recommended. ANL should engage system manufacturers. It is clear that the model is capable of representing a huge variety of system conditions; however, it is not clear what the optimal system design is because the studies have not been combined.
- Several good accomplishments were achieved this year. Some—the neural network, for example—are not of the same value as others. To be trusted as generally as is required, this network really needs more data to learn from, and of course it cannot know when it is treating a system that it is improperly set up to handle.
- The nitrogen (N) model is useful, but it may need calibration at more H/N ratios in the feed.
- The studies to determine optimum Pt loading provide an interesting result and also offer an opportunity to validate the model by comparing with experiments. Model predictions on the optimum purge schedule could also be compared with experiment.
- The accomplishments and progress were significant this year and have been well presented. Comments regarding specific components include the following:
  - Model precision: It has been demonstrated that model and experimental precision are comparable. This is an important point for the model validation.
  - Stack: Collaboration with 3M to validate the model for nanostructured thin-film (NSTF) MEAs and stacks was increased.
  - Fuel management: Collaboration with 3M to validate the model for the anode subsystem and to propose a stack purge strategy was increased. It would have interesting to integrate the temperature evolutions during power variations into the N_2 dilution and purge strategy.
  - Water management: The performance of planar humidifiers using Gore's sandwich membrane structure was integrated. The researchers have proposed some improvements.
  - Dynamic performance and drive-cycle simulations: The researchers presented some interesting data on optimum Pt loading, optimum system efficiency, and optimum fuel cell system operating parameters regarding cost. Some surprising results are that the optimum Pt loading (0.25 g/kW) is higher than DOE's

targets and the optimum system efficiency (45%) is lower than DOE's targets. For the last point, the heat exchanger area constraint in a car should be taken into account in the model.

Question 4: Collaboration and coordination with other institutions

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

- Interactions with 3M (stack) and Gore (humidifier) are fairly clear (they provided input data for the model). Honeywell's involvement is explicit.
- The collaboration with component suppliers and dissemination teams appears to be very good and relevant to achieving the project goals. Enhancing the collaboration with other component suppliers would be appreciated to not develop too specific of a model.
- By necessity, this project is highly collaborative with major suppliers of fuel cell materials and subsystems. Furthermore, the well-considered models produced by this project feed into other efforts to determine the actual costs of production for high-volume goods.
- This project has numerous and appropriate connections and collaborations that lead to value for DOE. In addition to the mentioned collaborations, ANL is also working with others on vehicle-level simulations and bringing the value in this project to those projects.
- This project does have sufficient partners to do what it is doing, but not enough to study more broadly applicable issues (e.g., different system designs, different system components, different hybrid vehicles, and durability studies).
- There is a strong level of interaction with several industry partners on particular technology improvements. The project would benefit from a wider cross-section of industrial collaborators, perhaps outside of the U.S. DRIVE Partnership community.

Question 5: Proposed future work

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

- ANL has good plans that will continue to support key needs in the program. A bit more model development on balance of plant (BOP) would help.
- This project looks like more of the same instead of doing what is recommended here, which would have a broader impact than the intended plans.
- More validation on real systems should be pursued. The effect of durability on system trade-offs should be studied thoroughly, including taking the degradation of stack and BOP into account.
- The PI has a firm grasp on what future work will serve the Program well, including his enthusiasm for adding durability attributes into the models and his extemporaneous answer to another reviewer's question.
- A GCTool model of fuel cell systems for forklift applications may not be necessary, because forklifts are now commercial products.
- Establishing closer collaborations with original equipment manufacturers (OEMs), including those outside of U.S. DRIVE, would be beneficial.
- The researchers should identify other suppliers in addition to 3M and Gore for inclusion of novel cost-saving technologies as these technologies are developed.
- The future work proposed is in accordance with the general goals of the project. Validating, calibrating, and documenting the stack model is important because it is for the validation of air, fuel, thermal, and water management models. To do that, closer collaborations with the OEMs and component suppliers need to be established.
- Supporting SA, Inc. in conducting its high-volume manufacturing cost projections and collaborating in life-cycle cost studies are appropriate to ensure global system optimization for cost, performance, and durability.
- Incorporating fork-lift applications is fine.

Project strengths:

- This project has a strong analytical foundation.
- ANL conducts very valuable modeling work, which is validated by tests.

- This project has a long, documented history, excellent team, fine models to base off of, and good responses to suggestions for improvement.
- The open system model is useful for helping to assess different design and system operation options, including cost impact. This project has just enough collaborations to establish at least one system configuration.
- This project is highly focused on DOE's needs for having an accurate and current fuel cell model available to answer fuel cell system-related questions.
- ANL provides valuable and necessary input to the cost modeling of others.
- Results are clearly communicated and suboptimization problems (humidification, purge, etc.) usually provide interesting insights.
- This project provides essential intelligence to the Program and provides great value for its budget. The richness of the data and the careful analytics are exemplary for a modeling project. This work demonstrates a superlative level of technical competence.
- The main strength of this project is the development of an independent analysis to assess design-point, part-load, and dynamic performance of fuel cell systems. This model takes into account many critical technical issues and appears helpful to guide the cost analysis investigated in other projects.
- The collaboration with different OEMs and component suppliers is good.

Project weaknesses:

- ANL lacks access to the newest confidential information from component makers.
- It is a challenge for ANL to keep up with the latest designs being considered by the OEMs, which of course all have their own unique design.
- Assumptions on the stack and system are dependent on specific system and BOP design. No verification of the model against real applications has been pursued.
- Integration of stationary systems should start now.
- The experimental validation of fuel cell systems or systems integrated in cars is not presented.
- As collaboration with 3M has been increased, the MEA and stack modeling may become too specific and not applicable to other suppliers.
- This project is not focused on studies that can have a major impact on the key Program barriers: cost and durability.
- This project includes a very limited component set to date (i.e., NSTF only, externally humidified systems).
- ANL needs more partners to include more component and system-design options.
- There is too much focus on subtle system operating strategies (e.g., H_2 recycling purge strategy), and not enough on higher-level and more broadly applicable studies such as optimal fuel cell power rating.

Recommendations for additions/deletions to project scope:

- ANL should broaden the set of collaborators beyond the U.S. Council for Automotive Research and existing suppliers.
- It is not necessary to include forklift fuel cell systems in future studies, unless the rationale is clearly laid out.
- The researchers should investigate fuel cell systems at higher temperatures, such as 95°C–120°C for automotive systems.
- The model should be validated through comparison with fuel cell systems or vehicle data.
- The modeling of stationary systems should be investigated.
- Collaboration with other component suppliers should be enlarged so as not to become too specific.
- ANL should conduct and disseminate more studies on complete vehicle optimization, specifically optimum fuel cell power rating versus battery rating. Although there were some slides in the backup this year, it is not clear what is being done here—for example, what the battery cost assumptions are and whether projected future battery pack costs are included. Some collaborations between the DOE Vehicle Technologies Program and this project seem warranted to obtain good battery assumptions. For validation (which the PI claimed would be challenging), ANL should collaborate with the projects examining FCEV field data (i.e., the National Renewable Energy Laboratory).
- ANL should analyze how a system will start up from low ambient temperatures, but not necessarily sub-freezing (using the low-temperature performance shown in the backup slides for NSTF). For example, it was

unclear how long it will be before an FCEV will have acceptable power output when started from 30°C. ANL should show the percentage of rated power availability during various start-up operating profiles.

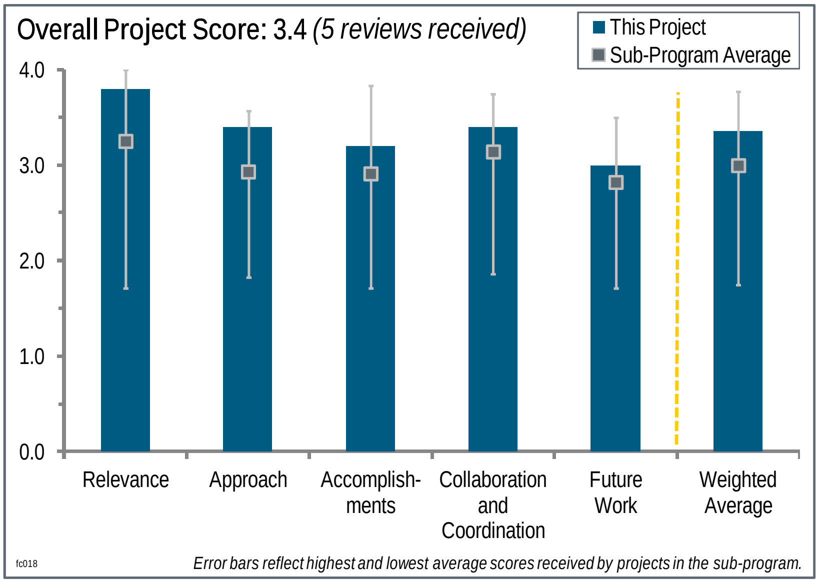
- ANL should analyze MEAs other than those with NSTF. For example, conventional MEAs (Pt/C) with low loadings (0.1 mg/cm^2 on the cathode) now offer performance on par with NSTF, but the performance variation with operating conditions is very different (e.g., temperature sensitivity).

Project # FC-018: Manufacturing Cost Analysis of Fuel Cell Systems and Transportation Fuel Cell System Cost Assessment

Brian James; Strategic Analysis, Inc.

Brief Summary of Project:

The goal of this project is to conduct a process-based cost analysis of stationary, light-duty automotive, and bus fuel cell power systems. The cost analysis can be used to assess the practicality of proposed technologies, determine key cost drivers, and identify the most fruitful research paths to cost reduction, thus providing insight for directing research and development (R&D) priorities for industry as well as for U.S. Department of Energy (DOE) targets.



Question 1: Relevance to overall DOE objectives

This project was rated **3.8** for its relevance to DOE objectives.

- The cost analysis is an important tool for the deployment of fuel cell technology.
- It is imperative for the assessment of technology advancing and target settings in the DOE Hydrogen and Fuel Cells Program (the Program).
- This is absolutely relevant for the DOE objectives because the analysis sheds light on the state of the technology.
- This project provides a valuable output, which satisfies the Program's need to show the drop in fuel cell system cost as the technology gets closer to market. Inclusion of other applications is useful to provide insight into R&D in other parts of the Program.

Question 2: Approach to performing the work

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

- It is good to leverage widely collected data, including suppliers' information.
- The cost analysis addresses all relevant aspects of the technology.
- The approach is thorough and well thought out. This project has good interactions with industry and Argonne National Laboratory (ANL) to provide coordinated outputs. There is significant value generated from the key parameter sensitivity analysis.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- There is very little missing in the analysis.
- This project has a great methodology of analysis. The system designs need to be touched up.
- For the transportation fuel cell area, not enough accomplishment can be seen. Updated technical accomplishments should be incorporated into the cost analysis, such as various materials and process selection of bipolar plate technology. For membrane electrode assemblies, platinum alloy dispersed on carbon (state of

the art) should be analyzed as well as nanostructured thin-film options as a reference. It is unclear why air-compressor cost is so sensitive for the entire system cost. It is basically off-the-shelf technology.

- This project has shown rapid progress on the stationary fuel cell system analysis. It is useful to know that reformer balance of plant, rather than the reactor itself, is the key cost driver. This is the type of value and insight that is typical of this project. The automotive design seems to have stabilized. However, the researchers should prepare to make substantial changes/improvements in the next few years as additional pre-commercial and commercial fuel cell electric vehicles are introduced and innovations are revealed. It is very useful to know that the knee of the curve occurs at 50,000 units per year. As a result of this analysis (if validated by original equipment manufacturers [OEMs]), there should be a concentrated push to enable the market to reach this cost tipping point (incentives, etc.). Federal hybrid electric vehicle incentives from 2006 were sustained to a level of approximately four times this production volume per manufacturer. The tornado chart results were very interesting.

Question 4: Collaboration and coordination with other institutions

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

- Strong industry and laboratory collaborations continue to make this project successful.
- Relevant partners and components have been addressed in order to obtain realistic cost projections.
- It is good that this project incorporated ANL system models for a performance baseline. It is good to communicate with OEMs.
- There should be more opportunity to communicate with other suppliers to increase technical options to be used in the analysis.
- The principal investigator (PI) has done an excellent job communicating with OEMs.

Question 5: Proposed future work

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

- The project should keep watching the U.S. DRIVE Partnership target and assumptions to update the transportation fuel cell cost model.
- Exploring the differences between light-duty and bus fuel cell systems will provide additional insight and perhaps lead to some ideas about how to leverage both of them to reach the 50,000 production volume goal. Medium-duty trucks should be included in the analysis at the same time as the fuel cell buses.
- The assumed performance of the fuel cell technologies should be stated (beginning and end-of-life performance). There is significant uncertainty about the lifetime of the low-cost metallic bipolar plate. If the low-temperature polymer electrolyte membrane (PEM) fuel cell has to use graphite-based bipolar plates, the impact on cost should be included in the risk analysis. The higher value of excess heat for high-temperature PEM and solid oxide fuel cells should somehow be included in the analysis.

Project strengths:

- The methodology is very good.
- This project has excellent solid investigations on cost and very sound results giving valuable guidance.
- The systematic methodology of the cost model and the accumulated database are strengths of this project.
- Clear, methodical analysis leads to a high confidence in the results from this project, which is made even more robust by the added sensitivity analysis. This project gives very interesting insights into what the major cost drivers are for the fuel cell systems analyzed. These results are useful for helping guide where DOE should put future investments focused on cost reduction.

Project weaknesses:

- Some process modeling would benefit the analysis.
- With more than one application included in the DOE Hydrogen and Fuel Cells Program Annual Merit Review (AMR) presentation, it appears to be hard to cover all of the work in one presentation. Perhaps break this up

into two (or more) AMR presentations for fiscal year 2013. No reviewer comments from last year appeared to be addressed in the back of the presentation, perhaps because of the shift in companies.

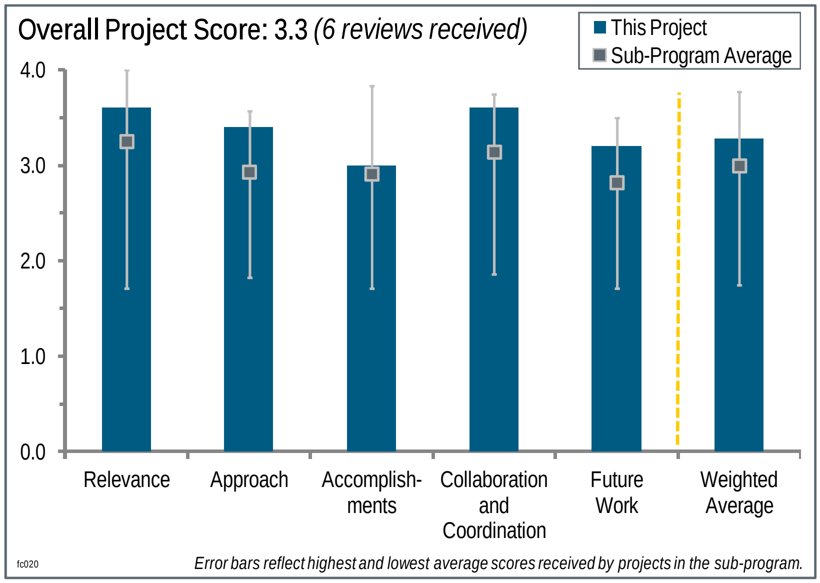
Recommendations for additions/deletions to project scope:

- It is recommended that the researchers look at medium-duty trucks as an interesting application in parallel with fuel cell buses.
- This project should add an industrial review for each system type. This way, one can align the project with performance projections and the best practices of industry. In addition, this project should add metrics to show how this system cost correlates to other similar systems (on a cost/lb basis). For example, vehicles today tend to cost approximately $12/lb of final product. This type of gross comparison will show if the PI's estimates are in the ballpark of manufacturing cost estimates. It would also be very helpful to know how much steel, plastic, copper etc. is in each system, and what the raw material cost is relative to the system cost.

Project # FC-020: Characterization of Fuel Cell Materials

Karren More; Oak Ridge National Laboratory

Brief Summary of Project:



The objectives of this project are to: (1) identify, develop, and optimize high-resolution imaging and compositional/chemical analysis and preparation techniques for the µm-to-Å scale characterization of the material constituents comprising fuel cells; (2) understand the fundamental relationships between the material constituents within fuel cell membrane electrode assemblies; (3) integrate microstructural characterization within other U.S. Department of Energy (DOE) projects; (4) apply advanced analytical and imaging techniques for the evaluation of microstructural and microchemical changes to elucidate microstructure-related degradation mechanisms contributing to fuel cell performance loss; and (5) make capabilities and expertise available to fuel cell researchers outside of Oak Ridge National Laboratory (ORNL).

Question 1: Relevance to overall DOE objectives

This project was rated **3.6** for its relevance to DOE objectives.

- This project has a continuing history of providing insight into fuel cell functions using the newest techniques of microscopy.
- This project is an imperative analytical tool for major technical problems of automotive fuel cell materials and their materials research.
- This project is key to providing an improved understanding of the performance of catalyst materials.
- This is a valuable, core project in the DOE Hydrogen and Fuel Cells Program (the Program). It provides broad access to electron microscopy for projects studying fuel cell materials, in particular the structure and distribution of catalyst particles.
- This proposed effort focuses on using advanced electron microscopy tools for understanding the structure morphology of membranes and catalysts for fuel cell applications. A very wide group of collaborators are included in this effort and represents a concerted and focused attempt to understand some specific phenomenological effects, including degradation. It is very appropriate for the overall DOE effort in the area of fuel cell development for automotive use.

Question 2: Approach to performing the work

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

- It is good to develop the analytical method for ionomer/polymer electrolyte membrane (PEM) degradation. This area should be emphasized. For catalyst work, it is questionable whether it is worthwhile to keep working on platinum (Pt) nanoparticle analysis. It would be good to shift the project focus to novel catalyst structures, such as nanostructured thin film (NSTF), core shell, etc.
- This project provides a high level of "value added" because it goes beyond simple access to electron microscopes. The expertise made available at ORNL, particularly in the area of sample preparation, creates an efficient and productive resource. Having a common electron-microscopy resource for many research projects

capitalizes on the advantages that specialization offers, "leveraging" the knowledge gained in one research project for the benefit of many.

- In general, the applications are done at the state-of-the-art level and are needed at that level (good use of the high-end resources). Of course, there are some questions this method can and cannot attack, but it is useful and powerful for the right projects. The balance of what is done could be improved upon: in situ PEM analysis is probably the most important thing the researchers could do, and development of this technique could be a higher priority.
- The approach is well thought out and appropriately designed for the slated objectives. This year's effort is specifically tailored to understand the dissolution and migration of Pt particles as a function of various operating conditions. It must, however, be recognized that the measurements are very straightforward and that there is no complicated combination of tools and techniques in this effort.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- The usefulness of the results is hard to judge because very few results were presented.
- In accordance with the stated objective, the accomplishments are good and on target.
- Slides 5–18 demonstrate that outstanding progress has been made this year in areas of interest to the U.S. DRIVE Partnership's Fuel Cell Technical Team (FCTT) and industrial collaborators.
- This project has made good progress for ionomer degradation analysis, and a good start on atom probe tomography to characterize NSTF catalysts.
- This project has developed valuable insight into how Pt moves in accelerated stress tests (ASTs), which is important because if mechanisms are different, then the AST may not be general or dependable.

Question 4: Collaboration and coordination with other institutions

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

- This project is working and adding value to many of the best projects in the DOE portfolio. There are many active and valuable collaborations, not simply occasional conversations.
- The collaboration with partners seems good. However, it was not possible to evaluate the success of the partnerships with all the collaborators, most likely due to the time constraints of this presentation. As indicated in slide 19, the collaborations appear to be widespread across the community and progressing well.
- This project is inherently collaborative because it provides an expert technical resource to non-ORNL collaborators. The presentation demonstrated an impressive list of university, industry, and government partners (slides 2, 19, and the technical results slides).

Question 5: Proposed future work

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

- A better translation of the investigations into applying the results should be attempted.
- It is suggested that the researchers focus on the development of new analytical tools (e.g., atom probe tomography). It is also good to focus on ionomer analysis.
- This project has valuable and appropriate plans with good prioritization of the tasks. It would be good to see more in situ fuel cell work.
- The proposed future work (slide 20) demonstrates informed planning to increase the relevance of the project. In particular, the plan is to reestablish a capability for in situ liquid transmission electron microscopy (TEM) after consultation with the FCTT.
- The proposed future work is in good measure with the previous results. The attempts to understand the coarsening of the particles and migrations in the surrounding structures, such as the membrane, are in line with what is required for durability studies. Also, the stated objective of understanding ionomer interactions with catalysts is important. Some further details would have been helpful in understanding how these measurements would further advance knowledge from the current state of the art.

Project strengths:

- This project has good collaboration with many parties interested in this work.
- This project is providing a strong capability of electro-scanning analysis.
- This project has a long history of excellent work, the best quality tools, and is inventing technique over time to push what can be done.
- This project provides technical competence and relevant experience in the field of electron microscopy for the benefit of a broad range of projects within the Program.
- This project reflects the development of a unique user facility at ORNL for performing cutting-edge electron microscopy on interfacial structures used in PEM fuel cells. In this context, a wide network of collaborators has been established across the community of academia, industry, and national laboratories.

Project weaknesses:

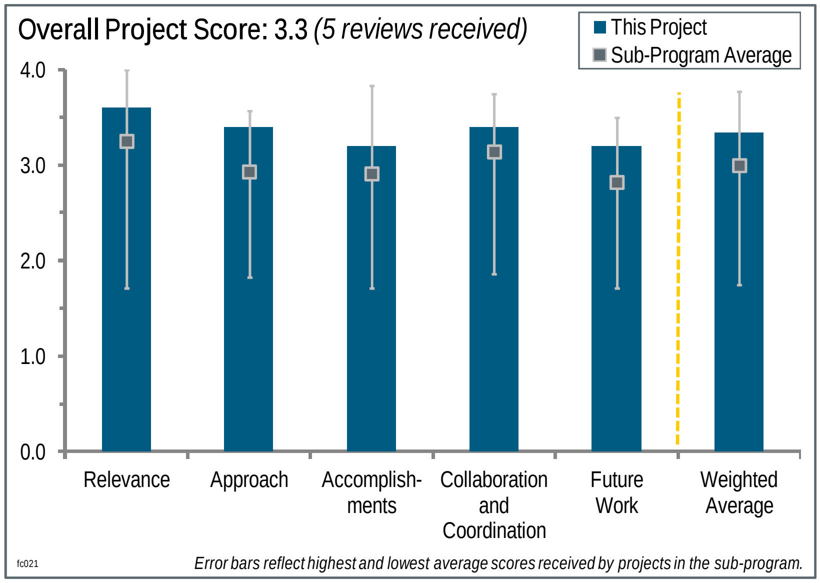
- The principal weakness is that most of the results are “expected.” This effort therefore does not significantly push the envelope of science. This is a problem with all technique-driven programs, which often get drawn into a technique looking for a problem rather than the reverse.
- The objectives of this project include, “make capabilities and expertise available to fuel cell researchers outside of ORNL.” This was on slide 3 in all capital letters. However, in the response to the 2011 reviewer comments, the researchers seemed to backtrack on this, saying, “We do not perform ‘the community’s’ routine microscopy.” Rather, their primary goal is to listen and provide answers. Further, they are also tasked with identifying and developing proper techniques. It is unclear what their balance between research and service is supposed to be, or whether or not they are at the intended ratio, but their project manager can easily review this and make adjustments as necessary.

Recommendations for additions/deletions to project scope:

- Development of the in situ liquid TEM is of great importance, considering the nature of the interface being investigated in this program. This should be emphasized in the future.

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

Muhammad Arif; National Institute of Standards and Technology



Brief Summary of Project:

In this project, the National Institute of Standards and Technology (NIST) will develop and employ an effective, neutron-imaging-based, non-destructive diagnostic tool to characterize water transport in polymer electrolyte membrane fuel cells. NIST will: (1) provide a research and testing infrastructure to enable the fuel cell and hydrogen (H_2) storage industries to design, test, and optimize prototypes for commercial-grade fuel cells and H_2 storage devices; (2) make research data available for beneficial use by the fuel cell community; (3) provide a secure facility for proprietary research by the industry; (4) transfer data interpretation and analysis algorithm techniques to industry to enable entities to use research information more effectively and independently; and (5) continually develop methods and technology to accommodate rapidly changing industry/academia needs.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.6** for its relevance to DOE objectives.

- This project addresses important issues.
- It is probably impossible to achieve a water management project goal without good neutron imaging in real time of real fuel cell hardware. This facility provides a unique capability to DOE and industry researchers.
- The development of this technique for use with fuel development is interesting. If the technique helps understand water transport within a cell, it may meet the requirements of DOE. However, it is unclear how this expensive equipment can be used in cell design for most companies.
- The development of advanced diagnostic tools, such as those at NIST's neutron imaging facility, and making them universally available to researchers and fuel cell developers, provides a critical need to those developing technology to meet DOE's fuel cell targets.
- Neutron imaging is a powerful tool for probing into an operating fuel cell. It can provide valuable information as to the location and movement of water in an H_2/air fuel cell. The NIST facility is well equipped and the NIST personnel are well qualified to carry out the proposed research.

Question 2: Approach to performing the work

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

- This project provides valuable analytical service.
- The approach appears sound, but the end use is unclear. The usage costs are also unclear.
- This project fully addresses barriers by allowing imaging in steady state and, most importantly, during transients. Other good aspects include the environmental chamber for freeze testing; the continually improving resolution; and the integrated approach, which allows both fuel cell components and stacks to be tested.
- NIST recently completed an upgrade of their facility, which includes higher-resolution imaging capability. With the development of these higher-resolution capabilities, the error and uncertainty analysis becomes even more

important, and it would be nice to see more attention to those areas. The new cold neutron capabilities should prove valuable for freeze start/tolerance studies.
- The NIST approach was primarily focused on building hardware components for the imaging of fuel cells and fuel cell components. It was not clear if the NIST researchers also contributed to the interpretation of fuel cell imaging experiments and the planning of future fuel cell experiments.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- The accomplishments are interesting; however, the end use was not clearly evident.
- NIST provided an interesting expansion of tools and important insights for fuel cell developers.
- Limited data were collected at NIST's facility over the past year as the facility was upgraded. It will take some time before one can assess whether or not these upgrades will provide value. Still, many users continue to use NIST's facility as part of the their fuel cell research and development effort, including several that were highlighted in the DOE Hydrogen and Fuel Cells Program Annual Merit Review (AMR) presentation, so clearly the facility continues to provide a great value to fuel cell development in the United States.
- The new hardware installed included a full-scale fuel cell test stand. With 0.10 micron resolution, a 25-fold increase in resolution was achieved. The goal of 1 micron resolution is still to be achieved, but the path to 2 micron resolution was achieved. New capabilities of imaging with cold neutrons are being designed and a new standardized high-resolution test cell fixture was built.
- A majority of the slides dealt with the hardware and test cells that are available or that were developed/constructed at NIST. For example, results were shown to demonstrate that the prior 250 micron resolution has been improved to 25 microns (a 10-fold enhancement in resolution), with an eventual goal of a 2 micron spatial resolution. While such work is important, how neutron imaging is helping researchers better understand fuel cell operation and the role of water during fuel cell power generation is of the most interest. In this regard, seven slides were shown of fuel cell data (of a total of 24 slides in the entire presentation), including a Los Alamos National Laboratory (LANL) study of non-precious metal catalysts, a University of South Carolina (USC) study on transient membrane hydration and conductivity, and a University of Tennessee (UT) heavy water/light water contrast radiography study. More explanation of how the neutron imaging results were improving fuel cell operation or improving understanding of fuel cell operation would have been valuable. It was unclear how important the fuel cell data shown at the AMR were.

Question 4: Collaboration and coordination with other institutions

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

- The list of collaborators is impressive.
- This project has excellent collaborations with industry, national laboratories, and universities that are hard to criticize.
- The nature of this project is to develop advanced tools to meet customers' needs. NIST has clearly been successful in this area, as evidenced by the numerous projects that have effectively been carried out at the facility.
- There were excellent collaborations with other institutions; the NIST facility relies on other researchers who need NIST's neutron imaging capabilities. It was a bit disappointing to see that there was not a closer connection between the collection of data at NIST and the interpretation of the results at LANL, USC, and UT. It was not clear if there were face-to-face discussions of the neutron imaging results by NIST and these outside collaborators. It was unclear how closely the NIST researchers worked with their outside collaborators, and who will choose the next round of experiments—the outside collaborators (in which case the NIST people are just operating equipment) or a collaborative team of in-house and outside researchers.

Question 5: Proposed future work

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

- The proposed follow-on work is interesting and appears to be in keeping with the project.
- NIST is continuing to develop a world-class facility; 1 micron resolution, when achieved, will be a world record.
- The focus on the higher (1–2 µ) resolution could help provide significant learning about transport issues within thin electrode and membrane layers. To date, several studies conducted at NIST have had to use components that are much thicker that those used in actual systems. However, these high-resolution studies will need significant time to run and will require substantial uncertainty analysis to determine accurately the component interfaces, whose roughness is often greater that the proposed high-resolution detector. The planned efforts to increase the field of view while maintaining resolution are certainly warranted.
- The AMR presentation lists future work, but it does not connect such future work to the needs of fuel cell researchers. For example, one future task is to measure the water content in an operating fuel cell membrane electrode assembly with a resolution of at least 5 µ. It is unclear why is this important, under what fuel cell operating conditions such data will be collected, what questions will be addressed in such experiments, how such experiments will advance the understanding of fuel cells, and how it will ultimately help us reach DOE's technical targets. It is unclear if the 5 µ resolution is a NIST scientific goal, or if such a resolution will give fuel cell researchers the information they require to answer fundamental questions regarding the performance of fuel cell components and the methods of fuel cell operation.

Project strengths:

- The strengths are the knowledge and dedication of the project members.
- Lots of great work was still accomplished, despite the reactor being off-line for much of the year.
- The expertise of the neutron imaging people at NIST is unquestioned.
- Interesting data have been collected during the past year and numerous collaborations have been developed/cultivated.
- This project has a very strong collaborative effort with the fuel cell community and dedication to meet customers' needs. NIST has a unique, world-class facility that allows researchers to measure water profiles in situ that they would not be able to do elsewhere. The solid team at NIST continually strives to increase its value and build its customer base.

Project weaknesses:

- The weaknesses are the lack of apparent direct and cost-effective applications to cell and system design.
- The type of analysis NIST and its users do requires significant error analysis to support many of the conclusions that are generated from the experiments that are done there. It was not obvious from the presentation how much effort NIST has spent on these analyses. It is very important that the users of NIST's facility fully understand the limits of its capabilities.
- The connection between the imaging results and improving the fuel cell operation/improving the composition/structure/performance of fuel cell components is tenuous. It is unclear if this is an academic/scientific exercise or if the neutron imaging work is critical to fuel cell development. This aspect was not adequately addressed. More/better interpretation of imaging results should be included in next year's AMR slides. Collaborators should be present in the audience during the presentation to answer questions regarding the rationale for performing the imaging experiments and the interpretation of the imaging results.

Recommendations for additions/deletions to project scope:

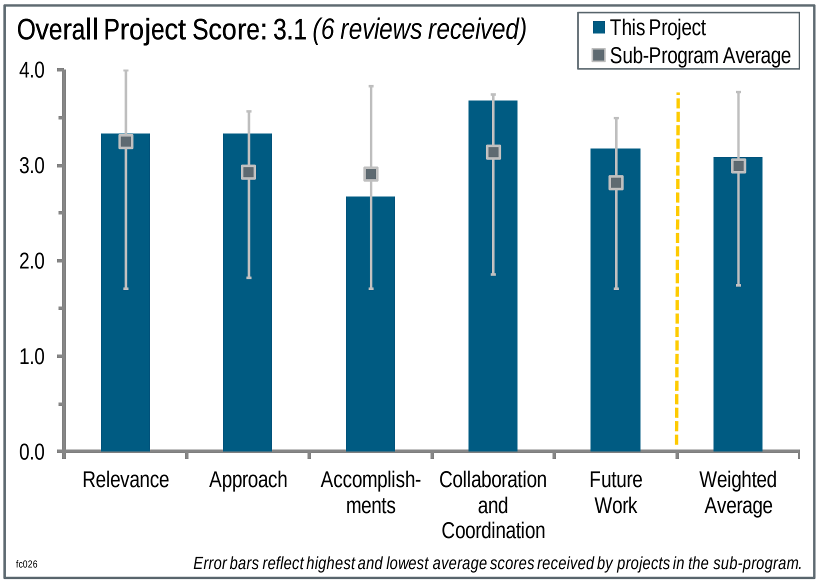
- The project scope seems appropriate, with the one exception of the additional effort spent on error and uncertainty analysis and educating the facility users on the limits of the techniques.
- As presented, the end use is unclear. Clarification on how these data are to be applied and the availability of this device to other manufacturers would greatly enhance the acceptance of this project.

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

Adam Weber; Lawrence Berkeley National Laboratory

Brief Summary of Project:

The objectives of this project are to: (1) build a fundamental understanding of transport phenomena as well as water and thermal management at low and subzero temperatures and (2) elucidate the associated degradation mechanisms due to subzero operation. Improved understanding of transport phenomena, water and thermal management, and durability at subzero temperatures will enable mitigation strategies to be developed to deal with degradation, thus allowing for the U.S. Department of Energy (DOE) targets to be met with regard to cold start, survivability, performance, and cost.



Question 1: Relevance to overall DOE objectives

This project was rated **3.3** for its relevance to DOE objectives.

- This project fully supports the water management and degradation (due to freeze) objectives of the DOE Hydrogen and Fuel Cells Program (the Program).
- This project is improving fundamental understanding of issues affecting not just cold starts, but also performance in polymer electrolyte membrane (PEM) fuel cells.
- Understanding operation and water management at low temperatures where flooding may occur is important, especially for thin electrode structures such as nanostructured thin films (NSTFs).
- PEM fuel cell start-up under cold or freezing conditions is very important for automotive applications. It is important to characterize, understand, and mitigate any damage that may occur with multiple cold/freezing start-ups.
- Understanding subzero start-up was a critical factor for fuel cell electric vehicle commercialization; however, it appears a number of original equipment manufacturers (OEMs) have solved this matter down to -20°C. It is unclear how relevant this project is today, especially with its very narrow focus of only on 3M materials.
- This effort is very well aligned with the goals and objectives of the automotive fuel cell effort of the Program. Understanding the freeze thaw effect of a membrane electrode assembly (MEA) is crucial to enabling this technology for applications. Because the interface is complex—comprising a proton conducting membrane; a gas diffusion media; and an interface composed of porous conducting support, ion conducting polymer, and catalysts—it is imperative to have a holistic approach as indicated in this effort where advanced tools and methods, electrochemical measurements, and computations are combined to provide specific indicators on the effect of freezing conditions in this three-phase interface.

Question 2: Approach to performing the work

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

- Expanding the analysis to examine dispersed electrodes in addition to the NSTF has broadened the potential impact.
- The approach shown on slide 5 is outstanding and is a very good balance of experimental, modeling, and state-of-the-art analytical support. Focusing only on an NSTF catalyst, however, is the major negative of this project.

- This project has a very strong synergistic use of modeling and experiments. The model used is multiscale and includes all of the physical phenomena that are important. The use of experimental data for critical mechanisms related to cold operation and failure is critical. The researchers made a good choice of baseline cell assemblies.
- The top-level approach of addressing freeze and low-temperature operation through a combination of modeling and experiments is the appropriate way to address these complex issues. A quad serpentine cell is probably not the best choice for the baseline cell, because this design is not ideal in terms of water management.
- The approach of using advanced tools and methods, such as GISAX, capillary pressure, DSC, and electrochemical measurements (such as standard polarization and high-frequency resistance, with detailed computation) is comprehensive and very well designed to understand this complex three-phase interface system. The principal investigator has made very good progress in implementing this strategy, as manifested in the results shown.
- Multiple approaches are being employed to characterize and model cold or frozen water effects in simple PEM fuel cell configurations. One wonders if it is possible to employ some additional approaches to better determine where the water/ice is in the PEM fuel cell and to determine its amount in the different areas. For example, it might be possible to introduce a fluorescent dye into the water, if this will not complicate the fuel cell functions.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- Very good progress has been made, as indicated by comparison with the results shown in 2011. Strong and close collaboration between various parties involved in this project is well indicated.
- The overall progress appears good, but there does not seem to be a clear progression to an identification of the key cold start “choke points” in the PEM fuel cell system yet. The inclined sessile drop measurement of hydrophobic and hydrophilic surfaces is good.
- The model validation, and subsequent application, to understand the implications of the measured properties is key. Experimental characterization, particularly catalyst water uptake, is filling knowledge gaps, but the lack of modeling results to demonstrate the sensitivity of measured properties is a concern.
- Good progress was made during the year, but the one criticism is that the team has left a lot to do in the last year to conclude the project. The excellent durability of NSTF in freezing conditions was demonstrated. The study of water uptake in catalyst layers and membranes was very nice.
- The accomplishments to-date are more or less in line with the well-established approach and work plan. What is not clear is why the performance of the 3M MEAs is so poor at lower temperatures (40°C). While subzero start-up is the focus of this project, it appears these MEAs are too poor for even a room-temperature start-up. It is unclear whether this a material issue or a hardware issue; either way, it needs to be addressed. The analytical work done on MEAs post freeze-cycling is very interesting.
- Good progress has been made in component characterization and determining the fundamental properties of the materials. The catalyst layer freeze durability results are interesting. More details regarding the conditions of freeze/thaw cycles would be appreciated. Durability is expected to be highly dependent on the condition of the catalyst layer prior to freezing. The difficulty with obtaining reproducible results between laboratories/cell-testing sites has slowed progress. Model convergence has also delayed achieving milestones, and convergence issues are a concern. This should be given top priority moving forward.

Question 4: Collaboration and coordination with other institutions

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

- This project has excellent collaboration with national laboratories, universities, component manufacturers, and OEMs.
- This project has an excellent set of collaborations designed to cover the range of key programmatic needs.
- This is an outstanding team that has been organized, and the work from each partner is clearly illustrated.
- The teamwork and coordination seem strong; it would be helpful to more clearly identify which members are responsible for the individual tasks and accomplishments.

- Collaboration between the partners is good. A collaboration with an OEM or fuel cell manufacturer experienced with water management and freeze issues with conventional plate and dispersed platinum (Pt) catalysts may be beneficial to insure testing is being done under relevant conditions for freeze and restart.
- Outstanding collaborations are indicated among Los Alamos National Laboratory (LANL), 3M, United Technologies Research Center (UTRC), and Pennsylvania State University. LANL data on fuel cell durability is tempered with 3M supplied materials and tools, and methods such as DSC from Pennsylvania State University. Modeling and parametric analysis is closely coordinated between UTRC and Lawrence Berkeley National Laboratory.

Question 5: Proposed future work

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

- The future work looks reasonable.
- Testing of NSTF with alternative anode gas diffusion layers (GDLs) is important because 3M has seen substantial benefits with different anode GDLs.
- The proposed future work is in line with the progress made. This proposed effort is well in line for making significant contributions to knowledge on freeze tolerances in such PEM interfaces.
- It is great to see that the NSTF with the PtNi alloy will be tested. A very comprehensive component characterization will be achieved, especially as a function of water content. The development of a transient model will be vital.
- The future work plan is acceptable, provided the following two criteria are well understood; otherwise the output of this project will not have so much value to the fuel cell community. The researchers should work to understand why the 3M MEA is so poor at 40°C–55°C, and implement some more benchmark data with traditional Pt/C electrodes.
- There have been a number of studies of water leaving GDLs, both ex situ and in operational cells. It is not clear what the experimental studies will add, though a thorough analysis of the available information and improved model formulation could be valuable.

Project strengths:

- The project features a good multidisciplinary and collaborative approach.
- The project has assembled and excellent team, and has excellent project management and capabilities.
- A strength of the project is its multidisciplinary, multilength-scale combined theoretical and experimental approach.
- The project is well organized, and the team is thoroughly investigating fundamental properties and physical processes.
- The materials properties studies (catalyst water uptake, DSC of freezing in catalyst layer, droplet adhesion force) are excellent.
- This project is well designed to answer the complex question of durability mechanism under freeze conditions. Recognizing the role of freezing in altering the nature of a three-phase interface, as in the case of a conventional PEM fuel cell, this effort combines advanced tools and methods such as SAXS, DSC, and HFR with hierarchical modeling of transport and kinetics. Good progress has been made in implementing this strategy with good collaborative effort.

Project weaknesses:

- This project has no weaknesses.
- It would be nice to see a larger variety of MEA configurations.
- This project focuses too much on 3M NSTF electrodes. The fuel cell data is too low and needs to be understood.
- This project has some model convergence issues, which is a serious concern and must be solved for the project to be successful. The modeling is crucial to understanding the experimental results, and without a robust, validated model there will be little advancement.

- The principal weakness of this effort is that it does not adequately deal with freeze cycle transients, which would manifest itself in a stack. Most of the effort is designed for single-cell-level work; however, there is a complete set of issues that are stack level, which this proposed effort does not address.

Recommendations for additions/deletions to project scope:

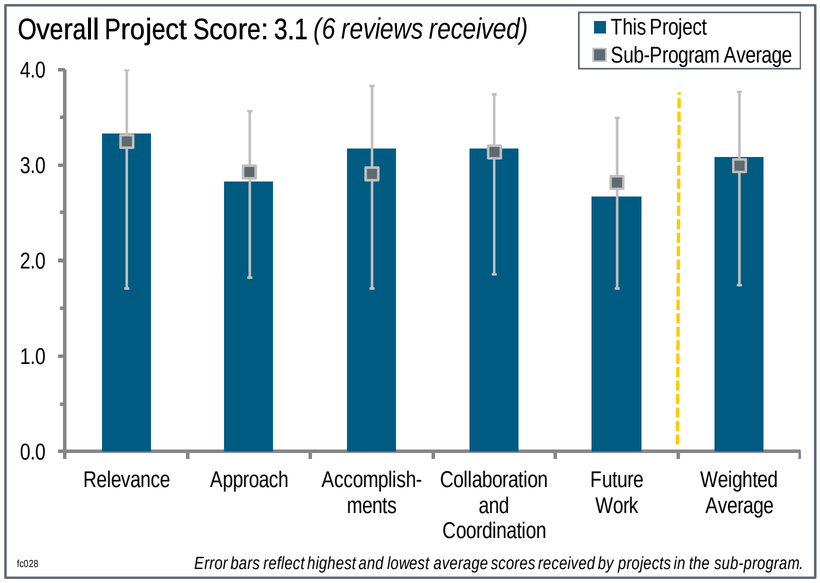
- This project should expand to Pt/C, at least to benchmark and try to improve cooler IV performance.
- The researchers should try to find some better ways to track the water/ice locations and its content in the PEM fuel cell in a cold-start.
- DSC work does not provide the same type of information as IR thermography. There may be other analytical methods that would provide data regarding pore structure and filling with ice or water, such as X-ray tomography to see water/ice interface or advancing ice surface.
- While a close collaborative effort is manifest in this work, the role of 3M seems to be simply related to being a source of electrode materials; its contribution in terms of validating some of the results generated in this work would be more beneficial to the overall program.

Project # FC-028: Transport Studies Enabling Efficiency Optimization of Cost-Competitive Fuel Cell Stacks

Amedeo Conti; Nuvera Fuel Cells

Brief Summary of Project:

The objective of this project is to optimize the efficiency of a stack technology for fuel cells by using a combination of high current density with low platinum (Pt) loadings to meet the U.S. Department of Energy (DOE) 2015 cost targets. A model capable of predicting high-current-density operation in different architectures is the central deliverable of the project. The high-temperature operation has been explored in both single-cell and full-format stack testing to address requirements.



Question 1: Relevance to overall DOE objectives

This project was rated **3.3** for its relevance to DOE objectives.

- The project is focused on achieving fuel cell cost by optimizing design and operating envelopes.
- The project is very relevant, but raising the efficiency of a specific stack may not help other systems that are likely to become commercial.
- The program target cost and performance values for a polymer electrolyte membrane stack are relevant to DOE's objective. The targeted values are focused on national goals.
- The project addresses barriers and technical targets that are critical to the Hydrogen and Fuel Cells Program. When successfully executed, this project could have a large impact in the understanding of water transport within fuel cell stacks.
- A validated multiphase model that is sensitive to controllable material and design parameters is critically needed. The main question is if enough information will be released to enable other developers to exercise the same optimization of stack efficiency.
- This project is well aligned with the DOE Hydrogen and Fuel Cells Program. The key to developing low-cost fuel cell stacks is to be able to use models to predict the performance achievable under different operating regimes. This allows industry to conduct the relevant trade-offs to understand the technical feasibility and cost of each of the different system components (including stacks).

Question 2: Approach to performing the work

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

- This is a sound approach with a clear path forward that employs the strengths of the participating partners.
- The approach is conventional, a combination of design, modeling, fabrication, and testing. The basic thesis was to address target (goal) parameters by increasing current density levels to high values.
- The temperatures and pressures used should be reevaluated, keeping in mind the practical values for radiator size and heat rejection.
- The project team's approach of developing models based on single-cell data and validation at the stack level is sound. Good mapping of operational parameters was carried out for the higher-cost membrane electrode assembly (MEA) case to form the baseline against which the lower-cost MEA could be compared.

- Modeling and comparing open flow field and land/channel architectures establishes the flexibility of the model. Focusing on cost and optimizing designs and operating conditions to meet cost is a unique approach and could reveal weaknesses in the approach of focusing on performance first and cost later.
- The program approach seems comprehensive. However, it is not constrained by the heat rejection target. It seems that this target is regarded as a “nice to have,” and the optimization has continued without this as a constraint. This renders the technology useful for only niche applications. DOE must focus on major market penetration.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- The basic target, performance based on a metric of current for each milligram of Pt catalyst, was demonstrated. There was no convincing evidence that the cost target was addressed or achieved.
- Good progress has been made on the project, and it is clear that the MEA technology developed and showcased in this work has the potential to meet the DOE performance targets. What is less clear is whether the technology has the capability to meet the durability targets. This is not achieved through stable operation at one operating point for a number of hours. It should be assessed effectively with a meaningful accelerated stress test (AST).
- Several new techniques were developed to experimentally determine key parameters important for fuel cell modeling that did not show any agreement in the literature. The presented new techniques show that the team is creative, understands its research topic, and is making exceptional progress. Experimental results indicate improvement in material development to control water management in the stack.
- Performance and modeling targets of the project have been met at low loadings. The projected cost target of $15/kW can be met with low platinum group metal (PGM) loading and high current density in a four-cell, full-format stack. Durability data should be presented, especially at higher temperatures. Q/iTD findings should be reported.
- The model predictions are very impressive. It seems the model can predict open flow field and channel performance very accurately. If this model is this robust under many other operating conditions, it is state of the art. The lack of published results about this model begs many questions, though. A full disclosure of the model in a peer-reviewed forum is strongly encouraged.

Question 4: Collaboration and coordination with other institutions

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

- The project partners appear to be engaged and contributing well to the technical output.
- The research team is adequate for the scope of the project. Input from the auto industry is received through the U.S. DRIVE Partnership’s Fuel Cell Technical Team.
- This is a good team with industry and university partners. There is a clear separation of tasks between partners that requires a significant amount of communication and team work to accomplish the project goals. The progress shows that this collaboration was executed successfully.
- The collaboration seemed to involve vendors and suppliers. It was impossible to discern the roles of each partner, other than it appeared that the university partner was involved in testing and modeling, and that the MEA partner supplied fuel cell hardware.

Question 5: Proposed future work

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

- More details about the material optimization should be disclosed. It is unclear where the magical improvements in efficiency were coming from.
- The team needs to address how the model will be made publicly available for other groups to gain access to and use. The future work on materials optimization should be preceded by AST evaluation of the lower-cost MEA technology.

- The future work section is in line with the proposed approach. It was unclear whether the model would be expanded to a full stack model. More demonstration is needed that show the project made progress in improving the addressed technical targets. No information was shared with respect to a cold start-up time.
- There are five months left. Modeling future work will exercise and validate the model over a broader envelope of temperature, PGM loading, and cell architecture. The model will be published. Experimental work will focus on improved MEAs.
- The project is 90% complete and the future work was essentially to close out the project, show final stack test results, and publish the report. One cannot tell if the results presented have value for future commercial fuel cell hardware.

Project strengths:

- The team is competent.
- The perspective received from a “cost first” approach is valuable.
- This project has good linkage between single-cell and stack data, and it has solid input from collaborators.
- This is a comprehensive project that includes fundamental aspects as well as a development aspect with the target to understand and improve water management.
- This project has strong collaboration with suppliers and universities, and it has demonstrated a clear improvement in Nuvera’s product performance.

Project weaknesses:

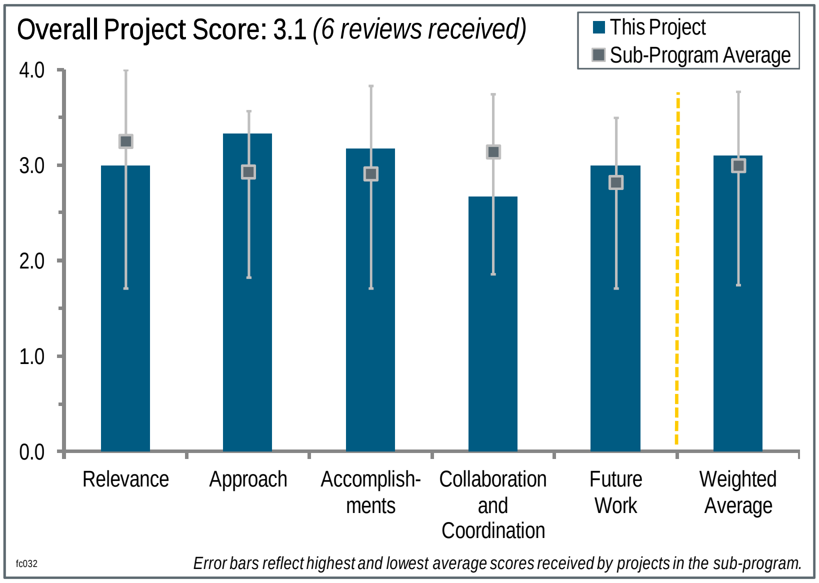
- This project considers performance in isolation of durability.
- It is not clear how the DOE-funded technology improvements will transcend to the broader fuel cell development community.
- The data was not really convincing. There was concern (expressed in previous DOE Hydrogen and Fuel Cells Program Annual Merit Review comments) that the proposed operating temperature was too low. Therefore, operating temperatures were increased, and it was apparent that it was the intent to go to even higher temperatures. It is true that the balance of plant, especially the thermal management components, get large and costly when operating at low-temperature (room temperature) stack. However, there is reason not to get too hot, because then, for automotive applications, considerable energy loss occurs because the stack must be heated, and start-up energy eats away at the anticipated fuel economy. Temperatures in excess of 85°C usually are not necessary. Even though there was considerable concern expressed about temperature, there was no description of thermal management or any estimates of the cost of that hardware. It would have been appropriate to present information that the earlier review team was correct or not so correct by showing estimated designs and costs for thermal management subsystems for a range of operating temperatures. There was no suggestion of experimental error in measurements. Some data suggested very limited durability, and indeed there is a calculation showing that the stack operates best when the water balance is at net zero (the ratio of so-called back diffusion and electro osmotic drag fluxes is one). This is usually true, as others have shown. However, it usually has proven difficult to park the stack at that point, even for a short time. That concept has not been shown to be useful.

Recommendations for additions/deletions to project scope:

- More details about the material optimization should be disclosed.
- Use of state-of-the-art electrocatalysts that will also help raise the efficiency is suggested.
- The team should demonstrate how start-up/shutdown behavior can be improved with the generated results.
- The team should conduct limited work on ASTs leveraging other project information or capabilities prior to committing funds to further design optimization.
- There is still some time left on the project. One would like to see a series of replicated experiments, even polarization curves. Data at one fixed temperature is fine; there needs to be some limited measurement of durability. Even a run of operation for 24 hours could address those concerns.

Project # FC-032: Development of a Low Cost 3-10kW Tubular SOFC Power System

Norman Bessette; Acumentrics Corporation



Brief Summary of Project:

The objectives of this project are to: (1) improve cell power and stability of the cell building block within solid oxide fuel cells (SOFCs), (2) reduce the cost for cell manufacturing by improving processing yield and productivity while decreasing material consumption, (3) increase stack and system efficiency, (4) meet system efficiency and stability goals through prototype testing, and (5) integrate to remote power and micro combined heat and power (micro-CHP) platforms to allow short- and longer-term market penetrations.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.0** for its relevance to DOE objectives.

- This project is advancing fuel cell technology without relying on a hydrogen (H_2) infrastructure.
- Certain project aspects such as cost reduction and micro-CHP and other applications align with the DOE Hydrogen and Fuel Cells Program and fully support DOE's research, development, and demonstration objectives.
- This work supports DOE's goal to integrate the fuel cell technology into remote power and micro-CHP platforms to allow short- and longer-term market penetrations.
- The project is addressing several DOE goals, including cell and stack power density, cell and system cost reduction, system efficiency, and system durability and lifetime.
- This project has been structured to be responsive to the DOE Office of Energy Efficiency's (EERE's) Fuel Cell Technologies Program (FCT Program) goals and objectives. Originally it was in the Solid State Energy Conversion Alliance program (a government-industry partnership supported by DOE's Office of Fossil Energy), and more recently it moved to the FCT Program, where it has been underway for four years and is approximately 95% complete.
- This program seeks to reduce costs in distributed SOFC power plants. It seems only tangentially related to EERE missions, because it only appears to address very niche markets that seem more appropriate for the military. Some of the progress could be brought to bear on larger SOFCs, although the tubular technology is generally not regarded to be cost effective at larger scales, so the alignment in this respect is not great.

Question 2: Approach to performing the work

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

- This project is focused on key barriers, such as cost reduction and performance stability.
- This project is addressing the appropriate critical barriers of power density, durability, systems integration, and cost.
- The technology development barriers have been clearly identified, and actions have been taken on each.
- The approach taken in this project is to focus on cell performance improvements and improving manufacturability at the cell level, then addressing issues of stack assembly and cost reductions.

- The program has approached cost reduction in credible ways by exploring high power densities, reducing the number of components, improving Takt time, and improving life-cycle costs through durability improvements.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- This project has made good progress toward the objectives of cost reduction and performance stability.
- This project made significant progress early on increasing power density for tubular geometry (although it is still less than planar geometries), and since then it has increased durability (10,000 hour test), worked on systems integration and development of recuperator and cooling, and minimized process steps to reduce cost.
- Significant accomplishments have been achieved during the past year. Current density has been increased to meet project goals with stable, long-term operation. Significant reductions in cell processing times have been achieved, balance-of-plant (BOP) parasitic power losses and the number of fuel cell modules were reduced, and commercial units have been placed in the field (nine months in the field with more than 90% reliability).
- It looks like the project team has made great strides in improving manufacturing cost and quality for the fuel cell stacks, and it has started work on BOP costs. Field demonstration units look like they will deliver the performance needed, but the main question is on durability.
- Researchers were able to maintain voltage stability while increasing current density from 150–250 to 350mA/cm^2 over the life of the program. There is now a fully automated cathode and barrier layer spraying in the manufacturing process. The team also developed a tri-sinter process for tubes, and thermal insulation cost was reduced by 65% with an 85% potential reduction number of parts cut in half.
- The progress on durability seems good, with demonstrated stability at constant power hold, thermal cycling, and variable ambient temperature conditions. However, no life-cycle targets were provided, so it is hard to understand where they are with respect to requirements. The progress on cost reduction is even harder to assess because no information is provided on what the target cost is, nor how the demonstrated cost reduction activities are correctly pareto-ed to address it. The presenter did refer to having cost models, but this was not included in the presentation. If this information is business sensitive, the researchers should use pie/bar charts showing relative magnitudes but omitting values. Related to this is the reduction in Takt time for processing the tubes. While a substantial fraction was reduced, the processing time is still measured in days. It seems that very large production volumes would require huge capital investments, so understanding whether this is feasible is critical.

Question 4: Collaboration and coordination with other institutions

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

- This project collaborates with Ariston Thermo Group (Ariston).
- There are some collaborators, but it is not clear from the presentation what their roles are.
- The project is working with eight strategic partners. They are making units for the military, and some 36 units have been fielded. The 91% availability is good.
- There is an Italian partner (Ariston) that is obtaining a European Certification (CE) marking for a micro-CHP system.
- There does not seem to be any participation by partners, though the overall score should not be negatively impacted because of this.
- Acumentrics claims a number of strategic partners, but it is not clear that they played any significant role in the present project except possibly as a sponsor or customer. The project is heavily focused toward bringing a product to market, and it seems reasonable to maximize the in-house effort as much as possible.

Question 5: Proposed future work

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

- The proposed future work for this project is appropriate.
- The project ends in fiscal year 2012, but the proposed future work seems like a logical extension of the work to date.

- The proposed work focuses on durability, cost reduction, and transfer to markets indicative of a maturing technology.
- The project is essentially complete and the remaining effort will support micro-CHP development, which appears to be reasonable.
- Without understanding the magnitude of the gaps, it is hard to assess how the articulated future work will address these.
- The future work is consistent with program goals: complete RP-20 micro-CHP integration and continue cost reductions on each product platform.

Project strengths:

- This project has made good durability progress.
- This project has been making steady progress over time.
- This project is developing a maturing fuel cell technology that does not rely on H_2 infrastructure.
- This project has strong in-house technical, engineering, and manufacturing capabilities.
- This project is focused on key technical barriers, the development of systems and products, and prototype demonstration.
- It looks like Acumentrics has made great strides in reducing the manufacturing costs of its stacks and that it has a plan of attack for BOP components. It is very promising that the combined heat and power system is on track to get a CE mark.

Project weaknesses:

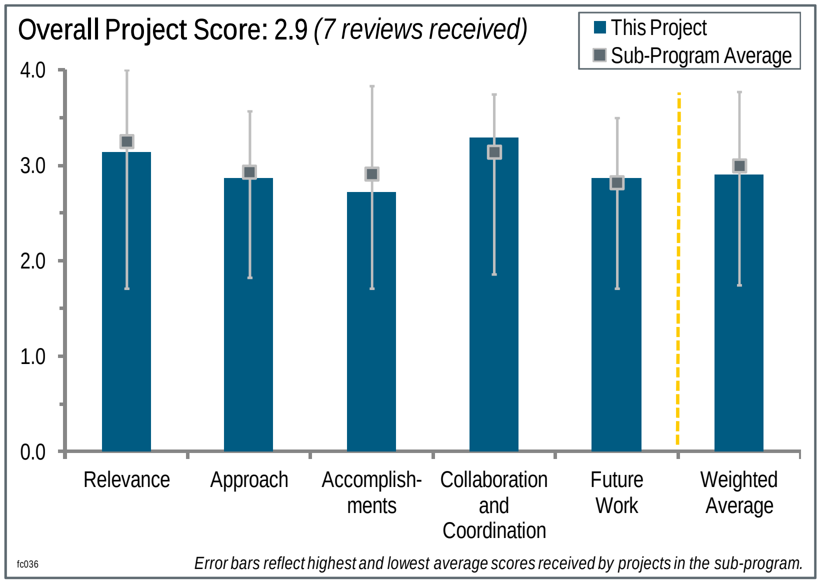
- This project’s metrics/targets are not well defined.
- This project could use further power density improvement.
- The rate of progress seems appropriate, but it might have been faster.
- This project needs to better articulate its requirements and how current/future measures go toward meeting them.
- The required durability for a home appliance has not yet been fully demonstrated, adding to the risk of demonstrating the combined heat and power system.

Recommendations for additions/deletions to project scope:

- This project appears to be near completion. Remote natural gas applications at the wellhead seem to be a good emerging market. The natural gas composition at the wellhead needs to be investigated for compatibility with fuel cells.

Project # FC-036: Dimensionally Stable High Performance Membranes

Cortney Mittelsteadt; Giner Electrochemical Systems, LLC



Brief Summary of Project:

This project identifies pathways to dimensionally stable membrane (DSM) support fabrication. Initially, the project investigated various approaches to identify a scalable and cost-effective route to fabrication. Currently, the project is pursuing three fabrication routes: (1) ultraviolet curing of polymers between a mold and a backing substrate, (2) mechanical deformation via mold pillars, and (3) precipitation of polymers on a mold using a non-solvent.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.1** for its relevance to DOE objectives.

- Durable, high-performance, and low-cost membranes are critical to achieving the overall the DOE Hydrogen and Fuel Cells Program targets.
- This project addresses key barriers in the Program related to durability and the cost of membranes.
- Developing low-cost, dimensionally stable polymer electrolyte membranes (PEMs) is critical to the Program and is also beneficial to hydrogen (H_2) production via electrolysis.
- DOE's objectives include fuel cell cost and fuel cell durability. If successful, this project could impact goals posted for those objectives.
- This project addresses the durability and cost of room temperature to medium temperature PEM fuel cells. This also applies to high-temperature PEM fuel cells. With modifications, it could also apply to solid oxide fuel cells.
- This project aims to increase membrane durability by adding new support layers with micro-fabricated openings. This directly relates to increasing durability. The project also aims to address cost barriers, but direct evidence of cost reduction was not presented.
- Development of new membrane mechanical supports should be a relatively low priority for the Program. Adequate mechanical supports have already been developed by several stakeholders. At this point, improving ionomer properties should be the highest priority in membrane research and development, but this project lacks an ionomer development component (understandably so, given the relatively small size of this project).

Question 2: Approach to performing the work

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

- This project is using an innovative approach that enables low tortuosity and high-temperature stability (120°C).
- This is a nice approach to manufacturing fuel cell membranes with increased mechanical durability.
- Using a strong web and optimizing its geometry to contain an ionomer could be an enabling technology for using polymers that have good conductivity but no strength/crossover resistance.
- This multipronged approach is beneficial and increases the likelihood of success. The mechanical deformation process should work for a wide variety of materials and should be easier with thinner supports. The materials selection appears limited for the ultraviolet (UV) micro replication approach and could involve a substantial amount of work to develop a UV curable thiolene with appropriate mechanical properties.

- The project comes down to building a supported membrane to achieve a strong but thin ion exchange separator component. In fact, almost all useful fuel cell membranes are supported, so the project, which focuses on supported membranes, brings no novelty. This issue then comes to lower cost and higher performance. The approach described were tasks in manufacturing engineering that strive to identify relative costs for three conventional manufacturing approaches.
- The 2DSM approach is inherently limited by the hole size. Fabricating supports with extremely small holes is difficult. The 8 micron holes indicated as the lower limit of hole size are still large enough that a significant quantity of ionomer will not be active (i.e., the ionomer layer on the parts of the support away from the holes will not carry any significant current density and is essentially wasted). The inhomogeneous distribution of the support within the membrane (support-free layers above and below the support) is also a concern in terms of the creation of internal stresses during relative humidity cycling. The approach to support fabrication, which involves the parallel development of three possible fabrication techniques, is good, though more details about the mechanical deformation procedure would be appreciated.
- State-of-the-art membranes already meet durability targets. This project does not show a significant ability to reduce costs over current membrane supports. This project therefore does little to enhance membrane technology over what is already available. Although developing new supports is interesting, there seems to be little likelihood that these supports will be utilized in future membrane electrode assemblies (MEAs). None of the techniques presented has high porosity (more than 50%), which will make reaching conductivity targets more challenging. Also, the in-plane dimension of the nonconductive support layer is on the same magnitude as the through-plane thickness. This may lead to non-uniform catalyst utilization, which may decrease catalyst effectiveness and reduce MEA durability. Routes to mass production are not presented (and are not easily envisioned) for micro-replication and mechanical deformation. It is not certain if any of these techniques can be reduced to practice for less than $\$5/m^{-2}$.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- This project has made good progress on all three parallel methods.
- Some progress was apparent, but it is obvious that the early results are not fully convincing that promise has been demonstrated. Indeed, this micro-fabrication remains a tough task.
- Even though the approach is nice, the lack of progress and accomplishments was disappointing. It is not clear if this was related to funding or the process to manufacture the membranes.
- Membrane supports were fabricated on a bench top level and possible interesting candidate techniques were identified. It is hard to say at this point in the process whether or not these materials will be able to meet target goals once incorporated into ionically conducting membranes.
- A number of ways of achieving the same result have been demonstrated in this project, indicating the approach is valid. The approach did not get rated a 4 because the most favored approach, mechanical deformation, was minimally discussed, because it is proprietary, and so it is difficult to appreciate the validity of this approach.
- The project is progressing well in terms of developing viable fabrication pathways for each of the three techniques. The mechanical deformation technique seems to be lagging behind the others though, with support porosity still far too low. The lack of a schedule with milestones makes it difficult to assess the actual progress versus what was planned. The projected costs are of some concern. The lowest projected cost for the supports is $\$20/m^2$, which is equal to the cost target for the entire membrane; however, the principal investigator (PI) indicated that further reduction would occur with volume.
- Three potential pathways for DSM-support fabrication were developed and all were found to be viable and have the potential for scale-up. The supports were thicker than optimal for PEM fuel cell use. The three methods appear to have been demonstrated with fairly thick (approximately 10 microns) supports. They should be demonstrated with thinner supports (approximately 5 microns and possibly less), which would be used in approximately 15 micron or less membranes. This may prove to be more difficult for some of the approaches. The mechanical deformation samples shown had square holes, which may concentrate stresses at the corners. It is not clear if this would cause problems or if the mechanical deformation technique can provide round holes. The phase-inversion casting process results in porous supports. It was mentioned that thermal treatment can reduce this porosity to about 25%, but the temperature needed and the effect of the porosity on the mechanical

properties of the support was not fully discussed. Current thiolene's mechanical properties were not substantially better than Nafion.

Question 4: Collaboration and coordination with other institutions

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

- This project has very productive collaborations with industry and academia.
- The partners and degree of collaboration are appropriate.
- The collaboration seems to be mainly with vendors.
- The work was well distributed between partners and the collaboration shows synergy.
- This project had a great set of partners that brought a lot of expertise from industry and academia with a nice role.
- It appears the collaboration between the project partners is working well. Collaboration with a PEM ionomer/membrane supplier could prove beneficial in the next step.
- Giner, University of Massachusetts, Impattern Technologies, and Colorado Photopolymer are an effective team. More active participation by an end user might be helpful, although it appears General Motors, Ford, and others are aware of this work and have been giving inputs.

Question 5: Proposed future work

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

- This effort is well focused on achieving the program targets.
- This project is continuing to improve preparations in reasonable ways. Through-plane conductivity work is laudable and should be correlated to all preparations; correlation of the crossover of reactants for all preparations would be interesting.
- The future work path is not clear, and one cannot tell how these supports will be optimized or when ionomer impregnation will be incorporated. The path to reach DOE's goals is not specifically identified.
- The need to down-select to a process and optimize will be critical in the future work, which is believed to be in the last year. MEA fabrication and fuel cell qualification will be key to validating the DSM and should have started before now.
- The future work described sounds appropriate. Given the pace thus far, and recognizing the remaining problems with some of the fabrication techniques, actually achieving all the described tasks remaining is doubtful.
- There is still much to do before the project is complete. Some of the candidate specimens are just now being prepared. These test pieces are the reinforcement and will need to be built into useful membranes. The loaded ionomer parts will then need testing. No credible pathway forward was apparent in the presentation.
- The three methods appear to have been demonstrated with fairly thick (approximately10 micron) supports. They should be demonstrated with thinner supports (approximately 5 microns and possibly less), which would be used in approximately 15 micron or less membranes. The ability of the support to reduce X-Y swelling and chemical and mechanical durability and performance of the membranes should be demonstrated at these thinner thicknesses as well.

Project strengths:

- This project has a strong team and good results.
- This project addresses cost issues for proven 2DSM technology.
- The techniques used for support development allow for a wider variety of materials to be used as supports.
- The membrane and electrode assembly is an important cost issue. This activity works to lower cost.
- This project provides a good membrane that is both dimensionally stable and has the potential to be lower in cost. The proposed manufacturing process seems to be very scalable.
- Pursuing three processes in parallel increases the chances of achieving both the performance/durability targets as well as the cost target.
- Giner Electrochemical Systems is leveraging the expertise gained from work on an earlier DOE project. The relative simplicity of the 2DSM is a strength in terms of ease of fabrication and support/ionomer integration.

Project weaknesses:

- This project has no weaknesses.
- Weaknesses include the low porosity of supports, high cost compared to current available supports, and support geometry that may not maximize durability.
- The inactive regions of membrane ionomer due to shadowing by the support and the possible mechanical problems due to a lack of support distribution throughout the membrane are weaknesses of this project.
- It seems that not much progress has been made since last year. Also, converting these DSM materials into a complete MEA for fuel cell qualification would have been good to start before the last year. Also, more details on the potential cost of such systems would have been nice to see.
- There should have been early work on the reinforcement-ionomer merger. There are many issues with adhesion, mechanical and thermal stability, chemical incompatibility, etc. that are potentially challenging. Much effort has been spent on getting the strengthening component; however, that is just a piece of the target.

Recommendations for additions/deletions to project scope:

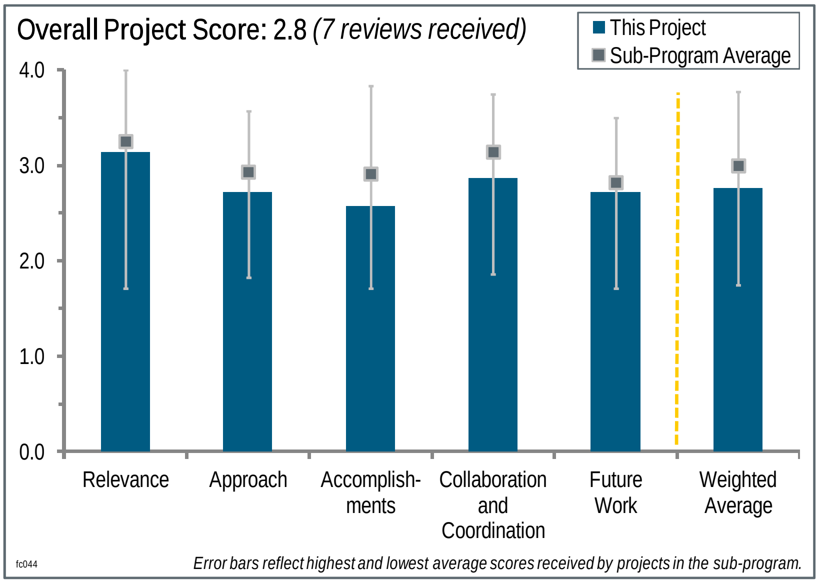
- Performance evaluation data is very important to have before completion of project.
- Possible additions include modeling the current distribution through the pores of the membrane and how this will affect electrode utilization, and load cycling durability of MEAs made with new supports.
- The three methods appear to have been demonstrated with fairly thick (approximately 10 micron) supports. They should be demonstrated with thinner supports (approximately 5 micron and possibly less), which would be used in approximately 15 micron or less membranes. The ability of the support to reduce X-Y swelling should be demonstrated at these thinner thicknesses as well.
- It is too late in the project to change the scope, but for future work the PI may want to consider developing a current density distribution model, which could be used to design a support structure that minimizes effective membrane resistance while maximizing mechanical strength. Such a structure would have to be much more streamlined than the current 2DSM structures.
- The project needs to consider a basic “test” of the concept. It makes sense to pay for a few laser-drilled polymeric test pieces to be made and then to load those specimens with ionomer. (Yes, this test specimen will cost “too much,” but it is not the car part.) The researchers should boil that ionomer-plastic component in water for a few hours, and see if things stay together, then test conductivity. This need not be as thin as the projected component. It makes no sense to move forward with a polymer that cannot work. There also should be some tests on polymer stability and the effects of constituents released from the polymer (the reinforcement), as well as the effects of those constituents on fuel cell durability. Nafion, in its commercial applications, is sold as a reinforced composite using Teflon, which is not a cheap material and not very strong. It can be assumed that Teflon was chosen because of its inertness and that Teflon “debris” does not foul the electrochemical cell. There should be some suspicion about thiolene as a fuel cell component. Certainly sulfate is OK, such as sulfonic acid. However, reduced sulfur compounds tend to poison electrochemical processes and low-cost, commercial polymers are probably not going to be useful in fuel cell components.

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

Eric Brosha; Los Alamos National Laboratory

Brief Summary of Project:

The objective of this project is to develop a ceramic alternative to carbon material supports for a polymer electrolyte membrane (PEM) fuel cell cathode. Ceramic supports require enhanced resistance to corrosion and platinum (Pt) coalescence; can preserve positive attributes of carbon such as cost, surface area, and conductivity; and are compatible with present membrane electrode assembly (MEA) architecture and preparation methods. Goals for the ceramics include possessing the required surface area; fostering high Pt utilization; and exhibiting enhanced Pt-support interaction, adequate electronic conductivity, and corrosion resistance.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.1** for its relevance to DOE objectives.

- This project aligns well with DOE's objectives.
- This project supports all three critical DOE objectives: durability, cost, and performance.
- This project is very relevant because it address carbon support corrosion/oxidation issues. These are major outstanding issues with the durability of fuel cells with respect to voltage degradation. While there are system mitigation strategies that can be put in place, in the longer term, a stable material is required to reduce overall system cost.
- The stated goal is to get rid of carbon because, "in order to make durable ceramic for fuel cells we need to get rid of all carbon." This statement is not necessarily true, and perhaps prevention of carbon corrosion is a better objective. Getting rid of carbon would be great, but it is unlikely that any material will meet carbon's cost and electrical/electrochemical properties.
- This project clearly addresses the durability of dispersed platinum group metal (PGM) catalysts by replacing carbon with an oxide. Whether such a material substitution can address the cost and performance barriers is highly speculative, because the very nature of the enhanced conductivity properties that such oxides must meet will make it harder for nanoparticles to perform as well on a conductive support. The limitations of specific activity and Pt dissolution under high voltage cycling of particles with small radii of curvature will still be present.
- Corrosion-resistant supports for fuel cell catalysts may not be essential for adequate durability because system controls may adequately mitigate stressful conditions, such as start-stop and hydrogen (H_2) starvation. However, corrosion-resistant supports would simplify the requirements on control systems and balance of plant, and could lessen the durability/reliability impacts of control-system malfunctions. Therefore this work, while not necessarily required for the primary path to affordable, durable fuel cells, could contribute to the development of more robust systems. Non-carbon supports are sufficiently difficult to develop that some work to develop them should continue, even if they are not deemed to be the primary path to mass-produced fuel cells. If carbon supports come up short despite all efforts to mitigate the severe stressors, it would be good to have non-carbon supports available as "plan B."

Question 2: Approach to performing the work

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

- The technical approach used in this project is adequate and well defined.
- It is necessary to show that material (ceramic support) screening for corrosion resistance and electrical conductivity may be based on the state-of-the-art carbon support.
- To be successful, Los Alamos National Laboratory (LANL) will have to develop a carbon-free support that still has a large enough surface area. The synthesis will also have to be scalable and the supports will need to fit into an existing MEA.
- It is unclear why proceeding with C-Mo_2C was a go decision.
- With Mo_2N, it may always be a trade-off between having no free carbon and having a high surface area. Both are needed.
- The down-selected approaches have many promising properties among them. There is still some concern that some approaches still involve the use of carbon to get either conductivity ("black" TiO_2 phase) or durability (carbides). This appears to have been born out of the results from the initial accelerated stress test (AST) measurements.
- Although the work presented is good, there was insufficient information to establish whether a structured approach is being used. Theory and modeling are used to support the experimental approach, but it is not clear how much they are guiding the work.
- The correlation of the high-level targets into specific support requirements has not been made clear. For example, it was not clear if there was a minimum conductivity required for the support, or if there is a stability that should translate into meeting the targets. It is not clear if a gated process has been used for down-selection of support materials. The characterization is stated as ongoing, but limited information on, for example, support conductivity has been shown.
- One of the stated goals is to have a support material with comparable conductivity to carbon, which is metallic. Unfortunately the nanoscale oxides and carbides selected, such as the oxygen-deficient titanates and MoC, are prone to hydration and possibly oxidation in the fuel cell cathode environment. The basic question is whether a metallic interface can be preserved between the Pt catalyst and the support, and between the supports in the fuel cell environment. From the electrochemistry, it appears that LANL has very highly resistant electrodes. Thus, there should be much more thought about how to maintain interparticle metallic conductivity and more of an emphasis on surface, rather than bulk, properties.
- The overall primary approach toward generating conductive supports with good corrosion resistance and adequate surface area through templated synthesis (polymer-assisted deposition [PAD]) is, in theory, a good one, because most other syntheses of conductive support candidates do generate quite low surface areas.
- The project has properly placed increased emphasis on electrochemical evaluation of materials this year, but it still needs to improve its methodologies, or at least its reporting metrics. Instead of reporting "Kinetic currents at 0.8 V," LANL should use the standard metric of A/mg PGM at 900 mV RHE. Rotating disk electrode (RDE) PGM loadings should be adjusted to give between 0.5 mA/cm^2 and 1/2 of the limiting current at 900 mV.
- More emphasis needs to be placed on direct measurements of conductivity after exposure to relevant conditions. It was unclear if Ti_4O_7 maintains adequate conductivity after exposure to oxygen reduction reaction (ORR) conditions. It is unclear if nitrides and carbides stay in that form, corrode, or turn into insulating oxides, and if the surfaces of nitride or carbide particles get passivated by an oxide that decreases conductivity to an unacceptable level.
- It seems doubtful that the theory is advanced enough yet to be able to predict adequately whether electronic conductivity will be maintained through the surfaces of these supports when they are in contact with acidic electrolyte over the relevant potential range. This is a central issue in the development of non-carbon supports.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- Good progress has been made so far toward the project and DOE's goals.
- It is necessary to identify the reason for poor fuel cell performance.
- Very good progress has been made. A significant amount of high-quality results have been generated.

- Overall, good progress has been made in electrochemical evaluation by RDE and MEA, even if the results are not that good. At least LANL is showing conclusively that their supports do not work.
- LANL has established improved stability for supports when compared to carbon supports. LANL has also determined that residual carbon is an issue resulting in reduced stability.
- LANL has attempted incorporation into MEAs and MEA testing, although performance is poor due to integration issues.
- The surface area stability of the AST of the zero-carbon-containing oxide (Mo_2N/Pt) shows excessive (60%) Pt electrochemical surface area lost after 10,000 cycles. This may be a general concern because Pt dissolution mechanisms from repeated oxidation and reduction are not so support-dependent and will remain a concern with any dispersed nanoparticles.
- The difficulty of making dispersion ink with very different types of catalyst particles should not be underestimated and may prevent the ability to deduce solid conclusions about the performance or durability of the catalyst particles in MEAs. The best fuel cell performance shown is still an order of magnitude away from state-of the-art Pt alloy/C performance (0.2 vs. 2 A/cm^2 at 0.6 V under 30 psig H_2/air). This is a huge gap to overcome just by electrode processing.
- This is a very difficult area in which to work, and one must expect most of even very well chosen systems to fail, but one might have expected a bit more systematic work to address questions of conductivity and durability.
- LANL has shown that it is difficult, and perhaps impossible, to get carbon-free Mo_2N through polymer-assisted deposition, and it has shown that the significant carbon content contributes to a lack of durability under severe carbon-corrosion tests. This is an accomplishment, even though the outcome is not positive.
- It seems somewhat late in the project to just now be starting attempts to synthesize carbon-free Mo_2N by converting MoO_3, though this is a good idea.
- More corrosion data should be generated and shown, and the issue of loss of conductivity due to the formation of thin layers of insulating oxides on the surface of the conductive particles when exposed to realistic ORR conditions needs to be addressed.
- To maintain conductivity of TiO_2 under ORR conditions, one probably needs metallic dopants rather than just growing an oxygen-deficient oxide. Doping was mentioned in the presentation, but it appears that no data on doped titania were presented. Last year's DOE Hydrogen and Fuel Cells Program Annual Merit Review presentation for this project improperly listed NbO_2 as a conductive oxide, but Nb can be an effective dopant in TiO_2.

Question 4: Collaboration and coordination with other institutions

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

- LANL has limited collaboration.
- LANL has good collaboration with partners.
- LANL's activities are well coordinated among the team members.
- Collaboration with an existing company in the fuel cell area could give ideas for a path forward.
- Collaboration between the laboratories in the project appears adequate. No-go materials are coming from different laboratories.
- The computation work seems to be concentrating on Pt/oxide interactions rather than the more critical surface conductivity issues, but the latter may still be intractable.

Question 5: Proposed future work

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

- An XPS evaluation of the surfaces of these ceramics would be very beneficial.
- The planned work on characterization and fuel cell and durability testing is appropriate.
- The proposed future work is adequate and laid in accordance with the project objectives.
- It is suggested that LANL focus on fuel cell performance before going to the durability assessment.
- Some attempt should be made to characterize surface properties with transmission electron microscopy and XPS, although neither is an in situ method.

- The future work planned appears consistent with the prior years' work, but it is not encouraging that it will overcome the very poor performance and substandard durability that the new materials have shown to date.
- The continued emphasis on totally carbon-free supports seems justified, as when carbon is present it is hard to tell whether Pt supported on the non-carbon phase is electrochemically active.
- The planned attempts to make fine-grained MoO_3 and convert it to the nitride seem wise.
- The return to TiO_2 work is appropriate, but major emphasis should be placed on metallic doping to maintain conductivity.
- Increased emphasis is needed on measuring electron conductivity after exposure to ORR conditions.
- Activity and durability should be measured in MEAs, where conductivity issues become more challenging.

Project strengths:

- Good institutions and personnel are involved in this project.
- A bold approach to change how fuel cell MEAs are built is a strength of the project.
- LANL identified promising supports with techniques used to produce high surface area.
- LANL has a very good team. Each team member brings relevant expertise to the project.
- This is an innovative idea, especially if a non-carbon-containing support with high surface area can be obtained.
- The dedication to get the carbon out rather than using carbon additions to patch up inadequate electron conductivity is a strength of this project.

Project weaknesses:

- Having to add carbon to try and generate the conductivity lost is a weakness of this project.
- Poor performance may require a Pt loading increase in catalyst layers.
- LANL did not show a clear screening or modeling link to support properties/characterization.
- The original concept of using oxide particles as supports for nanoparticles to improve Pt surface area stability is a weakness of this project.
- There is not enough emphasis on controlling and characterizing the interparticle resistances, which are likely to govern the resistances of the electrodes.
- The future of such support is unclear.
- The high cost of the PAD process is also a weakness.
- LANL needs to improve the volume and quality of electrochemical testing. The planned testing in MEAs as well as RDEs is critical; conductivity issues are harder to deal with in MEAs.

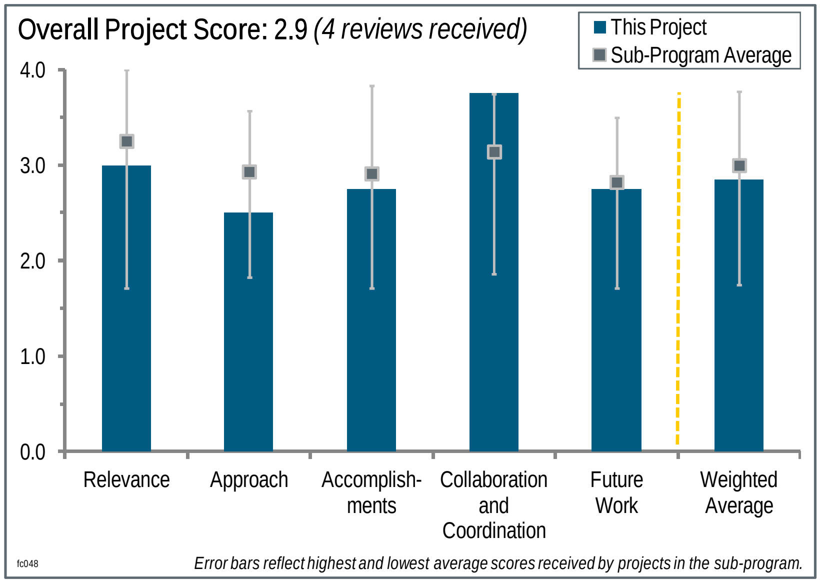
Recommendations for additions/deletions to project scope:

- LANL should incorporate surface analysis of the materials using XPS.
- LANL should include comparisons to most stable commercially available carbon supports.
- LANL should provide structured and gated approaches to selecting and developing supports.
- The electrical conductivity of the electrocatalyst should be measured and included as one of the parameters that can be correlated to the performance of the MEAs.
- The researchers should add direct testing of conductivity in support powders after exposure to relevant ORR conditions.
- LANL should try to initiate a calculational effort to address bulk and surface conductivity issues and not just Pt-binding energies.
- It is not clear that LANL will be able to demonstrate any significant path to overcoming the major barriers addressed by the project. However, the argument can be made that this body of work is justified to answer those very questions about the utility, or lack thereof, of metal oxide supports for dispersed nanoparticle electrocatalysts. It was unclear if going back to the titania supports would really improve the large performance gaps.
- LANL needs to make metallic surfaces, put Pt on them, and then characterize them. LANL could develop a bulk metallic oxide electrode and decorate it with Pt. Some sort of coating to control the oxidation of the oxide/carbide surfaces could be used. The oxide/carbide surfaces need to be characterized.

- LANL should put a very thin layer of their oxides or carbides on carbon. Then they would have all of the benefits of carbon (cost and high conductivity), but eliminate the Pt-C contact, which is the trigger for the carbon degradation.
- LANL's results show that carbon is a pretty handy thing to have in an MEA.

Project # FC-048: Effect of System Contaminants on PEM Fuel Cell Performance and Durability

Huyen Dinh; National Renewable Energy Laboratory



Brief Summary of Project:
Core project objectives are to: (1) identify fundamental classes of contamination in polymer electrolyte membrane fuel cells, (2) develop and validate test methods, (3) identify the severity of contaminants, (4) identify the impact of operating conditions, (5) identify poisoning mechanisms, (6) develop models/predictive capability, and (7) provide guidance on future material selection. Successful completion will increase performance and durability of fuel cells by limiting contamination-related losses and decreasing overall fuel cell system costs by lowering the balance-of-plant (BOP) material costs.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.0** for its relevance to DOE objectives.

- The project seems relevant to DOE objectives and targets, but there is some question as to the relevance of those to hydrogen (H_2) technology.
- This project will allow the manufacturer to select the material of construction (primarily for ancillaries), knowing the impact on performance/durability.
- This project tackles the very important issue of contaminants that are introduced by either the components used for the stack or the BOP or by the cathodic air flow. The potential H_2 impurities are not considered.
- Over the course of the years of fabricating fuel cell electric vehicles, developers have commonly run into failure modes that stem from the release of materials within the system, whether it is from elastomers or metal cations from the BOP, or the release of similar contaminants from bipolar plates and seals. A project that seeks to address contamination from within the system is relevant to development. What compromises the probability that the end results of this project will be relevant to any individual developer is the low probability that an individual contaminant studied will be the same that actually affects a developer. Developers "pick their poisons" by which material sets are down-selected for the various systems and stack components. The premise of this project creates quite a challenge to actually find system-induced contamination that will be experienced universally by developers.

Question 2: Approach to performing the work

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

- The approach is based on the material screening of commercially available materials and those that are available as improved non-commercial qualities from different manufacturers.
- There is some question as to whether the leak tests are truly representative. While it is known that there needs to be some acceleration to get appreciable leak amounts, it is not known what the acceleration factors truly are, and thus this needs to be addressed.

- The specific concentrations of contaminants and model compounds will depend on fuel cell design, so the results will not be directly applicable. The trend in material classification, however, will hold, and the results will help stack designers to choose appropriate materials.
- Ideally, the approach should include studies at low platinum (Pt) loadings that reflect what the status of the technology will be near commercialization. The cathode loading used in this project is 0.4 mg Pt/cm^2, which is much higher than program targets. It is difficult to see how the project will eventually draw conclusions regarding the different contaminants tested. Voltage loss is measured versus total organic carbon (TOC) and concentration, but it is unclear how the project intends to derive conclusions from these data. There would have to be some comparison between an accelerated concentration and a realistic concentration to see what the acceleration factor is, and whether unacceptable degradation would occur during vehicle lifetime. It is unclear (1) how the realistic concentration is determined, and (2) whether the accelerated concentration level causes degradation modes that would not be experienced at the lower realistic concentration.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- This project still needs to develop correlations between ex situ analyses and in situ performance losses, but the project is well on target at this time.
- The project is on target and many tests for many materials have been accomplished. There is a question as to the importance of the dosing concentrations on performance and whether the concentrations are too high.
- Species being released from polymers have been determined; select species were investigated in depth in parametric studies; studies that include a variety of operating parameters are ongoing; and different levels of degradation have been identified and materials have been classified.
- In terms of a gross number of experiments (660), it is evident that the project has done a considerable amount of work. The problem with the project is what has been reported, which does not provide direct conclusions regarding any particular contaminant. Acceleration levels are not reported; all that is reported is voltage loss versus a given TOC and concentration. Some questions remain for each contaminant. It is unclear what the mechanism of degradation is, if the degradation is recoverable at a lower concentration, if contaminants crossover to the anode, and what the mechanism of recovery is, if one exists. It might help to see the Zytel voltage loss plotted versus TOC and concentration. Silicone degradation trends have already been acknowledged in the fuel cell research community.

Question 4: Collaboration and coordination with other institutions

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

- This project has no issues with collaboration.
- The project is large, but it seems well coordinated with a good team. BOP material suppliers, however, are one stakeholder group missing from the team.
- The project consortium is well tailored and consists of academia as well as industry. A wide selection of samples of different manufacturers has been acquired and tested; hence the project is well interconnected with industry.
- The collaboration between the National Renewable Energy Laboratory and General Motors (GM) appears to be strong. GM has helped considerably in the survey of possible contaminants and in developing the criteria by which contaminants are selected for study. The contributions of the other organizations (3M, University of Hawaii, Los Alamos National Laboratory, University of South Carolina, and Colorado School of Mines) are described broadly in one slide, but it is not apparent exactly how their individual contributions advance the project.

Question 5: Proposed future work

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

- The future work is sound.

- There is a lot of good proposed work, which may be a bit overambitious. It should be better prioritized toward more examination of durability and identification of critical parameters. It is not clear that good correlations exist for the proposed contaminants and screening tests. It would be good to focus on some ex situ parameter estimations, such as membrane conductivity and rotating disk electrode testing with the different model contaminants instead of full cell testing initially.
- The project plan does not address the combination of contaminants/model compounds. The testing matrix would become unmanageable, though. It might be interesting, based on material selection recommendations, to check the effect of a combination of contaminants; one could imagine that some material selection could have a negative impact (more than just additive) on the fuel cell performance.
- What is described for future work is fair, and hopefully the project has the statistical prowess to be able to do all of what is described. Considerable data mining will be needed to draw conclusions about the mechanisms involved for each contaminant, and how the effects of each contaminant may vary with concentration and operating conditions. The project has 18 months remaining from the time the slides were due. The question exists as to whether in that time the project will both be able to determine mechanisms for degradation and then be able to enter them into a model that predicts the effects of contamination. It would be interesting to know if there is a model that has been validated for non-contaminated performance, which can then be modified to account for contamination.

Project strengths:

- The screening used is a quick route to good results.
- This project has a strong team with the appropriate tools/techniques, and it should be successful.
- The capability to test both ex situ and in situ with the various expected contaminants is a strength of this project, along with a good team.
- This project has demonstrated the ability to perform an incredible number of experiments in a short period of time. The project has an extensive knowledge base about possible contamination and about which contaminants are likely to exist through fuel cell system operation. The project has interactions with numerous stack manufacturers: GM, Nuvera, and Ballard. The project has been able to establish experimental methods to treat both liquid- and gas-phase contaminants.

Project weaknesses:

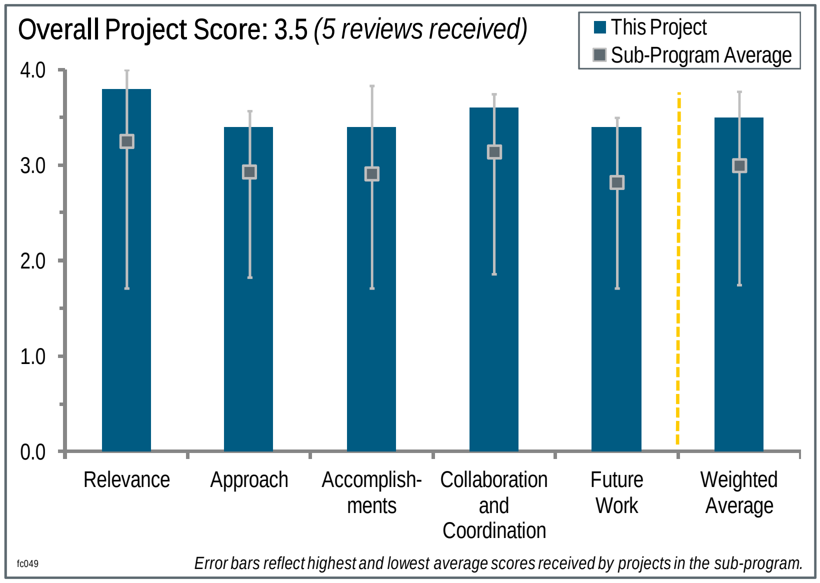
- Very high levels of contaminants are used in this project.
- It is not clear if clear trends or validated simple tests exist and will be generated in this project. It is also unclear as to the output of the screening back to the manufacturers.
- The fact that each contaminant is studied individually may be a weakness, but one needs to start somewhere in understanding the impact of material selection and their contaminants on fuel cell performance.
- The project has had difficulty drawing specific conclusions about contaminants and providing some degree of direction as to how the gathered data might eventually be used by manufacturers to drive component selection. The project needs to identify contamination mechanisms that reach beyond what is already understood (e.g., silicone constituents cause membranes to get brittle and greater amounts of cations reduce membrane conductivity).

Recommendations for additions/deletions to project scope:

- This project investigates lower concentrations of impurities, which are more likely to be present, and at what level critical contaminants are likely to occur.
- Something needs to be done to address acceleration factors and what concentrations are to be expected within the cell. Something like a generation amount versus a removal amount is required to see if various contaminants accumulate in the different fuel cell materials.
- If a baseline model does not exist, it may be worth considering whether developing a predictive model is within the scope of the project. The project should strive to observe trends among contaminants that feature the same functional groups. Some of the prior presentations from this project seemed to indicate that this direction was being followed, but this year’s presentation did not make that seem clear enough. It would be good to add some indication of how contaminants might be selected for long-term durability testing. If time does not permit, perhaps long-term durability testing should be removed from the project.

Project # FC-049: Development of Micro-Structural Mitigation Strategies for PEM Fuel Cells: Morphological Simulations and Experimental Approaches

Silvia Wessel; Ballard



Brief Summary of Project:

The objectives of this project are to: (1) identify and verify catalyst degradation mechanisms, including (a) platinum (Pt) dissolution, transport/plating, carbon-support oxidation and corrosion, and ionomeric thinning and conductivity loss, and (b) mechanism coupling, feedback, and acceleration; (2) correlate catalyst performance and structural changes through catalyst layer and gas diffusion layer (GDL) properties; (3) develop kinetic and material models for aging with a macro-level unit cell degradation model, micro-scale catalyst layer degradation model, and molecular dynamics degradation model of the Pt/carbon/ionomer interface; and (4) develop durability windows of operational conditions, component structural morphologies, and compositions.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.8** for its relevance to DOE objectives.

- The project goals are relevant to DOE's objectives.
- Durability is the most important issue with getting cars on the road. It also paves the way to further reduce Pt loading, and hence serves cost.
- It is necessary to move toward commercial stacks if the United States intends to compete in the fuel cell arena. Ballard knows well the cost, performance, and durability requirements and the project targets national goals.
- This is an important modeling effort aimed at understanding in very fine detail the reaction and decay mechanisms in a polymer electrolyte membrane (PEM) fuel cell.
- Studies of the durability of PEM fuel cell systems and materials are critical for defining the electrode properties and operating conditions that will enable these systems to meet the operating lifetime target of 5,000 hours for the automotive application.

Question 2: Approach to performing the work

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

- The approach used is very good. There is a balance of theory and experiment.
- This project is directly tackling the issues of cell durability and performance by using very detailed analysis methods, which appear to meet experimental results.
- Ballard brings a new design and evaluation technology with the intent of demonstrating a new fuel cell stack. There is considerable analysis, with lots of technical variables. The team's approach is "conventional" (design, build, test), but that is the correct pathway.
- The portions of the project that are performed at Ballard, specifically the model development and sensitivity analysis, appear to have a sound approach and have made substantial progress in the past year. The overall approach of integrating multiple characterization techniques of the various electrode components does not appear to reach the objectives or add value to the approach.

- Because the materials are almost defined in PEM technology owing to longstanding research and development, the issue of microstructural mitigation is key for further advancement. The detailed approaches address the most important issues of catalyst degradation mechanisms and the correlation of microstructural changes and performance, as well as developing kinetic models and material models for aging. Operating conditions are considered in order to identify operating windows other than just keeping developing materials against electrochemical barriers. The targets are precisely defined and ambitious. The project combines cutting-edge approaches, such as molecular dynamic modeling, microstructural modeling of the GDL, and experimental verification in order to identify and understand the mechanisms for degradation, create design curves for degradation, and find operating windows in which the degradation is tolerable or negligible.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.4** for its accomplishments and progress.

- The accomplishments so far are promising. However, it is unclear to what extent this model will be available for other fuel cell developers to use once it is complete, or whether it will be so specific to Ballard technology that it will be of lesser value to others.
- The project began in 2010, and it is far along in its progress. There is emphasis on electric contacts in modeling, which is an important stack issue. This project is still evolving, building on testing results, and it shows good probability that new designs will improve performance and durability.
- A very complex molecular dynamic model of the Pt/C/ionomer interface has been completed and the one-dimensional membrane electrode assembly (MEA) model has been completed and verified. The experimental investigations have been completed, except for the interface characterization and the property changes of aged GDLs and catalyst layers. The overall progress is in line with the project plan.
- The progress shown has been very good and key milestones have been met. Model-based simulations have been experimentally validated. The results on carbon corrosion and Pt dissolution seem to be quite similar to other projects. One issue is the question of model discrimination and parameter estimation. It is not quite clear how many adjustable parameters there are in the model, and how the other parameters have been estimated.
- The project has made significant and valuable progress in developing and validating the microstructural and unit cell performance model. Significant progress was also made in the evaluation of the effects of Pt loading on initial performance and degradation rates, electrode composition, materials properties, and operational conditions on durability. It was not clear from the presentation what accomplishments have been made by the project partners in the past year and what characterization has been performed beyond electrochemical surface area, cell performance, crystallite size, and Pt in the membrane. For example, it is unclear if the University of New Mexico performed the XPS studies of carbon-corrosion mechanisms.

Question 4: Collaboration and coordination with other institutions

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

- The team is quite strong and the presentation was integrated with contributions from the various partners.
- This project has a strong industrial lead provided by Ballard with very specific research contributions. All project partners are renowned or at least well known in their field.
- Collaborations are very strong and bring in many key institutions in the industry. This is reflected in the quality and depth of the work.
- It may just be the way the material was presented or the limited time for the presentation, but there does not seem to be significant contributions from the project partners.
- There are strong partners in the team, and Ballard seems to have its role well characterized. Ballard seemed focused on durability and appreciates that durability is much related to stack operation and maintaining the stack in safe operating parameter space during the entire stack lifetime. Certainly, other successful fuel cell stack manufacturers have also appreciated that a few minutes of operation under adverse conditions may turn out to cause performance degradation that might be observed after a few years of sensible operation.

Question 5: Proposed future work

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

- The proposed future work is appropriate.
- The future work is sound with the project goals and appropriate to achieve the goals.
- The project needs to deliver a stack with demonstrated performance, and it looks like that goal will be achieved.
- The proposed future work shown in the presentation is vague, especially in terms of what future experimental investigations will be performed.
- The future work will provide important insights into addressing performance and durability barriers. However, the sole focus on Pt dissolution and carbon degradation misses the fact that most fuel cell systems in practice fail for reasons other than degradation, such as mechanical crossover failure of the membrane. It is a rare fuel cell that meets its end of life solely due to degradation of performance. Perhaps it is beyond the scope of this project, but it would be good to see some work done to address the actual reasons that fuel cells fail in practice.

Project strengths:

- This project has a very robust analysis.
- This project has a strong consortium that is working toward resolving the critical issues of a real application.
- The emphasis on combining insights from theory and experiments and efforts taken toward model validation is a strength of this project.
- The team is excellent. Ballard demonstrated they know the problems and the proposed sensible solutions to address those problems.
- The project strengths are the careful and extensive experimental studies by Ballard and model development and validation at Ballard.

Project weaknesses:

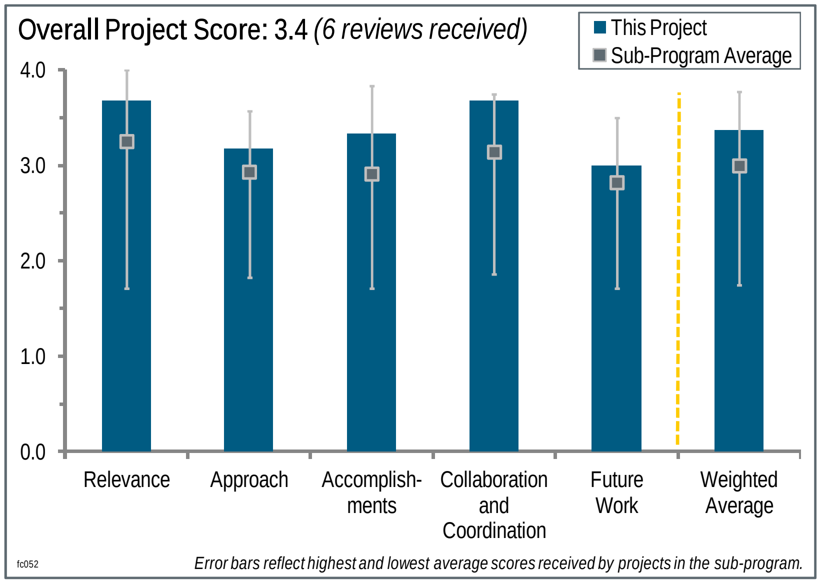
- This project has no notable weaknesses.
- This project is missing key failure modes experienced in practice.
- The lack of evident contributions from project partners and the lack of post-mortem materials characterization are weaknesses of this project.
- Not a weakness as such, but it is important to ensure that overlap with other national laboratory projects (Los Alamos National Laboratory, Argonne National Laboratory) is avoided. It seems like 2–3 projects have similar subsets of partners with different lead institutions. Some input on parameter estimation and sensitivity analysis should be provided.
- There are not really any weaknesses. However, Ballard is a leading stack producer and has sold many stacks within the global marketplace. Because those stacks are proprietary, both in design and performance, it is hard to judge the actual value of this improved design. This makes an informed analysis of the value of this activity difficult.

Recommendations for additions/deletions to project scope:

- The researchers should add other failure mechanisms, such as crossover, poisoning, and membrane humidity issues.
- The researchers should add the effect of temperature on carbon-corrosion mechanisms to the project.
- The concept of a “durability window” is excellent (much like the “engine map” of an internal combustion engine).

Project # FC-052: Technical Assistance to Developers

Tommy Rockward; Los Alamos National Laboratory



Brief Summary of Project:

Los Alamos National Laboratory (LANL) provides technical assistance to fuel cell component and system developers as directed by the U.S. Department of Energy (DOE). This project includes testing of materials and participation in the further development and validation of single-cell test protocols. This project also covers technical assistance to Working Group 12, the U.S. Council for Automotive Research (USCAR), and the USCAR/DOE U.S. DRIVE Partnership's Fuel Cell Technical Team (FCTT).

Question 1: Relevance to overall DOE objectives

This project was rated **3.7** for its relevance to DOE objectives.

- This project serves as a vital link in technology transfer and technical assistance.
- This project is relevant "by definition" because the DOE Hydrogen and Fuel Cells Program (the Program) gives LANL direction. The portfolio described this year is more relevant than last year's efforts.
- This project has historically been well aligned. Because the work is not defined in advance, it is impossible to be sure it will remain so, but it seems highly likely. By offering DOE an independent judgment of methods and material function, this project is critical to the goals of the Program.
- This project is useful for generating the reliable benchmarking of fuel cell components at the domestic and international levels, and for providing technical assistance to the fuel cell and component developers to overcome technical barriers and achieve the goals set by DOE more quickly.
- The sharing of technical expertise developed by the national laboratories with support from DOE is an important and necessary activity for moving fuel cell technology forward. This project provides a great service to industry.
- The relevance of the developer support project was much improved versus last year. This particular year, the project focused more on tasks that addressed more essential questions related to fuel cell performance and durability. Some examples of the more fundamental efforts include: (1) analysis of in-plane currents during start-up/shutdown, (2) discovering improved mass transport with nanotube-incorporating microporous layers (MPLs), and (3) studying the durability of TreadStone plates. The study of plate surface energy versus performance serves as an example of efforts that overlap with work that has been done within the community. Consensus has already been reached that highly hydrophilic plates would be advantageous.

Question 2: Approach to performing the work

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

- The description of how work flows in and the nature of the output was good.
- The project integrates very well with other DOE efforts and the U.S. DRIVE FCTT.
- The technical approach used in this project is adequate. LANL intends to use the standardized fuel cell test procedures to evaluate a variety of stack components objectively to address the main commercialization barriers: durability and performance.

- The approach is a little hard to classify except to say the tests done on the membrane electrode assembly (MEA) or plate level are done at small scale. This is a good and fast approach, though it can miss stack effects. In some past cases, the technology used in the rest of the system was not state of the art, and LANL could miss impacts on the newer systems if they differ from the older ones in response. This issue can only be addressed by providing LANL extra funds to update their equipment, and, if need be, for compelling the makers of novel state-of-the-art components (catalysts, membranes, etc.) to supply LANL with research samples.
- Because the project is dependent upon what developers bring to the project, the approach would not be fairly analyzed based on the particular efforts of the past year. In this project, the evaluation of the approach depends upon how discerning LANL is in understanding whether the tasks suggested from the outside are worthy subjects of investigation or just efforts that duplicate what has already been done. This past year, the project showed some improvement in this regard. There remain some questions regarding the hardware used for the investigators' work. It is worthwhile to contemplate whether the use of quad-serpentine flow fields is still part of the right approach in work that addresses components that may be used in automotive contexts.
- The approach builds on the expertise developed at LANL and other institutions, which is of great benefit to the emerging fuel cell technology. The study of a hydrophobic treatment of bipolar plates appeared questionable. UTC Power has used hydrophilic bipolar plates for more than 10 years. Early work at LANL suggested that thin film transport of water, rather than water "bubbles," was the best method for moving water from the system. The wicking techniques developed in the 1980s by Englehard supported hydrophilic bipolar plate concepts. The researchers should be aware of past results in open literature.
- The start-up/shutdown protocols are in the patent literature. It was unclear if this study attempted to repeat that work and if so, why.
- Testing metal bipolar plates is important. Gottesfeld at LANL conducted some similar corrosion studies that were not referenced in this work. Anode plate corrosion at the outlet is interesting, but the researchers did not say what local potential (electrolyte potential) was needed to develop this type of corrosion.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- A significant amount of high-quality results has been generated in a timely manner.
- Some interesting work was conducted this year, particularly the start-stop studies and the bipolar plate work.
- A good amount of work was completed in many different areas. The understanding gained in the two plate projects was very helpful in understanding corrosion and water management. The other work was extremely helpful to the principal investigators (PIs) involved.
- The correlation between Ballard and Nancy Université data was not explained. For the MPL material studies, characterization of the material and evaluation of performance versus either a previously tested baseline material or a DOE-relevant material, as well as communication of quantified properties, could be improved.
- LANL showed another example of how defects in MPLs enable greater water flux from the cathode catalyst layer by incorporating carbon nanotubes in the MPLs. It would be interesting to see images of the MPLs, but this work serendipitously adds to a growing understanding of water transport. The in-plane currents shown from the start-up/shutdown experiments show that the inlet of the cell contains more in-plane current than the outlet upon start-up. This result could suggest cell designs that could mitigate degradation.
- The study of hydrophilic plate treatments showed higher performance with greater hydrophilicity, which agrees with other studies. The project used a quad-serpentine plate design with small outlets, which may have exaggerated this conclusion. The study of the TreadStone plates showed, under somewhat accelerated circumstances (older membrane that likely has high HF release), where contamination may come from (wetter parts of the cell) and with what metals (iron [Fe], titanium [Ti]).
- The researchers have systematically demonstrated expertise in evaluating several polymer electrolyte membrane (PEM) fuel cell components. The application of the techniques developed by LANL and others has advanced the understanding of the chemical and physical properties of PEM fuel cells. There was no discussion of hydride formation on Ti-coated plates. It appears the researchers did not consider the formation of hydrides (hydride formation has been observed in electrolysis systems).
- It is not clear why the hydrophobic treatment of graphite bipolar plates was undertaken, unless it was requested by an outside source.

- The segmented plate study provides insight into the corrosion mechanisms occurring during start-up and shutdown.

Question 4: Collaboration and coordination with other institutions

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

- The project activities are well coordinated among the team members.
- Evidence is good for strong collaboration with DOE and with the other companies.
- LANL has a large number of critical people with which to collaborate, a very helpful group that supports success in other DOE contracts.
- The team has a good breadth of organizations, including international organizations. Information sharing and communication with other DOE activities is excellent.
- The project is built around collaboration between national laboratories, academia, and industry. This is a great example of cooperative research and development to resolve technical problems.
- The project is entirely dependent upon collaborations, so by necessity the collaborations are close and wide-ranging. The list of collaborators reveals that there could be some improvement in incorporating stack developers. Ballard is the only stack developer listed.

Question 5: Proposed future work

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

- LANL did not show a future work slide.
- The proposed future work is adequate and laid in accordance with the project objectives.
- As described, the future work is appropriate for now, because it seems to be dependent on future guidance.
- This is not really a fair aspect to grade this project on, because the future is not entirely in the researchers' control; rather, they attack the problems DOE directs them to address.
- As future work is determined by DOE, it is difficult to determine the scope of planned projects. An indication of trending topics for assistance or specific problem areas with which LANL may be best able to assist along with better communication of LANL's capabilities, with DOE agreement, may assist in gauging future plans for this project.

Project strengths:

- LANL has good interaction with the industry.
- The expertise and cooperative efforts between universities, LANL, and industry are strengths of this project.
- LANL demonstrates an impressive analytical capability, particularly with regard to the elemental mapping capabilities and protocol development.
- LANL is well equipped to do a variety of tests in support of the major DOE research thrusts in fuel cells. LANL is flexible, skilled, and dedicated.
- This project has an excellent team, approach, and results. This is a great idea to have one of the most credible teams in the fuel cell industry available to help developers meet the developmental targets for their fuel cell component technologies.
- LANL can collaborate with a wide variety of developers, suppliers, and universities. LANL has shown the ability to work on catalyst layers, gas diffusion layers (GDLs), metal plates, and many other components. A high level of testing throughput exists at the PI's location. The PI has deep experience in fuel cells.

Project weaknesses:

- There may be room for a little more autonomy on LANL's part.
- Some indication of DOE guidelines for selected projects would be beneficial.
- This project has insufficient funding to expand capabilities and have more developers involved in the project.
- Some of the evaluations appear to be unaware of previous literature, patents, or industry results.

- LANL often uses cell formats that are outdated (e.g., 50 cm^2 quad-serpentine flow fields). The project is at the mercy of collaborators to find quality problems to tackle.
- There is always the potential for the equipment (and especially the parts of the system that are not being tested) to fall out of date, and it is key that the researchers’ work remain funded at a level where they can continually update systems to reflect the state of the art in fuel cell design and a wide range of MEAs, GDLs, balance of plant, etc. as may be needed to answer some question of interest to the Program.

Recommendations for additions/deletions to project scope:

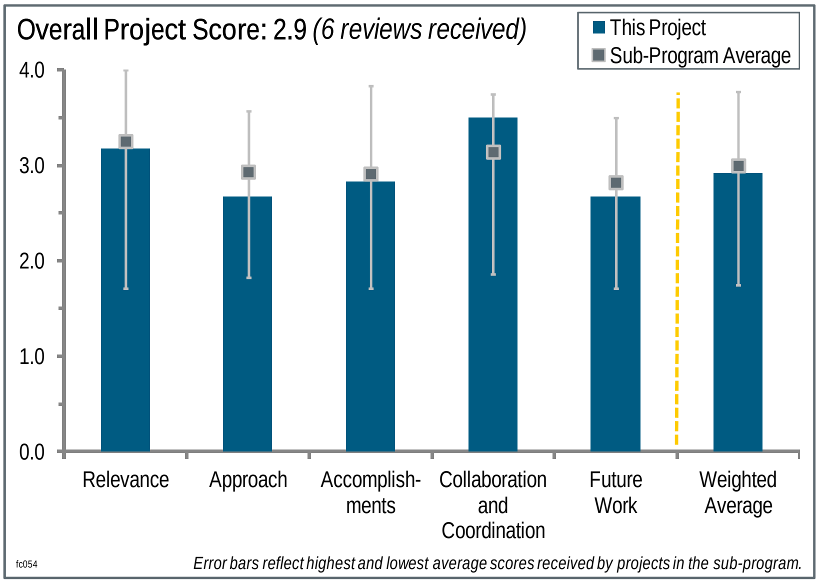
- DOE needs to provide more funding for this project.
- This project should be continued and expanded to include other projects funded by the Program.
- LANL should outline themes or topic areas where they see opportunities to discuss with the FCTT. Then, LANL could work along those lines for a longer period of time with less direction being necessary.
- Because the next tasks for the project were not shown, it is difficult to provide commentary here. That said, LANL needs to stay aware of recent publications so as to avoid accidentally duplicating results that are well known or well understood.

Project # FC-054: Transport in PEMFC Stacks

Cortney Mittelsteadt; Giner Electrochemical Systems, LLC

Brief Summary of Project:

The objective of this project is to improve understanding of the correlation between material properties and model equations for polymer electrolyte membrane (PEM) fuel cell stacks. The project will: (1) supply model-relevant transport numbers, (2) stress the model by developing different materials with different transport properties, (3) determine sensitivity of fuel cell performance to different factors, and (4) guide future research.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.2** for its relevance to DOE objectives.

- Water transport and the interaction with thermal management are important factors in meeting performance goals.
- This project relates directly to DOE's needs for understanding and increasing mass transfer effects within fuel cell components.
- This project is directly approaching several barriers with the intent to reach DOE's targets for cost and stack efficiency. Both of these targets are essential in moving PEM fuel cells toward commercialization.
- This project addresses several of the identified DOE technology barriers with a focus on stack performance, including cold start-up and stack power density and efficiency.
- While transport (water transport in particular) is critical to PEM fuel cell stack performance, this project is not particularly innovative; very similar research was already performed in past DOE Office of Energy Efficiency and Renewable Energy projects. Thus, the project's relevance to the overall objectives of the DOE Fuel Cell Technologies Program appears limited. It is also not clear what the main goal of this research is and how its ultimate success versus failure can be defined.
- This project addresses fundamental characteristics that are essential to every fuel cell model, particularly those associated with the membrane. These include the water diffusivity and electro-osmotic drag coefficient. At this time, many fuel cell models are attempting to address condensed water. This project also seeks to do this by understanding how the tortuosity and porosity of the gas diffusion media affect water flux across the gas diffusion layer (GDL). Developing a model is necessary for understanding how to improve power density while not compromising low-temperature performance. Greater power density lowers stack active area, which is the most powerful component toward reducing the overall cost of a fuel cell stack.

Question 2: Approach to performing the work

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

- The approach for this project includes developing models, improving inputs for models, using the models to study sensitivity of fuel cell performance to various factors and, finally, using model results to guide research activities.

- The membrane characterization is providing valuable information. The intention to address effective GDL transport properties is good; the presented and planned work looks to result in only a characterization study with some potential flaws.
- This project approach represents a good, though not very novel, combination of experiments and modeling. At the present stage, this project should be focusing more on fuel cell stacks and less on routine testing of components in single cells.
- The approach is sound because it combines cost analysis, material development, and in situ testing with the development of a model that will be made public. It remains a little unclear to what extent the published model will be useful for third parties, because the details of the developed materials and the stack architecture will not be published.
- The approach seems to have both modeling and experimental components, but it is unclear how much each one relates or feeds into each other within this project. It is unclear how new model development is occurring and if there is transfer besides just the material functional properties. The method development is good, although pretreatment conditions should be noted for the membranes. It was not clear what the main driver for the plates is.
- The project’s approaches for measuring water diffusivity through the membrane and the electro-osmotic drag coefficient are good and have been noted in the past for their novelty and the need that they fill to address these properties for more contemporary membranes. The methodology behind the porous media aspect of the model is missing. What is described is how the MacMullin number is calculated. However, this leaves unresolved how the mass transport losses are calculated from the MacMullin number for different operating conditions. There is some question as to whether water transport in the model is a function of GDL thermal conductivity, which other groups claim. This project is a good use of a variety of flow field designs and materials for model validation.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- Very good progress has been made toward the program targets in material development, experimental results, and the development of the model. It remains unclear if operations at high temperatures are sustainable over extended time periods. Results should be created or shared that indicate at what temperature operation is sustainable.
- The project has made good progress during the past year. A major milestone was accomplished on time and another is 60% complete (due 8/15/12). Progress has been made on identifying new membrane materials and incorporating them in membrane electrode assemblies (MEAs) for testing, and a new technique for simultaneous water uptake and diffusivity was devised.
- The team is progressing on schedule and the work seems to be well integrated. The membrane characterization, in particular, continues to show interesting and valuable results. The presented model validation studies met the team’s targets, but they were not a stern test. The modeling work shown only demonstrated (averaged over a cell) that the effective transport properties plus that model can match performance and (averaged over the cell) crossover, and not at transport-limited operating conditions.
- The new membranes appear promising, but there is no data on durability. Overall, the experiments seem to be interesting and done well, but some more theoretical discussion of their applicability is required along with the scope of the properties. Some of the model/data comparison is better with the new transport data, but some is not as good. This project is a good use of the different systems to test the model and the various segmented cells.
- Several claimed achievements appear to be about phenomena and ideas relatively well established in the field. For example, what makes the research in slide 7 an achievement is not clear. Both the decal transfer approach and fabrication of MEAs using hydrocarbon membranes have been well known and established in literature. Similarly, the technique of hydrogen saturation and electro-osmotic drag measurements in slide 10 is not especially new. Several bullets in that slide seem to re-state the obvious. Proton conductivity data in slide 6 may have been mislabeled; the numbers for Nafion, especially at low operating temperatures, seem much too low.
- Results on water diffusivity differ from those in the General Motors (GM) project by a factor of about 3–4. The GM project claims that a decrease in diffusivity at higher lambda (as is shown in this project) is due to a “device resistance.” Projects should check data with each other. An excellent match was shown between models and

experiments for all cells studied. It would be interesting for the project to report whether the outlet relative humidity (RH) for the cells in the validation efforts managed to exceed 100%. While the model-to-experiment match is impressive, there should be some analysis to understand whether wet conditions in the cells are really being achieved, or if the models are only accounting for dry conditions. Water transport numbers given for anode-to-cathode and cathode-to-anode should match. Model convergence needs to be slightly improved.

Question 4: Collaboration and coordination with other institutions

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

- There appears to be good collaboration and coordination between the projects.
- Excellent collaboration among the members of a well-qualified, highly interactive team has benefited this project.
- This is a strong team involving organizations with complementary skills. The roles of the team members are well defined.
- This project appears well coordinated and executed. Information and results are apparently moving among the team members effectively and being utilized.
- This project has an excellent mix of partners from industry, academia, and national laboratories. There is a clear separation of tasks that requires a significant amount of exchange and interaction between partners. The progress made shows that this required interaction is managed successfully.
- With the exception of some membrane property measurements, it is difficult to see how Virginia Tech membranes are being integrated into the project, or whether they are necessary for the development of a transport model. Ballard GDLs are contributing to the measurement of GDL properties. Good use was shown for GM flow fields in model validation, as well as thin metal plates from Tech Etch. The University of South Carolina was evidently very involved with both experiments and modeling. With the possible exception of Virginia Tech, all collaborators were well integrated into the project.

Question 5: Proposed future work

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

- The plans for the future work appear to be a reasonable continuation of the activities already underway.
- The future work aligns well with performed work and the program approach. The future work should additionally contain durability studies on either single cells or stacks at the proposed elevated operating temperatures.
- On a high level, the plans are good. Effective transport in "wet" diffusion media, and eventually moving on to short stacks and coupling thermal effects, is important for demonstrating sensitivity of performance to the measured properties in experimental hardware and in the models.
- The testing of membrane properties at lower temperatures should be done, because many fundamental aspects might change with temperature. Model validation at other conditions such as higher and lower humidity levels would be good to see. It is not clear as to whether transient modeling is going to be done.
- At the present stage of the project, future research should focus on final deliverables and concentrate on transport in stacks, as promised by the project title. Proposed work between now and the project end date in August 2013 remains focused mostly on component development and testing.
- The future work focuses on expanding the analysis to new materials. However, it may be interesting first to consider whether expanded yet realistic operating conditions might reveal inadequacies in the model. Inlet RH has been varied in the model validation exercise, but it is not clear if the model has really been run at wet conditions. Low temperature may be of interest, as well as freeze conditions. The project may also want to consider to what extent the model is capable of responding to changes in the catalyst layer parameters. Assumptions for the catalyst layer should be shown.

Project strengths:

- This project has a good team and comprehensive approach.

- This project is a solid re-examination of phenomena and materials that, to some degree, have already been studied previously.
- This is a strong, highly organized team working well together with good publications listed.
- The measurement of membrane properties with novel fixtures and the results that show the matching of models with experimental results are strengths of this project. Most of the collaborators have been integrated to work on something useful in the project.

Project weaknesses:

- This project has only a moderate level of innovation. There is also a disparity between work done and the stack-focused research originally promised.
- No information was shared about long-term operation at high temperatures. While the model is shared with the public, the results of the hardware development do not seem to be shared. This reduces the value for the fuel cell community.
- It is not clear as to what the objective of the project is. It could be the experimental setups, the model, the data, or the new materials; more focus may be required. It is not clear whether the key and critical properties are being measured. Some sensitivity analysis using the model is required.
- This project needs to report in greater detail the methodology behind the model treatment of water transport in GDLs. There is relatively little discussion of catalyst layers and how the model would respond to changes in catalyst layer properties (e.g., platinum loading, ionomer/catalyst ratio, etc.). This project team needs to better understand why there are differences between its measured water diffusivity and what is measured in other projects. There need to be better definitions of wet and dry conditions (beyond just what is given for inlet RH). If wet and dry conditions are varied more, it would be interesting to see if the model still matches the experiment.
- There was little publication or presentation in the open literature related to the accomplishments presented. The results need to be communicated more effectively to the community at large, particularly the membrane characterization. The modification and analysis of the diffusion media needs more inspection. For the substrates, the characterization is valuable, but there is no reason to expect a single MacMullin number relation to describe materials with vastly different microstructures and corresponding tortuosities. Some of the “papers” have significant amounts of binders and particulates. More importantly, the multilayer structures require analysis to make the effects of individual layers clear, unless a single layer is dominating. It is also not clear if the model is capable of treating the multilayer media as an assembly of distinct layers with varying properties.

Recommendations for additions/deletions to project scope:

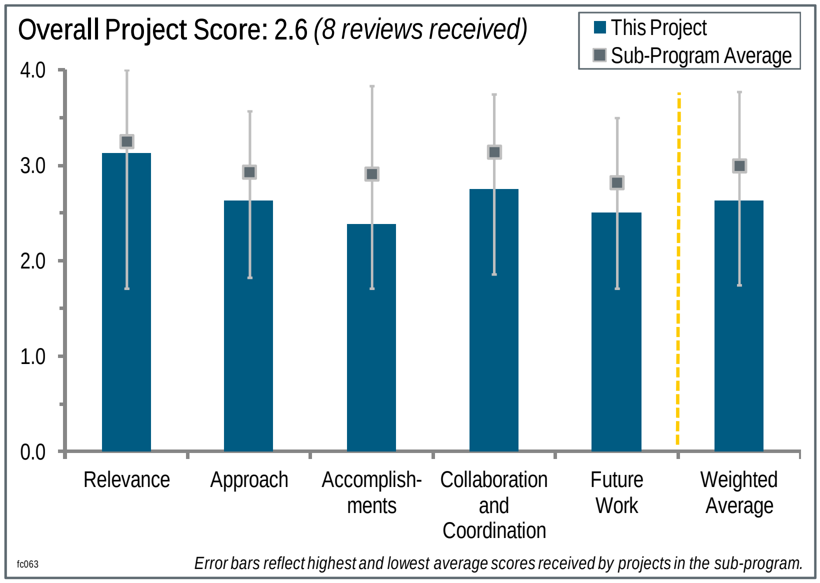
- This project should study the effect of long-term operation at high temperatures.
- The proposed work on cold start-ups should be done, as indicated in the project approach.
- Thermal conductivity of the new GDLs should be measured, along with membrane transport properties under liquid water. Capillary properties and breakthrough pressures of the novel GDLs should also be measured.
- With no connection between structure, chemistry (for the membranes), or materials and the effective properties, this is essentially a component characterization project. Incorporating some level of structure-property correlation to enable actual materials design would be valuable.
- This project should add more operating conditions at which to validate the model, particularly lower temperatures. The researchers should also report on whether thermal conductivity was considered and report on catalyst layer properties. Unless the model has already been validated at a wider range of operating conditions than those described, the project should not focus on new materials yet; the team should be sure the model works for baseline material sets.

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

David Mountz; Arkema

Brief Summary of Project:

Goals in this project are to: (1) develop ultra-thin membranes for fuel cells having low methanol crossover, high conductivity and durability, and low cost; (2) develop cathode catalysts that can operate with considerably reduced platinum (Pt) loading and improved methanol tolerance; and (3) combine the catalyst and membrane into a membrane electrode assembly (MEA) having a performance of at least 150 mW/cm^2 at 0.4 V and a cost of less than \$0.80/W for the membrane and cathode catalyst.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.1** for its relevance to DOE objectives.

- The project supports critical DOE objectives for direct methanol fuel cells (DMFCs).
- MEA work for portable power is in direct alignment with overall DOE objectives.
- The development of improved membranes, MEAs, and methanol-tolerant cathode catalysts are all relevant to DOE's portable power targets. The concept of a composite membrane to mitigate methanol crossover effects in a DMFC is not new. Previous attempts have had rather limited success.
- The project is relevant to the objectives of the Fuel Cell Technologies Program Multi-Year Research, Development and Demonstration Plan. The activities are aligned to DOE's goal. This project is focused on low crossover membranes and MEAs for DMFC application, which is very important for the commercialization of DMFC technology.
- Development of MEAs with better performance for liquid fuels is a critically important issue for DOE because ultimately liquid fuels will be much preferred to gaseous or liquid hydrogen (H_2). The work on reducing the methanol permeation is an important, although not particularly novel, approach. The catalysts development appears to have been a "bust."
- The program's overall objectives for the membrane performance and improved catalysts are good, along with the expected benefits in cost reduction and lower crossover. The project objectives provide a good link to the overall power density at the MEA level with a target loading, but the membrane objectives may be better targeted at a final MEA efficiency at the target operating point. The methanol permeability is not quantified under the operating conditions.
- The DMFC technology is an attractive one because liquid fuel is an easy way to store energy. To date, DMFC work is focused on two things: (1) making steady power from a high-energy-density methanol fuel and (2) achieving high power with lower than 2 mg/cm^2 of precious metal. Many have demonstrated short-term, high-DMFC activity, but the key issue with DMFCs is achieving a high steady state activity of the electrode that is measured after operating for two days and for continued operation up to or greater than 200 days. This project deserves a "3" because the authors show good relevance for some issues, but they said "lifetime" at their catalyst level was "in progress," yet this is the key issue.
- The project addresses DOE targets for performance, cost, and lifetime of portable fuel cell systems. The system targets are broken down to reasonable targets for the MEAs. Furthermore, the project addresses the important problem of methanol permeation, which is not directly related to DOE targets but indirectly influences

performance and cost. However, catalyst-specific power and platinum group metal loading are limited to the cathode, and no work is planned for the anode, though this may be equally important.

Question 2: Approach to performing the work

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

- The technical approach used in this project is adequate and in accordance with the set objectives.
- Both the composite membrane and the cathode catalyst choices have been questionable from the beginning. So far, there has been little, if any, evidence of the performance advantage over benchmarks brought about by the materials developed in this project.
- The approaches taken for the completion of all four tasks are adequate. However, it is not clear why the team took the approach of using palladium (Pd)-based co-catalysts to suppress the methanol oxidation on the cathode of DMFCs. It is very obvious that the blending of Pd into a Pt/carbon catalyst will create a mass transport limitation in the cathode. This approach is not very prudent for suppressing methanol oxidation at the cost of higher oxygen mass transport in the cathode.
- The approach of first developing and characterizing materials individually and then combining them is reasonable. However, materials characterization is focusing on performance and disregards durability almost completely. Starting long-term testing late in the project bears the risk of missing the project's target without sufficient time remaining for counter measures.
- The approach is good for membranes but poor for the catalysts. It appears that the original catalyst results were incorrect. This may be a just reward for proceeding with a project that has little or no basis in theory beyond faith that a different metal would work better. A more theoretically based approach would have avoided this. The membrane work appears to be based on a mechanical method of reducing methanol permeation. No basis is offered for selectivity. This is a pity, because the results appear to be encouraging.
- The approach is in four parts: (1) membrane, (2) catalyst, (3) MEAs, and (4) durability testing. The authors say catalyst work is 100% complete, yet they have not demonstrated steady catalytic response in a fuel cell. As long as the anode potential is greater than 0.1 V, then one is dealing with an H_2 anode that then "burns off" CO_2 at approximately 0.4 V to CO_2 and not a true direct methanol to CO_2 electrode at 0.1 V. It appears the team expects the membrane to solve all the problems. This does not seem to be a reasonable approach. The catalyst work should be considered incomplete until membrane and system durability are better.
- The overall approach of increasing the membrane stability and mechanical properties is good. However, the approach of cross linking the sulfonic acids may be problematic. As demonstrated in Arkema's data, incomplete cross linking of the sulfonic acid containing molecules can lead to significant loss of the ionic species over the operation of the fuel cell or increased electrochemically active impurities that can adversely affect the durability. The approach of lowering the crossover seems to be a more viable approach to improve the performance, whereas the improvements in the electrochemical performance may result from methanol-tolerant catalyst problems that are likely to occur with increased venting of unused methanol and potentially flooding of the cathode. Additionally, as water management is generally an issue in DMFCs, attention should be paid to the electro-osmotic drag (EOD) of the membrane, especially as the proportion of sulfonic acid groups are increased to decrease the membrane resistance. This may lead to increased water management issues and losses of performance and efficiency in the final MEA. For portable applications, it is important to decrease the air stoichiometry to increase the system level efficiency below the stoichiometric ratio of five used in the MEA testing. It is not clear how the approach of adding silica materials to the membrane provided a benefit in the crossover or other properties of the membrane, even if good dispersion is obtained.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- Eliminating part of the project that was not advancing made sense at the go/no-go review.
- The major accomplishments are two membranes and 50% lower Pt loading, which met the milestones.
- The performance is decent, but it all seems to be short term. The team needs to show good activity beyond two days of operation.

- The progress on the membrane selectivity is good from the empirical point of view, but the presenter gave no basis for the selectivity, which is a problem. The use of sol-gels has been done by Mauritz and others. The catalyst work is clearly a complete failure and the team has accomplished a "kill that" moment. A better rationale for why the Pd would work better would have been helpful, particularly because there are theoretical justifications for why Pt is better than Pd. This may serve as a warning to DOE to avoid empirical projects.
- The membranes achieved June 2011 milestones concerning area resistance and methanol permeation coefficient. The improvements to achieve the December 2012 milestone seem possible. The high sulfur loss, however, indicates poor membrane stability. MEA-performance targets are achieved with commercial catalysts; the catalyst developed in this project shows lower performance, and consequently work on the Pd catalyst development was stopped.
- The membrane optimization for the trade of the crossover and the area-specific resistance has made good progress. However, the data concerning the level of sulfonic acid group loss is concerning because this will lead to both a serious durability issue and a decrease in the membrane conductivity over time. The researchers do indicate that they understood the issues and expected to make progress toward resolving this. The MEA fabrication with the Arkema membrane appears to have made good progress in terms of power density in the lower methanol concentration. The performance seems to be in line with literature performance levels for similar membranes and electrodes. However, the data is gathered with a high-air stoichiometry (5X) on the cathode for both the perfluorosulfonic acid (PFSA) and the Arkema membrane. Additionally, Arkema does not provide any MEA data for the crossover flux (mA/cm^2) of their membrane compared to those they reported for the PFSA membranes at the same operating conditions. The catalyst data does not show any encouraging results in a practical MEA and appears to have been discontinued. It is not clear what benefit is expected to be gained by incorporating the silica materials into the membrane even if the dispersion issues can be resolved.
- Performance of the best membranes synthesized in this project show very little improvement over PFSA benchmarks once feed conditions are optimized for each polymer and membrane thickness. The membrane performance target should have been more demanding than the one used. Performance trade-offs between membrane resistance increase versus lowered methanol permeability should have been realized earlier. The choice of Pd-based "co-catalysts" for the cathode has been difficult to rationalize from the beginning, either from the point of view of methanol tolerance or oxygen reduction reaction (ORR) activity. Not surprisingly, the performance of MEAs with Quantum Sphere Inc. (QSI) Pd co-catalysts at the cathode has turned out to be much below that of MEAs with a "regular" Pt/C cathode. An increase in ORR performance with an increase in methanol concentration (slide 23) is stunning; it may indicate some serious problems with rotating disk electrode (RDE) experiments.
- Although the team had reported success in meeting the goals on membrane development in June 2011, the new, low-cost polyelectrolytes are highly leachable and may not be adequate to provide desired durability in DMFC conditions. The team needs to go back to the previous polyelectrolyte chemistry, which was not highly leachable, and attempt to address the membrane cost using polyelectrolyte chemistry. Moreover, the team needs to optimize the membrane composition to achieve at least similar, if not better, performance than standard PFSA membranes. The testing of Pd co-catalysts was done under extremely high air stoichs, which is inappropriate for portable DMFC conditions. The team should conduct their testing at air stoich of 2.0, which is close to that being preferred in portable DMFC systems. The team should also look into the leachability of a TPS additive. Given their small size and high acidity, which is expected to enhance their solubility in methanol water mixture, dissolution/extraction from the membrane matrix will result upon prolonged DMFC operation.

Question 4: Collaboration and coordination with other institutions

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

- Overall, the collaboration seems reasonable.
- The project activities are well coordinated among the team members.
- Arkema is teaming with QSI and the Illinois Institute of Technology (IIT). This is a strong academic-industrial team with proven accomplishments in catalysts and membrane developments.
- Obviously, the collaboration with the catalyst developer was not good, and the only other collaboration is with IIT. The knowledge from this project is not being disseminated satisfactorily.
- The partners are working in their respective fields of expertise. Coordinated interaction or collaboration with other experts is not mentioned.

- The team consists of good partners, including a university and catalyst company. However, inclusion or consultation with a national laboratory or DMFC portable power manufacturer could have helped the team to evaluate their membrane and catalyst under realistic DMFC operational conditions.
- While collaboration between Arkema and IIT appears to have yielded some promising materials, cathode catalysts from QSI have performed much below expectations. There seems to be very little information revealed between catalyst developers and the lead organization, which might have contributed to very disappointing results.

Question 5: Proposed future work

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

- The future work is fairly vague and appears to be more of the same empirical development.
- The proposed future work is adequate and laid in accordance with the project objectives.
- Developing MEA and testing durability is needed, but the lack of ongoing catalyst work is surprising, especially before long-term stability of this DMFC system has been determined.
- The future work described is aligned with the proposed work of the project. The team needs to consolidate on a membrane/MEA structure and then start the durability work quickly to ensure completion of the durability testing within the proposed period of the project.
- The direction of proposed research is unconvincing, especially in the MEA task. The proposed path forward does not seem novel, nor does it promise significant improvement in DMFC efficiency (the claimed objective of this project). Some work is unnecessary. For example, the task of "understanding the role of methanol crossover on performance" has been accomplished and published by now.
- The proposed future work is suitable to meet the December 2012 milestone in membrane development and the September 2012 milestone in MEA development. Work on durability is limited to testing. It remains to be seen if the proposed work to address the sulfur loss can be done without a negative effect on membrane performance, but the approach seems reasonable.
- The future plans for the membrane appear to focus on two key areas: (1) improving the membrane conductivity and lowering the crossover, and (2) increasing the durability by addressing the stability of the sulfonic acid polyelectrolytes. They do not discuss measurements of the membrane EOD, which may be an important factor in the overall performance of the MEA, especially at low-air stoichiometries. It is not clear how the silica additives will provide a benefit in the membrane selectivity for this type of membrane. It would be useful to look at the overall crossover of the MEA under operating conditions, as well as look at the methanol permeability to examine the behavior of the methanol crossover and selectivity. The MEA development objectives loom, but they may need to be tied to understanding improvements in the membrane behaviors to optimize the overall performance as well as evaluating improved electrode fabrication. As part of understanding the areas for improvement, it may be important to understand the effects of both the methanol crossover as well as the water crossover into the cathode, and the effects on the performance. Water management issues may be important to obtaining the optimum efficiency. More MEA durability measurements should be included in the future work; it was not clear this was an important part of the plans and seemed to be a weakness of the current target membranes (sulfonic acid group loss).

Project strengths:

- This is a strong team, with all of the resources needed to solve the problems.
- The goals of the project were in alignment with important DOE barriers.
- The membrane work is very strong, and the ability to make membranes and MEAs was demonstrated well.
- The partners are experts in the fields of membrane development and catalyst development.
- The team showed a great ability to produce and test a wide variety of membrane compositions.
- This project did good, reproducible membrane development work on the base membrane. MEA development looks promising.
- The team is well organized and capable of developing DMFC membranes and MEAs. The team is equipped with the necessary resources required for the success of this project.

Project weaknesses:

- There is a lack of preliminary indications that DMFC activity will be steady in this project.
- The approach is much too empirical. There is little justification given for why the researchers expect the approaches to work, and indeed one approach certainly did not.
- Catalyst development and characterization of catalyst-specific power are limited to the cathode; no work is planned for the anode, which may be equally important.
- The effort in this project appears disjointed, with insufficient progress achieved to date. MEA performance has been below expectations and some RDE data has been downright confusing.
- The team could have benefited from consulting with a national laboratory or DMFC company to determine adequate testing procedures for DMFCs. The team also needs to move quickly to consolidate on the final membrane and MEA structures to facilitate initiation of durability testing.
- Increasing the stability of two selected membranes by cross linking is a high-risk approach. Typically there is a trade-off between membrane resistance and cross linking by increasing the number of polymer cross links, usually resulting in the decrease of membrane conductivity. However, to achieve a performance target with a lower Pt loading, the membrane conductivity must be improved.
- The increased need of catalyst and MEA optimization is a weakness of this project. Issues with the sulfonic acid cross linking may provide a difficult approach to getting a uniform conductivity and good durability. Some data on the membrane electrosmotic drag would be useful, as would comparable measurements of the methanol crossover for PFSA membrane and the Arkema test membranes under the same conditions, preferably in an MEA at the same conditions that the electrochemical performance is measured in. This project did not have enough MEA durability measurements, especially given the highlighted issues with the approach of incorporating cross-linked polyelectrolytes and their potential leaching.

Recommendations for additions/deletions to project scope:

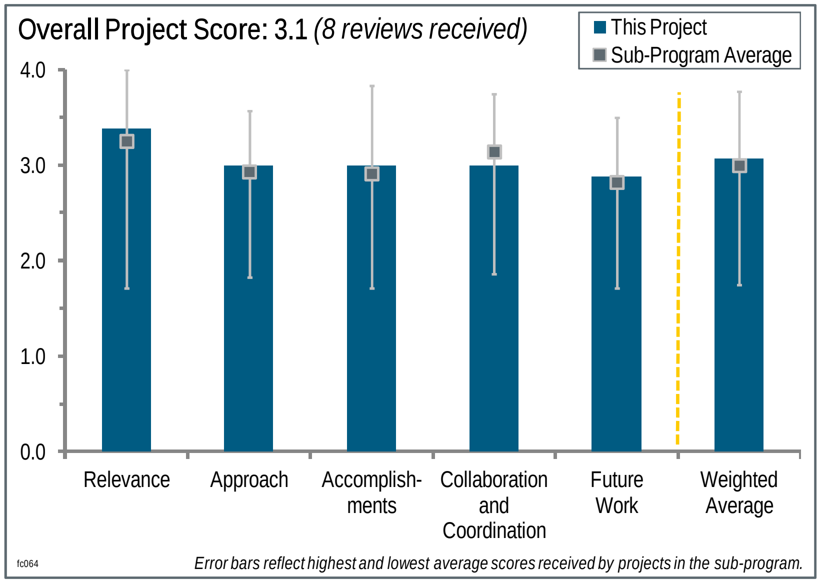
- This project needs to add some partners with some theoretical strength.
- Stopping the work on catalyst development was probably a good decision.
- It would be ideal to mitigate the catalyst agglomeration issue or find an alternative catalyst to use.
- The ongoing catalyst work should still be considered until the term stability of this DMFC system has been determined. Catalysts developed to date may have good short-term activity, but this activity may not last.
- The project needs the addition of a more intensive catalyst characterization with a clear understanding of the causes of the results obtained.
- The researchers should consider what benefits are expected from the addition of the additives (silica materials) to the membrane. They should also increase the measurement of the water crossover as part of the scope and metric to understand the water management of the MEA.
- Given the team's strength in membrane development and the time left in the project, the team should focus solely on task 1 and task 4. The team should drop task 3, specifically of the goal of 150 mW/cm^2 at 0.4 V. This goal seems to be a long shot, and presently there is no indication that the goal is achievable. By dropping task 3, the team will have more time to complete the other two tasks.
- Given the very poor outcome in the cathode catalyst task, there seems to be little justification for QSI's further participation in the project. To make the best possible use of the remaining resources, the focus of this project should shift to membranes with high conductivity and reduced methanol permeability. All MEA testing of performance and efficiency validation should be carried out using DMFC benchmark catalysts available commercially.

Project # FC-064: New MEA Materials for Improved DMFC Performance, Durability, and Cost

Jim Fletcher; University of North Florida

Brief Summary of Project:

The project objective is to increase membrane electrode assembly (MEA) functionality and internal water recovery to facilitate direct methanol fuel cell (DMFC) system simplicity, increase power and energy density, and reduce costs to address the U.S. Department of Energy's (DOE's) consumer electronics goals. This objective will be accomplished by: (1) improving the performance and durability of the University of North Florida (UNF) MEA to increase power and energy density and lower the cost; (2) developing commercial production capabilities to improve performance and lower cost; and (3) increasing catalyst stability to lower degradation rates and catalyst loadings.



Question 1: Relevance to overall DOE objectives

This project was rated **3.4** for its relevance to DOE objectives.

- This program addresses using liquid high-energy storage in a stable power generation system with modest cost.
- This project has high relevance to DOE portable power goals, but also in water management issues and liquid-fuel MEA design, which should be the ultimate goal for DOE fuel cell technology.
- Early applications such as portable DMFCs are critical to show that fuel cell technology is real and ready. There is also a real benefit over batteries regarding high-duration energy density
- The project addresses the simplification of the water management in DMFCs, which should ultimately result in cost, weight, and volume reduction in a fuel cell system.
- This project is very relevant for portable power to address DOE's 2013 consumer electronics goals. The plan is to transfer processes to Johnson-Matthey Fuel Cells Inc. (JMFC) to improve performance and cost, which will ensure industrial relevance.
- The project is relevant to DOE's objectives because it addresses the most important limitations of state-of-the-art DMFCs: low catalyst stability and high degradation rates, and poor water management and low energy density depending on a complex water management system.
- Water management is a key issue for the implementation of portable DMFCs. This project takes on improved water management using a passive cathode approach to reduce system complexity and size associated with active cathode water recovery.
- It is very challenging to achieve a commercially viable DMFC system for consumer electronics goals because of the balance-of-plant (BOP) requirements, high catalyst loadings, and the low power density achievable with DMFCs. Overall, the team has demonstrated a terrific DMFC system; however, it is unclear whether this is a commercially viable solution for consumer electronics. Unfortunately, 300 W-hr/L will not be competitive against current consumer products. Even the 2015 goal of 800 W-hr/L seems a little low for making a significant impact toward replacing the current power solutions for the consumer electronic market. It would be nice to see how the cost is addressed in this work. For example, the current catalyst loadings and how much reduction in loading is expected to be achieved need to be defined, the cost of the MEA compared to current DMFC MEA projections needs to be projected, and the work being done on the BOP to ensure a smaller

footprint to enable a significant improvement in system energy density needs to be outlined. It is difficult to correlate the colors on the weight/volume breakdowns to the actual component; it would help if a letter were also assigned so that it is clear which pie wedge corresponds to which component.

Question 2: Approach to performing the work

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

- This is a novel concept.
- This project uses a clever membrane and system design to maintain high system performance over 10,000 hours, which is impressive.
- This project has a good approach to increasing the specific power density by simplifying the system. Focusing on catalyst reduction late in the project bears the risk of missing the project targets.
- The approach is good in that it pulls together a number of developed components and tests the water management and liquid handling properties. The diagram of the cell is, however, unintelligible and makes no sense. It is unclear if this was deliberate.
- The addition of a liquid barrier to the cell design allows for a simplified DMFC. It also reduces the oxygen diffusion at the cathode, and thus reduces the performance of the MEA as compared to one without the liquid barrier. The performances are approaching the DOE targets, but are not there yet and the project has no specific plan (or time, since there are only a few months left on the project) to reach those targets.
- Designing direct methanol systems with a water-balanced operation through system/component engineering is an approach that has seen significant investment, including failed attempts at commercialization by at least two companies (MTI Micro Fuel Cells and Polyfuel). The likelihood that the investment of this project, focusing on minor optimization of components, will lead to a commercial or close-to-commercial device is low.
- Overall, the project follows a good engineering approach that addresses the major attributes required to advance DMFC technology. Specifically, the incorporation of the Polyfuel membrane and stable anode catalyst from JMFC combined with the barrier layer approach to simplify the system provides a very good approach with a reasonable probability of success of meeting DOE targets. The project team addressed durability and operational issues. It is not clear how the project team is addressing the integration issues for the stable anode catalyst.
- The presentation materials were well organized and presented clearly. Having the system in hand was also excellent. The critical barriers of water management, durability, and methanol crossover were the key focus, and it appears that very good progress was made. The only area that did not seem to progress well was the anode catalyst development at Northeastern University (NEU); it was not clear what the critical issues were and how the team was going to address them in their "optimization" work. It would be helpful to see some electrochemical impedance spectroscopy of the NEU MEAs. Because the partnership of JMFC more than makes up for this aspect of the program, it is still fair to give a rating of 4 rather than a 3.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- This project has great results going against difficult metrics.
- The optimization of individual components, such as the catalyst layer and membrane, has led to improvements in fuel cell performance.
- Achieving a commercially viable DMFC system for consumer electronics is very challenging. While the progress is excellent, it is not clear if all the barriers (specifically the BOP and stack weight) can be overcome such that 800 W-hr/L will be demonstrated.
- The progress made in the project to reduce off-state degradation and improve catalyst durability is good advances. The use of a thicker membrane is rather obvious and seems like it should have been used earlier. The performance improvements relative to DOE's targets has been modest.
- Achieving 10,000 hours of continuous DMFC operation in a portable package (not a laboratory bench test) is very impressive. The reason this is not "outstanding" is that the issue of deactivation when the system is "off" is now an issue. Short circuit deactivation is an issue. These issues can be dealt with as is indicated in

slide 16. What is impressive is the team has overcome the Achilles heel of DMFCs (the catalyst) by clever membrane and system development.
- Excellent progress has been made on the water management but progress has not been so great on understanding the behavior of the electrode layers, as demonstrated by the poor performance of the NEU catalyst. There is no evidence given for poor conductivity in the catalyst layer. This needs to be probed since this will be necessary for future work. The variability in the catalyst's performance indicates there is a long way to go in understanding how the catalyst layer works.
- The team has produced an MEA capable of supporting a small system, close to meeting 2013 targets. The researchers have achieved 20% improvement in MEA performance and 10,000 hour durability demonstrated, with some operational issues identified and mitigated. They have also achieved a significantly lower crossover for a given cell resistance with their UNF HC membrane, and they can get the same performance with a 45 µm membrane, but with significantly reduced methanol crossover. Fuel efficiencies of >90%, and overall efficiency of approximately 30% were achieved.
- Good progress has been made in membrane optimization regarding methanol crossover. Also, the degradation rate in discontinuous operation was improved, but the cumulative (real operation) time should be increased for the long-term experiments. A degradation rate of 34 µV/hr seems to be not good enough to meet DOE's targets, which are 3,000 hours or 5,000 hours for 2015. The degradation rate was measured in an on-off mode only with 200 hours of real operation.

Question 4: Collaboration and coordination with other institutions

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

- This project has good collaborations with JMFC and NEU.
- Collaboration with JMFC has resulted in improved MEA performance and scalability of the process.
- The UNF/NEU/JMFC team is very strong and they are using their strengths to effect outstanding results.
- The project has a reasonable level of collaboration, including JMFC to ensure industrial practicality and promote commercialization.
- There is a good cooperation with JMFC and NEU. The partners are working on their respective fields of expertise.
- The work suffers heavily from the lack of involvement with a systems integrator, a role that is being attempted by a university and leaves questions about whom ultimately would manufacture these devices should the project be successful.
- The NEU contribution appears disconnected. The technical transfer to JMFC is good, but then this appears to limit the extent of the collaboration possible. This seems to be a pity, because there is a big problem to overcome in understanding the catalyst layer operation that more collaboration might help with.

Question 5: Proposed future work

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

- The future work is good as far as it goes, but it needs a more fundamental approach to catalyst layer design and development.
- The proposed work includes a reduction of catalyst loading, stack testing, technical transfer, and anode development, all of which are laudable next steps.
- The future work is just a continuation of what has been done so far, which is appropriate considering the amount of time left in the project.
- The proposed work seems suitable to meet the targets in the remaining time, except the catalyst loading reduction and the improvement of the MEA with NEU catalyst.
- The project is almost complete. The technology transfer to JMFC and continued integration work is appropriate. It is not clear if issues with performance for stable catalysts will be overcome within the time frame, though.
- It is not clear how much NEU contributes to the success of this program. The technical plan for optimizing and improving the performance of the NEU catalyst was not presented and therefore cannot be evaluated properly. The researchers should partner with a group that can help reduce the BOP size and weight.

- The future work is somewhat disconnected with very little synergy between the tasks of each participant and too broad to lead to substantial advances in any specific area. MEA development seems to be the primary need so that performance capable of meeting out-year targets can be approached. The stack testing should be limited; it is unclear who the consumer of commercial MEAs would be, or if they should be a focus at this time, and the value of NEU's catalysis work has not been demonstrated and does not seem to be as important as the focus on barrier layers.

Project strengths:

- This project has a strong team.
- This project has good focus on and success with the water management issues.
- The team has demonstrated systems and long-term durability of DMFCs.
- This project provides a new approach to the simplification of the BOP components.
- This project takes a practical engineering approach with strong linkages with an industrial partner to enable commercialization.
- This project has made excellent progress toward showing the benefits of fuel cells over batteries. The researchers have achieved or exceeded most DOE metrics. This is the only company at the DOE Hydrogen and Fuel Cells Program Annual Merit Review with a working fuel cell system.
- MEA, stack testing, system operation profiles, and the partnership with JMFC are the strengths of this project. The fact that the researchers have demonstrated a nice system with excellent durability is another strength.
- The project idea to use a water barrier layer to minimize the system components is an interesting concept. The project partners have great experience and expertise in their respective fields, especially in catalyst development.

Project weaknesses:

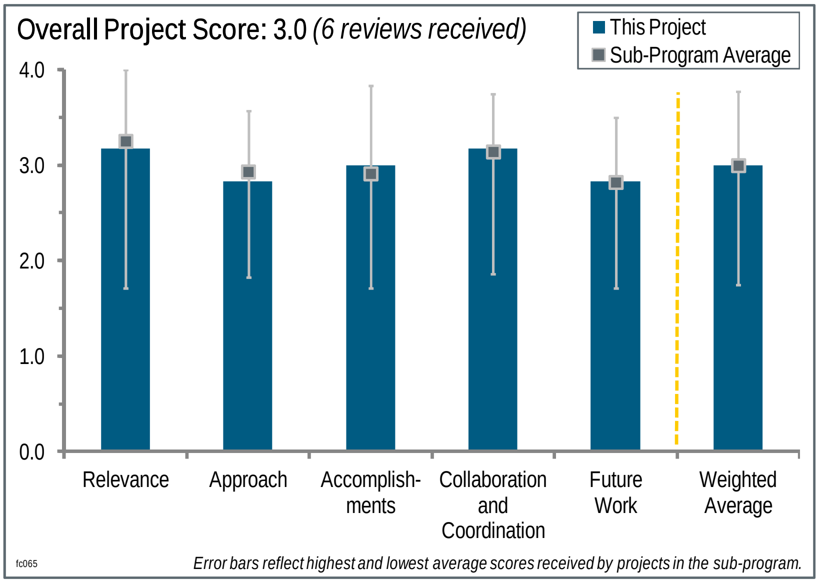
- This project has no weaknesses.
- The work at NEU appears weak, and the lack of a BOP partner seems to be a weakness.
- The diagram of the water management system is confusing. The approach to the catalyst layer design is too empirical.
- The MEA integration issue is not understood and the approach to understanding this is not outlined.
- The project is more procedural oriented. To solve the degradation problems, a more scientific approach would be helpful. It is unclear if the development will help to commercialize this kind of DMFC system with a passive water recovery system.
- The largest weakness of the project is the lack of a system integrator/manufacturer. It leaves major concerns for what the path forward is for findings from the project, assuming a high level of success. An additional concern involves how far the temperature can be pushed and whether or not current thermal gradients in the system will prevent higher temperature and therefore more-efficient, higher-power-density operation.
- At temperatures greater than 50°C–60°C, water will be lost through vapor diffusion through the liquid barrier. This has two primary consequences: (1) limiting the operating temperature/tolerance to temperature upsets, and (2) not eliminating water storage and pump, but only reducing their sizes, resulting in a lower power density than claimed today.

Recommendations for additions/deletions to project scope:

- This project should add more collaborators to help develop a basis for catalyst layer design.
- The project team should focus on the optimization of the system operating parameters and the conditions during on/off. Mechanically “sealing” the cathode is one promising approach.
- Considering that the methanol cross-over is a minor component in DMFC performance losses at the fuel cell operating methanol concentration, it would be interesting to have a perfluorosulfonic-acid-based MEA for comparison of performance.
- Focus on catalysis will only result in modest improvements in performance and work by others, including Los Alamos National Laboratory’s project on DMFCs, which has made advances that this team could apply without using resources. The team should focus just on optimization of water management and increasing temperature of operation while maintaining water balance.

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

Jean St-Pierre; Hawaii Natural Energy Institute



Brief Summary of Project:

The objective of this project is to identify and mitigate airborne contaminants that adversely impact system performance and the durability of fuel cells. The project includes contaminant studies, real-world operation and mitigation strategies, model development and application, and outreach. The Hawai'i Natural Energy Institute (HNEI) will identify the contaminants contributing to losses in fuel cell tests and will include in situ and ex situ tests to determine the loss mechanisms.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.2** for its relevance to DOE objectives.

- Studying the effect of airborne impurities on fuel cell performance is important.
- Airborne impurities have significant impact on fuel cell performance, especially on the cathode, making this project highly relevant to the objectives of the DOE Fuel Cell Technologies Program.
- This type of understanding is absolutely critical to meeting all the fuel cell system, membrane electrode assembly (MEA), and MEA component targets simultaneously in actual commercialization.
- This project is quite relevant because it continues to be important to understand the effect of air contaminants on fuel cell performance and durability. It is also relevant to develop or design mitigation strategies.
- Impurity testing is quite important for characterizing possible fuel cell performance degradation mechanisms. A comprehensive analysis of impurity effects is needed to prevent failure of early fuel cell demonstrations. A number of early adopter projects have already been impacted by a failure to understand the role of contaminants. These include premature failures in high ammonia and sulfur gas environments, fuel cell degradation due to system contaminants, and inadequate feedwater quality.
- Studying the impact of airborne contaminants and developing mechanisms is critical to extending the life of the fuel cell system in real-world applications. The concentrations used to study the impact of the contaminants on fuel cell performance works well for screening purposes. However, the use of high concentrations could lead to false "positives," hence it is recommended to crosscheck the impact at realistic concentrations for a few select contaminants.

Question 2: Approach to performing the work

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

- The approach is somewhat empirical; however, it does identify potential contaminants.
- The approach is good and straightforward and includes identification, contaminant studies, and modeling. It seems that the contaminant studies will not continue after the third year.
- It is not clear if the approach misses, for example, environmental impurities that are entrained in the inlet air streams, not as gases, but as micro-particles such as road salt in the upper Midwest.

- The approach is sound, but more work is needed to establish allowable concentrations into a fuel cell system to minimize voltage loss. Further, the interactions between contaminants are not being currently addressed; any potential negative interactions need to be identified.
- The effects of impurities on, for example, catalyst loading, should have been explored not only to reveal the mechanisms, but because operation at low loadings can often show exaggerated effects. The relationship between multiple exposures' reversible decay and non-recoverable performance should be explored in at least one of the 1st tier impurities.
- The approach used in the project is clear and is likely to be effective. While somewhat involved, the selection criteria for the 1st and 2nd tier contaminants have already proven to be quite effective. Avoiding duplication of other contamination-related projects is a plus. The wording of go/no-go decisions (slide 7) is confusing, i.e., more appropriate for milestones rather than decision points. It is not clear what the go/no-go decision points really are.
- The approach is fundamentally flawed, which is odd, given the strong team. Both UTC Power and Ballard have extensive experience operating bus fleets in various parts of the world, including the United States, Canada, Germany, and China. In addition, UTC Power has extensive experience with phosphoric acid fuel cells for stationary applications. It is not clear why the contaminants were not selected based on the knowledge base of these two partners.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- The researchers continue to make good progress in evaluating the Tier 1 and Tier 2 potential contaminants.
- Excellent progress was made on screening the contaminants using the two down-selection criteria and testing those in a fuel cell.
- HNEI has made good progress to date and has a good publication record. The researchers need to provide more details on which performance model is being used and at what level the impurities are being dealt with. It is unclear if surface area measurements (electrochemical surface area obtained by CVs) are being made to assess the effect of each of these contaminants.
- This project appears to be meeting its targets. In some cases, more in-depth understanding of the causes of the observed performance losses would be welcome. Development of predictive models for various contaminants is needed. This project would benefit from earlier development of mitigation strategies.
- HNEI has tested 19 Tier 1 contaminants and four Tier 2 contaminants. However, much of the data analysis appears lacking. The conclusions: "Higher concentrations are worse," "Higher current densities are worse," and "Lower temperatures are worse" could have been made before testing. The voltage recovery work lacks analysis. Knowing which contaminant can be recovered from is important, but understanding why recovery from some contaminants is possible but not others is lacking.
- The results generated are no doubt very reliable and the models that result will be useful. It is not clear that the data are sufficiently large to make the models as reliable as they could be. Also, long-term exposure to any one impurity at low levels, repeated exposure to higher levels of any one impurity, or long-term exposure to multiple impurities could manifest in different long-term decay rates that in turn are dependent on the type of catalyst alloy or loading in use. Some preliminary investigation of these effects with key impurities would be good to see if further funded research in that direction is warranted.

Question 4: Collaboration and coordination with other institutions

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

- The established collaborations are working well.
- The workers are collaborating effectively with many partners.
- The collaborations appear good and valuable, with actual data and designs being transferred.
- It is not clear what, if any, work was performed under this project by UTC Power and Ballard. The work in slide 13 looks like it was performed by C2E2. The other interactions with General Motors and Nuvera are good.

- This is a well-integrated project involving strong industrial partners that should ensure sufficient validation of performance loss data and the use of relevant test protocols. Participation with organizations other than partners in the project has been very good.

Question 5: Proposed future work

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

- Planned work on metal plates and contaminants should be coordinated with this work.
- It is unclear when the segmented cell work is going to be performed.
- The future work plan is good and well thought through. There should be gradually more emphasis on mitigation strategies or a means of preventing performance loss that cannot be reversed.
- The proposed work to understand the interactions between contaminants is critical. Further, the impact of contaminants at lower loading of platinum (Pt) catalysts is needed. Work to establish a tolerance limit for the contaminants is needed.
- The future work proposed could be better focused by continuing to build on the more basic investigations of impurity type effects on MEA performance and performance decay models. The impact of non-reversible decay from repeated exposures/recoveries should be explored because that is what will happen in the real world, and models based on short-term exposure/recovery will not anticipate longer-term effects. Other researchers have seen that after repeated recovery steps, non-recoverable permanent damage can be done to the catalyst surface structure, which is what will determine the long-term durability. Finally, it all gets worse at lower catalyst loadings. So at the minimum, the types of tests being done with the Tier 1 and 2 impurities should be done on state-of-the-art PtCo/C alloy catalyst electrodes at the DOE target loadings of <0.125 mg/cm^2.

Project strengths:

- This project has good techniques, collaborations, and methodology.
- This project has many collaborators to screen families of potential contaminants, and the work progresses on schedule.
- HNEI and the principal investigator are performing extensive experimental work and publishing results for use by the fuel cell community.
- This is a well-executed project that stands a good chance of determining the impact of some common contaminants on cathode performance.

Project weaknesses:

- The data are treated with a somewhat empirical approach rather than a more chemical interaction type of modeling.
- This project needs to better explain the overall objective of the project and provide more details on model development.
- Most of the analysis presented is qualitative or semi-quantitative. More value would be added if the individual mechanisms of the contaminants are understood.
- Limiting the scope of the impact of impurities with just a single functional impact mechanism that is independent of catalyst type, loading, or prior exposure history is a weakness.
- A certain weakness of the approach used in this project is the disconnect from the very rich literature database on strongly adsorbing unsaturated and aromatic hydrocarbons, many of which have been the focus of this research. Literature can be of help in determining both the mechanism of the electrode performance loss and mitigation strategies (if at all available). Increasing the understanding of the contamination process for different pollutants should be accelerated.

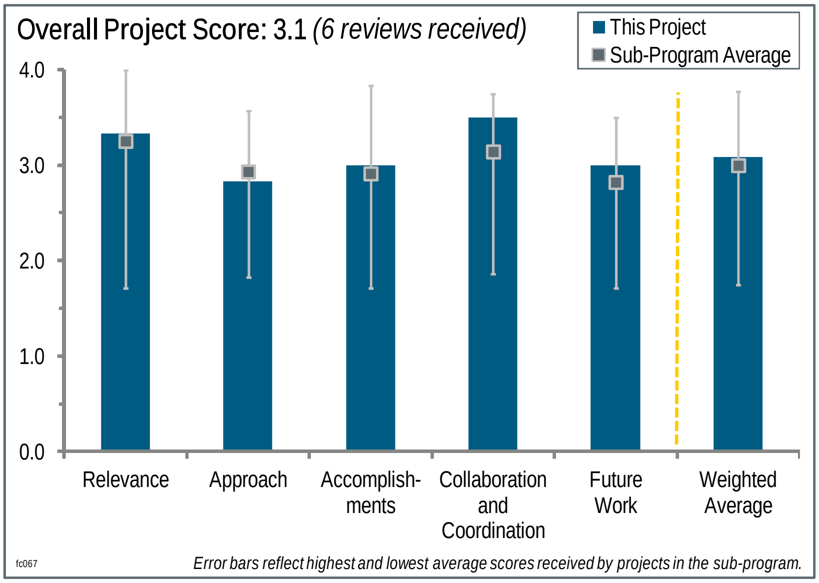
Recommendations for additions/deletions to project scope:

- This project should specify tolerance limits and contaminant impacts at automative-relevant Pt group metal catalyst loadings.
- This project should include more in terms of understanding the fundamental degradation mechanisms at work.

- More chemical/electrochemical diagnostic work to identify the mechanisms of contaminant impact on the performance is needed.
- This project scope should include a clear statement as to whether performance recovery is possible or whether filtering is the only option. Such a recommendation should include concentration limits relevant to either approach.
- HNEI should add a focused task to look at the long-term effects of repeated exposure and recovery steps, using one of their worst offending impurities on an MEA with a cathode that has a loading of only 0.1 mg/cm^2 and a very thin (approximately 10 µm) membrane.
- Bin impurities based on: reversible catalyst contaminants, irreversible catalyst contaminants, membrane contaminants, contaminants contributing to other performance loss, and contaminants with no deleterious effects.

Project # FC-067: Materials and Modules for Low-Cost, High-Performance Fuel Cell Humidifiers

Will Johnson; W.L. Gore



Brief Summary of Project:

This project seeks to develop a durable, high-performance water transport membrane and a compact, low-cost, membrane-based module utilizing that membrane for use in automotive, stationary, and/or portable fuel cell water transport exchangers. More-efficient, low-cost humidifiers can increase fuel cell inlet humidity, which reduces system cost and size of balance of plant (BOP), improves fuel cell performance, allows for reduced fuel cell stack size, and improves cell durability.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.3** for its relevance to DOE objectives.

- Successful development of this humidifier would help systems meet overall DOE goals.
- Humidifiers are important for meeting high-temperature automotive targets, although they are ancillary to specifically quantified targets in the technical plan.
- Durable, efficient, and low-cost humidifiers are critical to optimizing both thermal management systems and fuel cell stacks. This project addresses critically important objectives for fuel cell commercialization.
- For the automotive application, the continued development of a membrane humidifier is key. The case was not presented for the level of need in the stationary or portable application.
- The project supports DOE's research and development objectives, in particular for the BOP, by developing more-efficient, low-cost humidifiers. This will lead to reducing system cost and size of BOP, improving fuel cell performance, potentially decreasing the size of fuel cell stacks by running under wetter conditions, and improving fuel cell durability.
- This work is extremely relevant to the DOE Hydrogen and Fuel Cells Program (the Program). However, this type of work is expected to have occurred without DOE funding because it is a core-competency effort of the original equipment manufacturer (OEM). The benefit to the Program is that it sheds light on humidifier technologies. It may be more beneficial to benchmark humidifier technologies and publish findings than to help individual companies' humidifiers. By benchmarking humidifier performances, competition will be spurred and both automotive OEMs and humidifier manufacturers will benefit. OEMs will have a clear picture of product availability, and humidifier manufacturers will have a better idea of areas they need to improve to stay competitive.

Question 2: Approach to performing the work

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

- Technically, this approach is very good.
- Stability issues show that a broader range of materials should be considered before the down-select step.
- The combination of using a custom membrane material well suited for the application with a simple and cost-effective module design is a good approach.

- The approach is consistent with the project aims, including starting with material improvement of the new Gore membranes optimized for high performance and low cost and then design and optimizing the humidifier design with the subcontractor, dPoint.
- The focus, to date, has been on materials evaluation. The project needs more emphasis on the module (underway) and the integration into the system to better understand the effects of the edges of the operating envelope, start-up/shutdown, etc. For example, earlier testing would provide feedback on how best to test the membrane.
- The goal of developing a low-cost, passive humidifier module is good. The campaign to understand performance and durability of such a module is also good. The breadth of the program is a bit disappointing because it focuses on one specific proprietary material and one specific proprietary module design. This limits the program value for the industry.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.0** for its accomplishments and progress.

- The project is proceeding through its targets.
- Good progress has been made so far. The work put into identifying the degradation mechanism caused by the formation of anhydride species reducing the accessible number of SO_3 sites was appreciated.
- The data provided were helpful in understanding the performance and challenges of this humidifier. The team seems to understand the importance of durability and the criticality of evaluating the impact of environmental contaminants. The fundamental information on degradation mechanisms was good.
- The Gore material has shown promise, but there are significant issues with degradation. The presenter appeared to downplay a potentially fatal issue as shown in the data from General Motors (GM). The laboratory testing does not appear to simulate actual conditions; for example, the water quality that will be achieved in an operating engine.
- It is not clear that there is a viable path to overcoming the limitations of the current material. There is also a concern about the cross-flow design of the module. The claimed 96% effectiveness is physically impossible except in counter-flow designs.
- This project has made excellent progress toward objectives with main accomplishments: understanding of the source of durability loss—chemical changes in PFSA (as shown with the GM experiment) in accordance with the literature. It appears reversible with acid heat treatment, but this will not be possible to do in a system. There is also the risk of contamination (e.g., from sodium, magnesium, and potassium). The membrane acts as a filter and may protect the fuel cell stack if there is no other filter in the system. The module performance is consistent with single cell, and ex situ testing shows a loss of performance at 80°C of 20%–30% over 5,500 hours. The sub-scale module design is complete and the sub-scale prototypes are built—some first results would have been appreciated. Module cost is estimated to be approximately $100 at high volumes, achieving DOE targets. It appears two times the cost of tubular humidifiers.

Question 4: Collaboration and coordination with other institutions

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

- This project has an appropriate mix of collaborators.
- The company is working with automotive OEMs and is acting on feedback for device specifications.
- The prime contracting team is a little thin, but outside testing by automotive OEMs and modeling by Argonne National Laboratory (ANL) brings an important balance to the project.
- There is a good amount of collaboration for a project of this size. The collaborator dPoint seems like a good choice for module development and testing.
- It was very good to include the end user in testing. The module development should have begun earlier, even if the material had not yet been optimized.
- This project has appropriate collaboration with a product integrator (dPoint), with automotive OEMs (in particular GM), and ANL (to have a link with the system modeling of Gore membranes in humidifier modules).

Question 5: Proposed future work

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

- The future work is adequate for this project.
- This is a reasonable work plan, given the current barriers.
- This is a good plan. The results of the durability testing and the final full-scale module are eagerly anticipated.
- There is no opportunity to feed back the data from module testing (or module integration into a working fuel cell system) into the membrane development.
- The proposed work is in accordance with the project goals and the time remaining for the project. It will allow for complete membrane durability testing, the scale-up membrane manufacturing for final module build, finished sub-scale validation testing, and a final full-scale module.
- Given the specific nature of the materials and module design being developed with funding from this program, more effort to distill learning more broadly useful to the industry at large would be valuable. More targeted poisoning studies with likely environmental contaminants would be valuable.

Project strengths:

- The Gore material has advantageous performance parameters.
- There is a good balance of partners and collaborators, a good focus on performance and durability, and a good exploration and explanation of fundamental mechanisms.
- The combination of using a custom membrane material well suited for the application with a simple and cost-effective module design is a good approach.
- The objective is still worthwhile and the overall approach has been reasonably well structured. The materials expertise brought to bear on the problem is impressive.
- This project has many strengths. The approach of starting from an existing membrane material, which has been improved during the project, was appropriate. The collaboration from the beginning with an integrator was relevant. It has been appreciated to investigate the degradation issue and to understand the mechanisms; it may lead to future improvements.

Project weaknesses:

- The module design should have been better reported at this final stage.
- There is not enough emphasis on meeting all of the application requirements, including solving the degradation issue. This project needs better feedback of module and system testing into the material development process.
- The commitment to a narrow family of materials at an early stage seems to have boxed in and limited the project. It is also unclear how well designed the module is.

Recommendations for additions/deletions to project scope:

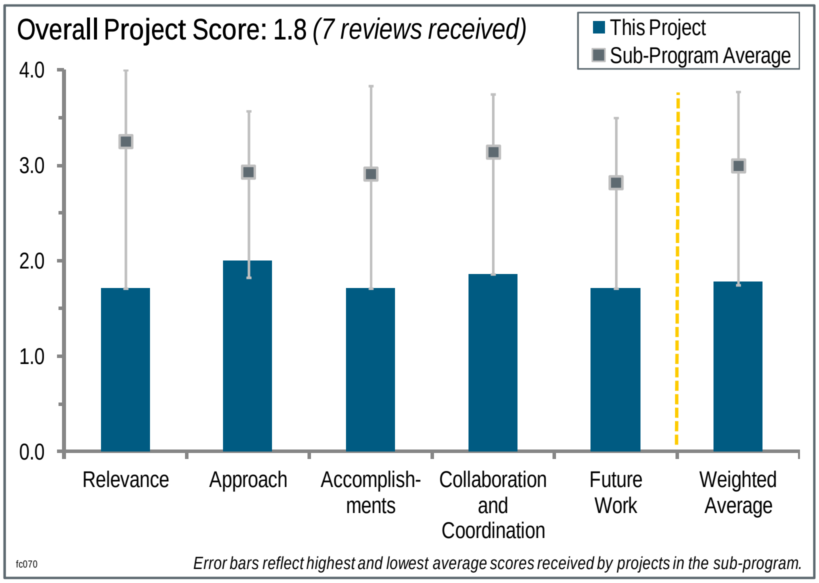
- This project is near completion; no changes are recommended.
- There are no recommendations this near to the completion point in the project.
- Some specific and targeted testing of the impact of environmental contaminants on durability and performance, and perhaps some collaboration with projects FC-048 and FC-065 would be useful.
- There are no particular recommendations, because the project is at its final stage and the results are in accordance with the aims. However, it would be interesting to investigate the behavior at higher temperatures such as up to 95°C, as some automotive systems are rising to such temperatures.

Project # FC-070: Development of Kilowatt-Scale Coal Fuel Cell Technology

Steven Chuang; University of Akron

Brief Summary of Project:

The overall goal of this project is to develop a kilowatt-scale coal fuel cell technology. The results of research and development (R&D) efforts will provide the technological basis for developing megawatt-scale coal fuel cell technology. For fiscal year 2012, the project objectives were to test the effect of operating conditions (temperature, voltage load, and concentration of CO, CO_2, and H_2O) on the performance and energy efficiency of the coal fuel cell, and to investigate the integration of coal fuel cells in series and parallel stack configurations.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **1.7** for its relevance to DOE objectives.

- It was not clear from the poster how the work was addressing DOE Hydrogen and Fuel Cells Program (the Program) objectives.
- The project is focused on the use of a fossil fuel, and thus not well aligned to DOE's Office of Energy Efficiency and Renewable Energy (EERE) objectives.
- Although this project supports DOE research, development, and demonstration objectives, it is not clear why it is part of the Program.
- The results of this R&D effort will provide the technological basis for developing megawatt-scale coal fuel cell technology.
- This project does not bring new or unique information to the fuel cell community. Fuel cell operation on coconut coke/Petcoke has little value and apparently fuel cell operation on coal gasification products was never done.
- The project is a poor fit for the EERE Fuel Cell Technologies Program. Coal-fired generating systems are being considered for very large installations where the economics are more favorable. Coal and other solids handling equipment are expensive per unit material handled in the smaller sizes.

Question 2: Approach to performing the work

This project was rated **2.0** for its approach.

- The approach is good. The researchers are working on increasing anode performance and understanding carbon reactivity.
- Although the project had overly ambitious objectives, the approach is reasonable. The approach is laid out to address the barriers.
- The project should focus on addressing key technical issues, such as performance and performance degradation, cell component stability, lifetime, etc. for direct operation of solid oxide fuel cells (SOFCs) on coal.
- The primary objective of the project was to demonstrate and characterize fuel cell operation on coal at the kilowatt-output level. Apparently this was never done and easier-to-use substitutes for coal were used instead.

- There was no obvious connection between the work and the technical barriers it claimed to be addressing. From the chemical analysis performed, it appears that coconut-based coke is not chemically similar enough to real coal or coke to be an appropriate substitute for laboratory experiments.
- The approach is unfocused and cannot lead to long-term evaluations of electrode activity, because the project is nearly over. The approach appears to avoid any activity that would lead to progress in feeding and handling coal in an SOFC. It is not clear how the project will lead to megawatt-scale fuel cells that operate on coal. Handling of ash constituents, including contaminants, which is usually the limiting factor in long-term durability in coal-fired power plants, has not been addressed.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **1.7** for its accomplishments and progress.

- This progress has made some accomplishments, but no breakthroughs.
- The progress appears far from achieving the objectives of developing kilowatt-scale coal-fueled SOFCs.
- Little progress has been made toward the original goals and objectives of the project.
- Poor performance has been demonstrated. The cells show 12% degradation after only 100 hours (even when operated on methane instead of coal). Current densities are far too low to make an economic system. While the objective of the project was to develop kilowatt-scale fuel cells, the project was limited to fuel cells with only tens of milliwatts. Too much of the work has been devoted to exploring SOFC materials composition and tape casting methods, which duplicates work done elsewhere in the Solid State Energy Conversion Alliance (SECA) program.
- The accomplishments appear modest. The performance of coconut coke and Petcoke is not very good. Performance of coal would likely be worse due to the release of numerous volatile contaminants, including sulfur. The performance of the unit cell needs to be improved before any thought of scale-up should be considered. Performance degraded more than 10% after about 100 hours when the cell was fueled with methane. Performance can be expected to degrade even more when coal is used.
- It is hard to believe that all five tasks are equally at 90% complete. The results are obtuse, making it difficult to assess the outcome of the project or their relevance to the overall project or to the Program needs. The plots on slide 7 provide an example of this. Although the label under the left-hand plot asserts increases in methane, carbon monoxide, and carbon dioxide, the plot is labeled with “m/e.” Thus, the reader must first assume that the molecules only lose one electron in the mass spectrometer and must then calculate molecular weights while also trying to interpret the graphs. Although the slide asserts, “water could be a good substitution for hydrogen in coal-based fuel cells due to its similar performance and higher rate of coke gasification reactions,” the right-hand plot on slide 7 actually supports a 15%–25% decrease in current density, and thus, performance.

Question 4: Collaboration and coordination with other institutions

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

- The project partners’ engagement with the project was not made clear.
- There was little apparent technical interaction with the collaborators on this project.
- The collaboration was with the Ohio Coal Development Office, FirstEnergy, and Coal Fuel Cell.
- The financial support of the collaborators is good, but there should be more collaboration with other developers of SOFC technology from the SECA program to help solve some of the anode/cathode/electrolyte composition and fabrications problems.
- There is evidence of collaboration with an end user, but it is not clear what role they played in the project. Collaboration with a major fuel cell developer would have been much more helpful in determining the direction of the research.
- This project has good collaboration with the Ohio Coal Development Office (state), which focuses on the fundamental research for the determination of the fuel cell efficiency, and with FirstEnergy Corp (industry), which addresses practical issues of the fuel cell stack scale-up.

Question 5: Proposed future work

This project was rated **1.7** for its proposed future work.

- This project is essentially complete and no further support for this project is recommended.
- The principal investigator has a clear vision for the next steps within the project, but, with the exception of durability testing, the steps still do not appear to address the stated barriers.
- Any additional work should focus on coal injection and fly ash removal (the unique aspects of the project), and defer trying to scale up to kilowatt size until better performance is demonstrated.
- This project needs to focus on addressing key technical issues of fuel cell operation on coal. It is too early to evaluate coal injection and fly ash subsystem and design, or fabrication and testing of a kilowatt-scale demonstration system.
- The proposed future work needs to be completed to demonstrate the feasibility of this approach. However, the project is nearly over with not much to show for the effort.
- The proposed work should advance development by further testing the coal injection and fly ash removal units, demonstrate the long-term performance and durability of the fuel cell stack in series and parallel configuration, and further test a small-scale (<10 kW) coke/coal fuel cell system.

Project strengths:

- The researchers have an excellent understanding of the barriers.
- The direct coal utilization fuel cell concept is a strength.
- This project has good leverage in using funding obtained from the State of Ohio (Ohio Coal Development Office).
- Using coal directly in an SOFC offers the potential to facilitate carbon capture from what is otherwise a fuel burdened with significant greenhouse gas potential. It looks like the project team has made some modest steps toward that goal.

Project weaknesses:

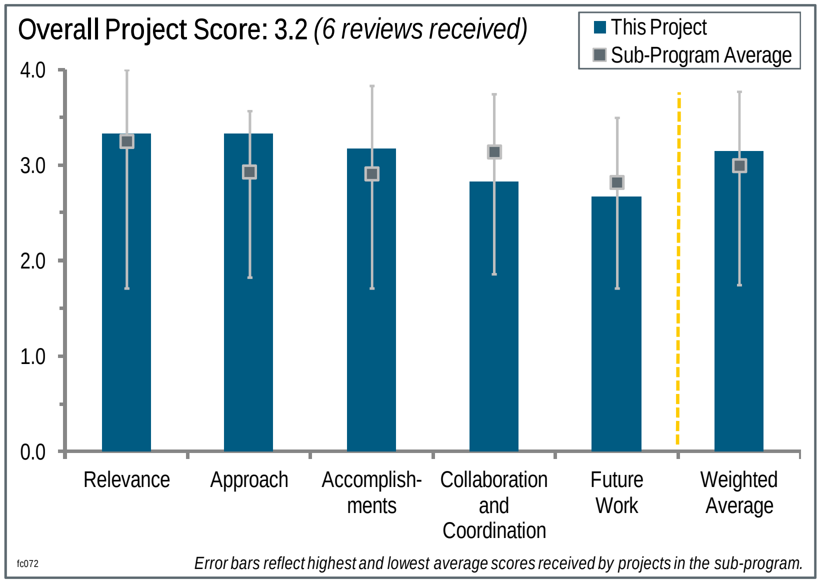
- This project is a very difficult, long-term R&D effort.
- The fuel cell performance presented to date does not justify further investment in this pathway.
- There is a lack of focus and expertise/experience on SOFC technology. Collaboration with SOFC developers and SOFC experts is needed and the metrics/targets are not defined.
- The primary objective of this project, fuel cell operation of coal gasification products, was never done or attempted. Apparently, because of impurities the coal has streamed, cleaner but meaningless substitutes were used.
- The project lacks relevance to the EERE fuel cell development effort. Only a small amount of progress is evident over the course of the project. Expectations were too ambitious and the team lacked a major SOFC developer to guide the research in fruitful directions.
- Poor performance has been demonstrated. Cells show 12% degradation after only 100 hours (even when operated on methane instead of coal). Current densities are far too low to make an economic system. While the objective of the project was to develop kilowatt-scale fuel cells, the project was limited to fuel cells with only tens of milliwatts. Too much of the work has been devoted to exploring SOFC materials composition and tape casting methods that duplicate work done in the SECA program in the Office of Fossil Energy.

Recommendations for additions/deletions to project scope:

- The project team needs to indicate, and preferably demonstrate, how a direct-coal fuel cell can provide performance that would make an industry-scale system worth investing in. It looks like this program is a long way away from that goal, and it is not clear how one would get there from here.
- The project completion date is May 31, 2012, and almost all the funds have been spent so there is little opportunity to change project scope. Nonetheless, any additional work should focus on coal injection and fly ash removal (the unique aspects of the project), and the team should defer trying to scale up to kilowatt size until better performance is demonstrated. Too much of the work has been devoted to exploring SOFC materials composition and tape casting methods that duplicate work done elsewhere in the SECA program.
- Considering the technical status and the progress to date, the researchers should delete tasks 4, 5, and 6.

Project # FC-072: Extended Durability Testing of an External Fuel Processor for SOFC

Mark Perna; Rolls-Royce Fuel Cell Systems (US) Inc.



Brief Summary of Project:

The overall objectives of this project are to: (1) conduct long-term tests in relevant environments for the three fuel processor subsystems that support operation of the 1 MWe solid oxide fuel cell (SOFC) power plant, including synthesis gas, start gas, and desulfurizer; (2) determine long-term performance of catalysts, sorbents, heat exchangers, control valves, reactors, piping, and insulation; (3) evaluate the impact of ambient temperatures on performance and component reliability; and (4) determine system response for transient operation. Project objectives from May 2011–2012 include the following: (1) complete desulfurizer subsystems testing in an outdoor facility; (2) evaluate sorbent vessel construction materials for the desulfurizer and reactor components for synthesis gas; and (3) perform start-gas subsystem testing.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.3** for its relevance to DOE objectives.

- This project fully supports DOE’s Hydrogen and Fuel Cells Program (the Program) and research, development, and demonstration objectives.
- Fuel processing for stationary fuel cells is directly relevant to DOE’s objectives. The focus on SOFC is narrow.
- The current project investigates the stability of SOFC under trace sulfur contaminants. Fossil fuel contains sulfur-related impurities. It is hard to reduce the H_2S level to less than 3 ppb. The current project is critical for SOFCs to operate under such impurities, especially for a long term.
- This project supports the Program through the development of fuel processing subsystems for a 1 MW SOFC for distributed generation. Specifically, in fiscal year 2012 the work involved an evaluation of the durability and performance of two critical balance-of-plant components: a catalytic partial oxidation (CPOX) fuel processor and a desulfurizer for a pipeline natural gas feed. Both the overall system and key components are being investigated.
- In this project, an external fuel processor is being developed and tested for potential use with a MW-scale SOFC operated on pipeline natural gas and, later, on alternative fuels, such as biogas. Stationary fuel cell systems for power generation or combined heat and power generation are expected to provide opportunities for early market entry of fuel cells. Successful demonstrations of natural gas reforming for this application will help to support the commercialization of SOFC technologies.

Question 2: Approach to performing the work

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

- This project should focus on testing under realistic conditions.
- The concept eliminates the need for on-site hydrogen storage. A process block diagram would have helped with understanding the three parts of the system.

- The start-up cycles are targeted to the temperature fluctuations, which accelerate the sulfur poison. This acceleration test method reveals the H_2S poison mechanism in cooperation with post-test analysis.
- The approach is sound. The subsystems have been developed over many years and have now been tested over significantly long time periods (the desulfurizer for 8,000 hours). Physical and chemical signs of degradation for key components have been/are being evaluated. All of this is necessary prior to a full-up power plant demonstration in the future. This will occur at some future date when the fuel cell stack becomes available; this is projected to be in 2013.
- The external fuel processor has been configured as a combination of three subsystems, each of which can be designed and tested independently under operating conditions appropriate for each. The synthesis-gas subsystem (for normal operation) is based on a CPOX, and it will be tested for up to 1,200 hours, including 10 start-up/shutdown cycles, and at operating loads of 10%, 50%, and 100% of design flows. The start gas is a non-flammable or weakly flammable reducing gas, primarily for heating the fuel processor system; this subsystem will be tested for 200 hours. The desulfurizer subsystem reduces 1–10 ppm sulfur in feed gas to less than 0.1 ppm using oxy-desulfurization followed by sulfur sorption; this subsystem will be tested for 8,000 hours.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- This project has made excellent progress toward the targets.
- All technical goals will be met by the end of the project; however, cost is not addressed.
- The project has been completed very well with outdoor test demonstrations.
- The major accomplishments in the past year are that the start-gas subsystem was tested for multiple start-ups, its durability testing plan has been prepared, and durability testing of the desulfurizer subsystem has been completed. Spikes of up to 200 ppb were observed at low-load operation, while mostly the measured sulfur levels were less than 40 ppb. The only items left to complete are the durability test of the start-gas subsystem and some post-test analyses.
- The accomplishments of this project are somewhere in the good-to-outstanding range. Major program accomplishments are the demonstration of the desulfurizer subsystem for 8,000 hours of operation, and a synthesis-gas subsystem for 1,000 hours. The start-gas subsystem testing (200 hours of operation) is in progress. This desulfurizer demonstration was a major program milestone. Investigators are concentrating on durability and performance; they appear to have abandoned start-up and shutdown time and transient operation, although this is less critical in most stationary applications. The investigators have achieved their target of <100 ppb of sulfur in product streams for the desulfurizer subsystem, while the DOE program goal is <10 ppb. The desulfurizer can produce sulfur spikes of at least 200 ppb during low-load operation. This could create problems for the SOFC unless sulfur-tolerant anodes have been developed in the Rolls-Royce (RR) system. They have not addressed this issue. The investigators suggest the use of corrosion-resistant, nitrogen-strengthened stainless steel for commercial units, which is a good idea. Finally, the RR target is 95% system availability, while the DOE goal is 99%. Modern competitive molten carbonate fuel cell systems are at 98% availability, creating an advantage in this area (however, SOFCs have many advantages over molten carbonate fuel cells in other areas).

Question 4: Collaboration and coordination with other institutions

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

- This project collaborated with RR fuel cells.
- The project is performed with the cooperation of the Ohio Department of Development and the Stark State College Fuel Cell Center with students' involvement.
- No stationary fuel cell suppliers, other than RR itself, are involved. No collaboration with gas suppliers or fuel processing catalysis companies is evident. The only paper or presentation was from the 2011 DOE Hydrogen and Fuel Cells Program Annual Merit Review. The fuel cell community does not benefit from the RR results.
- There has not been a lot of external collaboration. Interaction with the Ohio Department of Development is financial and siting. The relationship with Stark State College involves mostly education/training. There are no interactions with other private industry, because the technology is RR proprietary.

- This project involves active collaboration among the project lead, RR, the Ohio Department of Development, and Stark State College. Thus, this collaborative effort includes industry, state/local government, and an institution of higher learning—an ideal combination.

Question 5: Proposed future work

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

- The future work will contain the final report.
- The work for the remainder of the project is appropriate.
- Investigators will complete the degradation assessment of the desulfurizer and start-gas subsystem. Following the start-gas test, key components will be examined for chemical or mechanical degradation. A full-up test with the RR solid oxide system (large, but not specified size) is planned. The future work is not contingent on DOE funding.
- This project is nearing completion. The only activities left are to complete the durability testing of the start-gas subsystem, conduct post-test analyses, and prepare a final report on the project. Outside and beyond the scope of the project, the team plans to operate an SOFC on the product gas from the fuel processor, if the necessary funding can be arranged.

Project strengths:

- This project has well-defined targets and plans and a focus on testing and demonstration.
- This project has a very capable and collaborative project team, and a systematic work breakdown structure and program plan.
- This project developed an approach for testing external fuel processors for SOFCs. The desulfurizer subsystem has been evaluated for up to 1,000 hours. Material and catalyst disabilities were also investigated.
- A strength of the project is the demonstrated durability of subsystems and key components. RR considers the development of fuel cell systems for stationary power to be a key future product line and is capable of funding this work going forward.

Project weaknesses:

- The integration with SOFC operation is a weakness of this project.
- Spikes of high sulfur in the product line could cause problems unless RR has developed sulfur-tolerant anodes for their fuel cell. This project is also late in meeting milestones.
- Two reviewers said this project has no weaknesses.

Recommendations for additions/deletions to project scope:

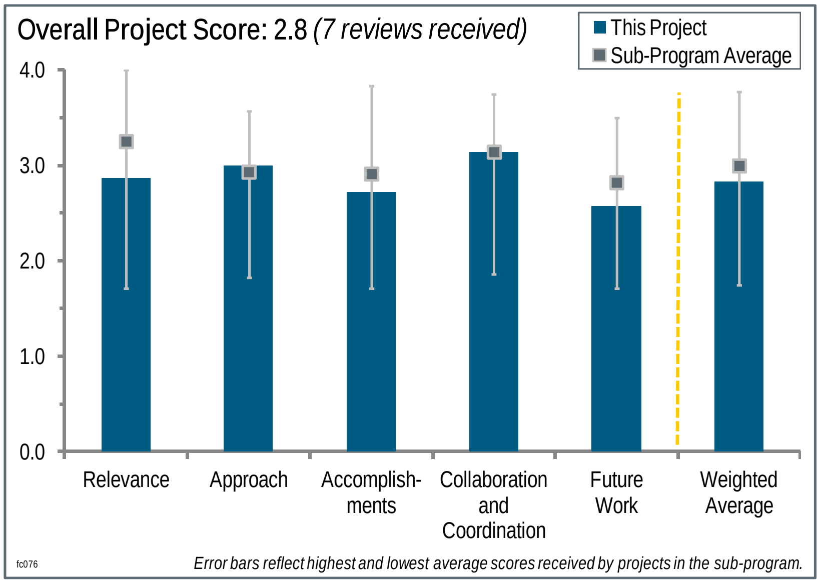
- The Program should continue to follow the work.

Project # FC-076: Biomass Fuel Cell Systems

Neal Sullivan; Colorado School of Mines

Brief Summary of Project:

The main objective of this project is to improve the durability and performance of solid oxide fuel cell (SOFC) systems while lowering costs. The objective is focused on three tasks: (1) develop materials and architectures to improve SOFC durability under biomass-derived fuels, (2) develop biogas fuel processing strategies for SOFC integration and develop low-cost ceramic microchannel reactive heat exchangers for fuel reforming, and (3) provide modeling support for tasks 1 and 2 using computational fluid dynamics (CFD) and chemically reacting-flow tools.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **2.9** for its relevance to DOE objectives.

- This project is targeted at improving the durability, performance, and cost of SOFC systems. It also seeks to employ wastewater products as a hydrogen (H_2) source.
- The project is well aligned with DOE's goals of increasing the durability and efficiency of SOFC systems. The project supports DOE's goal of lowering the cost of SOFC systems.
- This project generally supports the goals and objectives of the DOE Hydrogen and Fuel Cells Program by studying the potential for using bio-waste as a fuel for fuel cell power generation.
- The project goal is to improve the robustness of hydrocarbon- and biomass-fueled SOFCs and systems. The project goals are relevant to the DOE goals of increasing the durability, performance, and transient operation of SOFC systems and decreasing the cost of balance-of-plant (BOP) components.
- The major merit is the durability of SOFC stacks when operated with biogas. The BOP integration serves to increase performance. The durability of the BOP is an issue when operated with biogas, which always contains detrimental contaminants. Indirectly all of these points contribute to lowering cost.
- The goals are to improve the durability of advanced materials, improve control strategies, decrease costs, and develop low-cost integrated reactive heat exchangers. It is unclear how this development fits into overall DOE research, development, and demonstration goals.

Question 2: Approach to performing the work

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

- The approach is well thought out and focuses on overcoming barriers such as the cost and durability of SOFCs.
- The proposed approach is logical and systemic, combining both experimental and modeling tasks designed to extend and enhance SOFC performance on biomass fuels.
- Collaboration between the tasks has improved. The cooperation between modeling activities and hardware development can still be improved.
- The approach is focused, it creates next-generation SOFC materials and architectures, and it utilizes microchannel-reactor technology for tight thermal integration.

- One major approach is improving the materials and manufacturing technologies of SOFCs. The other major approach is finding new techniques to reform biogas products so that they can be better utilized in the SOFCs. Modeling supports both of these.
- The project consists of three tasks. The approach in task 1 is to develop materials and architectures to improve the durability of the tubular barrier layer and anode supports. The approach in task 2 is to develop bio-fuel processing strategies and low-cost ceramic, microchannel, reactive heat exchangers for fuel reforming. The approach in task 3 is to provide CFD modeling support for tasks 1 and 2.
- The ceramic approach is backed by outstanding competence of CoorsTek Inc., and as such, it is sound. The approach already comprises the cell itself and BOP components, such as a heat exchanger and a reformer based on the heat exchanger technology. The latter is designed for reforming all of the fuel gas because no external water is supplied for pre-reforming, and the CO_2 originating from the biogas production is used for CO formation. This approach is outstanding for biogas-operated SOFCs. Developing control algorithms in this program, which otherwise operates on single cells and BOP components, seems premature, particularly because the systems design cannot be tested at this stage or in the near future. The targeted systems size of 1 kW can be understood at this stage of development from a ceramic point of view. For biogas applications, bigger targets should be considered (e.g., in the 100 kW range). One kW is more of a portable application that can be powered with any other, probably bottled, gas.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- This project's accomplishments have been fair. The researchers have demonstrated extended operation of the first tubular barrier-layer SOFC and developed ceramic microchannel reactors for methane steam reforming.
- A demonstration of 50 hours of operation of the perovskite-based tubular anode supports has been performed. The biogas reforming strategies have been completed. Progress has been made on the fabrication and modeling of ceramic microchannel heat exchangers.
- Significant progress has been made toward creating a more durable tubular SOFC by integration of perovskite barrier layers. Significant progress was made toward the development of strategies for biogas fuel processing at wastewater treatment facilities. Significant progress was made toward designing a cost-saving microchannel heat exchanger for fuel processing.
- For the project ending in September of this year, the work in task 1b (perovskite-based anode support) seems far from a positive result. Thermomechanical modeling of ceramic heat exchange is still missing, but it is essential for the evaluation. Pressure drop in the microchannel reactor is critical in system operation. In addition, comparison to a steel heat exchanger is missing and reforming results should be compared to thermodynamic equilibrium.
- Progress seems to be slow and the project is almost finished (85%). The modeling does not seem to follow the experiment very well and the results on the microchannel heat exchanger do not offer much new information. The real challenge for microchannel technology is for the reforming of biomass feed streams where fouling/plugging the channels with impurities, particulates, and delamination of the reforming catalyst is likely to be a serious problem. The flowchart for the system at the Denver Wastewater Treatment Facility is generic and trivial.
- Task 1 has been completed. Tubular perovskite barrier layers were synthesized and integrated with the CoorsTek Inc. SOFC. The project demonstrated >12 days of continuous operation with biogas. The project is extending efforts to perovskite-based SOFC cells with SLT-based anode support and a Ni-YSZ anode functional layer. In task 2, the project developed and validated models to guide the definition of external-reforming operating windows. The principal investigator (PI) demonstrated that the SOFC electrochemical performance on CPOX-O_2 and steam-reformed biogas can match the performance with humidified hydrogen (H_2). The PI measured the performance of a ceramic microchannel heat exchanger over a broad range of operating temperatures and showed that the addition of rhodium to a steam-reforming catalyst improves the conversion of CH_4 to H_2. In Task 3, the project integrated the ANSYS-FLUENT CFD model with the CANTERA model for chemically reacting flows. The integrated model was used to optimize the backing-side conditions for reactive testing and to determine the set points for high CH_4 conversion. The project is extending the high-fidelity flow models to develop rapid linear models for predictive controls that can be used to meet

load demand while satisfying the constraints. The PI is conducting systems analysis to evaluate the integration of an SOFC system with the Denver Wastewater Reclamation Facility.

- Great progress was made on the SOFC tube by integrating a new SLT barrier layer and achieving a reasonably good areal resistance, the generally low areal resistance of tubular designs considered. The barrier layer test results should be extended to a minimum of 2,000 hours in order to obtain reliable results, because some impurities have the tendency to accumulate and hence only become obvious after extended operating times. The ceramic heat exchanger and reformer are great developments. Using microchannel technology for this allows for high-systems integration. On the systems level, the pressure drop in microchannels should be considered, since it may lead to exaggerated compression losses in the system if the channels were very small. On the systems integration side, early efforts should be started to connect the ceramic devices to the piping, which was a major hurdle for integrating ceramic heat exchangers for industrial applications about two decades ago and which finally made industry abandon that pathway. Further information on the gas-tightness of the ceramic BOP components would be important. Based on the developed models, operating conditions for the reformer could be figured out that allow for biogases use at good efficiencies in the SOFC. Considering the budget, the multiple tasks involved, and the duration of the project, the results achieved are outstanding.

Question 4: Collaboration and coordination with other institutions

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

- The collaboration between CoorsTek Inc. (Golden, Colorado) and tubular SOFC is fair.
- CoorsTek Inc. is a partner in this project and supplies materials and SOFCs.
- This project has coordinated collaboration well between a university and an industrial partner.
- Further collaboration outside CoorsTek Inc. and the Colorado School of Mines (CSM) is missing.
- This project has good collaboration with CoorsTek Inc. The collaborations could have been expanded to include expertise in biomass reforming, which is ongoing at many locations currently.
- This project has very good collaboration with CoorsTek Inc., a major United States ceramics manufacturer. CoorsTek Inc. receives no funding for their activities on the project.
- The fact that a strong industrial lead from CoorsTek Inc. was combined with the scientific approaches of research institutions is the strength of the project. All partners performed their work with high proficiency.

Question 5: Proposed future work

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

- The project ends in September 2012 and is 85% complete.
- The proposed work contributes well to the goals.
- The planning should have more detail with well-defined objectives.
- It is not clear what the remaining work is; there should be more detail explaining how the remaining work can be accomplished in the remaining few months.
- The PI has laid out plans to complete each of the three tasks in the remaining few months of this project.
- The plans are built on achieved progress and may lead to the development of a new, more robust SOFC architecture.
- This project is almost over; in the remaining time effort should focus on the experimental components of tasks 1b and 2b and the researchers should work closely with the existing partner.

Project strengths:

- This project has a strong industrial partner.
- The academic knowledge of CSM and the industrial expertise of CoorsTek, the largest ceramic company in the United States, is a strength of this project.
- This project has a good combination of academic knowledge and industry experience. There is good correlation between modeling and hardware testing.
- This project has a strong industrial connection. The ceramic microchannel work may find applications in a number of different industrial areas.

- This is a multidisciplinary approach that involves catalysis, engineering, and advanced modeling. This project uses CFD modeling as a guide to design heat exchangers/fuel processors.
- This project has a combination of a strong industrial lead, high ceramic competence, and a strong research. Developing a perovskite anode is a good approach for combating contaminants instead of painfully cleaning the biogas to extreme levels.

Project weaknesses:

- This project has no weaknesses.
- This project could be making progress at faster rate, because it is three years along.
- This project's progress is slow and it lacks an experimental validation of models developed in the project.
- This project lacks a clear definition of the deliverables. The PI has not explained how CoorsTek has benefited from this research.
- Tubular SOFCs are not durable enough to tolerate sulfur, and incorporating additional equipment required to eliminate impurities may increase the cost of the total system.
- The project has not evaluated whether the stack and heat exchanger concept used has the potential to achieve the performance and cost goals.
- The project is very broad and the target of developing 1 kW and considering biogas reforming are not fully in line. The project can be understood as a scouting project on what is possible to achieve using ceramic approaches. In that sense, the broad approach can be understood as a strength.

Recommendations for additions/deletions to project scope:

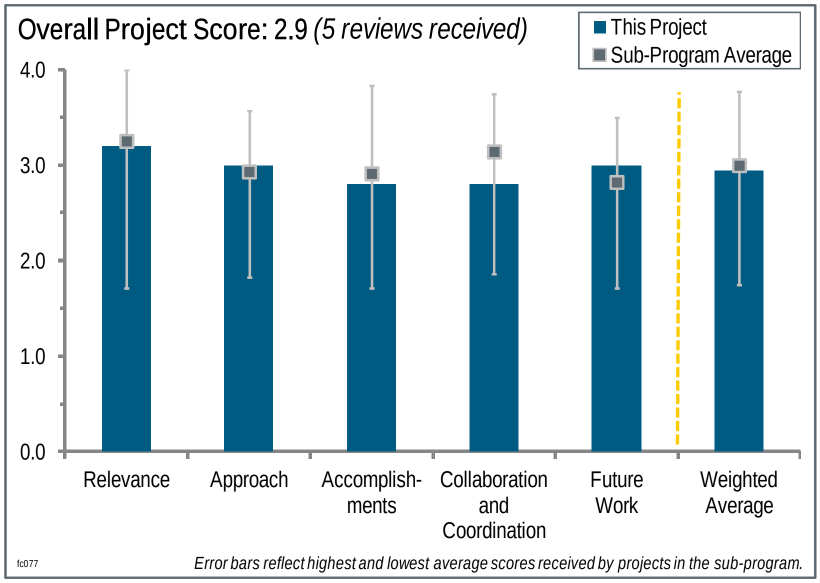
- This project should maximize experimental results in the time remaining in the project.
- This project should concentrate on real stack tests using biogases.
- The researchers should conduct a long-term test of the developed ceramic tubes under biogas (i.e., >2,000 hours), and integrate the ceramic heat exchangers with the tubing at an early stage.

Project # FC-077: Fuel Cell Coolant Optimization and Scale-up (plus work under SBIR III project)

Satish Mohapatra; Dynalene

Brief Summary of Project:

The goals of this project are to: (1) develop a flexible, instrumented fuel cell coolant system; (2) experimentally qualify coolant properties and performance through steady operation durability testing, non-operational durability testing, load cycle impact testing, and advanced coolant accelerated qualification testing; (3) determine corrosion inhibition efficiency of coolants; (4) increase surface charge of nanoparticles; and (5) conduct long-term testing and demonstration.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.2** for its relevance to DOE objectives.

- Long-life, low-conductivity coolants are needed for meeting DOE's fuel cell goals.
- There are already coolants that are suitable for the DOE Hydrogen and Fuel Cells Program (the Program). This project is not a high priority for the Program.
- It would be useful to develop a specific coolant that meets the requirements of automotive fuel cell systems, but there are also other methods to deal with this issue.
- The project supports DOE's research and development objectives, in particular for the balance of plant by developing a new fuel cell coolant. The main innovation is not using a resin filter, which reduces cost, weight/volume, maintenance needs, and the pressure drop.
- This technology is very creative and a good fit for the Program. In industry in the past there was almost no adequate selection of coolant systems and development of a new one was needed. Performing work such as this in the public arena will help other coolant manufacturers to endorse and develop the ideas of additives for optimal fuel cell operation.

Question 2: Approach to performing the work

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

- This is a very creative and effective approach.
- The project has been well structured from Phase I to Phase III. Returns from field trials should be added and reported regularly in Phase III.
- It is not clear how this approach (nanoparticles) will work outside the laboratory in the automotive application. There were significant issues with test results at 120°C.
- This is a reasonable approach, though it is not clear that the scale-up process is near optimal. More details about the methodology used would be helpful. Higher-temperature operation remains somewhat of an open issue.
- The use of nanoparticles to enhance the thermoconductivity of fluids is not uncommon in the industry. Typically nanoparticles use surfactants to prevent settling or flocculation. This needs to be described in the system. The approach did not seem to include tests at low-temperature operation.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- The company has shown a pathway to develop an effective coolant.
- Test data has been generally positive, but incomplete, especially at higher temperatures. No automotive end-user data was presented, although there are indications that testing is underway.
- Because scale-up is a significant objective, it is important that the path be established to much larger production rates. It is not clear that the current manufacturing technique will scale well. Otherwise, this project has made solid progress.
- The accomplishments and progress were good this year and have been presented correctly. The fuel cell coolant was optimized up to 80°C operation, and scale-up to 100 L has been completed. Improvement for operation at higher temperatures is needed. To do that, better mechanism understanding is requested. The testing facilities have been completed, but long-term testing has not been presented. The corrosion inhibitors were validated in short-term testing using immersion and electrochemical methods. Plastics and gaskets contribute more to conductivity increases than metals in pure water. This has to be taken into account in the use of this coolant depending on the system design. But further investigations using the developed coolant are needed to qualify it.
- The degradation of the fluid at high temperatures needs to be understood. Fuel cells are moving to a higher temperature of operation, so thermal stability is important. The conductivity in some cases appeared to increase over the baseline with the addition of the nanoparticles. With a nanofluid, settling and flocculation are important, particularly over time. These characteristics need to be understood over the same range of temperatures that the fuel cells will experience. The presenter mentioned that the nanofluid separates when frozen. This would concentrate the nanoparticles, which would increase the pore clogging and channel clogging as well as concentrate the surfactants to a level of concentration that may impact gaskets and other components, so chemical compatibility needs to be tested.

Question 4: Collaboration and coordination with other institutions

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

- The inclusion of a fuel cell company to the team is extremely important to validate the project.
- This project collaborated only in the tests. It is not clear that the testing covers the most critical parts of the future operating parameter space.
- The project is limited in scope and funding, and as such, no end-user data is presented, but it is obviously critical to the success of the concept.
- The company is working with fuel cell manufacturers, thus it is following system performance specification flow-downs that will allow it to integrate with field systems.
- Collaboration is correct, but it should be enlarged in particular with systems producers in order to improve the industrial feedback on the approach and the experimental field return.

Question 5: Proposed future work

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

- The completion plan is acceptable.
- The manufacturer is looking at the right tasks to further deploy their coolant.
- The proposed future work is a good start. They need to expand beyond ambient temperature testing for this system.
- There is still much work (durability testing) ahead, but there is limited funding to feed back the results of the testing into the coolant development.
- The future work is correct. At this stage, cost targets and achievements should be presented. Testing this coolant in different system applications (automotive, forklift, stationary) would make is easier to better appreciate the potential of this product.

Project strengths:

- This project is an interesting concept.
- The innovation of the cooling concept and combining water-based materials to achieve the goal are both strengths of this project.
- This project is taking an interesting approach to improving the liquid coolants for fuel cells. The project seems to be well organized.
- This is an innovative approach that may lead to a significant positive impact on the fuel cell system in terms of mass, weight, and maintenance by suppressing the resin filter. The different materials present in a fuel cell system have been taken into account.

Project weaknesses:

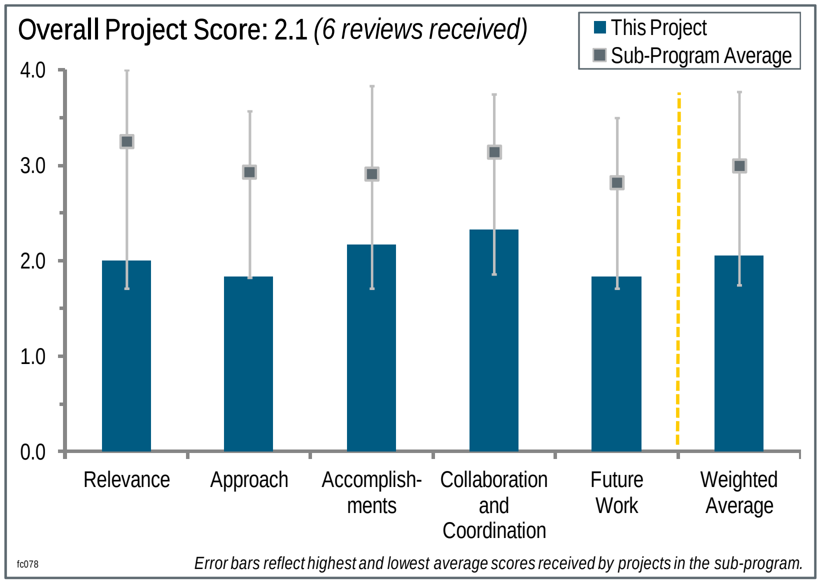
- It is not clear that the scale-up path will be adequate for large production volumes. It is also not well communicated or justified why water-based coolants are required.
- This project needs a better definition of success, especially for accelerated testing. It is not clear if material tests indicate whether the coolant meets requirements or is simply better than the alternatives.
- No long-term data was presented. To facilitate the comparison, the results presentation could be more homogeneous, particularly with regard to the units. Sometimes the durations are in weeks and sometimes they are in hours. A better understanding of the degradation mechanisms should be carried out.
- Freeze-thaw characteristics need to be investigated. Of particular concern is the separation of the materials that was shown in the discussion portion. This was not addressed in the future work section and should be added. The surfactant chemical compatibility with the materials and particularly the seals needs to be considered. While the surfactant concentration is low, the materials will be in contact for long periods of time (years), and during freezing there is a separation of the components, which would concentrate the surfactants in the seals.

Recommendations for additions/deletions to project scope:

- Considering the constrained budgets for the Fuel Cell Technologies Program, this seems to be a low priority.
- The researchers should work to understand the effect of conductivity increases at higher temperatures (105°C–120°C) in order to ensure the 5,000 hour target with <2 µS/cm measures the thermal conductivity effect. The researchers should also investigate the effect of the potential gradient in a high-voltage stack over time to see if it has any effect or not on the nanoparticles and corrosion inhibitors stability. The researchers should measure the conductivity variations in various materials with the developed fuel cell coolant as they have been doing with ultra-pure H_2O at 80°C for three weeks, and then focus the investigations on the materials that present the highest interaction.

Project # FC-078: 21st Century Renewable Fuels, Energy, and Materials Initiative

Joel Berry; Kettering University



Brief Summary of Project:

The goals of the project include: (1) the development of an improved high-temperature polymer electrolyte membrane (PEM) fuel cell membrane capable of low-temperature starts (<100°C) with enhanced performance; (2) the development of a 5 kWe novel catalytic flat plate steam reforming process for extracting hydrogen (H_2) from multi-fuels and integrating them with high-temperature fuel cell systems; (3) the development of an improved oxygen (O_2)-permeable membrane for high-power-density lithium (Li)-air batteries for ease of use and reduced cost; (4) the development of a novel, high-energy-yield agriculture bio-crop (Miscanthus) for alternative fuels with minimum impact on the human food chain; and (5) the extension of the math and science alternative energy education program to include bio-energy and power.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **2.0** for its relevance to DOE objectives.

- The project is relevant to the objectives of Fuel Cell Technologies Program (FCT Program) Multi-Year Research, Development and Demonstration Plan (MYRDDP). The activities are aligned to DOE's goals. This project brings a very wide variety of initiatives directed toward many different areas of alternative energy projects. This program seems to be very broad and covers very diverse areas of science.
- This project has five separate tasks. Two of the tasks (developing fuel cell membranes and catalytic plate reformers) are directly related to DOE Hydrogen and Fuel Cells Program (the Program) goals, while the other three (high-power-density Li-air batteries, high-energy-yield bio-crop, and alternative energy education) are not relevant to the Program.
- This project has four different unassociated parts. Only the high-temperature membrane is relevant to the fuel cell components portion of the FCT Program. The steam reforming reactor has a slight relevance to H_2 production, but this type of design does not appear relevant to the goals of the hydrogen production portion of the FCT Program. It is unclear why battery development is being supported by the FCT Program. The other portion should belong to DOE's Biomass Program.
- This project involves the development of several technologies, including high-temperature PEM membranes, a flat plate steam methane reformer, Li-air batteries, feedstock for biofuels production, and education. The first two support the Program's objectives. Batteries and biofuels production belong in the Vehicle Technologies and Biomass Programs, respectively. Education could be said to be a Program activity.
- The project plans to develop: (1) a high-temperature fuel cell membrane, (2) a steam reforming reactor, (3) a high-power-density Li-air battery, and (4) novel high-energy-yield agriculture bio-crop. Task 1 is relevant to the Program. The battery part might be of interest to the DOE Vehicle Technologies Program and the bio-crop development may be relevant to the Biomass Program. While the overall project supports the DOE Office of Energy Efficiency and Renewable Energy's goals, certain tasks do not appear to be relevant to the Program.

Question 2: Approach to performing the work

This project was rated **1.8** for its approach.

- There are interesting areas of research here, but it is not clear why there are such diverse topics in one project.
- The approach of this project is unclear and appears scattered due to the four unassociated pieces of the project. The membrane portion objective appears to "increase" conductivity and enable a high-temperature operating PEM with low-temperature start-up; however, it is unclear how the researchers are approaching the low-temperature start-up. Overall, there is almost no detail in the approach section.
- The approach of separating the projects into several different tasks to cover multiple areas of development is the right way to manage different scientific developments required for the success of this project. The technical barriers for each development pathway are addressed properly; however, a little more description would have been better. Very few details were given for the Li-air battery and "Biofuel from High Yield Energy Crop" approaches.
- Insufficient detail was presented to determine how well the relevant barriers are addressed in the two tasks related to the Program. The barriers to the development of the high-temperature membranes are not identified. The membrane work does not appear to address the barriers to membrane durability or the operation over an operating range, which includes water condensation. The leaching of phosphoric acid and Sulfonated-Polyhedral Oligomeric Silsesquioxane (S-POSS) are not addressed. There may be some impact on improving mechanical properties and the durability of polybenzimidazole (PBI)-phosphoric acid membranes by adding the S-POSS. The reforming work does not identify the barriers for fuel reforming. Barriers, such as reformer durability, do not appear to be addressed. The issue of lower surface area on a plate reactor compared to a pelletized bed reactor is not addressed. A non-platinum-group-metal catalyst is used, which should address the reformer cost issue.
- The investigators propose to tie together several different research objectives into a strategy for power generation. The biofuel generated from Miscanthus will "feed" the flat plate reformer, which will provide H_2 to a PEM fuel cell (for which the project is developing a membrane for low-temperature operation). When the whole system is used as an auxiliary power unit, the Li-air battery is used to provide power while the fuel cell "gets up to speed" in about 20 minutes. All the technologies under development in this project have significant barriers. Solving any one of them involves a major effort, and trying to do all of them is probably not possible with the funding provided. The project gives the appearance of starting from scratch in several different technology areas. There is no interaction with external resources that have dedicated many years and lots of research and development investment in the areas of research involved in this project, except for membrane development, which Michigan Molecular Institute has experience in. Frankly, it is not obvious at all why the researchers are developing an Li-air battery when there is so much work going on in this area around the world. The novelty of the materials and design they are pursuing is not clear. A similar comment can be made about the development of Miscanthus as a high-yield bio-crop. A lot of work has been done on it at places such as the University of Illinois. The data shown in figure 14 are taken from the literature. The process by which they convert biochar to E85 is not described. There is no plan for commercialization.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- Not enough results were presented to interpret whether the objectives are met, such as electrochemistry using the membrane and testing with standard activity and durability test protocols.
- Progress since fiscal year 2011 has been limited. A different membrane and synthesis pathway has been followed, which will take some time before it is fully developed and tested, although some promising conductivity results are given for 10% S-POSS. Results presented for the Li-air battery in O_2 versus dry air are a somewhat obvious results. The experimental results for the fuel reformer are expected. The computational fluid dynamics model used to design the reformer is not described. There is no attempt to relate the SEM results to the Li battery performance, and the anode and cathode reconstruction in cycling is well known.
- No lists of publications were provided. Using carbon for the air electrode of an Li battery makes no sense. This is also irrelevant to the stated goal of developing a better O_2-permeable membrane for high-power-density Li-air batteries. The durability of the membrane that has been developed is not clear. The principal investigator (PI)

informed this reviewer that the researchers are waiting for independent confirmation of the conductivity of these membranes. This data will be useful. Two of the four graphs in the reformer section are a repeat of 2011 data.

- Slide 17 indicates that the researchers have presentations, publications, and patents, yet none are listed anywhere. There is one slide on the membrane development, with data only at 90°C, 120°C, and 150°C. It is confusing why there is no data for low temperatures (i.e., approximately 20°C), because one of their main objectives is for low-temperature start-up. It appears they have made a total of seven membranes. For the reformer, the presenter did not show data relevant to the fuel conversion and selectivity to H_2 production, and nowhere did the presenter state what fuel was used.
- Overall, good technical accomplishments in all five different developmental pathways had been achieved. However, from the reported results it seems that the remaining part of the project will be difficult to complete by June 2012 when the project ends. It is not clear whether an extension of the project had been requested by the team or not. A good portion of in-situ evaluation work is needed for Li-air batteries and high-temperature membrane areas. For Li-air batteries, O_2 performance is reported, while battery performance in the air is the goal of the project and is not finished. Similarly for high-temperature membrane development, the high-temperature fuel cell performance evaluation is not completed.
- The fuel cell membrane work was able to improve on the conductivity the researchers obtained with PBI-phosphoric acid by doping the PBI-phosphoric acid with S-POSS, but this project has not improved on the conductivity of PBI-phosphoric acid reported in the literature at high-temperature conditions. In discussions with the researchers, they claimed to have improved the mechanical properties of the PBI-phosphoric acid membrane, but data for mechanical properties was not shown. It is unclear from this work whether S-POSS will improve conductivity of standard PBI-phosphoric acid membranes. There was a typo in the poster that suggested increasing the fuel-to-steam ratio increases H_2 production; however, in discussions with the researchers they revealed that this is a typo and it should have read increasing the steam to fuel (or steam to carbon) ratio increased H_2 production. Discussions also revealed that this is a microchannel plate reformer. Microchannel plate reformers have been investigated by Pacific Northwest National Laboratory in the past and there did not appear to be new results here.

Question 4: Collaboration and coordination with other institutions

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

- This project seems like it is several independent projects run without any coordination.
- The project appears to be a collection of five separate tasks with little collaboration between the groups.
- This project has good collaboration, but is there not a national laboratory or business partner that could give a good perspective on the work.
- The researchers have three different institutions involved in this project, but there appears to be no collaboration between them, as they are working on totally different things.
- The team consists of good partners, including universities and research laboratories. The inclusion of a national laboratory could have helped the team to get access to better research facilities and resources.
- The PIs have formed collaborations with Michigan Molecular Institute and Saginaw Valley State University; however, the project is not well coordinated overall. It appears that investigators proceed mostly independently on the separate components of the project. The investigators do not appear to have reached out to other institutions and private industry with long histories of membrane, Li battery, and fuel cell development. Such collaborators could help the project move ahead more quickly. There is no current industry involvement, which could lead to commercialization. The PI reports just having contact with a private company, Global Energy Innovation, which is also located in Flint, Michigan.

Question 5: Proposed future work

This project was rated **1.8** for its proposed future work.

- Given the project is 95% complete, there is too much future work. It is unclear how the researchers plan to accomplish all of it.
- The work is not focused well enough and would require a significant amount of funding to achieve meaningful results for all goals.

- This project should slow down on the biofuel and Li-air battery and concentrate on developing membrane electrode assemblies and testing their results on the high-temperature membranes, which show some promise.
- It is impossible to tell what is different in terms of the membrane development future work and what was done in the past. The other three tasks are not relevant to the FCT Program's MYRDDP.
- The future work described is aligned with the proposed work of the project. The team needs to complete the cell performance evaluation for Li-air batteries and high-temperature fuel cells quickly because the project completion date is only a month away.

Project strengths:

- This project could make progress in membrane development.
- The team is well organized and capable of evaluating such diverse areas of science. The team is equipped with the necessary resources required for the success of this project.
- The membrane task and reformer task are relevant to the Program. The membrane work could improve the performance of high-temperature PBI-phosphoric acid systems.

Project weaknesses:

- The tasks of the project do not seem relevant to the Program.
- This project is spread too thin over some interesting areas. More collaborators or a less-broad range of topics addressed would have achieved more results.
- The project does not appear to have specific targets or milestones for performance. Much of the project is unaligned with the Program's goals.
- This project is too diverse. This will make it difficult to achieve significant progress in any one technology area, let alone all of them with enough resources to bring it together at the end. The project coordination is poor, and it does not leverage work that has been done or is ongoing in other institutions/private industry. There is also no industry involvement.
- This project is very diverse and a project duration of two years is very little for the completion of all the proposed tasks in the project. Despite this limitation, the team did a commendable job of pulling together a functional working group between three different institutes and completed the majority of the tasks within the proposed time.

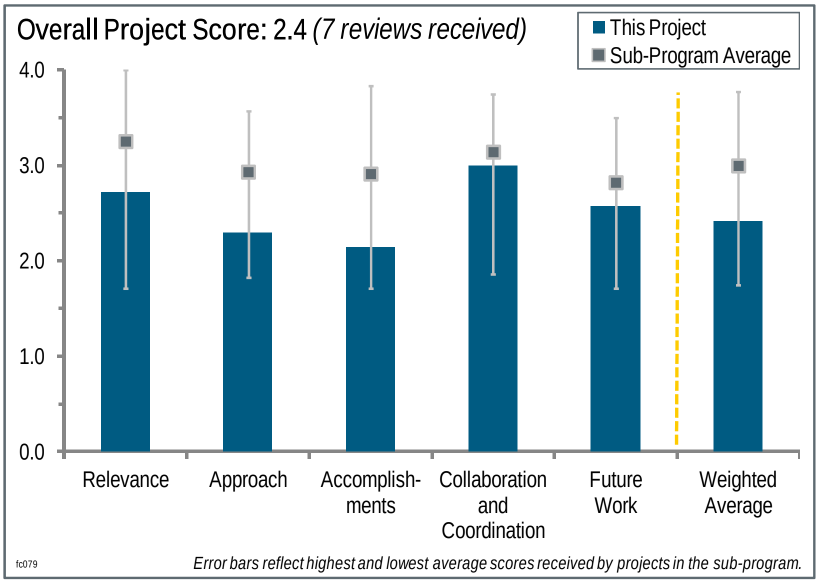
Recommendations for additions/deletions to project scope:

- This project should not be continued.
- Everything but the high-temperature membrane work should be eliminated.
- The bio-crop and Li-air batteries are unrelated to the Program goals and objectives and should be dropped.
- In the future, DOE should allow more time for such diverse multi-area development projects to allow successful completion of the project in time.

Project # FC-079: Improving Fuel Cell Durability and Reliability

Prabhakar Singh; University of Connecticut Global Fuel Cell Center

Brief Summary of Project:
Goals of this project were to develop an understanding of the degradation processes in advanced electrochemical energy conversion systems, and to develop collaborative research programs with industries to improve the performance stability and long-term reliability of advanced fuel cells and other power generation systems. The technology objectives focused on advancing fuel-cell-based power generation systems architecture, including renewable hybridized energy conversion and storage; developing and testing novel cell and stack structural and functional materials; and gaining a fundamental understanding of chemical, mechanical, electrochemical, and electrical processes.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **2.7** for its relevance to DOE objectives.

- Most of the topics reported on are in-line with DOE's objectives.
- Overall, the objectives are addressed through a very wide range of interactions with industry partners.
- The project is essentially 10 independent projects with varying levels of relevance to DOE's research and development objectives.
- This project is relevant to DOE's objectives; it is critical to understand the degradation processes and mechanisms for electrochemical systems, especially with the long endurance needed requirements.
- The project seems to cover several different areas and have some general objectives, but it is hard to understand how the teams help each other and what specific goals they want to meet.
- This is an atypical project in that although it can properly cite relevance to the cost, performance, and durability barriers, it looks at all types of fuel cells and therefore is not likely to be able to address the three barriers simultaneously for any one type, such as polymer electrolyte membrane (PEM) fuel cells for transportation.
- The focus on fuel production and processing involved with bio-gas scrubbing, desulfurization, and nanostructured support for acid-based electrolyzers are significant steps in advancing fuel cells. The work related to gas diffusion layer (GDL) manufacturing will definitely be an integral part of membrane electrode assembly manufacturing in the future as well.

Question 2: Approach to performing the work

This project was rated **2.3** for its approach.

- The approach here is to broadly address durability and reliability from a system perspective. Many different systems are considered (PEM fuel cells, molten carbonate fuel cells [MCFCs], and solid oxide fuel cells [SOFCs]). This broad approach may not be efficient.
- This project is too broad and the approach lacks a plan that ties the various work streams together. Although each work stream is somewhat organized in its topic area, these are niche areas of research. This seems like a buckshot into the fuel cell research space.

- The large number of small, disconnected projects significantly lowers the potential of meaningful impact resulting from any specific project. Teaming professors with industrial partners helps to provide guidance to projects.
- The approach is well defined, along with the roles and responsibilities for the team members. The presentation did not define go/no-go decision points for the tasks; this would have been helpful to determine the progress to date of meeting targets. It is recommended that go/no-go decisions be presented at the next review to evaluate the basis for the down-selection and scale-up efforts. A modeling effort for tasks 2–4 would have been helpful; modeling could reduce the number of tests to be performed and provide a quick evaluation of catalysts, materials, and electrode architectures.
- The approach is very broad and ranges across all technology aspects of all fuel cell types. It therefore has the danger of not being able to complete anything meaningfully for any one type. But the researchers' unique collaborations and method of execution saves the day on this point. There is also the opportunity for more broadly based mechanisms that apply across the board to come out because of the close networking possible among the co-located C2E2 faculty and students and be understood.
- While the project is very ambitious in tackling multiple problems with various principal investigators (PIs), it feels as though the focus has been a bit diluted in the end. The main goal of the project was to improve the durability and reliability of fuel cells. While the majority of the efforts have been focused on understanding the various processes and the modes of improvements in them, the clarity regarding the big picture is lacking. Maybe a focus on more than one type of fuel cell system might have led to the confusion. A unified focus attempt at incorporating the improvements to demonstrate the overall improved reliability and durability will be very useful. Also, developing a metric to qualify the improvements achieved in terms of life, cycleability, and/or cost would be very useful.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **2.1** for its accomplishments and progress.

- Some general results have been achieved, but there is no significant progress.
- The accomplishments are somewhat obvious and redundant relative to the other, more focused, programs.
- Progress so far is difficult to judge because each task is so isolated. It would appear that, overall, only two out of eight subtasks are on track to deliver to their stated objectives. These are 2.1 and 2.3. The others are struggling or failing to be relevant.
- An interesting example of how basic mechanisms may apply to widely disparate fuel cells for subtask 1.3 may be the result of nickel (Ni) precipitation in the MCFC electrolyte matrix, analogous to the platinum (Pt) precipitation in PEMs due to high voltage cycling.
- Out of the 10 different thrust areas, only a few have results that show any progress toward overcoming barriers or making progress. The work on MCFCs is interesting and represents an area lacking in current research and development. Most other work has a hard time competing at this scale with current state of the art. The work on fuel reforming catalysts (subtasks 2.2 and 2.4) are two of the more promising thrust areas.
- Little data was presented, accomplishments were made in generalities (promising performance was observed) and data was not presented to back up accomplishments. At this phase of the project bench-top testing should have been underway, and this final year should be focused on optimizing the several systems being evaluated. The presentation does not describe in detail the scale-up efforts (what sizes, what metrics will be evaluated, for how long, etc.). There seem to be too many unanswered questions in each of the four tasks. It is unclear if the goals of the project can be met with the proposed future work.
- While progress has been made in some areas, there is still scope for significant progress to be made. The future work listed by the PIs will be a good start. Furthermore, developing metrics for performance, stability, and durability related to the improvements made in the manufacturing/material design needs to be considered. For example, the motivation for the design of improved GDL needs to be established in terms of a durability measure. Examples include finding out what changes during degradation of the GDL/microporous layer (MPL), how the X-ray tomography addresses these, and how these are dependent on operating conditions. Similar questions must also be posed and addressed for other systems, such as mechanistic understanding of MCFC matrix stability/high-performance electrode development using soluble polymers.

Question 4: Collaboration and coordination with other institutions

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

- Having industrial partners for each task is a great way to keep the project relevant.
- A strong collaborative team has been assembled and tasks seem well coordinated among team members.
- Several groups and institutions have been involved in the project. The collaboration and coordination is very limited.
- This project seeks to use the DOE project to seed collaborations with a wide range of industrial partners. It is assumed they are successful, but perhaps a national laboratory or two could also be involved.
- Each subproject has coordination between a professor and an industrial participant, although industrial researchers or the mechanism of interaction is not clear. Additionally, there is almost no synergy among the 10 thrusts and how the co-location of these 10 projects is somehow beneficial is not evident. The project has had two publications to date.
- This is an excellent project to demonstrate the multiple collaborations between both academia and industry. There are many players and clear communication between the various institutions is visible in the progress made. The isolation between the various subtasks is due to the nature of this project and makes the degree of collaboration lower to a certain degree. This could be improved by identifying commonalities between various subtasks and working on implementing the improvements across the subtasks under a single roof.
- The wide breadth of collaborations and the way one faculty member and a post-doc are focused on a key issue for each collaboration would appear to improve significantly the odds that some good value will be created for each of the “customer–collaborators” involved. The close collaboration on a problem that the collaborator chose and is partially funding is also a benefit and increases the chances of success for each case because the “collaborator” has a bigger stake in the outcome.

Question 5: Proposed future work

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

- There were many projects funded for the amount of DOE investment. There is a good probability of at least one being meaningfully successful.
- In many cases, it is not clear how the future work will result in a clear deliverable that is needed by the development community.
- In most cases, future work may lead to small incremental improvements but is unlikely to result in meaningful advances in any given area.
- The future work plans may result in some improvements for each task. The team should prioritize its tasks to achieve more significant progress.
- Generally, there are good plans to go ahead. There appears to be no plan forward for subtasks 1.4, 3.1, and 4.1. The relevance of the plan for subtask 1.2 is not clear; it does not follow from the reported work.
- Plans are defined for the next year, but they lack metrics of what needs to be accomplished in this final year to meet objectives. It is not clear that all future plans can be realistically achieved, because most scale-up efforts have not started or just began. Too many unanswered questions still need to be addressed. An example is on slide 20; future work for subtask 1.3, material solubility, dissolution/growth mechanisms, and additives should have been evaluated at this point in the project. The same goes for slide 26, subtask 2.1, and the list of 11 future work efforts that need to be accomplished.
- In terms of task 1, while understanding the GDL characteristics using synchotron and neutron radiography adds value to the design of GDLs, it is not clear how this will impact the durability or reliability of the GDL/MPL. It is unclear how they will quantify the improvements and if they have considered the implications of the ionic liquid gel-based designs on water/gas transport properties. It was also unclear if this has been investigated, either using modeling and/or in literature. This was not clearly expressed in the motivation for this work. While suggesting that additives will be tried and tested, the PIs do not clearly say the reasoning behind or the selection criteria for these additives. Also missing is the link regarding the mechanism and stability issues in terms of Ni solubility. It was unclear if these are independent events and, if so, why the Ni solubility was focused on as part of previous work. It is an interesting approach to try and influence the microstructure of the catalyst layer using the synthesis procedure. However, the PIs do not address the issues related to the formation of films on the

catalyst particles and the potential impact of such films on resistance to both mass and electronic resistance. It seems unclear how the potential benefits achieved with soluble polymer are different from varying the loading of the binder in the catalyst layer. There is significant literature regarding such things. In terms of task 2 and 3, the future work scope for task 2 seems reasonable and follows the desired initial objectives. There still needs to be a clear definition of how each of these tasks has potential implications toward the fuel cell's operation.

Project strengths:

- This project covers a wide range of topics.
- There are many diverse projects centered around one department.
- PEM fuel cell and phosphoric acid fuel cell (PAFC) tasks are focused on critical development needs and seem well aligned with industrial partners.
- The relative co-location of these researchers along with a strong fuel cell presence in Connecticut is a project advantage.
- This project has full engagement of a wide range of industrial partners and tackles challenges from a complete system engineering perspective, as well as fundamental materials.
- A large number of PIs focusing on specific projects has shown significant accomplishments in the past year. Involving the industrial partners with every subtask ensures that the research is always related to the measurable properties qualified by the industry.
- This is a strong team with well-defined tasks, roles, and responsibilities. The program outcomes are impressive with several patents and publications/presentations to date. If successful with the tasks at scale-up stages, the team has a strong industry base to transition techniques and technologies into manufacturing.

Project weaknesses:

- This project has no focus.
- This project has too many tasks listed to be accomplished in the three-year effort. The proposed future work for the final year is not realistic.
- This very broad approach may defocus efforts. One wonders if it is possible to deliver to all 10 tasks at the end of the project. There is no coordination; each project is completely independent of each other.
- Several tasks are not well aligned with the near-term needs of the Hydrogen and Fuel Cells Program. The work is very scattered and there is no obvious plan to tie things together.
- The lack of a unifying vision or connection between the 10 project thrust areas makes this project equivalent to 10 single-investigator National Science Foundation projects. The lack of depth and disconnection of these 10 areas makes reviewing the project challenging.
- The project aims to solve problems in MCFCs, SOFCs, PEM fuel cells, fuel production, electrolyzers, and PAFC systems. While at the outset this type of variety offers significant opportunity for cross-collaboration, this type of effort is found missing. There are no clear measurables listed in terms of how the accomplishments of this project impact the fuel cell stability, reliability, and operation.

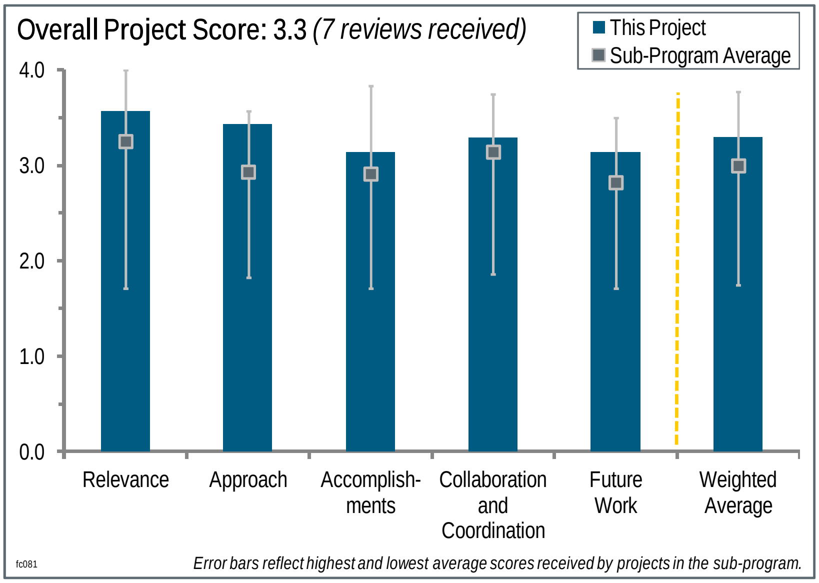
Recommendations for additions/deletions to project scope:

- This project should prioritize its tasks.
- The value of this project could be shown with a system-level demonstration that utilizes the knowledge from several tasks.
- This project needs to increase coordination between the subtasks. The work for Pt/WC focusing on HER is a low concern for electrolysis and should be redirected to OER.
- The future work for subtask 1.2 does not make sense—it does not follow from the reported work. It is assumed that activities will cease in subtasks 1.4, 3.1, and 4.1, because there is no future work planned for these subtasks.
- Tasks 1 and 2 need to be re-evaluated. The model effort should be expanded to make predictive decisions of matrix supports. Subtask 1.4 has a high impact on the industry; this task should be a priority. It is recommended that task 2 be focused on feasibility and efficient fuel processing approaches instead of desulfurization techniques.

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

Jennifer Kurtz; National Renewable Energy Laboratory

Brief Summary of Project:
This project focuses on three main ideas: (1) achieving benchmark state-of-the-art fuel cell durability by developing a snapshot of state-of-the-art fuel cell durability, uniformly applying analysis methods to data accumulated in a laboratory, and obtaining independent assessments and the status of state-of-the-art fuel cell technology; (2) leveraging analysis experience by utilizing analysis methods, experience, and data from fuel cell field demonstrations and comparing laboratory and field data; and (3) collaborating with key fuel cell developers to provide feedback, investigate factors affecting fuel cell durability, and study differences between laboratory and field durability.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.6** for its relevance to DOE objectives.

- The project is essential to benchmarking the progress of fuel cell systems over time and across industries.
- The objective is to get an independent assessment of the state of the art for durability by collecting information from system manufacturers.
- Collecting field and laboratory data on fuel cell durability is very important to assess the current status of fuel cell technology. The project will help the DOE Hydrogen and Fuel Cells Program (the Program) plan future goals.
- It is very relevant to the goals and objectives in the Fuel Cell Technologies Program Multi-Year Research, Development and Demonstration Plan. Real-world data is important from the industrial point of view, so it should be utilized well.
- This project addresses a key barrier—fuel cell durability, especially real-world durability, which is known to often yield much different results than in the laboratory.
- This is a technology validation project, in contrast with a technology development project. It gathers data from laboratory tests of fuel cells to provide situational awareness on the technology readiness of real-world stacks, and how they degrade over time. While this may be important for strategic decision making within the Program, in and of itself it will not overcome technical barriers.
- This project provides an important service to people running durability tests who do not have the tools for thorough analysis of their results. The composite results, while somewhat interesting, are not of great value because the details of the durability tests being compared (materials set, test protocol, stack design) are not available.

Question 2: Approach to performing the work

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

- The website is a good idea.

- The project features an excellent scheme and database where the all of the data are consolidated and easy to access.
- The project has a good approach and is excellent with respect to budget effectiveness—it leverages already available tools and obtains data from developers at no cost. It provides outstanding "bang for the buck."
- The approach is sound and aimed at getting progressively higher levels of information through the years and comparing field data to laboratory data. Data is aggregated to promote distribution without infringing on intellectual property (IP).
- The National Renewable Energy Laboratory's (NREL's) approach to measure and project time to 10% voltage loss and a high power point is reasonable. There are many limits to the approach, one of which is that the electric current that NREL uses to track degradation is not based on the design point of the stack/system. Thus, a current is selected that was able to run during the entire durability test even though it is quite possible that life would be much shorter at the actual designed peak power point. Also, the root cause of the degradation mechanism cannot be captured using this approach.
- Although this is beyond the control of the project, the project only collects the data that industry partners voluntarily provide. On one hand, the project data constitute an existence proof. However, this methodology creates a selection bias that may tend to overstate the readiness of fuel cells for field service. Furthermore, data from laboratory tests have a bit less relevance than would data from field tests, where, for example, environmental effects come into play.
- Life projection based on an available data scatter is tough, and this project has leveraged its previous work to make sense of seemingly random data. The electrode life is projected near the maximum power level; it is unclear if the degradation rate at lower power (25% of maximum) and the corresponding life projection is different.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- From last year, the number of data included has increased significantly. Data has been properly "sliced" to understand projected life as a function of stack type (short, full-scale, in system) and cycle type (steady, cyclic, accelerated). A significant gap between field testing and laboratory testing has been identified. The task of finding correlations between field and laboratory data will be very challenging.
- NREL did a nice job of breaking down the results from different platforms, test configurations, and test protocols. One new development in 2012 is the breakdown among tests run using steady-state, duty-cycle, and accelerated protocols. Otherwise, this project seems to continue to do similar analyses using the same methodology as it has done in years past. As NREL compiles more results over the years, the comparative analyses are becoming more informative.
- Within the limitations discussed in the previous two questions, the presentation demonstrates that the project is being executed competently. The project is accumulating a growing volume of data, the data are accessible through Internet-based tools, and the results are well integrated with other technology validation activities.
- The team has made significant progress in summarizing new data and creating histogram plots. It might be helpful if DOE could leverage other original equipment manufacturers (OEMs) to share data to increase the impact of this work.
- While the data in slides 14 and 15 are very useful for the Program, the project needs to include more data sets to be of greater value. The investigators should plan to incorporate a cost factor if possible, because durability and cost targets need to be met simultaneously. At a minimum, a catalyst loading and membrane thickness should be noted, if possible. In addition to 10% voltage degradation, membrane failures should be tracked.
- The technical progress is good. Although it is a tough challenge, the data from the laboratory have been qualitatively correlated with the real-world data gradually. Semi-quantification of their correlation is expected in the next step. The accuracy of decay prediction is to be enhanced more.
- There is a substantial amount of new data since last year. The new website is great; however, it would be outstanding if more developers and more applications (e.g., buses) can be included.

Question 4: Collaboration and coordination with other institutions

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

- The project features good outreach to developers.
- This project is inherently collaborative. It gathers performance data from fuel cell developers and publishes its results openly.
- Ten of 22 fuel cell developers provided data. The project team should strive to increase this number. The website is an effective way to collect feedback from companies abroad and in the United States, and it should be better publicized.
- NREL has been collecting data and providing analysis from 10 fuel cell developers. It would be nice to see some testimonials (even if they are anonymous) to get a feel for how much value the developers get from this type of analysis.
- It might help to reach out to foreign OEMs to gauge the level of progress in other parts of the world.
- A further increase of collaborators, especially fuel cell developers, is expected in terms of validity of the analysis.
- The fact that 50% of fuel cell developers are participating voluntarily is good, but even higher participation rates should be the goal. The project has established an excellent reputation for confidentiality, so developers that have chosen not to participate should be revisited. Also, the team should explore collaborators outside the United States.

Question 5: Proposed future work

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

- The proposed future work (slide 18) is well considered.
- There are good plans for the budget.
- Continuing the project is a good plan, but the team should be more aggressive about getting more participants for more field data.
- Cultivating existing collaborations is perceived as an important way to increase further the relevance of the project to the whole industry. The investigation of the decay modes goes beyond the project objectives, and it probably should be addressed by a larger team.
- Some of the future planned work could certainly improve the value of the project. NREL plans to specifically analyze stacks and cells run using DOE durability protocols. The most value should come from NREL's plans to investigate specific aging parameters on fuel cell durability, including start/stop and soak time. For this type of analysis to truly be of value to others besides the fuel cell developers that share their data, more details about the operating conditions must be disclosed.
- It would be useful to think about if there are any membrane-level diagnostics within the existing data set to gauge the life of the membrane in real-world applications. It will be useful to show a summary of the rated power density (W/cm^2) of the aggregated fleet and compare it to the DOE target of >1 W/cm^2.
- The future plan is basically acceptable. Voltage loss should be predicted not only for the rated current density, but also for other current densities, such as low and maximum current densities. In automotive use, low current density is average in general use, and maximum power corresponds to hill climbing. Therefore, various points are necessary for results that approximate "real world."

Project strengths:

- Strengths of the project include its sound approach, how the website allows everyone to provide information, and its focus on IP protection.
- NREL is uniquely set up to compare durability data sets from a variety of fuel cell developers for a range of applications. Without this project, such comparative analysis would not be available.
- The project provides performance benchmarks and situational awareness for the Program on technology readiness. The openness of the project and its integration with other technology validation projects are pluses.
- The project provides good value to DOE for the money.
- Strengths of the project include the theme, real-world data analysis, scheme, and network.

- The project features excellent leveraging of existing assets and obtaining data for “free.” There is good dissemination of results, especially with the implementation of the website.

Project weaknesses:

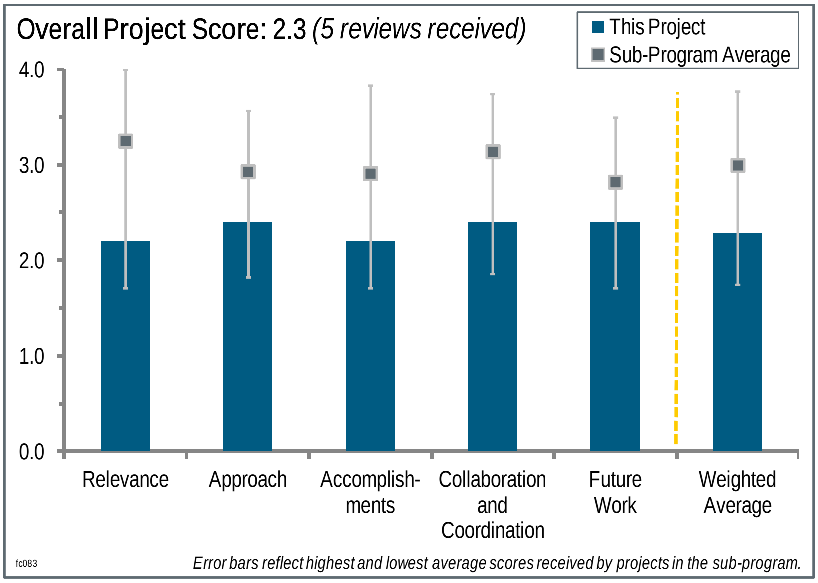
- The task of finding a correlation between field data and laboratory data is very challenging. The pool of data providers is still limited.
- The accuracy of the decay prediction is an area of weakness.
- There is not enough field data; the team needs more in order to add more applications and more results on current applications that are already being reported on.
- A lack of detailed information about the durability tests (operating conditions, load cycles, material sets, and stack design) renders the composite analysis of little value to those besides the fuel cell developers whose data is being analyzed. Of course, it is unlikely that this will change, because developers prefer not to disclose such information.
- The project is inherently limited by selection bias and by the unknown differences between the degradation of fuel cells undergoing laboratory tests and that of those in field service.
- The project is entirely dependent on voluntary contributions, and hence the data may not be from the current generation of hardware.
- Contacting developers to get data is an area of weakness. A bottleneck occurs because participation is voluntary. Maybe the DOE-funded projects can be pressured into providing the data as part of their work scope. This is outside the scope of the project’s principal investigator, but it may be accomplished with support from the Program.

Recommendations for additions/deletions to project scope:

- Feedback of better accelerated stress test protocols and other requirements based on real-world data.
- In order to estimate the difference between laboratory data and field data, it would be helpful to the Program to collect data on environmental conditions (air quality, temperature range, etc.).
- There is potential for much more information to be gleaned from analyses of the provided data sets. For example, if NREL could report trends of durability with events (e.g., voltage cycles) or operating conditions (e.g., average temperature), that could provide valuable information to the fuel cell community.
- The team should solicit data sets from outside of the United States (e.g., Japan for stationary applications). It should be noted that many of these Japanese systems include U.S. stack technologies. Foreign fuel cell developers who attend the Program’s Annual Merit Review meeting should feel “guilty” unless they participate. The team should also add transit bus applications. If there are not enough different bus fuel cell developers to provide anonymity, the team should ask fuel cell developers if they would be willing to share data without anonymity.

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

Chris Ainscough; National Renewable Energy Laboratory



Brief Summary of Project:
The main objective of this project is to build a modeling tool to optimize fuel cell attributes, including control parameters, and system and component sizes for unique individual building characteristics. The tool will add user flexibility for different building, fuel cell, financial, and control characteristics. The tool can be used to minimize life-cycle cost, lifetime greenhouse gas (GHG) emissions, and installed capital costs of fuel cell installations. The project will characterize the largest segments of the U.S. building inventory for use in the tool, leveraging the Commercial Building Energy Consumption Survey (CBECS); characterize building control systems and advanced control strategies for integrating fuel cell systems and building control systems; validate the model outputs against real-world data from stationary fuel cell installations; and determine the set of most-favorable system sizes and types to achieve national GHG emissions and energy demand reductions.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **2.2** for its relevance to DOE objectives.

- This project is relevant to DOE's goal of developing fuel cells for combined heat and power (CHP) applications. The stated objective is to build a tool for optimizing fuel cell attributes, including control parameters and system and component sizes for buildings. The scope of the tool extends to minimizing the life-cycle costs, lifetime GHG emissions, or capital costs of fuel cell installations.
- This is a very useful tool for market studies, as well as for fuel cell original equipment manufacturers (OEMs) that are evaluating potential customers.
- The relevance and how the project will support the deployment of fuel cell technology for CHP are not clear. There are no industrial CHP partners.
- There is always a great need for tools of this type to help end users and developers understand how fuel cells can aid them. For this work to be useful, it must be validated against real-world data, the assumptions used in the models must be clearly specified, and the model's limitations must be clearly explained.
- The project has some relevance to the DOE Hydrogen and Fuel Cell Technologies Program goal of fostering the commercialization of fuel cells in stationary/CHP applications. This project appears to fit better with the Market Transformation sub-program or even the Systems Analysis sub-program. It is not clear how completion of the model will aid in system optimization unless specifics on various design approaches are included in the model. It appears that the model is at a higher level and not specific to any one design that can be optimized.

Question 2: Approach to performing the work

This project was rated **2.4** for its approach.

- Working closely with buildings experts and leveraging other work by the National Renewable Energy Laboratory (NREL) for financial calculations is a good approach.

- One "grand unifying model" for all community buildings is appealing. However, the validity of the model and approach was unclear, given the high level of uncertainty on estimates and the number of unknown variables.
- The project has taken the approach to develop a high-level model that captures the interactions between the building loads and the fuel cells. The approach is clearly spelled out and benefits from work being carried out in other activities. In developing the Buildings Module of the model, the project team is integrating the building-related data (building types, locations, vintages) that are available from the NREL Electricity, Resources, and Building Systems Integration Center (ERBIC) and the 2003 CBECS. The project has University of California at Irvine (UC-Irvine) as a partner to develop the Control Strategies Module for refining and validating dispatch strategies. The Manufacturing Costs Module will be developed using the data developed by Lawrence Berkeley National Laboratory (LBNL) and Strategic Analysis, Inc. (SA) in related projects. The Economic Module is based on the financial calculations method implemented in the Hydrogen Analysis (H2A) model. The model has a Grid Pricing Module to reflect the differential prices in winter and summer as well as peak and off-peak times. The Natural Gas Pricing Module uses data from the U.S. Energy Information Administration forecasts.
- The approach of using NREL's buildings program is extremely important for this work to be successful. It is not clear how the heat is being utilized. The electricity rates are clearly set up. The heat value and demand were not described. Ramping the reformers for load following and the solid oxide fuel cell itself will dramatically impact the lifetime of the reformers and stacks. This needs to be considered. It is not clear how operations and maintenance costs are considered. This will be a large cost for the system, particularly for a system that is constantly ramping.
- The model will include fuel cell characteristics such as minimum and maximum power, temperature of available waste heat, and a ramp rate that determine the power and heat available in the next time increment. The partners will partner with UC-Irvine to develop and validate dispatch strategies for various classes of buildings. It is important that the model does not demand that the fuel cell be cycled on and off over short time intervals. Without significant tailoring of the submodels by each developer, the utility of the model may be limited to a high-level view of where a specific fuel cell system may be a good fit, and not necessarily a design optimization tool. It is not clear if cooling loads are being considered, which may be a better fit for a significant portion of the building stock in the United States. The approach emphasizes relatively small CHP systems for which there are not a lot of existing systems with which to validate the model. A few larger-scale systems exist, but their relevance to smaller systems is unknown.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- The project is following the project description; however, the project fails to communicate and demonstrate how this supports DOE goals.
- It seems like there has been significant progress in the development of the tool, but the most important step is validation against real-world data.
- The investigators have analyzed many different building types and processes. The model appears to have a great deal of flexibility, which is necessary. Clear documentation on how to use this flexibility will be needed.
- In fiscal year (FY) 2012, NREL has developed a graphical user interface; incorporated models for phosphoric acid, molten carbonate, polymer electrolyte membrane, and solid oxide fuel cells of different power ratings; and laid out a framework for accepting manufacturing cost data. The project is still in the process of developing the modules and did not discuss specific results.
- The actual start date of the project is not clear, even though it is listed as October 2011. If this is so, progress appears reasonable in the relatively short period since the project's beginning. However, the same comment appeared in the reviewer comments from the 2010 DOE Hydrogen and Fuel Cells Program Annual Merit Review. So neither the start date nor the end date is clear. It is unclear when the model will be available to fuel cell system developers. Some progress is evident, primarily arising from input from other existing projects.

Question 4: Collaboration and coordination with other institutions

This project was rated **2.4** for its collaboration and coordination.

- The project features good collaboration with other offices at NREL, UC-Irvine, and LBNL.
- There is a mix of industry and national laboratories that would be able to provide validation of the model output.
- It is unclear whether the approach has been accepted by fuel cell CHP companies.
- The principal investigator (PI) listed NREL ERBIC, LBNL, SA, and Battelle as collaborators, and UC-Irvine as a partner. The LBNL (FC-098), SA, and Battelle efforts are new initiatives. The LBNL and Battelle projects are still formulating models and have not yet contributed data to the NREL project.
- The collaboration appears to be good. The project team is working with NREL ERBIC, UC-Irvine, LBNL, SA, and Battelle, as well as fuel cell system OEMs. These interactions should enable the project to take advantage of existing work and avoid duplication of effort.

Question 5: Proposed future work

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

- It will be exciting to see the results of model validation against real-world data, and model publication.
- The project seems just to continue the ongoing model work without addressing or improving the relevance, approach, or collaboration.
- The remaining proposed future work for FY 2012 calls for expanding the control strategies and building types, providing input to the 2012 CBECS, and generating initial estimates of fuel cell sizes and building inventory for use by the new LBNL and Battelle projects. The proposed work for FY 2013 includes expanding fuel cell types, optionally continuing UC-Irvine work, implementing Design of Experiments capability and speed improvements to dispatcher code, validating models, and performing detailed optimizations.
- The future work plans are clear and cover the areas needed for a successful project. It is not clear how much additional effort and time are required to produce a product that can be used by developers. It would be good if a fuel cell system developer could exercise the parts of the model that exist and provide feedback on the implementation of the submodels into the overall model.

Project strengths:

- The approach and plans are well laid out.
- The approach and plans are generally well laid out.
- Working closely with buildings experts and leveraging other work by NREL for financial calculations is a good approach.
- This is showing good collaboration between DOE offices. The investigators are leveraging a lot of work being done in the DOE Building Technologies Program with the fuel cell work.

Project weaknesses:

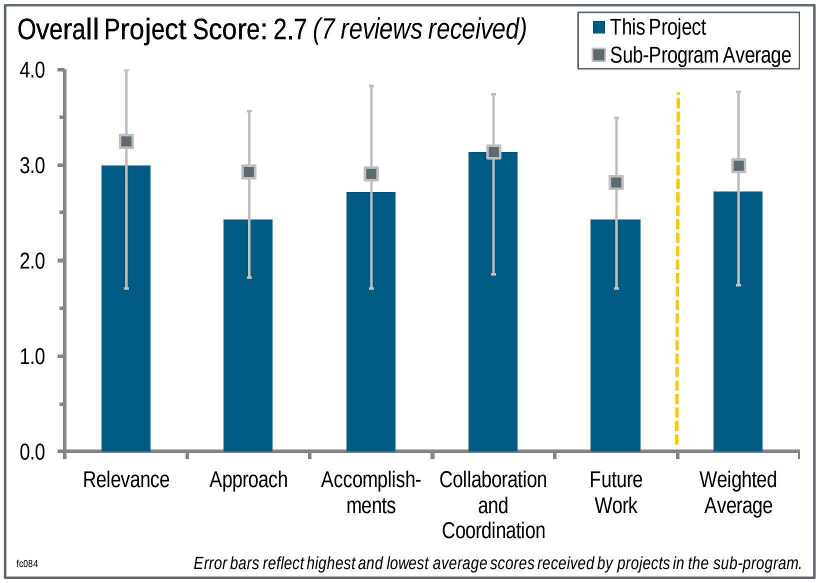
- At best, the project will produce a high-level model that is likely to be more useful for the system analysis team than the fuel cell team. To date, the project has not presented many useful results. The PI plans to generate some rough estimates later this year to focus the LBNL cost efforts. The first useful results are likely to come in FY 2013.
- It would be nice to clearly show the heat requirements and, perhaps, to allow the user to set the heat duty similar to how the user can define the electrical needs.
- The model appears to be suited more for a high-level or policy analysis rather than a design optimization tool. Its utility to fuel cell system developers is unclear. A collection of building energy needs and patterns may be more helpful to developers; they can put in their own system designs and characteristics.

Recommendations for additions/deletions to project scope:

- The investigators should add fuel cell system developers to the team. Maybe they should just concentrate on compiling building characteristics for use by system developers.
- The project should be terminated if the approach is not acknowledged and supported by the companies making CHP systems based on fuel cells (e.g., UTC Power, Ballard, and ClearEdge Power). These companies should also be the ones that define the major challenges for the deployment of CHP in commercial buildings, and the project should address and analyze these challenges.

Project # FC-084: WO3 and HPA Based Systems for Durable Platinum Catalysts in PEMFC Cathodes

John Turner; National Renewable Energy Laboratory



Brief Summary of Project:
The objective of this project is to improve electrocatalyst, membrane electrode assembly (MEA) durability, and activity through the use of platinum/tungsten oxide (Pt/WO_3) and heteropoly acid (HPA) modification to approach automotive polymer electrolyte membrane (PEM) fuel cell activity (0.44 mA/mg of Pt) and durability targets (5,000 hours/10 years). The project seeks to: (1) enhance Pt anchoring to the support, thereby suppressing Pt electrocatalytic activity loss under load cycling operations and enhancing electrocatalytic activity; and (2) lower support corrosion for increased durability under automotive start-up/shutdown operation and reduced Pt agglomeration and electrode degradation.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.0** for its relevance to DOE objectives.

- The project is focusing on addressing important barriers, using its proposed catalyst supports.
- The goal of catalyst durability and performance is addressed in this research.
- These objectives do address the stated DOE objectives. It is not clear, however, how the performance is to be improved by these two approaches.
- The durability of cathode catalyst support materials is one key in the quest to fully implement PEM fuel cell technology. As the principal investigators (PIs) proposed, one way to overcome carbon limitations would be to use conductive metal oxide supports such as WO_3 and SnO_2. Although metal oxide supports have potential, the PIs provided little evidence that the type of oxide they are developing is the right way to go.
- This project addresses DOE targets and goals and is especially focused on durability enhancement. The project evaluates a candidate for robust catalyst supports as a way to replace the widely used carbon supports, which are known to corrode. This could turn out to be an important contribution to achieving durability and performance targets.
- Goals for the project are well aligned, but the approaches used may not be. For example, it is unclear if atomic layer deposition (ALD) will ever be scalable to mass production at a reasonable cost. The presentation discussed a "large-scale" ALD process, but the process still needs improvement and a cost projection. Both WO_3 and HPA systems appear to add several steps to the manufacturing process.
- Because the project is intended to enhance catalyst activity and durability, the relevance of the project is high. The greatest strides that can be made toward commercializing fuel cell vehicles are those that can be made by lowering the loading of precious metal electrocatalysts and improving durability.

Question 2: Approach to performing the work

This project was rated **2.4** for its approach.

- Anchoring the Pt onto the support may impact durability, but it is unclear how this will improve performance. The poor conductivity of WO_3 is known, but it does not seem to have been considered by the PI. This will make it difficult to reach baseline performance, as indeed shown in this review.
- This research has the potential to increase catalyst durability. There does not seem to be a way in this research to address the barriers of cost and performance. If anything, these materials so far seem to show decreased performance and increased cost to state-of-the-art materials. Because the support is not conductive enough, carbon is needed in the ink. This adds further levels of complication to creating proper Pt/carbon interaction above that of traditional inks.
- The project is well planned and each participant represents an important contributor to the overall success of the project. However, it is not clear if the PIs developed the best method for establishing the active surface area. Due to a problem with CO oxidation at low potentials, the CO-stripping approach should be avoided. Although utilization of copper (Cu) UPD may overcome some of the limitations; the assessment of charge from Cu UPD stripping peaks is usually not easy and the method should be revisited.
- There are considerable synthesis and measurement tasks involved. The laboratory tasks seemed a little “casual.” There was no apparent attempt to replicate work and to demonstrate the repeatability of the results. It is true that substoichiometric oxides of heavy metals (such as tungsten) are hard to characterize. The chemistry is complex. But that complexity requires a well-defined set of replicated tasks with thoughtful control of contaminants and process conditions.
- It is clear that some progress has been made, but the overall approach is still questionable. Given that carbon corrosion is a significant barrier to lifetimes, it is unclear if it makes more sense to completely eliminate the carbon in favor of an oxide support (as is being pursued by other teams that presented in the meeting). The half-way approach used here of deposited Pt on an oxide, but then mixing the oxide/Pt catalyst with carbon, may eliminate or reduce some pathways to failure; however, the carbon will still see very high potentials during transients/start-up/shutdown, and it will still corrode, leading to failure. The Pt is intact on the WO_3, but it is electrically isolated because the carbon-conductive path is gone, and it is not going to work. The team has looked into using more corrosion-resistant carbon nanofibers. It is hard to determine the benefit of adding the oxide support if a more corrosion-resistant carbon is also required.
- ALD seems to be an expensive technique for producing nanoparticles. Usually the cost of ALD seems most worthwhile when coupled with the anticipation of producing higher specific activity thin films. Pt/SnO_2 has been tried before, and it is noted in the literature. The approach to this project should clearly state how the Pt/SnO_2 systems studied here are differentiated from what has already been explored. The usage of HPA is potentially risky due to the possibility of leaching; however, the reward could be high if HPAs help to stabilize other components. Pt nanoparticles on various forms of WO_3 have been tried in the literature. It would be more interesting if something could be done to put Pt conformally on the WO_3.
- The approach may have an opportunity to improve the durability, but the support conductivity issue may limit its activity. If the conductivity could be improved by the proposed approach, then the durability may become an issue again. This is a general problem, though. However, the project has not come up with a unique solution so far.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- Synthesis has been accomplished and performance and durability have been assessed.
- Progress has been shown in the synthesis and feasibility of new supports. Some progress has been made on showing increased durability using new supports. No data was presented to reduce cost or increase performance.
- These electrodes could be very interesting. The chemistry has proven complicated and somewhat difficult to understand fully. However, progress is being made as the synthesized materials are characterized.
- There still seems to be too many approaches and not enough capability to look at each one successfully. It was very surprising to see the SnO_2 work; this was a distraction.

- The progress in this project is successfully presented and participants have clearly demonstrated a strong team effort that has been capable of efficiently addressing DOE Hydrogen and Fuel Cells Program objectives. However, there is neither comprehensive discussion about the conductivity of oxides nor a critical charge transfer from oxide to Pt nanoparticles. These results would allow better understanding of key parameters that are responsible for the improved stability of cathode materials.
- The graphitized carbon nanofibers were used to increase the conductivity of SnO_2 and WO_3. They also assisted with improving SnO_2 activity. The major issue with HPAs is usually with respect to avoiding washout in the presence of water. The investigators managed to immobilize HPAs on carbon and demonstrated that the HPAs did not boil off the carbon. The activity for most materials did not meet the targets. The activity reported for materials with HPA was within noise of the baseline. Poor activity was shown for WO_3. Some stability was shown for HPA- and SnO_2-containing materials. Other studies (Masao et al., 2009) have noted Pt/SnO_2 stability at higher potentials. Just like with alloy catalysts that contain base metals, it would be good to see whether the stability is maintained at lower potentials.
- The ALD approach provides good Pt precursor deposition on the support; however, the final Pt particle size and its distribution do not show significant advantages over the colloid approach. Meanwhile, the colloid approach is very practical and scalable.

Question 4: Collaboration and coordination with other institutions

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

- The comprehensive collaboration in this project may result in promising progress.
- A good mix of key industry partners and academia has been established.
- Little collaboration has been shown with industrial partners.
- The team involving the National Renewable Energy Laboratory (NREL) and university researchers seems fully functional. Tasks are assigned.
- The future role of TKK looks to be very important regarding finding answers to questions of scalability and cost. The university interactions look well coordinated.
- The achieved accomplishments from this project indicate a well-balanced, synergistic effort among participants. Indeed, the success of this project relies on the highly diverse and organized team, which is well coordinated by NREL as a lead institution and John Turner as a lead PI. The role of the participants can be recognized by their contributions in their own expertise that add to the overall success of the project.
- There appears to be clear evidence of how most collaborators are contributing to the project. Nissan has contributed relevant accelerated stress test (AST) protocols. Fuel cell testing is still to come. The Colorado School of Mines has contributed perhaps the most interesting aspect of the project: the immobilization of HPAs on carbon. 3M's contributions are not very clear, although this may be a consequence of the fact that fuel cell testing has not begun in earnest. The University of Colorado-Boulder has contributed the ALD work for Pt/WO_3.

Question 5: Proposed future work

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

- There seems to be too great an emphasis on WO_3 rather than HPA, despite significant progress in this area. It is unclear if there is a potential to continue the Pt/SnO_2 work with TKK. It looks promising.
- The future work is focused on the durability barrier. Cost and performance are not addressed beyond using alloy catalysts.
- In this phase of the project, the PIs should have a more aggressive approach in designing a new generation of conductive and durable metal oxide supports. The PIs may want to go beyond WO_3.
- The "future" was not clearly defined. There was an indication that one task was to "scale-up" ALD, but the details of the proposed route to do that were not described.
- The ALD approach cannot be a viable way to make a catalyst that meets DOE goals.
- Regarding item 2, both the ALD and wet chemistry approaches still need significant work. A choice is to be made in December, which is only six months away. It is unclear whether the project team will actually make a decision in December, or if it will be argued that both approaches are still too immature for a decision to be

made. The team should stick to its plan and down-select. Yes, it is possible that down-selecting could be a mistake, but failure to do so will prevent real progress. The team should drop item 3. Until there is significantly more evidence in both rotating disk electrode (RDE) and MEA testing of the benefits of this approach using Pt, the team should not waste time looking at Pt alloys. The reviewer agrees with items 1 and 4.

- While some conductivity gains have been shown with the addition of the graphitized carbon nanofibers, mass activity is still not at the Pt/carbon baseline. Given this, it would be interesting to understand what remains in the systematic conductivity study. Because other work has produced higher activity for Pt/WO_3, it might make sense to reproduce some of the other work, and be able to draw from it metrics associated with conductivity. Decisions should certainly be made soon about whether to continue with Pt/WO_3. HPA-containing materials should enter into cells, just as described in the future work. Other scale-up efforts may not be as worthwhile due to low activity. It may be worthwhile to put 12% Pt/SnO_2+C into a cell for durability testing.

Project strengths:

- The project has put together a good team that has various strengths.
- Key industrial partners suggest that materials that show promise can be commercialized rapidly. There is a good balance between national laboratory, university, and industry efforts.
- New supports may lead to more-durable catalysts. The project team is taking time to make sure it has adequate electrochemical tests to measure progress toward the barriers.
- The development of durable (but not optimized) metal oxide supports has been demonstrated. Improved wet chemistry Pt deposition and ALD deposition seems to be applicable on WO_3 systems. The supports have shown modest improvements in durability over Pt/C catalysts, which clearly justifies the approach of this project. Highly collaborative team efforts have produced a valuable outcome from this project. The participants are well-known experts in the field of fuel cells, and they bring highly diverse expertise.
- It is important to keep looking for tougher catalyst supports, and “non carbon supports” may be necessary. This project is seeking such materials. It makes sense to keep that search active. The PI is excellent and has a good comprehension of the tools needed to make progress.
- The use of HPA-immobilized materials may be interesting to study from a durability perspective. The presence of Nissan provides for automotive-relevant ASTs. RDE methods appear to be well established. Some measure of durability has been shown for materials containing HPA and SnO_2.

Project weaknesses:

- The project does not directly address cost and performance barriers.
- Some clearer metrics and milestones might help.
- The team needs to improve the conductivity without leading to poor durability.
- It is unclear why so much time was wasted developing NREL’s own test protocol. This just makes it difficult for others to compare these results with their own. That is why DOE has its own protocols. The lack of consideration of the commercial feasibility of the synthesis processes makes scale-up difficult.
- The PIs need to explain how, specifically, they plan to improve metal oxide performance, and what methods they will apply to deposit Pt-alloys. It is unclear which type of alloys would be used, as well as the atomic ratio between Pt and alloying components. The reviewer hopes this would not be PtCo or PtNi, because other groups are also focusing on the very same alloys. Furthermore, the SnO_2 system should not be taken into consideration. The method for assessing the active surface area should be improved.
- So much time was spent on synthesis development and technique development, when the fundamental benefit of the approach is still not clear. It is hard to see this route resulting in a real game-changing advance.
- The project is essentially using a conformal method (ALD) for applying nanoparticles to a support. The project is looking at a number of catalyst systems—Pt nanoparticles on both WO_3 and SnO_2—that have already been explored in the literature. High oxygen reduction activity has not yet been realized for any of the materials.

Recommendations for additions/deletions to project scope:

- The ALD approach should be carefully evaluated. It should not be encouraged.
- The project should quickly move to either develop Pt/WO_3 that resembles literature activity or eliminate this aspect of the project. Fuel cell durability should be immediately studied for catalyzed HPA and SnO_2 supports.

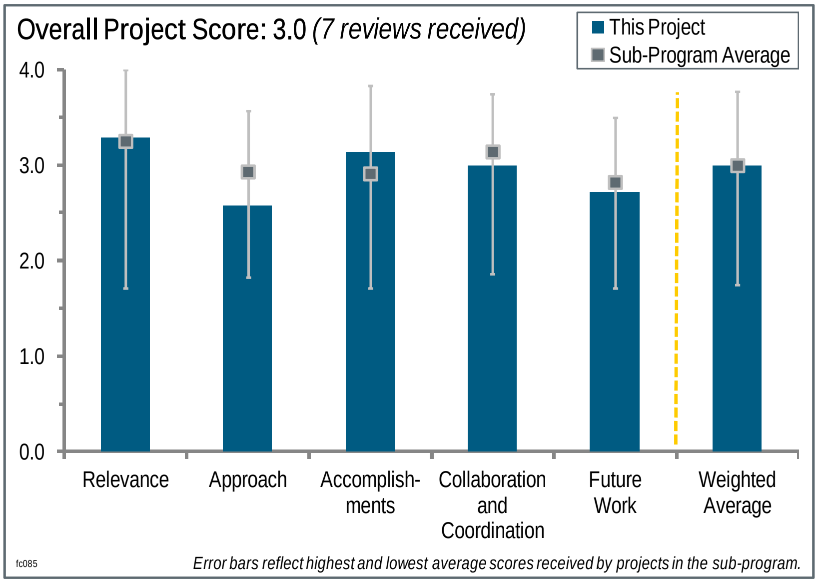
- If there is little hope to increase activity over current materials, the future work for 2013 should be pushed forward to use higher mass activity catalysts and see if they can be incorporated onto support structures.
- The team should continue with the Pt/SnO_2 work. Perhaps the ALD scale-up should not be done, because it is not really going to scale-up to industrial volumes. The team should broaden the HPA work in order to understand the protonic conduction of this material and seek to see if ionomer can be reduced within the catalyst layer or if Pt utilization can be increased.
- The team should take one of the WO_3 synthesis routes, focus on finding a way to deposit Pt on it with comparable mass activity to baseline catalyst, and demonstrate enhanced stability with that system. The HPA approach is still questionable; it is unclear whether Pt is on HPA or on carbon. Combining HPA with WO_3, when neither is well understood, is not likely to be successful.

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

Vijay Ramani; Illinois Institute of Technology

Brief Summary of Project:

Research objectives of this project are: (1) to develop and optimize non-carbon mixed conducting materials with high corrosion resistance, high surface area, and high proton and electron conductivity; and (2) concomitantly facilitate the lowering of ionomer loading in the electrode through enhanced performance and durability and by virtue of the surface proton conductivity of the electrocatalyst support. Addressing the issue of electrocatalyst and non-carbon support stability will help meet operational longevity, electrochemical area loss, and electrocatalyst support loss.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.3** for its relevance to DOE objectives.

- The project goal of developing a corrosion-resistant catalyst support is important to reaching the overall DOE objectives of implementing automotive fuel cell power systems.
- Improving support corrosion will have meaningful impacts on fuel cell durability and issues involved with start/stop cycling.
- This project addresses fuel cell durability, which is a major barrier to the adoption of fuel cell technology. The focus on catalyst support durability is relevant to automotive systems, which are subject to multiple starts/stops that corrode current catalyst supports. This support corrosion has been identified as a major source of performance degradation.
- The stability of catalysts/supports under polymer electrolyte membrane (PEM) fuel cell operating conditions involves complex processes that need to be resolved in order to fully implement PEM fuel cell technology. The work related to establishing the stability of catalysts on metal oxide supports will definitively be one important step in finding a new class of more durable cathode materials.
- Carbon corrosion is a real problem, and replacing the carbon is one of the few solutions. However, modifications of start/stop procedures and the use of catalysts such as nanostructured thin film are making it less relevant.
- This project started in September 2010 and is now technically in its second year. The principal objective of this project was to develop a stable support material for PEM fuel cell operation. The benchmark for this was set as durability measurements, which would reflect lower than 40% ECA loss or less than 30 mV per 100 hours tested at 1.2 V, both in accordance to General Motors' protocol. The project aims to do this using model mixed metal oxide (MMO) supports principally comprising RuO_2-SiO_2 (with and without sulfonic acid functionalization), RuO_2-TiO_2, and SO_4^{2-}/SnO_2 materials. Its relevance to the DOE Hydrogen and Fuel Cells Program is good, considering the fact that currently very few carbon supports are able to meet the DOE targets for corrosion resistance.
- The most pressing need toward meeting cost and durability targets for the commercialization of hydrogen (H_2) fuel cell electric vehicles is the development of high-activity, durable electrocatalysts for oxygen reduction. The project seeks to provide durable electrocatalysts through the use of metal oxide supports. In terms of cost, the

project seeks to lower the use of ionomer by adding proton-conducting species to the catalyst support. This is not likely to be as effective as developing higher activity.

Question 2: Approach to performing the work

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

- Work is directly aimed at addressing start/stop durability, which is a critical barrier. Testing methods directly address stop/start issues. The approach of using nonconducting, inexpensive SiO_2 and decorating it with conducting oxide particles made from more expensive materials will decrease costs compared to using the conducting oxides alone, and it may allow costs to be competitive with carbon if durability is improved. Utilizing a proof-of-concept with the RuO_2-containing system and the use of alternative cheaper conducting oxides (indium tin oxide [ITO]) is a good strategy that mitigates risk if high amounts of RuO_2 are needed. Durability testing using separate start/stop cycling protocols (1.0–1.6 V) and operational cycling protocols (0.6–1.0 V) do not take into account that the system operates between start/stops and that the catalyst surface oxidation state is changed, which may impact start/stop durability. In the start/stop protocol, the platinum (Pt) surface is always oxidized. Others have seen an effect from including a cycle going to lower voltage during stop/start cycling, and it may be important to include such a cycle in catalyst support testing. When testing alternative catalyst supports, catalyst-to-support mass ratios generally have been kept the same as for Pt/C. Due to the higher density of metal oxide supports, this will result in thinner catalyst layers and is likely to result in poorer mass transport and flooding under some conditions. It would be better to keep the volumetric ratio of support to catalyst constant to provide a better comparison to Pt/C, because the catalyst thickness would be more comparable and mass transport issues should then be more similar. The Pt-support mass ratio does not relate to a physical parameter that affects performance, so there is no reason to use this as a parameter for standardization.
- The principal investigators (PIs) proposed the synthesis of "model" MMO supports that would be used as a model support to investigate the feasibility of preparing non-carbon mixed/conducting supports. The approach, involving transformations of knowledge from model oxides to real oxide-based supports is, in principle, desirable. However, RuO_2- and SnO_2-type oxides have already been studied in electrochemistry. In order to learn more, the PIs must develop and use new characterization methods.
- The investigators are taking the logical path by proving the stability of the base material first; trying different base materials; and then trying to improve the catalyst material by controlling catalyst particle size, etc. Catalyst stability is the next biggest question, and the PIs are addressing that with the proper testing. At the end of last year there were a lot of comments on the use of ruthenium (Ru) as a support, which the investigators vigorously defended; however, in a completely qualitative way. They should have better prepared for this question; there is great skepticism about whether RuOx is a suitable support, so they need to first convince people of this. The authors also mentioned that they are using RuOx as a template to explore other metals, titanium (Ti) for example, but those do not seem to be working very well.
- The approach taken involves using MMO supports, which engender mixed proton and electron conductivity while maintaining high resistance to corrosion. In order to achieve this slated objective, the project is using RuO_2/SiO_2 composites with and without sulfonic acid functionalization, as well as RuO_2/TiO_2 and SO_4^{2-}/SnO_2 composites. The principal issue in the approach is that a comparison is being made between an MMO support and a typical carbon black with similar BET surface areas, but different particle size as evidenced by XRD Scherrer calculations. Because most of the Pt deposited on the MMO was larger, its ability to withstand catalyst dissolution tests better is not surprising. Careful comparison between Pt of similar particle size would be necessary to remove this artifact. This is evident from the similar loss of ECA between Pt/RuO_2-SiO_2 (1:0.5), which has particle size in the same range as 46% Pt/C (TKK). What was most interesting was the start/stop cycling results, wherein despite the potential excursion to 1.5 V (compared to 1.0 V in the catalyst dissolution test), the loss of ECA was lower. This clearly indicates that wetting of the larger Pt particles on the MMO was significantly better as compared to smaller-particle-size conventional 46% Pt/C (TKK). As a whole, in terms of approach, it is risky to use a transition metal that has the potential of electrodepositing onto Pt within the operating window of fuel cell operation. All of the data presented in this work does not simulate conditions in a fuel cell, especially during uncontrolled shutdown conditions where variations of reactant gas fronts create potential fluctuations not simulated in this study.
- The approach is to use stable but not conductive components, and functionalize/coat them with conductive components. The concept is good, but unfortunately it has only been demonstrated with expensive materials

(RuO_2) that also can corrode at the relevant high potentials. While the PI states in his rebuttal to last year's reviewer comments that RuO_2 is only a model system, the effort being put into this model system should be directed toward systems with the potential to meet the cost targets. It is promising that an automotive original equipment manufacturer (OEM) is performing cost analyses, but it would be nice if these cost analyses would be shared with the reviewers.

- The use of RuO_2 as a support material has cost and supply concerns, as noted by the presenter. While the presenter stated that RuO_2 is a model support material, it would seem to be a better approach to focus on the SnO_2 or ITO also proposed in the approach of the project. The importance of including ionic conductivity as an inherent property of the catalyst support is not clearly defined in terms of potential benefit; the target proton conductivity seems higher than it needs to be.
- The project has five different families of oxide supports. Of these five, three contain a precious metal (Ru), which may be a problem for cost. The project needs to incorporate a task for cost analysis. The project intends to thrift Ru by depositing RuO_2 onto a secondary support (TiO_2, SiO_2, or sulfonated SiO_2). There is some possibility that RuO_2 will not be stable for all of the potentials that a fuel cell cathode experiences. Sulfonating the catalyst support is an interesting development worth investigating. It may be beneficial for the rest of the program to understand if the 50 m^2/g BET surface area requirement was cascaded from the program targets.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- This work has made significant headway in a year in partnership with Nissan. However, actual fuel cell durability data is expected to determine the feasibility of this approach.
- The project has made good progress in the last year, both in Nissan's materials characterization and the Illinois Institute of Technology's (IIT's) materials synthesis. However, the focus should be on the development of cost-effective, stable materials rather than materials that have been shown to corrode in the potential range of the fuel cell cathode.
- Initial testing at Nissan North America showed much improved stability in stop/start cycling compared to carbon supports, showing only a 14% loss in mass activity (versus 55% for Pt/high surface area carbon)—approaching the DOE target of 10% mass activity loss in a test more severe than the DOE-suggested test. Initial results suggest that the sulfonated SiO_2-RuO_2 system has good protonic and electronic conductivity. A measurement technique is needed to demonstrate protonic conductivity in the presence of a good electronic conductor (i.e., after the RuO_2 is deposited). An H_2 pump experiment might be one option. The baseline is a high-surface-area carbon support, rather than a more stable carbon (graphitized). Better characterization of the morphology of the SiO_2-RuO_2 support would help direct optimization of the support. It was not clear how the size of the RuO_2 particles compared to the SiO_2 particles; that comparison will determine how much RuO_2 is needed to form an interpenetrating network among the non-conducting SiO_2 particles.
- While progress has been made in some areas, the proposed and accomplished work is still way out of balance. For example, methods have to be established for monitoring desolution of cationic species from supported oxides. It is well established that Ru is not stable in RuO_2 form; the same is most likely true for tin (Sn) in SnO_2. Furthermore, a big concern is the low conductivity of these oxides, especially in combination with SiO_2.
- Progress is in line with the investigators' plans; they have done a good job showing the stability of the supports, now they need to show that these can actually act as decent catalyst support materials. There is not much improvement in ECSA loss under cycling data. Not enough justification is given for the surface modifying the supports; perhaps proton conductivity to the supports has been shown to be a problem. In the researchers' own testing, it appears not to be. No work has been done on costing, and they should have seen this as a question.
- The materials synthesized to date have shown advantages in terms of durability to potential cycling in the carbon corrosion region compared to traditional Pt/C catalysts. The performance of these materials and their durability in the Pt dissolution region offer no improvements compared to baseline materials, which currently fall short of performance and durability targets and will be required for commercially competitive materials.
- There has been impressive stability for RuO_2-SiO_2- and RuO_2-TiO_2-based catalysts over cycling, especially with higher SiO_2 content for RuO_2-SiO_2 and higher-temperature heat treatment for RuO_2-TiO_2. It cannot necessarily be said that this stability was expected. (Although some loss of stability with catalyzation could be expected, as well as the lower stability with higher Ru content.) Investigators need to do cost analysis to confirm that Ru would not drive up costs. Investigators need to examine Ru on the anode post-mortem. Nearly

all activities need to be revisited following a reduction in Pt particle size. However, it is recognized that the true focus of this project is activity, not durability. That said, durability improvements cannot preclude needed activity.

Question 4: Collaboration and coordination with other institutions

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

- Good collaboration is shown between Nissan and IIT.
- The collaboration between IIT and Nissan is clearly outstanding. Nissan is fully engaged in this project.
- The collaboration between Nissan and IIT appears to be working well. Nissan's testing should ensure that auto-relevant testing conditions are examined.
- Although good collaboration exists between the PIs, this proposal will benefit if a tried partner with appropriate characterization skills is involved in the realization of this project.
- The addition of Nissan should help, and it appears that Nissan will do more of the heavy lifting in the future.
- For a project of its size, the two key participants are adequate for the work scope. However, adding additional collaborators, particularly a catalyst supplier, would be beneficial.
- There appears to be just one collaborator: Nissan. Nissan is a good collaborator because it brings the automotive perspective to the work. Nissan helps with implementing accelerated stress tests and fuel cell testing. A few micrographs were shown for the RuO_2-TiO_2 catalysts—enough to show that TiO_2 was not completely covered by RuO_2. However, there is more that could be done to reveal aspects of structure, oxidation states, adsorbent species, coordination, etc. Past reviewer comments have questioned the project's access to materials characterization. While it appears the project has arranged for this at University of Illinois, Michigan State University, and University of Michigan, the results shown in this year's presentation reveal that more could still be done. Hopefully, this work is ongoing.

Question 5: Proposed future work

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

- There are far too many unsubstantiated, declarative statements of *what* will be done, but no explanations of how and why.
- The researchers' plans are addressing the next concerns, and testing at Nissan should help. They will have to show that these supports can support smaller particle sizes to increase mass activity.
- The future plans appear to be very workmanlike. No serious approach is suggested to understand the observed results.
- The near-term focus should be on extending the concept to less expensive materials sets, with a secondary focus on catalyzation and functionalization with proton-conducting groups.
- Work optimizing Pt nanoparticle deposition is critical to obtain higher performance and to get an apples-to-apples comparison of durability, because durability is related to Pt particle size. Work optimizing the catalyst layer is important. The higher density of these supports suggests that a much lower Pt/support mass ratio should be optimum, and likewise that a much lower ionomer ratio should be optimum. A more detailed cost study may be beneficial.
- Further optimization of the Ru-containing supports with SiO_2 and TiO_2 may lead to incremental improvements in observed properties; however, the gains expected based on the data to date give little hope that materials that can reach DOE performance targets will be made. The focus on the optimization of Pt nanoparticles is unfortunate within the project, because this effort is better spent studying additional supports. A catalyst supplier would have been beneficial in this regard.
- The most important thing that the project has to do at this stage is to get the Pt particle size down, so as to make the activity acceptable. This appears to be covered under the bullet about optimizing the Pt introduction. At the moment, durability appears to be on a rotating disk electrode (RDE) basis, but this needs to be repeated in a cell for RuO_2-SiO_2 and RuO_2-TiO_2 catalysts. This work also appears to be going forward.

Project strengths:

- Having an OEM as a partner is a strength of this project.
- The focus on the metal-oxide supports chosen is bold and compelling. There are some good examples of lessons learned from RuO_2.
- The project has shown the potential of using mixed proton-electron conducting element for improving support corrosion resistance. This is done using Ru-, Ti-, and Sn-based metals and silicon-based proton-conducting supports. Good corrosion resistance is shown for some of these model systems.
- One strength of this project is the collaboration with an automotive OEM with extensive experience in membrane electrode assembly (MEA) fabrication and testing.
- Corrosion-resistant supports have relevance, and studying mixed ionic/electronic-conducting materials is a novel area that has not seen much effort.
- The synthesis of new oxide supports appears to be a strong part of this project. Other strengths include the collaboration with an automotive OEM and the ability to perform RDE measurements and screen for durability.

Project weaknesses:

- The focus on Ru-containing materials is an area of weakness.
- The development of new supports is rather slow, and the PIs may not have enough time to deliver the promised results. While the PIs had many of the needed skills, they did not have all of the skills necessary; it would be desirable to add the skills needed for characterization.
- Principal weaknesses are: (1) the use of Ru as a model; this is not only expensive, but it has dubious stability under potential cycling conditions of a fuel cell, especially the onerous start-up and shutdown steps; and (2) the approach does not compare similar systems for proper conclusions to be made. Particle sizes of Pt and the support BET surface areas are different; these differences could also be responsible for some of the observed results.
- The comparisons between the novel catalysts synthesized and the baseline Pt/C are unclear for relevance, particularly for cycling durability, because these tests are normalized for initial surface area and involve very different particle sizes. The presentation submitted for review contained 80 slides not including reviewer-only slides. The presentation itself was long and made focusing on key findings difficult. The supplemental slides, which were supposed to be limited to five, numbered 50 and were not reviewed thoroughly and could not be evaluated for potential impact or importance.
- Materials characterization to identify catalyst structure, oxidation states, etc. still remains a weakness. The premise of using a precious metal in the support will always raise some questions about cost. The current inability to avoid generating large nanoparticles of Pt has caused low mass activity measurements.

Recommendations for additions/deletions to project scope:

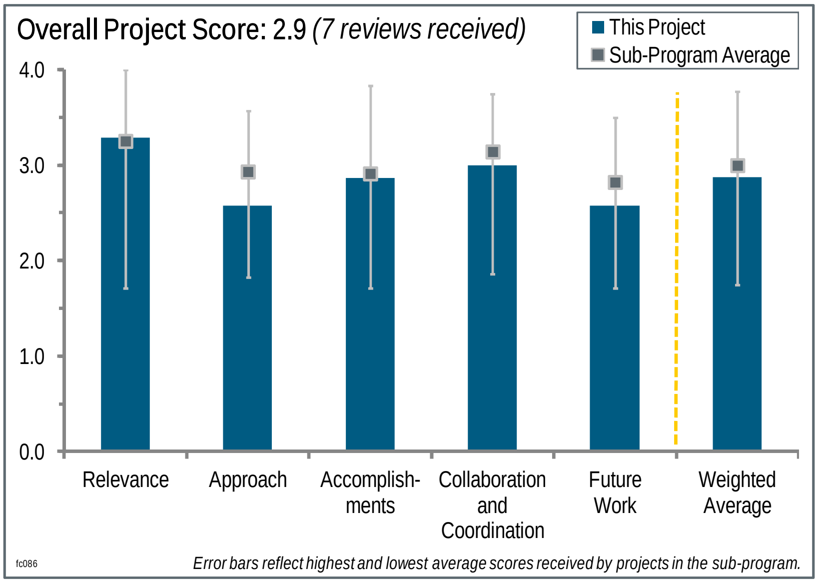
- A more detailed cost study may be beneficial.
- The team should use graphitized carbon with larger Pt crystallites as reference to truly extract the role of the MMO in providing stability. Also, proper fuel cell durability tests need to be reported, especially with H_2/air operations under lower relative humidity conditions.
- The team should move its focus beyond RuO_2 supports. Side-by-side comparisons with Pt/C, including both graphitized and high-surface-area carbon supports with comparable Pt deposition, would be useful.
- For materials synthesis, there needs to be immediate emphasis on lowering the size of Pt nanoparticles for both RuO_2-SiO_2 and RuO_2-TiO_2 catalysts. Despite low activity, RuO_2-SiO_2 and RuO_2-TiO_2-based catalysts should be made into MEAs and durability testing should proceed in a cell. Although durability results from RDE are good, the relevant environment is an actual fuel cell.

Project # FC-086: Development of Novel Non-Pt Group Metal Electrocatalysts for Proton Exchange Membrane Fuel Cell Applications

Sanjeev Mukerjee; Northeastern University

Brief Summary of Project:

This project will develop new classes of non-platinum-group metal (PGM) electrocatalysts that will meet or exceed U.S. Department of Energy (DOE) 2017 targets for activity and durability in fuel cells. This will enable decoupling of polymer electrolyte membrane (PEM) technology from platinum (Pt) resource availability and lower membrane electrode assembly (MEA) costs to less than or equal to $3/kW. The science of electrocatalysis will be extended from current state-of-the-art-supported noble metal catalysts to a wide array of reaction centers.



Question 1: Relevance to overall DOE objectives

This project was rated **3.3** for its relevance to DOE objectives.

- Developing electrocatalysts for the oxygen reduction reaction (ORR) supports the DOE Hydrogen and Fuel Cells Program (the Program).
- Lowering or eliminating PGM and its cost is relevant to DOE objectives.
- It would be a technical breakthrough to develop inexpensive, non-precious-metal catalysts that perform as well as or better than Pt.
- Removing PGM from the cost of MEAs is in line with DOE objectives.
- The project is aligned with the Program and fully supports DOE objectives to decrease fuel cell costs and increase durability. This project aims to develop non-PGM catalysts and offers the potential to substantially reduce costs by removing Pt, one of the largest costs associated with PEM fuel cells, while still meeting performance metrics.
- Lowering the cost of PEM fuel cells is critical to ensuring their commercial success; catalyst cost is a significant portion of the MEA cost and this project intends to develop non-PGM catalysts, which should significantly decrease the cost of the catalyst.
- Non-PGM electrocatalysts are critical to the Program. The cost of an MEA made with non-PGM catalysts needs to be carefully analyzed, including the catalyst production, reproducibility, and cell performance control, so that the real cost can be compared with that of mature PGM catalysts.

Question 2: Approach to performing the work

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

- The approach is sound.
- The approach is based on using suitable complexes and reacting them with metal sources on porous carbon to generate catalytic sites in novel materials. Adequate characterization is applied.
- The approach includes fundamental modeling, mechanistic studies, ex-situ (spectroscopic) characterization, rotating disk electrode (RDE) testing, and cell testing. The principal investigator reported that the density

functional theory (DFT) calculations were performed in a vacuum. Drawing conclusions and direction from this is risky when the actual system includes oxygen and water.

- The project's reasonable approach leads to promising catalyst activity. The non-PGM-catalyst approach still has some uncertainty, such as catalyst active sites, corrosion issues, and surface hydrophobicity characterization, which may have a significant impact on the activity, and hence cell performance.
- The rationale behind the materials selected for development is not obvious; it is unclear what precedence in the literature led the team to select and design the material targets. There was no description of how the team was going to tailor catalysts and control the metal support interactions or control the reaction center's electronic structure. It is unclear what is different about this team's approach to understanding the mechanism of ORR electrocatalysis and mass transport compared to other research groups in this field.
- The approach is directed toward identifying the catalytic center in non-PGM ORR catalysts, improving the catalyst, and improving the electrocatalyst structure for mass transport. Identifying the mechanism and catalytically active site will allow for real advancements to be made in non-PGM catalysts. The computational studies have been idealized and have been vacuum calculations. In the cluster approach, the calculations appear to utilize an N-5 coordinate metal (with an imidazole group occupying one of the axial sites). The nature of the group in the axial position trans to the site where the reaction is occurring can have a significant impact on the reaction energetics because this impacts the electron density available for back bonding to the reactant at the reaction site. It seems unlikely that an imidazole group with strong electron donating character would be present in the real-life (pyrolyzed) system. Due to a higher concentration in these pyrolyzed systems, a weaker-donating -π system interaction (with the next layer of N-substituted carbon) appears to be more likely. A weaker donor is likely to have a significant impact on the bonding energy. Future calculations should trend toward more realistic systems, eliminate the imidazole, and move away from vacuum conditions if possible.
- The project focuses on the development of iron (Fe)-based catalysts for the ORR, and so far results have been equivalent to some of the best non-PGM-based catalysts to date. The project uses a combination of experimental data and molecular modeling, but with little evidence that the model and/or spectroscopically identified species are indeed the active catalyst sites. Some catalysts are tested using RDE and others are tested using the polarization curve; considering that RDE results do not always correlate well with fuel cell performance, it would be good to maybe use RDE as a screening tool and MEA testing as a validation.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- The team has developed electrocatalysts that are as active as the best ORR non-PGM-based catalysts.
- It would be interesting to know what the polarization curves are for Fe and FeCo at lower loadings than 0.6 mg/cm^2.
- Scaling up the catalyst production is very important to validating the approach and revealing more issues, as well providing valuable information for understanding the catalyst's intrinsic properties.
- The DOE 2010 target for performance has been met. The durability cycling results are encouraging. Other tasks seem to be on track.
- Two to three electrocatalysts displayed high activity for the ORR that is close to the present DOE milestone of 150 A/cm^3. Stability has to be improved for most of the catalysts. The mechanistic studies are based on several spectroscopies. These were skillfully used to elucidate several features of these electrocatalysts. The bifunctional mechanism is not strongly supported by the data. "A lack of Fe-Fe bond" is only a suggestion. When H_2O_2 desorbs from one site, it is not likely that it will readsorb on similar ones. RDRE should help in elucidating this mechanism. DFT calculations were used as an additional tool for reaching the conclusions.
- There is no technical data showing that either the precursors or the pyrolyzed materials were obtained as proposed. It is unclear what happened to the metal-organic frameworks described in 2011. There appears to be little recent progress as compared to 2011. Milestones are only slightly changed from 2011. The polarization data should have a control that uses a traditional MEA for benchmarking. Polarization testing conditions should be specified. RDE data and Tafel slopes should have Pt run as a control for benchmarking.
- The team made good progress toward understanding the catalytically active site via the delta-mu EXAFS studies, which have shown a correlation between the presence of Fe-Fe-bonded species and increased activity, and suggest a dual site mechanism. The project has developed non-PGM catalysts with good activity, 150

A/cm^3, but this is still significantly short of the DOE target (300 A/cm^3). The project has developed catalysts with good durability for a non-PGM catalyst over the catalyst durability cycle (0.6–1.0 V); however, durability is poor when subjected to catalyst support durability tests. The researchers have shown some of the losses are recoverable, but stop/start (support) durability needs to be improved.

Question 4: Collaboration and coordination with other institutions

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

- All collaborators participated in the research.
- The team is complete, and includes representatives from fundamental science, stack suppliers, and auto companies.
- There is no description of the nature and frequency of the team's interactions.
- The project features an excellent mix of collaborators.
- BAS might be interested in scaling up the catalyst, but there is probably not much involvement at this time.
- The coordination within the project is good. Coordination with the top experts in the field of non-PGM catalysts outside of the project is occurring.
- The project has strong collaboration with academic institutions. The team should work with more industry partners that will lead the team to gradually resolve the real industry concerns and issues.

Question 5: Proposed future work

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

- The future work is logical and rationally follows the progress made so far.
- Verification of the catalysts' activities and selecting the catalysts for further work is planned to be done using the RDRE technique. The proposed reaction mechanism should be checked using the same measurements. Other plans are a reasonable continuation of the results obtained so far.
- The work proposed for 2012 looks remarkably like that proposed in 2011. There was no description of how the researchers propose to improve mass transport, or even why they think mass transport is bad with the current electrode.
- The studies designed to identify the reaction mechanism should be given the highest priority of the proposed future work.
- Because it is very difficult to identify the active species for such catalysts, the planning proposed may lead to marginal improvements if the species identified are not the active catalyst(s).
- The team should emphasize the research and development work on the batch-to-batch comparison.

Project strengths:

- This project features a strong team and excellent in situ and ex situ techniques.
- The team appears to have access to a lot of really good equipment for testing and characterization.
- The effort to determine the reaction mechanism has provided key insights and is a strength of this project.
- Improvements in transport phenomenon and electrode nanostructures could be applicable to other systems.
- The modeling is strong, as it needs to be, because the actual reaction site is still not known with certainty.
- The project is very strong on research, characterization, and analysis.

Project weaknesses:

- The materials the investigators have chosen for development show little promise for meeting the DOE goals.
- The team needs to strengthen the collaboration with industry partners.
- The characterization of bulk catalyst by XP and the assumption that the main species identified is the catalyst site might not be accurate.

- It appears that the cost to make these materials would be very high, offsetting the cost for PGM cells, which are steadily reducing their PGM loading. These are currently at <0.1 mg/cm^2 Pt on the cathode and <.05 mg/cm^2 on the anode. It is unclear what the cost structure of these routes would be.

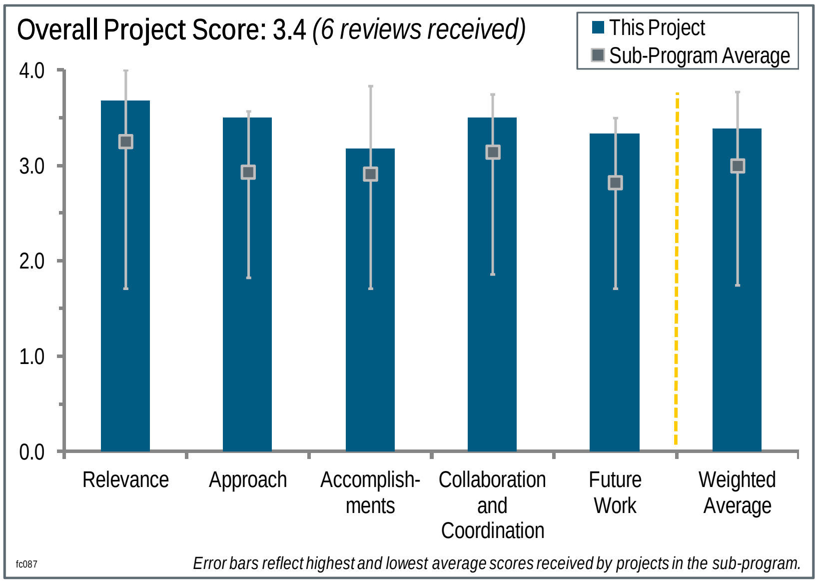
Recommendations for additions/deletions to project scope:

- The team should make additional efforts to verify the reaction mechanism, and perform more long-term stability tests of the catalysts.
- The team should make several larger batches of catalysts that are enough to carry out systematic investigation and cross-checking experiments.

Project # FC-087: High-Activity Dealloyed Catalysts

Fred Wagner; General Motors

Brief Summary of Project:
Project objectives are to: (1) demonstrate reliable oxygen reduction reaction kinetic mass activities; (2) demonstrate the durability of the kinetic mass activity against U.S. Department of Energy (DOE)-specified voltage cycling tests in fuel cells; (3) achieve high current density performance in hydrogen/air fuel cells adequate to meet DOE heat rejection targets and platinum (Pt)-loading goals; (4) scale up to full-active-area fuel cells, to be made available for DOE testing; (5) demonstrate the durability of high current density performance; and (6) determine where alloying-element atoms should reside with respect to the catalyst-particle surface for the best durable activity.



Question 1: Relevance to overall DOE objectives

This project was rated **3.7** for its relevance to DOE objectives.

- The project is well aligned with DOE goals on reducing the cost of polymer electrolyte membrane fuel cells' cathode catalysts and improving their durability.
- Increased catalyst activity and reduced catalyst cost is paramount to the DOE objectives. Dealloyed catalysts are a promising avenue to reach these objectives.
- This project represents an important contribution to the DOE Hydrogen and Fuel Cells Program (the Program). The main focus is placed on improving the durability and reliability of fuel cells by utilizing durable catalysts with improved performance and low Pt content.
- Objectives of the project show direct relevance to DOE's overall objectives of cost reduction (reducing Pt loading), increased durability (maintaining kinetic activity and high current density), and improved performance (achieving high current density with high voltages).
- The development of a cathode catalyst with high activity, performance in the high current density region, and durability is critical to the implementation of fuel cells for automotive applications.
- Catalyst cost is a significant barrier to meeting automotive targets.

Question 2: Approach to performing the work

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

- The project is well designed and sharply focused on DOE targets for catalyst activities and durability.
- The focus of this project has been improved by concentrating on nickel (Ni) and cobalt (Co) systems. The project is well designed and led.
- The project is well planned and each participant represents an important contributor to the overall success of the Program. The major barriers are successfully addressed, and a systematic approach has been pursued in achieving the projected milestones.

- The approach is focused and clear, and milestones and go/no-go decision points have been established. The project has gone through several iterative cycles of synthesis, evaluation, and characterization; the next step is scale-up and durability testing with large batches.
- The approach to improving catalyst activity is excellent, but there is the issue of improving activity and durability while also achieving high current density performance. As identified by the principal investigator (PI), a big risk with this approach is whether durability can be maintained or even improved while meeting the activity targets. Also, the project has gone away from copper (Cu) as the non-noble metal due to its tendency to plate on the anode and degrade anode performance, with the implication that Ni and Co are still leached from these alloys and transported to the anode, but not plated. Perhaps this leaching is then an issue for other components of the cell and system. It is unclear where these leached metals are ending up.
- The project demonstrates a feasible approach, which is relatively low risk compared to other approaches (e.g., non-PGM catalysts). Durability appears to be the key challenge, and some good concepts to overcome the issues to date have been formulated. However, it is not clear why the Pt-Co system is not more at the forefront as compared to the Pt-Ni system. Both should be considered.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- Half of the DOE targets with respect to catalyst durability and activity are already met. The project demonstrates steady progress toward overcoming barriers.
- Good progress has been made, but there is still a relatively strong risk that durability targets may not be met. The team is making acceptable progress for this point in the project schedule.
- There has been good progress on meeting the goals of durability and activity in the same sample, but it is not yet clear that any one sample will be able to meet all of the goals. It seems that the Co systems may be more likely to meet the goals, but the Ni systems have greater promise for high activity if durability issues can be overcome.
- The progress in this project was successfully presented and participants have clearly demonstrated a strong team effort that has been capable of efficiently addressing Program objectives. The concept of utilizing dealloyed catalysts has been implemented for several Pt-bimetallic alloys and was verified as a potent approach in pursuing catalytically active and durable systems with a lower Pt content. A number of characterizations were performed, ranging from RDE, MEA, electron microscopy, in situ EXAFS, etc. The synthesis of a Pt bimetallic system was performed at laboratory-level scale (2 g), and also for larger batches (100 g). All of that has contributed to achieving a better understanding of the key parameters that are responsible for the improved catalytic performance and durability.
- A six-month extension has been requested, and only 30% of the tasks are completed to date. Lost more than 60% activity at 30,000 cycles, not the 40% target. The team has identified the reason for the loss in durability, and it is verifying the hypothesis now. It moved away from Pt-Cu to Pt-Ni because of Cu dissolution issues and Cu deposition on anode. Also, the team is no longer working with Co; Ni should be more durable than Co. The membranes are still too thick at 25 microns.
- Milestone 1 was not achieved. Is it not implied in the milestones discussion that the different milestones should be achieved in one catalyst material at a specified catalyst loading. It is unclear why there is a milestone of achieving mass activity, another for achieving kinetic activity durability, and then another for achieving the two milestones in one material. The accomplishments are good, but there are still many issues to be solved in a short amount of time—the biggest being retention of enough non-noble metal to effect activity but not degrade durability.

Question 4: Collaboration and coordination with other institutions

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

- The project is well coordinated due to General Motors' (GM's) collaboration between industry and universities.
- The project shows good collaboration between all partners and features work done by both academia and industry.
- The coordination of the team is excellent. Contributions from all of partners were evident.

- The project features a nice team composition, which appears to be well coordinated.
- The achieved accomplishments from this project indicate a well-balanced synergistic effort among the participants. The success of this project in addressing critical barriers in fuel cells is due to the highly diverse and organized team, which is well coordinated by GM as a lead institution. Participants have contributed their expertise, which add to the overall success of the project.
- An outstanding team has been established with clear roles and responsibilities. The project's iterative process includes all of the participants. The project will have a strong technology transfer due to having the right team members from the universities characterization capabilities, the catalyst manufacturers, and the stack integrators.

Question 5: Proposed future work

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

- Barriers are clearly identified, and plans are focused on overcoming those barriers.
- The future plans are clearly defined and focused. The future plans are realistic in scope. The team did ask for a six-month extension. Mitigation strategies are in place. The team must still address membrane thickness and durability. Durability testing has already started.
- This project excels in adapting the focus and approach—as evident in the future work—to attempt to achieve all of the targets simultaneously with other materials and processes.
- The PI has shown that the team is clearly focused on addressing the major barriers remaining and has a good approach to do so.
- The proposed future work seems reasonable, but there do not seem to be alternative paths to take if altering synthesis and dealloying conditions does not increase Ni durability. The amount of alloying material should be calculated in relation to the amount of replacement of protons in the membrane. The high current density operation problem will be underestimated if state-of-the-art thin membranes are not used in testing. For instance, it is unclear what percentage of protons will be replaced in a 20-micron membrane, and what levels of contamination are considered acceptable and why. Additionally, polarization curves should be taken past 1.5 A/cm^2 to show the full effects of the cationic contamination. This should be done by taking the curves to true limiting currents, and also by doing some sensitivity of high current performance to air pressure.
- The accomplished achievements in this project are not in ideal accordance with the proposed future work. The diversity of catalytically active and durable systems in the proposed future work is not sufficient. Instead of focusing on alternate dealloying methods, more efforts are needed toward utilizing different alloys. This team has demonstrated great potential in combining expertise in MEA, RDE, dealloying, characterizations, and synthesis; however, only a few promising systems have been studied with a rather narrow distribution of constituents, mainly a 1:3 ratio. Additional focus is necessary to synthesize nanoparticles with even size and elemental distribution.

Project strengths:

- This project features a combination of advanced characterization techniques with state-of-the-art methods of fuel cell manufacturing and testing. The project has a solid fundamental background that allows the team to make fast go/no-go decisions.
- The team succeeded in showing high-activity catalysts that have a great chance to meet the DOE targets.
- The project features a well-organized approach with realistic goals and go/no-go decision points. An excellent team has been established to transition this technology from a concept to manufacturability/industry.
- Strengths of this project include the abilities of the team and the willingness to change approaches and materials to achieve the targets.
- The project team includes academics, component industry representatives, and original equipment manufacturer industry members. The approach is scientifically based and has merit. The project is focused on critical barriers and has good future plans.
- The team demonstrated the utilization of dealloyed catalysts that are different than the Pt-Cu system. The concept of strain that influences catalytic properties of alloy catalysts seems to be applicable not only to Pt-Cu, but also to Pt-Ni and Pt-Co systems. These materials have shown improved performance and durability over Pt catalysts, which clearly justifies the approach taken in this project. A highly collaborative team effort has

allowed this project to produce a valuable outcome. Participants are well-known experts in the field of fuel cells, and they bring highly diverse expertise in MEA, RDE, synthesis, characterization, dealloying, and EXAFS. Kinetic and mass activities have reached the projected milestones.

Project weaknesses:

- The synthesis work does not look as strong as the other parts of the project.
- One weakness is the understanding of the trade-offs between durability and activity. So far there is no great concern about high current density operation, but this will likely be addressed in the following year.
- Only 30% of the project is completed to date. A six-month extension has been requested.
- The project appears to be too focused on one alloy composition (Pt-Ni).
- The project is mainly focused on a few systems, instead of providing an evaluation for a number of dealloyed catalysts with a wide range of elemental ratios. In the current work, particle size distribution seems to be a significant factor that can compromise the quality of measurements and may divert conclusions and future work into the wrong direction. Having ultra-small and large particles at the same time can largely affect the dealloying process, and, therefore, elemental distribution in particles may vary. The lack of theoretical input is a missing part in the listed accomplishments.

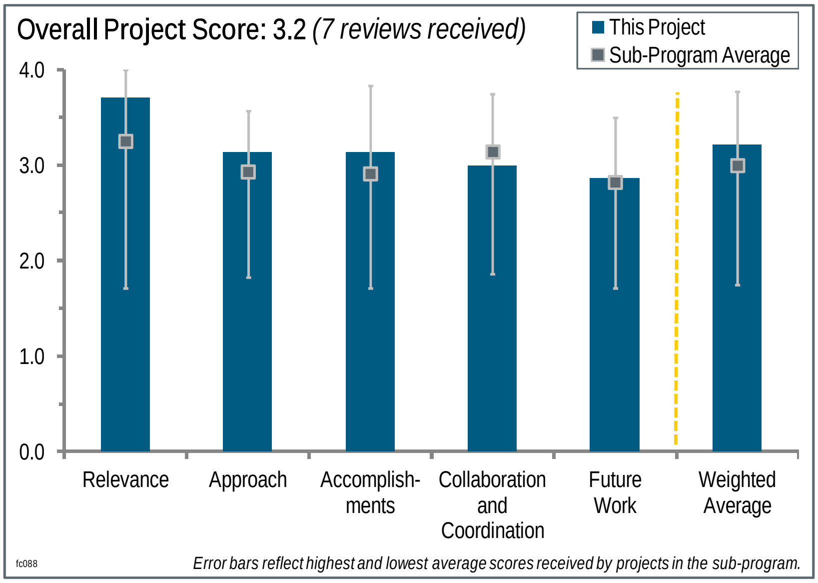
Recommendations for additions/deletions to project scope:

- It would be good to see more durability testing on other alloys (e.g., Pt-Co).
- Next year the team should move to evaluate the durability of supports. It would be interesting to complement a GM evaluation of non-noble metal diffusion time from the center of the particle to the surface by fundamental research. Durability as a function of particle size for dealloyed catalysts needs to be evaluated.
- More work on maximizing Co alloy activity may be necessary if Ni durability cannot be increased. Research seems focused on Ni systems that show greater promise, whereas Co systems may be closer to reaching DOE targets.
- This reviewer suggested that the team keep the project tasks and future work as is.

Project # FC-088: Development of Ultra-Low Platinum Alloy Cathode Catalyst for PEM Fuel Cells

Branko Popov; University of South Carolina

Brief Summary of Project:
This project will develop a high-performance, low-cost, durable cathode catalyst and support that is able to meet the 2017 U.S. Department of Energy (DOE) targets for polymer electrolyte membrane (PEM) fuel cells. The goal will be met through: (1) optimization studies of carbon composite catalyst (CCC) support, (2) development of advanced hybrid catalysts based on CCC support and platinum (Pt), (3) development of carbon nanocage (CNC)-supported Pt-alloy catalysts, (4) synthesis of corrosion-resistant hybrid supports, and (5) development of high-volume procedures for the synthesis of promising catalysts.



Question 1: Relevance to overall DOE objectives

This project was rated **3.7** for its relevance to DOE objectives.

- The development of Pt elctrocatalysts is critical for the DOE Hydrogen and Fuel Cells Program.
- The project is aimed at reducing platinum-group-metal (PGM) content and cost.
- The development of low-PGM catalysts is highly relevant.
- The nature of the work, the motivation for the study, and other aspects of the project are relevant to DOE objectives.
- Reduction of total Pt loading in the fuel cell stack below 10 g is vital for fuel cell electric vehicle commercialization. This project is clearly aligned with this target due to its development of ultra-low Pt alloy cathode catalysts.
- The project is focused on a critical issue, which is to reduce the cost of the catalyst and improve the performance and durability of the catalysts, the catalyst layers, and the membrane electrode assemblies (MEAs) that support them.
- Commercialization of fuel cells can only happen with a reduction of the catalyst cost, which will happen by significantly reducing the amount of Pt used or switching to a non-PGM catalyst. This project takes the approach of significantly reducing the amount of Pt at the cathode.

Question 2: Approach to performing the work

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

- The project builds on previous CCC non-PGM work by the principal investigator's (PI's) group.
- The approach is multifaceted with the investigation of two different supports: CCC and CNC. In both cases, the alloying of Pt with cobalt, copper, or nickel will eventually be employed to further increase activity.
- Development of CCC and modified TiO_2 supports for Pt appears to be a useful approach that has produced active catalysts.
- The experimental approach lacks detail in some places. A significant amount of data was presented; however, the presentation lacked clarity and focus. Almost no details were given on the approach used to synthesize the various catalysts studied in the project.

- The project has made impressive progress through its empirical approach. However, much is empirical and it is concerning that the unknowns are building up to a point where future progress will be stymied due to the existence of too many empirical optimizations that cannot be explained by scientific means.
- The project title emphasizes the development of a Pt alloy cathode catalyst, but a lot of work has been devoted to the development of catalyst supports in order to primarily address the durability of the cathode. The project addresses critical performance and durability issues. It is not always easy to compare the results obtained, because they were not run under similar conditions; in addition, it would be interesting to compare this project's results with a commercial catalyst on the developed support.
- The approach of combining high-activity Pt alloy catalysts with oxygen reduction reaction (ORR)-active supports is a sensible path toward high activity with low PGM loading. The use of conductive oxides as stable supports is also a good approach.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- The progress and accomplishments are outstanding. This project is a very productive use of resources.
- ORR performance, mass activity, and durability (for at least one catalyst system) have been achieved, per the project milestones.
- The PI shared many, many results, indicating a lot of work has been done. Some of the results are very impressive with very clear analytical results supporting RRDE and fuel cell results. Furthermore, the PI has clearly synthesized and validated a whole host of different catalysts in both an RRDE and a fuel cell. However, most, if not all, of the IV data is iR-corrected, which, while valuable, needs to be benchmarked against non-iR-corrected data, otherwise it is not clear how relevant the benchmark data is. NRE212 is often used as the benchmark membrane in MEAs, but for an automotive study, at least, NRE211 should be used. Not enough durability work has been performed to successfully demonstrate the durability of the alloy or support.
- The activity and durability of the Pt catalysts (0.1 mg/cm^2) modified by carbon and pyridinic nitrogen are quite good. The stability of Pt_2Ni requires additional characterization. If the Pt is covered by layers of graphitic carbon, it is unclear how oxygen (O_2) is reaching it. The properties of the Pt/TiO_2 catalyst,—particularly its corrosion resistance—are not sufficiently described.
- Many of the DOE targets seem to have been met. The June 2012 go/no-go decision point has not yet been met, but it seems likely to be achieved. It is not always clear at what conditions the testing is done. For example, Yonsei University seems to use fully humidified O_2 and not air, and data are reported on an iR-free basis. Real-world conditions would provide better information.
- The effect of the proprietary membrane on performance needs to be clarified. Because the results presented are iR-free, the membrane conductivity cannot account for the enhancement. The difference is most likely related to the electrodes. Hydrogen (H_2)/O_2 testing is appropriate for mass activity determination, but for MEA testing at high currents, gases should be H_2/air. The durability results of Pt/TiO_2 are encouraging, but higher initial performance is required. The high mass activity of the HCC catalyst is promising, but H_2/air performance at high current densities is still lacking. The Pt/C baseline used in the H_2/air results is below state of the art, but this is mitigated by the indication that the catalyst layer structure is still being optimized. The activity and durability of the PtNi/CNC catalysts are promising.
- It is difficult to evaluate progress in some areas because confusing, contradicting, or insufficient information was presented. For example, the results from MEA optimization studies for non-carbon supports were given. Durability comparisons were made between carbon and non-carbon (e.g., Pt/TiO_2) corrosion tests and related fuel cell performance. However, it was not explained how this level of performance is achieved from a non-conductive support. Moreover, no description of how the support was prepared was given. It is also difficult to ascertain progress when the PI does not always differentiate between old and new progress. For example, a JACS publication from 2009 (vol. 131, no. 39) showed the same corrosion data and figures for the Pt/TiO_2 catalysts that were presented as new work in this DOE Hydrogen and Fuel Cells Program Annual Merit Review and clearly not conducted in fiscal year 2012. This paper does give insight as to how the TiO_2 support was prepared, and it is a carbon-based route that raises the question of how carbon-free the non-carbon supports really are. The data presented by the PI conveys that the sample is composed of only oxide and Pt. As was seen in an earlier presentation by another PI working on non-carbon PEM fuel cell supports, a small carbon residual can significantly impact (i.e., add) electronic conductivity to the non-conductive TiO_2. This group also shared

data showing that even a small carbon residue also impacts durability negatively. The PI needs to explain the fuel cell performance and durability shown in light of probable carbon contamination.

Question 4: Collaboration and coordination with other institutions

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

- The results of the collaboration are clear, and the collaboration makes a substantial contribution to the project.
- The project's collaborations are good and include universities and auto manufacturers.
- There seems to be a competent team assembled.
- The University of South Carolina (USC) and Yonsei University seem to be working independently on the catalyst support, but coming together on the Pt alloy used.
- The PI states that Yonsei University and Hyundai are partners. Yonsei University's work is barely covered and seems to be quite minimal. Hyundai's funding was stopped in December 2011, and it will resume in February 2013. It appears that the PI is doing all of the work at USC with very little interaction with the partners.
- Collaborations are very minimal, which casts some doubt on some of the results. The development of more collaborative work with, for example, Los Alamos National Laboratory or Argonne National Laboratory for durability studies, and Northeastern University for the CCC work, would be very useful. This work needs to be reproduced by others. Furthermore, there is a need for greater theoretical underpinning, and the team should consider how to accomplish this.
- The collaboration with Yonsei University is valuable because the parallel development of alternative catalyst structures mitigates risk. The collaboration with Hyundai is valuable for hardware design and stack testing. Collaboration with a participant with better MEA development expertise could be helpful.

Question 5: Proposed future work

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

- For future work, the team plans to further improve the best systems. The improvements appear reasonable.
- The future work is focused on meeting go/no-go points and milestones, but there is little discussion on how they will be met.
- The future work looks reasonable. The USC tasks are presented in terms of goals; it would be good to see more details about how these goals will be achieved.
- The future work simply states future milestones, but not the approach that will be used at USC. The PI in this case did share plans from Yonsei University, but the PI made no mention of Hyundai's future planned work.
- The gaps between the project targets and the results today are well understood, and the future milestones present a path to achieving those targets. No alternative approaches have been highlighted if the durability issue of the support (carbon-based) or the performance of the catalyst on TiO_2 support cannot be addressed.

Project strengths:

- The project features reasonable performance for a low-Pt-based catalyst with encouraging durability.
- The basic approach is sound and the participants have extensive relevant experience. The project leverages earlier successful work on designing PGM-free catalysts.
- Obviously, many different catalyst types have been validated and tested at USC. Many of them appear to achieve the necessary performance levels. There is very good correlation of work between the analytical and RRDE/fuel cell results.
- This project features a very broad and quick verification of the catalysts' performance in MEAs.
- The project is attacking known areas in catalysis needed for the advancement of PEM fuel cells.
- One area of strength for this project is its very productive and apparently reliable measurements. Data is plentiful and demonstrates excellent progress.

Project weaknesses:

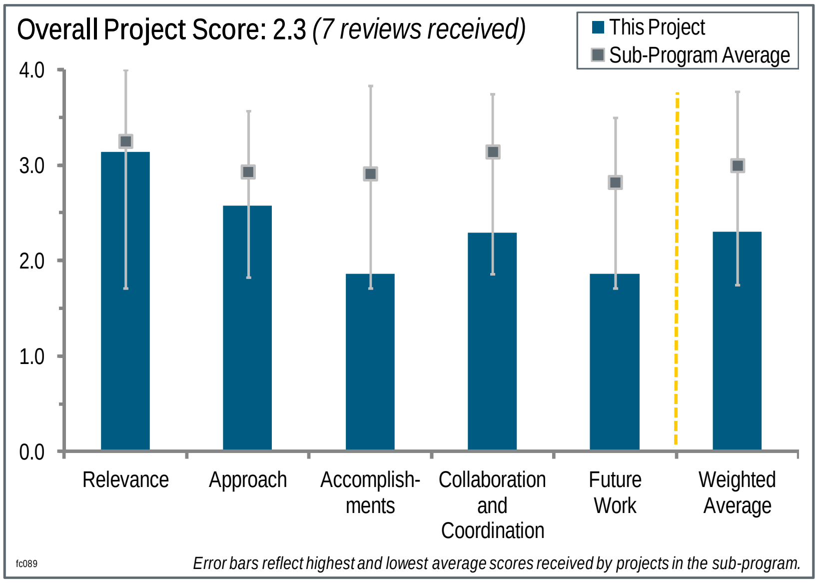
- There is poor interaction with the collaborators. The fuel cell data is all iR-corrected, which takes away from its value—the investigators need to show absolute data. The durability data is lacking when compared to the other data generated.
- The understanding of the behavior of some systems should be improved to make the future work more successful.
- One area of weakness was the confusing and unclear presentation of methods and data.
- The project was not reproduced by others—the team needs collaboration. The project needs better theoretical basis for the improvements.
- Very little data is available on the catalyst particle size before and after operation. More characterization is needed to understand better the degradation mechanism(s).
- The promising mass activity results are not all translating into high-performance MEAs, as of yet. It is not clear that the participants have the MEA integration skills to match their catalyst development skills.

Recommendations for additions/deletions to project scope:

- Additional characterization of the Pt/TiO_2 system seems necessary.
- The team should add more collaboration for both practical measurements and theoretical basis.
- The team should focus on one or two high-potential catalysts. It should also make better MEAs with NRE211 and show its absolute performance under a variety of conditions. Researchers should clearly show their future plans and get Hyundai involved again.
- The team should not make any changes.
- No additions or deletions should be made.

Project # FC-089: Analysis of Durability of MEAs in Automotive PEMFC Applications

Randy Perry; DuPont



Brief Summary of Project:

This project addresses gaps in the understanding of fuel cell durability and modeling of fuel cell performance degradation. The project focuses on durability at low relative humidity and during automotive cycling operation, and addresses short side-chain polymers (-O-CF2-CF2-SO3H). Three main objectives of the project are: (1) determine accelerated stress tests (ASTs) to be used to generate data for modeling of the individual degradation mechanisms; (2) develop an overall degradation model that correlates the stack operating conditions to the degradation of the membrane electrode assembly (MEA); and (3) develop MEAs with a design lifetime target of 5,000 hours with ≤7% degradation and that show a clear path toward meeting U.S. Department of Energy (DOE) 2017 technical targets.

Question 1: Relevance to overall DOE objectives

This project was rated **3.1** for its relevance to DOE objectives.

- This project is generally well aligned with the performance and durability goals for MEAs.
- Improving durability is critical for the commercialization of fuel cells. The project is defining ASTs, developing a degradation model that includes operating conditions, and working to develop durable MEAs.
- ASTs are potentially very valuable tools for assessing fuel cell materials durability; however, they do need to be validated.
- Developing a degradation model and the goal of achieving the DOE targets are critical for polymer electrolyte membrane fuel cell commercialization.
- The topic area and original intent of the proposed project was relevant, but for various reasons the project has not followed the proposed scope or added much value to the DOE Hydrogen and Fuel Cells Program.
- Delays in the start of this project perhaps hurt its timeliness; at the same time, the delays could be an advantage if the researchers could build upon the learnings of the other durability projects. Unfortunately, it was unclear from the presentation how the projects differentiate and relate to one another. This project seems to be using protocols that are slightly different from DOE's protocols. That can be an advantage and a disadvantage in comparison.

Question 2: Approach to performing the work

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

- The approach of looking at durability issues in Nafion-related membranes is good, as are the diagnostics and modeling to understand degradation.
- The approach appears to rely solely on a few materials and AST tests. The team dropped the development of a durable MEA. It is unclear why it took so long to get the Nissan subcontract in place. Nissan should be commended for providing so much work while not under contract.

- The researchers performed two types of ASTs—the test protocol developed by the U.S. DRIVE Partnership's Fuel Cell Technical Team (FCTT) and a Nissan protocol; however, it is difficult to evaluate the validity of the tests because no long-term degradation tests were performed. The value of the study without baselines is unclear.
- The proposed approach and intent of the project were useful and effective, but the actual execution of the project has been limited to contributions from Nissan using the Japanese fuel cell durability protocols and comparison to the DOE protocols.
- The approach of this project seems well thought out, with clear go/no-go points, although the "Approach" flow chart on slide 5 is not very helpful. There is a typo on slide 4, go/no-go 1, sub-bullet 2, which sets a target of 1 kW/cm^2. This should be 1 W/cm^2.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **1.9** for its accomplishments and progress.

- There does not appear to have been much development of mechanistic models or validation of the results.
- The project describes MEA testing, but little progress has been made toward the goal. The only thing that the researchers have really done is develop test plans.
- The amount of work completed and the results presented seem in line with expectations for this type of project. The details provided from the test protocols, especially from Nissan, were very informative and helpful in interpreting the results.
- This project is adopting the "Nissan/Japanese" fuel cell carbon corrosion start/stop protocol, and it shows data from approximately five catalyst-coated membranes. The MEA on slide 9 appears to have originated with a cracked catalyst layer. These are probably not the best samples to use, because the cracks have known degradation effects. Most results appear to have come from Nissan's own work, not as part of this project.
- The strength of the project has been in the background work performed by Nissan. The accomplishments to date have been limited compared to the proposed statement of work. Also, development of the model based on experimental/characterization results is not evident, and the application of the suite of characterization techniques listed is also not evident. Due to difficulty in establishing subcontracts, the progress of the project in general has been minimal, specifically the contributions from Illinois Institute of Technology (IIT).

Question 4: Collaboration and coordination with other institutions

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

- The project seems to have a well-balanced scope of work and good interaction between the partners. The partnering organizations represent a nice balance from academia to materials development representatives to original equipment manufacturers.
- The collaborations in the project are unclear. IIT is supposed to do the modeling, but nothing was shared until the reviewer-only slides; even so, the IIT work does not show if there is any real collaboration between IIT and other project partners. Nissan appears to have done most of the work to date, yet it was not under contract.
- 3M was a subcontractor in 2011, and it is no longer one. It is unclear whether there was a particular issue that precluded their involvement.
- The only progress made with Nissan involved deciding which ASTs to use.
- Good collaboration between DuPont and Nissan is evident. The collaboration with IIT has either not started or is minimal.
- The collaboration is weak because the partners were so late to get under contract. DuPont could have been more creative to get its partners going earlier. DuPont does get some credit for working with Nissan to develop test protocols.

Question 5: Proposed future work

This project was rated **1.9** for its proposed future work.

- The proposed work is fine, except for the lack of a model.

- The project needs more analytical work to understand the reasons for differences in degradation. Long-term drive-cycle testing also needs to be performed to validate the ASTs.
- There appears to be a tremendous amount of work to be done before the go/no-go decision this fall. It is unclear if the team will have sufficient time to gather enough data for the reviewers to make an informed decision.
- The planned future work seems to be on target to continue addressing DOE's objectives and is generally responsive to the detailed feedback provided by the FCTT and reviewers. Researchers should pay close attention to distilling the key learnings from their work and put those learnings in clear context in next year's report to the community.
- Future work includes beginning tests with HSAC. It is not clear from this project what the researchers intend to learn from this. Testing the effect of polymer morphology seems like an area that has been unexplored and one at which DuPont should excel. New work on gas diffusion layers and plates was mentioned in terms of post-mortem, but no information was provided on what type of testing/characterization was used, what type of plates, or what data the researchers hope to get and how to use it.

Project strengths:

- This project features good collaboration and a well-balanced team. The team employed good transparency of protocols.
- This project has good coordination between the partners.
- The original proposal was reasonable.
- The team and its complementary expertise is the project's strength.
- The industry lead provides relevant membranes.

Project weaknesses:

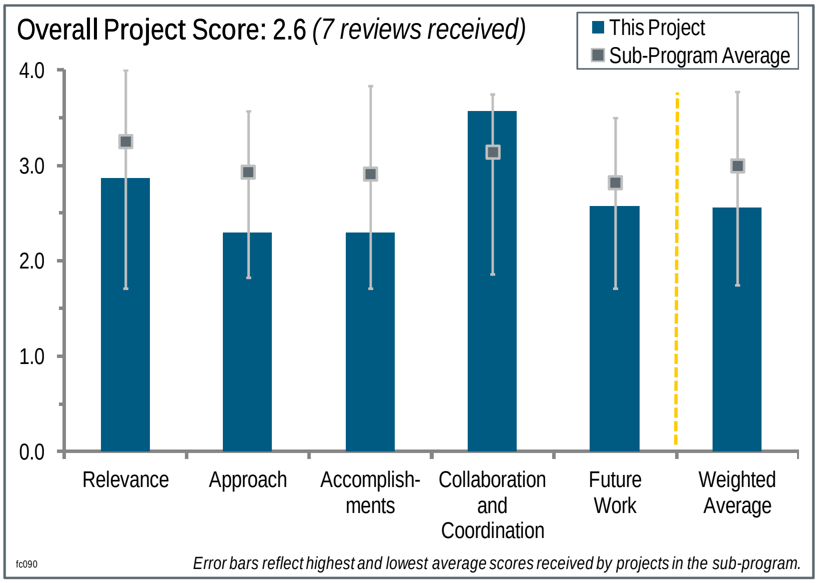
- The project needs to provide greater contextualization of what the team has learned that is important to the industry.
- The project should show real collaboration and how data, materials, and/or information are being exchanged to make a stronger project, rather than just being three independent projects.
- The project needs more modeling and validation.
- The project has not yet really started.
- A weakness of the project is the lack of coordination and development of the model based on experimental results, which has been hampered by subcontract issues, delaying the actual flow of funds to the partners.
- The project has not achieved any modeling despite being a modeling project.

Recommendations for additions/deletions to project scope:

- The researchers could be much more descriptive concerning the nature of the future work and what they intend to learn. The integration of the modeling effort appears to need work. The carbon corrosion work would be more valuable if CO_2 evolution data was reported with recovery sequences. Similarly, only electrochemical surface area was presented; particle distribution analysis is more valuable.
- The investigators should begin work on the effect of short-chain ionomers and model development immediately.
- With 80% of the project funds remaining, this effort should be significantly re-scoped to make it an effective use of DOE's investment. A partner with a practical model is needed, and the appropriate measurements and diagnostics should be carried out to get useful results from the model.
- There are no additions or deletions to make to the project scope.

Project # FC-090: Corrugated Membrane Fuel Cell Structures

Stephen Grot; Ion Power



Brief Summary of Project:

The main goal of this project is to pack more membrane active area into a given geometric plate area, thereby allowing both the targets of power density and platinum (Pt) utilization to be achieved in fuel cells. In support of this goal, the project will: (1) demonstrate a single cell (50 cm^2) with a twofold increase in the membrane active area over the geometric area of the cell by corrugating the membrane electrode assembly (MEA) structure; and (2) incorporate an ultra-low Pt-loaded corrugated MEA structure in a 50 cm^2 single cell that achieves the U.S. Department of Energy (DOE) 2015 target of 0.2 g Pt/kW, while simultaneously reaching the power density targets of 1 W/cm^2 at full power and 0.25 W/cm^2 at one-quarter power.

Question 1: Relevance to overall U.S. DOE objectives

This project was rated **2.9** for its relevance to DOE objectives.

- The project is relevant to reducing the cost of polymer electrolyte membrane (PEM) fuel cells by using a novel cell/stack design architecture.
- This is a nice project that addresses key barriers within the DOE Fuel Cell Technologies Program (FCT Program) related to costs and performance.
- Enhancing membrane and MEA performance is very important to reaching FCT Program targets.
- The team's two goals are laudable: (1) raise volumetric power density and (2) lower catalyst loading. The attempt to raise power density by geometrical change of stack is laudable, although the validity of achieving this goal by the team's approach has not been demonstrated—even preliminarily—after one year.
- Any method to decrease material costs while maintaining performance and durability would be highly relevant. However, the ability of the structures developed by this project to do so in a cost-effective, readily manufacturable way is questionable.
- The basic construct for a PEM fuel cell, bipolar plate, and frame has not been seriously questioned for decades. It makes sense to review such assumptions periodically. So, it is good to see that DOE is willing to explore basic stack design issues.
- New, "out-of-the-box" ideas are welcome to overcome barriers to fuel cell adoption, but this project has a low likelihood of being incorporated into automotive fuel cell stacks. It is not easy to understand how corrugated structures can significantly increase the system power per volume or significantly reduce the system cost for the same power. The corrugated structure greatly increases the complexity of the stack components. Durability will also likely be negatively affected.

Question 2: Approach to performing the work

This project was rated **2.3** for its approach.

- This is a nice approach to a novel, single-stack system that is early in the project.
- This is an innovative approach to enhancing fuel cell power density and Pt utilization.

- The task structure, schedule, and milestones were not presented, so the approach is not clear. The approach seems to be almost trial and error. Additional modeling (e.g., fluid flow and cell compression) would be instructive. The presenter did not discuss whether the adjacent cells are nested, pinched, or parallel. This is a critical design and assembly parameter.
- The approach is to raise the real versus projected area by corrugation. Calculations suggest this gain will persist in three dimensions, but this is not a "given," and therefore the premise needs validation by experiment as soon as possible.
- A corrugated structure was made, but the team gave no indication that a thought process was used to determine the optimal corrugated geometry. It is unclear why the aspect ratio between the frequency and amplitude of the corrugations was chosen. In order to maximize total surface area per square centimeter, one could build a corrugated structure with a very high aspect ratio. This would definitely have higher current density per cm^2 flat area, but it would not necessarily make a better power-to-volume or power-to-weight ratio in the stack. The price modeling needs more explanation. It is hard to see where the actual cost benefits are coming from. It is unclear what assumptions are being made. It would be interesting to know if the total amount of Pt used to reach the same system power is decreasing, and how this decrease in Pt is affecting voltage at maximum power and efficiency. Corrugated materials cannot be less expensive than non-corrugated materials. Also, system volume is shown to decrease, but the team did not present any real data showing how the size of these new systems compares to the size of the state-of-the-art fuel cell stacks. Investigations into metal flow fields show more promise than the corrugation work.
- In principle, this is a promising approach, but serious concerns remain as to how this approach could be implemented in a high-throughput manufacturing process. It will be much more difficult to achieve low tolerances in the corrugated structure than in a conventional flat structure. There may be issues with pressure distribution and contact resistance. Also, the 1 W/cm^2 target is an MEA target, which presumably applies to the real area of the MEA, so the corrugated structure would have no bearing on the achievement of this target.
- The basic assumptions made in the proposal were perhaps not well thought out. Considerable time and effort has been spent working on materials issues and determining how to build the gas diffusion layer (GDL) instead of wondering why it should be built. The critical issue of power density seems to be ignored, as was the issue of operating any stack with very low Pt loading with the expectation of having useful durability (ultra-low Pt loading and excellent durability tend not to go together.) There was also no apparent questioning about the complexity of manifolding, the issues of water removal, or thermal management. Likewise, the fluid dynamics of reactant flow appears to be overlooked.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- A cost-benefit study showed positive results (reduced cost) for the concept. Progress is respectable, considering the funding level.
- Most work has been on non-corrugated flat screens and projections based on estimations. Real effects such as compressive deformations (shorts by sharp points) have not been experimentally checked.
- Some thoughtful design and fabrication work has been completed. However, early results tend perhaps to show that the design will be difficult to accomplish.
- Because the project is only 25% complete, there are still a lot of questions to be asked, especially in the area of GDL and membrane corrugated manufacturing to allow more active surface area. Subassemblies have been built but not utilized.
- Progress has been made toward creating corrugated structures. Cost analysis with different Pt loadings has also been completed.
- Researchers developed very nice tooling to properly implement this concept. General Motors (GM) developed a very interesting cost model that will help focus the future effort toward the overall lowest cost.
- The project is still only 25% complete, and fuel cell testing with the corrugated membranes has yet to begin. Some progress has been made with GDL development and with the modeling effort. The lack of reference to any schedule, milestones, or decision points makes it difficult to assess actual progress versus the proposed work. The MEA testing results with flat cells are hard to interpret because they are plotted in terms of current (should be current density), and because the operating conditions are poorly specified. Not knowing the

experimental details makes the comparisons to the baseline materials unconvincing—it is not clear if these GDL materials really provide an improvement, or if the baseline is just bad.

Question 4: Collaboration and coordination with other institutions

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

- Strong collaboration was shown.
- The collaborations are good, covering screen and plate suppliers and an automotive original equipment manufacturer.
- GM, Graftech, Ion Power, and their vendors are all first rate.
- This team is very strong technically with significant collaboration with industry.
- This project features excellent collaboration with GM and various suppliers.
- The collaboration with GM is very valuable.
- The collaboration from GM apparently was focused on cost. It seems a little early to worry about cost; there is no firm evidence that the concept is sound. The other collaborations seem to be interactions with vendors.

Question 5: Proposed future work

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

- The proposed future work does not show a comprehensive plan of design.
- Fuel cell testing is critical to validating this concept.
- More detail is needed. Dates, milestones, and decision points are needed.
- A critical part of the project is single-cell testing, which is planned for the next period assuming that all of the cell parts (plate, GDL, etc.) can be fabricated and assembled.
- The proposed work involves making a corrugated GDL and testing it in a single cell. Stack work needs to be done as well to experimentally test the premise of lower catalyst and higher volumetric power density.
- The proposed future work is exciting, as the project team pushes toward working with the new subassemblies to manufacture the corrugated GDL and start single-cell testing.
- It appeared that the required funding to move this project forward is much more than is in the budget. The pathway forward was not described.

Project strengths:

- This project has a strong team.
- The project features a good concept and a great team to implement it.
- This project has a novel approach that will allow for more surface in the membrane (e.g., more active surface area), which could increase single stack performance. The cost analysis presented showed the potential for significant cost savings by using these corrugated systems.
- The metal meshes show interesting results.
- The idea is innovative and, if implemented successfully (a big if), could provide significant cost reduction.
- It is worthwhile to explore possibilities for new electrochemical reactor design, and this is one of those.

Project weaknesses:

- The physical (flow, structural) modeling is inadequate.
- The team did not complete enough engineering design work before bending the metal.
- This is taking a long time, and secondary issues such as the corrosion cost effects of mechanical stresses and lifetime should have been addressed by now.
- Because the subassemblies for GDL and membrane production have just been completed, more single-cell testing is needed. Mechanical compression, bonding/interface of cell setup, and uniform thickness are a concern going into cell testing.

- The assumptions made in modeling are not intuitive. It is hard to imagine how a corrugated structure is better than a structure that is running on a lower Pt loading but with more active area.
- The corrugated structure will be more difficult and more expensive to manufacture than conventional flat MEAs. Increased dilution of Pt across the MEA leads to higher peroxide production. Reactant distribution and water management may be more difficult.

Recommendations for additions/deletions to project scope:

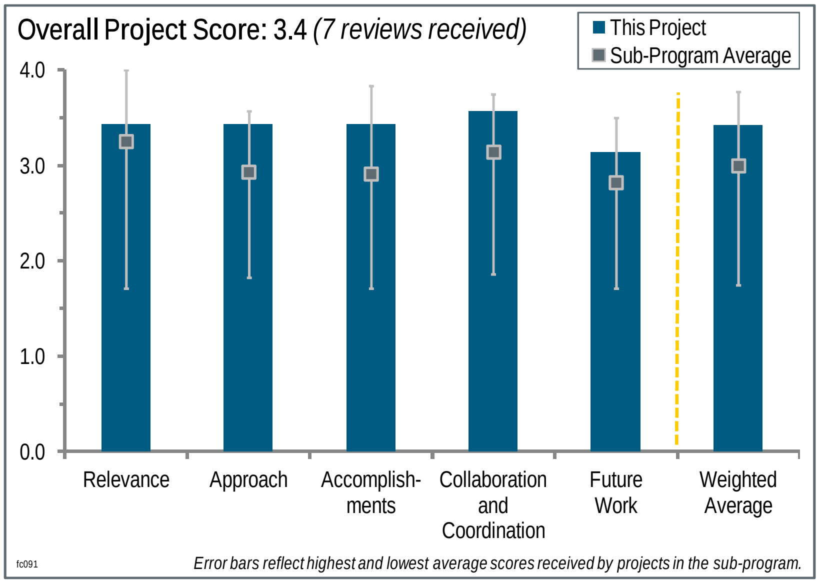
- Stack work needs to be done as well to experimentally test the premise of lower catalyst and higher volumetric power density.
- It is recommended that this project should continue in anticipation of results next year.
- Work on optimizing metal mesh flow fields seems to show more promise than corrugated geometry. A more fundamental and detailed understanding of these results would be more useful for the community.
- Doing some computational fluid dynamics to describe the flows of reactants, products, and heat makes sense. There also needs to be some thoughtful analysis on this design, even if the initial proposed advantage is not real. (That assumed that operation with a lower current density would result in higher voltage, even with very low electrode loadings.) There could be benefits in this approach. The Argonne National Laboratory team could be of good help here, and several issues might be quickly resolved. Clearly there needs to be some thinking about reactant crossover, and the implications of having much more surface area with the same number of electrons generated. At low current density levels, “i” is low, and thus iR is too. Perhaps crossover can be reduced to a low level. The GM help stressed costs. Costs are not the issue here. (Solid oxide fuel cell designs have for decades used corrugated cells, and that design was selected for higher durability.) The analysis needs to look at the complex set of variables. There are many; for example, if one works at 85% efficiency rather than 50% efficiency, there is much less heat to manage.
- The team should not make any changes.

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

Piotr Zelenay; Los Alamos National Laboratory

Brief Summary of Project:

This project's objective is to develop advanced materials (e.g., catalysts, membranes, electrode structures, and membrane electrode assemblies [MEAs]) and fuel cell operating concepts capable of fulfilling cost, performance, and durability requirements established by the U.S. Department of Energy (DOE) for portable fuel cell systems and to ensure a path to large-scale fabrication of successful materials.



Question 1: Relevance to overall DOE objectives

This project was rated **3.4** for its relevance to DOE objectives.

- The project is excellently aligned with DOE portable power and overall fuel cell goals.
- The project is relevant to meeting DOE targets for portable power.
- The project's multiple goals seem to be aligned with the overall DOE portable power goals.
- Catalyst and electrode work in particular are relevant to the DOE Hydrogen and Fuel Cells Program for portable power devices.
- This project fully supports DOE objectives related to direct methanol fuel cells (DMFCs).
- The project is relevant to the objectives of the Fuel Cell Technologies Program's Multi-Year Research, Development and Demonstration Plan. The activities are aligned to DOE's goal. This project is focused on the development of advanced materials, such as catalysts, membranes, electrodes, and MEAs for DMFC application, which is expected to fulfill the cost, performance, and durability requirements established by DOE, and is very important for the commercialization of DMFC technology.
- The project addresses the main DOE challenges of durability, cost, and performance for portable fuel cells. The project is aligned with the DOE goals and performance targets for portable fuel cells. The DOE performance targets have been modified since the project proposal and may present an opportunity to have a successful outcome even if the original proposed targets are not met.

Question 2: Approach to performing the work

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

- The technical approach used in this project is adequate and well defined.
- Overall, the approach is sound and focused toward addressing barriers.
- The work is a good balance of introducing new materials into DMFCs and working on enabling technology for next-generation fuels. Degradation studies indicating the change in crack formation in catalyst layers as a function of methanol concentration and voltage are useful.
- The approach is quite broad and covers catalyst research; innovative electrode structures; hydrocarbon membranes; research into the use of alternative fuels; and characterization, performance, and durability testing in multicell devices. The approach appears to be well balanced with numerous go/no-go decision points.
- The multidirectional approaches taken for the completion of all tasks are adequate. All of the technical barriers have been addressed appropriately. The responsibilities for anode, membrane, alternative fuel development, and performance/durability testing were given to the research teams with significant experience and strength in those respective areas of research.

- The overall approach seems to be a sound evaluation of the materials' performance and durability. The project team seems to be pursuing a range of different approaches to improving the performance of the subcomponents. Making gains in these materials for DMFCs should provide gains related to the performance and durability. The work in alternative fuels with dimethyl ether (DME) and ethanol provides an interesting alternative approach; however, the ability of these materials to meet the DOE objectives for portable power in a practical packaged design was not clear either due to fuel storage and use issues or uncertainty about their ability to meet the energy density targets.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.4** for its accomplishments and progress.

- The team has made significant progress in many areas.
- Very good progress has been made. The team has generated a significant amount of high-quality results.
- Significant progress toward project goals was made. There should be some sort of cost estimate on how use of the BPSH copolymers is going to lead to a lower MEA cost.
- The team appears to have made good progress in several areas, in particular in ternary anode catalyst development by showing good mass activity for the catalyst. Additionally, the work on understanding the degradation mechanisms looks promising as a mechanism to understanding the long-term performance of DMFCs for portable applications. While the work on catalysts for ethanol oxidation looks promising, it was not clear whether sufficient performance would be obtained for a practical MEA. The results for the DME looked promising, but it was not clear how the fuel could be utilized in a practical portable power system.
- The project team has made several achievements, including excellent improvements in anode potential with advanced catalysts, and successful scale-up to 100 g without performance loss. The anode research is on track to reach the target of improved activity of thrifted PtRu catalysts without a durability loss, and to reach the project goal of 150 mA/cm^2 at 0.60 V (DMFC). The work on ternary catalysts for ethanol oxidation is very promising.
- Los Alamos National Laboratory (LANL) has achieved numerous milestones to date. The mass activity milestone for a tin-containing PtRu catalyst was exceeded by 150%. Three multi-block copolymer membranes bettered the performance of Nafion® 212 in DMFC testing with a 55% reduction in methanol crossover. LANL achieved a DMFC fuel utilization of >95% at peak power. Good performance was achieved for direct ethanol fuel cells, but the stability of the catalyst needs to be improved.
- Significant achievements have been realized in membrane and anode developments. Impressive performance was observed with multi-block copolymer membranes. The gas-feed DME is showing little performance advantage in the mass transport region. However, overall performance efficiency, considering the parasitic loss incurred by pressurizing the system, should be considered for making comparisons between liquid- and gas-feed DME cells. The team has reported that the DME cell performs better than a DMFC at a voltage higher than 0.49 V. However, the team should consider their respective performances at higher current densities, where a practical cell will be operating; from the given data, a DMFC clearly outperforms a DME cells at 500 mA/cm^2. The team should calculate the parasitic loss associated with the back-pressure of a gas-feed DME system to make a true comparison between gas-feed DME and liquid-feed DMFC systems.

Question 4: Collaboration and coordination with other institutions

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

- Many good collaborators with key areas of expertise have been pulled into the project.
- The project features a large number of high-quality collaborators.
- The project activities are well distributed and coordinated among the team members.
- Collaboration is very strong with universities and a major catalyst supplier. The project team is strong; however, it is not clear how much funding and effort is placed with the collaborators.
- The team consists of a good mix of university, national laboratory, and industrial partners. The proposed collaboration with Oorja Protonics will also be very advantageous to the team because Oorja has commercial high-power DMFC system offerings, which will help the team obtain good information on the material/performance requirements for a high-power DMFC system.

Question 5: Proposed future work

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

- The future work described is aligned with the proposed work of the project.
- Reducing methanol crossover in the BPSH block copolymers by removing the 6F moiety and replacing it with hydroquinone may lead to higher interfacial resistance between the membrane and the electrode.
- LANL should propose a critical path that will lead to a viable product with commercial potential for at least one of the paths being investigated. This critical path will necessitate some form of go/no-go decision, which may lead to abandoning a promising research area to achieve a recognizable success by the end of the project.
- Even though the proposed future work is adequate and in accordance with the project objectives, it would be good to see the correlation between methanol crossover and catalyst layer cracking before the development of a mitigation strategy, since the cracks are formed in MEAs with Nafion®.
- The overall approach to further work looks good for the four broad categories listed in the presentation. The ternary anode catalyst improvements and durability analysis under practical stack operating conditions are important steps in understanding the benefits of the new anode material. The materials development regarding the membrane and catalyst is focused on the key improvements in durability and performance. For nanostructured catalysts, it would be useful to understand the trade-off of the potential performance improvements with any increases in catalyst cost. It would be useful to ensure that performance benefits were achieved in an MEA. As improvements in the DME oxidation are made, it would be helpful to understand how they could be incorporated into a portable power application that can meet the DOE portable power objectives.

Project strengths:

- LANL has made excellent progress to date. This project will significantly advance portable power fuel cell capability.
- The project features an excellent team and results. The catalyst synthesis correlates well with theory.
- The team is well organized and capable of developing DMFC membranes and MEAs. The team is composed of research organizations with adequate expertise and resources. Overall, the team is equipped with the knowledge base and resources required for the success of this project.
- This is a strong team that has carried out good basic and applied work to understand the fundamental mechanism for performance improvement and durability.
- The project features excellent work and significant achievements. The project combines fundamental approaches with MEA/fuel cell testing.
- One strength of this project is the excellent make up of the team. The slides contained a significant amount of organized information with highlights pointed out to make the reviewers' job somewhat easier.

Project weaknesses:

- The team is a combination of a large number of research organizations, which may be a management challenge for LANL.
- The project involves many different approaches. Because these are early fundamental data for some of the approaches, it is hard to see how some of these approaches are going to produce increases in power and energy density to meet the DOE goals, which are expressed at the system-level for portable power.
- Increased clarity on the practicality of some of the approaches is desired.
- The slides were too information dense. Too much detail on each slide made it difficult to follow during the presentation. The slides should contain one or at most two takeaway messages.
- More data are necessary regarding the membrane durability in the presence of higher concentrations of MeOH. In addition, the crack formation mechanism in the MEA catalyst layer needs to be better understood, whether it is caused by the membrane properties, catalyst, or the MEA preparation.

Recommendations for additions/deletions to project scope:

- LANL should continue to focus on methanol as a fuel.
- LANL should narrow the scope of the project to those paths with the most promise.

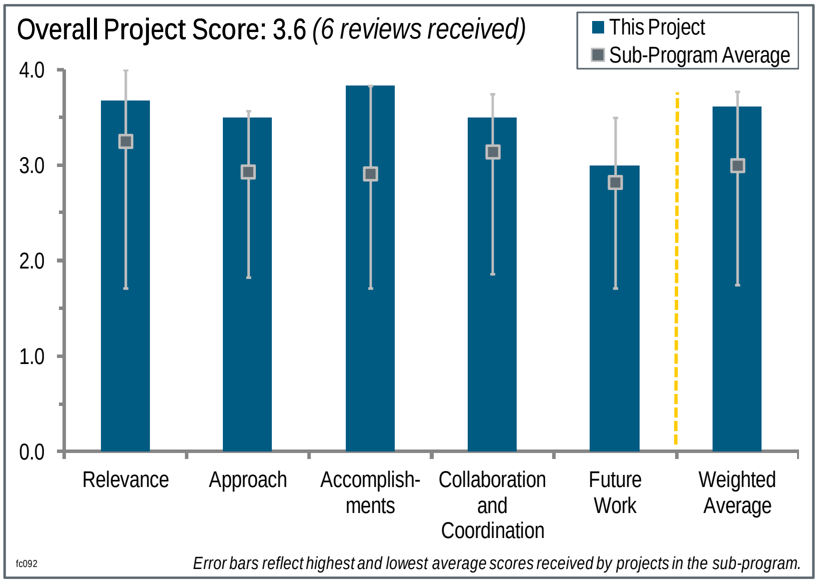
- Given the team's strength in direct fuel systems, the team should drill down to the fundamentals of DME fuel systems and determine the viability of the success of DME technology as compared to DMFC technology.
- LANL should carry out work to understand the potential benefits or trade-offs in the performance improvements and costs at the system level.
- Durability is a major concern. LANL should increase emphasis on understanding and mitigating durability effects. The team should also increase interactions with SFC Energy to ensure practicality and progress toward realistic goals and operating scenarios.
- LANL needs to investigate thoroughly the causes of the crack formation in the catalyst layer. It is recommended that Johnson-Matthey Fuel Cells Inc. be involved in the preparation of catalyst layers and the MEA assembly as a parallel path to the MEA preparation procedure developed by LANL.

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

Jon Owejan; General Motors

Brief Summary of Project:

The core objectives of this project are to: (1) develop a validated transport model including all component physical and chemical properties; (2) disseminate information about the model and instructions for using the model to the public, primarily through the project website; (3) compile data generated in the course of model development and validation to guide model physics, and publish it on the project website; and (4) identify rate-limiting steps and recommendations for improvements to the plate-to-plate fuel cell package.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.7** for its relevance to DOE objectives.

- Understanding transport and transport processes is critical to the commercialization of polymer electrolyte membrane (PEM) fuel cell systems for transportation applications.
- This project legitimately addresses multiple barriers, including performance, water transport, system management, and transient operation.
- It is very relevant to the goals and objectives in the Fuel Cell Technologies Program's Multi-Year Research, Development and Demonstration Plan. This program is important as a basic analytical and design scheme between materials and performance.
- The project is firmly in line with DOE goals and objectives with respect to understanding (water) transport in PEM fuel cells and its effect on performance and durability.
- This project is relevant and the results will be useful to the fuel cell community, but it is not clear that stack manufacturers or automotive fuel cell companies will be able to use these results. There is significant variation in the materials and design of stacks, and the one-dimensional models are not easily transferrable to other fuel cell stack systems.
- This project is well aligned with the DOE Hydrogen and Fuel Cells Program. Of key value is the ability to link changes at the membrane electrode assembly (MEA) structure level for low-cost design concepts to changes in sensitivity to the operating design window, both in terms of absolute performance and current re-distribution at the cell level. This allows industry to conduct the relevant trade-offs to understand technical feasibility and cost of each of the different system components (including the stack).

Question 2: Approach to performing the work

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

- The approach is almost perfect. Based on the experimental results, MEA performance is well predicted.
- This project employs characterization (which is mostly in situ transport measurements) to develop a down-the channel model.
- This is an ambitious but impressive attempt to measure and model all component resistances and overpotentials in the stack. If successful, this would be the definitive PEM fuel cell model.

- Extensive ex situ characterization and parametric in situ testing support the modeling effort for auto-competitive material sets. Most of the modeling effort is based on actual component data, such as thermal conductivity and component diffusion. The results are posted on a public website and component properties are public.
- The approach, as described, was very clearly laid out with responsibilities for each of the collaborators as well as when deliverables are expected to mature. One barrier that was assumed to have been addressed was “start-up and shutdown time and energy/transient operation.” It was not clear from the presentation material how this barrier was being dealt with; indeed, in the supplemental notes it was made clear that durability was not a focus. Transient operation was not described in any detail and neither was the expected impact of transient operation on low-cost designs, although some assumptions could be derived from the detailed assessments made in this body of work. Of particular note was the modular, bottoms-up approach to model development and experimental design where individual component elements could be investigated in the absence of other noise or design factors.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.8** for its accomplishments and progress.

- The experimental measurement techniques are exceptional. A huge database is also helpful for validation. This can be a model case of an MEA design scheme.
- This project has produced a significant amount of data. However, there is so much data and so little description about the data that it is difficult for a person outside the project to understand most of it.
- The model is based on physics and component properties, not fitted parameters. Extensive data are openly provided on components and their properties, and the model accuracy is reasonable. Parameters needing improvement have been identified and are being addressed.
- Components—such as the microporous layer—that are critical in determining the mass transport have not been evaluated and reported. It would be useful for ex situ characterization to be carried out, and details of the techniques and experimental setups should be made public.
- This projected completed baseline validation steps over two current densities and four temperatures and gained a comprehensive understanding of water permeability and film thickness effects on water motion. The project is achieving a comprehensive understanding of platinum (Pt), including loading, roughness, structure, and oxide coverage. The researchers did nice work with understanding water in the gas diffusion layer.
- This project has made great progress so far. The excellent experimental data sets help with model development, which has been progressing well given the timeline of project. Some improvement (see slide 22) would be desirable in the future, but overall the project is on track. It is critically important to underline the importance of structure-property relationships as developed in such an excellent way by this project team.

Question 4: Collaboration and coordination with other institutions

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

- This project has good, productive collaborations.
- The partners appear to be playing their positions on the team well.
- This project has excellent collaborations with component suppliers, national laboratories, and universities.
- The material supplier coordination seems clear. It appears there are small interactions with the university subcontractors.
- The team consists of automotive fuel cell users, materials suppliers, academic modelers, and characterization experts.

Question 5: Proposed future work

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

- The future work plan is very consistent with project objectives and deliverables.
- The material set for auto competitive is not justified, especially the choice of 15 wt.% Pt/Vulcan and I/C = 0.7.

- It appears that the project will be completed on time and the proposed future work all looks feasible. Most issues may occur in the two-phase water model.
- There are a few details in the future work section. The work on thin catalyst layers has not been addressed to date. It is difficult to ascertain exactly what type of characterization is going to be done.
- A second data set will be verified and the model will be improved by incorporating more physical component behavior data and specific component models to address known issues.
- The future plan is basically OK. In order for more accurate predictions, other measurement techniques in catalyst layers should be studied. This can be helpful when the model cannot predict experimental data, because there might be other key parameters related to kinetics/mass transport that are not identified yet (e.g., ionomer coverage on Pt, announced by Nissan).

Project strengths:

- This project is organized, systematic, and executed with engineering rigor.
- This project has good in situ testing diagnostics and correlation to operation.
- This project has a beautiful synergy between experiment and model and an extremely strong team.
- The access to characterization tools and technical data set generation capabilities are strengths to this project. This will lead to the development of good building blocks for other work, once complete.
- The exceptional experimental measurement techniques, quantitative analysis of the correlation between measured effective properties, and overpotentials/I-V performance are all strengths of this project.

Project weaknesses:

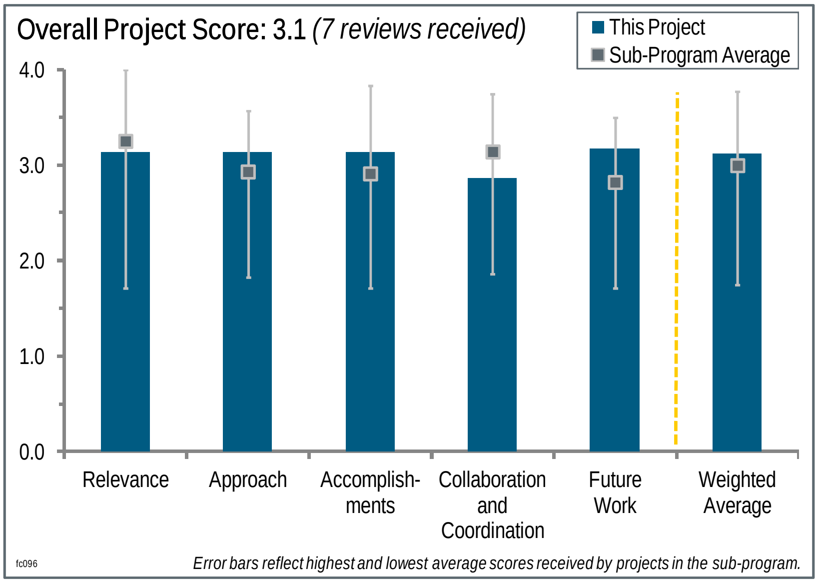
- This project currently has no weaknesses.
- The material set for auto competitive is not justified, especially the choice of 15 wt.% Pt/Vulcan and I/C = 0.7.
- Even at project end there will probably be some regimes of fuel cell operation that cannot be adequately explained by approach, but that is the nature of complex systems.
- There is no real linkage to the actual duty cycle cases, either stated or implied (i.e., there is not enough of a real-world spin on operational sensitivity, because most issues with low-cost MEAs are expected to occur at the extremes of the operation).
- The component characterization appears mainly in testing the transport properties in test stations. There is little in the way of information on how the surface structure affects the transport properties or correlation of fundamental properties to the transport, such as PSD. It is unclear how the photoacid dye information is being used, or what it means. There is a lot of terminology used in this presentation that is not defined, and it is difficult to determine what it is. It appears that the 1-d modeling case has a ways to go before the prediction of HFR is good.

Recommendations for additions/deletions to project scope:

- This project should include ex situ measurement system setups on the website.
- The researchers should develop the measurement techniques in the catalyst layer.
- The project team easily has the capability to extend the study to include durability/duty-cycle aspects that are vitally important to DOE's overall objectives. If this is not done in this project, it is important to link this to other groups where durability is a focus.
- Including thin catalyst layers, such as NSTF, and how the transport varies would be a valuable addition to this project. The graphs presented significant data, but there was not enough explanation and what was being measured was not completely understood (i.e., the multitudes of cases on various graphs). More detail is needed to make much of this information meaningful.

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

Quentin Ming; InnovaTek



Brief Summary of Project:

Objectives of this project include the following: (1) establish a design to meet the technical and operational need for distributed energy production from renewable fuels; (2) design, optimize, and integrate proprietary system components and balance of plant in a highly efficient design; and (3) demonstrate the technical and commercial potential of the technology for energy production, emissions reduction, and process economics.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.1** for its relevance to DOE objectives.

- This project fully supports DOE Hydrogen and Fuel Cells Program (the Program) and research, development, and deployment objectives.
- Biomass reforming integrated with a fuel cell is doubly relevant to the DOE Office of Energy Efficiency and Renewable Energy's mission. The applicability of the fuel processor technology to fuel cells other than solid oxide fuel cells (SOFCs) is not addressed.
- This project strongly supports the Program's goals and objectives with a focus on the use of non-petroleum renewable fuels such as biomass. There is an emphasis on meeting DOE performance and cost targets.
- This is a relevant project from the point of view of reforming hydrocarbons for use in fuel cells.
- The project appears to be addressing the issues of cost and durability. The project is not supporting the stated objective of shifting from fossil fuels to non-food biomass.
- The project is developing technology to produce electric power efficiently from domestic renewable resources and advancing fuel cell technology without relying on hydrogen (H_2) infrastructure.
- The project supports the Program's goal of achieving H_2 production from diverse domestic sources with distributed power demonstration by the second quarter of 2018. The project also supports fuel cell goals.

Question 2: Approach to performing the work

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

- The project focuses on critical issues and system demonstration.
- A systemic, logical approach involving design, review, and confirmation, as well as prototype testing, has been adopted for this project. It is heavily hardware oriented. The approach is very much results directed.
- This is primarily a straightforward engineering and design project to integrate a biomass fuel processor with a fuel cell. The individual subsystems already exist. No scientific breakthroughs are required. Modeling and simulation are effectively utilized. Process economics will be analyzed as part of the project. Transients are not being considered at this time.
- This is a rational engineering project that makes good use of DOE's resources. The focus on natural gas makes a lot of sense. Some pushback was registered during the meeting on this, which is unfortunate. Natural gas is

part of the President's all-of-the-above strategy, and it makes a lot of sense to consider it in the present circumstances.

- The approach to date has addressed cost well, and it is currently addressing performance. Durability has not yet been addressed significantly. The project team should use design for manufacturability much earlier in the design process so that the design starts with these principles in mind instead of trying to rework the design for lower cost later.
- The research is appropriately focused on the critical barriers of fuel processing catalysis, start-up transients, systems integration, and cost reduction. However, SOFCs are capable of internally reforming CH_4, and significant system efficiencies could be obtained if they optimized the fuel-processor-SOFC system for only partial external fuel reforming rather than a complete conversion to syngas.
- The overall approach of establishing a design to meet technical and operational needs for distributed energy production from renewable fuels should overcome the barrier of using SOFC power with renewable non-food biomass fuel. The technical objective is good: (1) develop a reformer that generates H_2 from non-food biofuels; (2) develop a highly efficient processing design of an integrated SOFC and fuel processor; and (3) prove the technology in a long-term field demonstration with a utility partner. The approach successfully used design reviews and third-party reviews in go/no-go decision points for manufacturing and assembly to lower costs. The design uses an efficient InnovGen fuel processor and Topsoe fuel cells. However, the fuel cell does not have an extensive testing record. The testing time of the 1.2 kW unit was not discussed.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- The project has achieved reasonable accomplishments, given the budget.
- The team has made excellent progress in system operation demonstration with biomass reformates.
- The project seems to be on schedule. The integrated system did indeed produce 1.2 kW grid power from biofuel and operated stably for about an hour, but no fuel cell performance data was presented.
- Progress has been good thus far, in the first third of the project. Technical milestones have been met. The initial, long-term testing of bio-kerosene reforming has been very successful, and the operation on biofuels producing power is impressive.
- The first-generation system appears to have proven itself out, but the real test will be with the second generation system, especially for durability. Process models were used to assess thermal stresses, which affect the durability, and operation, which affects the performance, of the InnovaTek reformer.
- The researchers are making good progress and have achieved milestones; however, the critical technical milestone (M4) to achieve 40% system efficiency is a few months after the Program's Annual Merit Review (July 2012), and as of now they have only achieved 27.5% (slide 31).
- The researchers' accomplishments are significant. They used simulation and modeling to successfully develop efficient and thermally integrated superior component/system designs with a microchannel heat exchanger and a fuel burner. They also developed an optimized catalyst for biofuel reforming, and fabricated and integrated proprietary system hardware, software, and catalysts. Additionally, they demonstrated 1.2 kW power from bio-kerosene and sent power to the grid for an undisclosed time.

Question 4: Collaboration and coordination with other institutions

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

- The project features collaborations with several companies and organizations.
- The team comprises a qualified entity for each of the subsystems.
- Collaborations have been excellent, including subcontractors and partners. The involvement and support of student interns from local universities is a very positive aspect of the project.
- The project received good input from the city of Richland. There should be more evidence that Topsoe is engaged.
- Although there are several external partners, it is not clear that InnovaTek is really collaborating with any of them.

- There are a number of good collaborations on fuel processing. However, it might have been preferable to collaborate with one of the few U.S. SOFC companies.
- The project features collaborations with Topsoe Fuel Cell; the city of Richland, Washington; Washington State University; and Pacific Northwest National Laboratory for biofuel energy technology services.

Question 5: Proposed future work

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

- Strengths of this project include its well-defined future work and work focus.
- The project features a logical, reasoned approach.
- Plans for future work are consistent with and supportive of the original project plan. Revision of the original goals and objective are not necessary, and the remaining technical and cost milestones are adequate to measure progress in the future.
- Analyzing the process economics is important, although the assumption of a 20-year life is questionable. For a continuous power system, this is four times longer than 40,000 hours of operation.
- The proposed future work is appropriate and should build on current performance by decreasing parasitics and improving integration efficiency.
- The planned future work is appropriate: pursue further technology improvements and system optimization, add additional fuel cell collaborators, perform analysis of process economics, and perform field demonstration and long-term operation.

Project strengths:

- This project’s strengths include its strong, motivated technical team and its supportive collaborators.
- The project’s strengths include its development of useful technology and focus on natural gas.
- InnovaTek has demonstrated integrated system performance that generates power.
- The plans and targets are well defined. Other strengths include the system approach and focus on addressing key issues.
- Strengths of the project include how it does not rely on an H_2 infrastructure to advance fuel cell technology, and the resultant efficiency gains.
- The project has strong collaborations and integration and system evaluation components.

Project weaknesses:

- There is little evidence of tight coupling with the fuel cell provider.
- The team should work with a U.S. SOFC manufacturer.
- The project appears to have abandoned fossil fuel displacement and renewable energy generation in favor of using relatively cheap natural gas as the fuel. It is not clear, though, whether InnovaTek’s natural-gas-fired system would be competitive with an internal combustion engine (ICE) combined heat and power system, much less utility-scale gas turbines.
- The team needs to focus more on integration of the two key components of the system—the reformer and the SOFC.
- It is unclear whether camelina is a sustainable supply of biodiesel. May be possible coking or plugging with a liquid fuel and the microtubular reactor, although an atomizer may solve this problem. The 67% fuel utilization is low and needs to be improved.

Recommendations for additions/deletions to project scope:

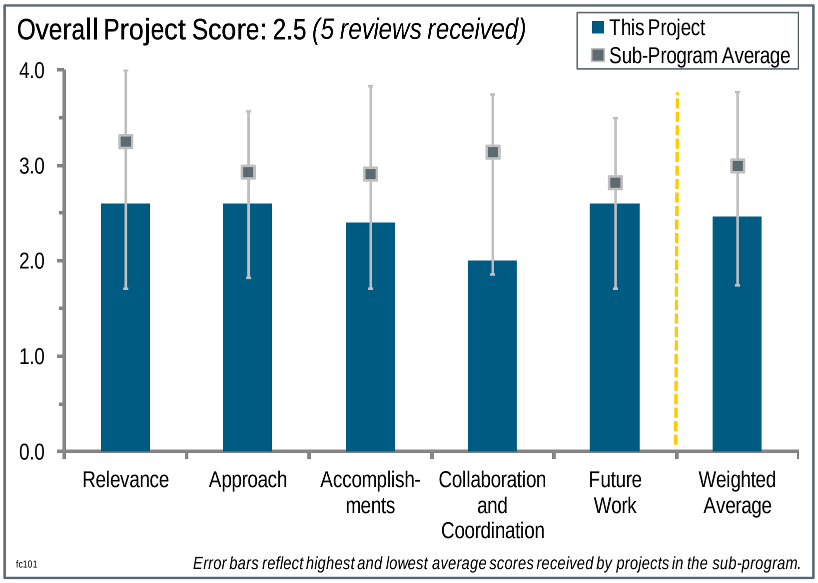
- Perhaps it would be interesting to consider internal reforming in place of the external reformer for some of the biofuels or for natural gas.
- The team should develop a better relationship with a fuel cell company.

- It would be good to know what market conditions are required for wider acceptance of a biofuel-fired distributed generator, and what the target lifetimes are of such systems, especially as compared to the expected lifetimes of ICE generators.
- The researchers should optimize the fuel-processor-SOFC system for only partial external fuel reforming, rather than complete conversion to syngas.
- The researchers need to use a Versa fuel cell. It has better performance and a better test record.

Project # FC-101: PEM Stationary Power Plant

Tom Skiba; UTC Power

Brief Summary of Project:
The project goals are to: (1) investigate the feasibility and value proposition of a 150 kW high temperature polymer electrolyte membrane (PEM) stationary fuel cell operating on natural gas reformate; (2) project durability and reliability of PEM fuel cell components; (3) conduct a preliminary systems analysis of a PEM power plant capable of achieving greater than 45% electrical efficiency; and (4) demonstrate an advanced fuel processing breadboard system capable of delivering a hydrogen - rich, low-CO (less than 10 ppm) reactant stream to the PEM stack.



Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **2.6** for its relevance to DOE objectives.

- Demonstrating a 150 kW PEM stationary power facility is highly relevant.
- This project is confusing. It seems very long (started in January 2004) and very expensive ($11.6 million from DOE) for the planned results (only a model).
- The project addresses the technical targets for 2017 listed in table 3.4.6 of the Fuel Cell Technologies Program's Multi-Year Research, Development and Demonstration Plan, and is therefore relevant to overall DOE objectives.
- It would be better if UTC Power could use their own resources to determine whether a 150 kW stationary PEM system is feasible and a good value proposition for the company. It is not clear that this project will have a significant benefit to the DOE Hydrogen and Fuel Cells Program.
- The appropriateness of this system is not completely convincing. UTC Power is already working on a very low-cost stack phosphoric acid fuel cell (PAFC), which is integrated with their systems. The market economics for PAFC systems heavily depend on their ability to generate useful (high-quality) waste heat. In this embodiment, the system cost "might" be marginally lower, because the stack is a lower-cost technology. However, cost may also increase, because this stack technology relies on more reformate cleanup. The system will not generate useful heat and any improvement in capital cost may be lost in lower revenue (no heat output). The market pull needs to be identified before jumping into such a system. The target market is unclear and there are numerous companies that developed PEM combined heat and power systems, only to find marginal sales (even when the product price was subsidized by shareholders and the government).

Question 2: Approach to performing the work

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

- This project seems like a reasonable approach, although not much is given about the details.
- The technical approach for this system design is impeccable and reflects UTC Power's ability to design and implement systems. The system is still in its early stages of development, and comments can only be made on the system process design.

- The approach in this project is mainly analysis, with some small single-cell tests planned. It seems that the industry has progressed well beyond this for stationary PEM systems, and evaluation of integrated hardware would be of much greater value.
- Investigating the feasibility and value proposition as a first step is a good approach; a go/no-go decision point. It is not clear that all of the milestones remaining can be reached in 2012, given the percentage complete as detailed on slide four. There is no integration with other efforts identified in the presentation and there are no technical collaborations identified on slide 21. While this may be due to intellectual property issues, use of the resources of other entities might help progress.
- The approach of using perfluorosulfonic acid (PFSA) membranes and trying to generate steam for the reformer from the stack is questionable. The energy penalty associated with compressing the low-pressure steam will be significant, and capital and maintenance cost for the steam compressor will drive up the cost of ownership. A better approach would be to use a membrane capable of operating significantly above 100°C, but this would probably have to be something other than PFSA.

Question 3: Accomplishments and progress towards overall project and DOE goals

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

- The system design has been performed, and technically the system is feasible.
- There do not appear to be a lot of accomplishments and progress for a project that began in 2004.
- The project started in 2004, and it appears the technical feasibility is the only milestone achieved in eight years. The progress has not been addressed in the brief, and it was difficult to address the accomplishments and progress. There were no timelines provided upon which to gauge progress.
- This project seems to have had many stop-starts throughout its history, and the continuity of results reflects that. The data and plots provided in the poster were pretty basic analyses and could be accomplished with much less investment than the cost of this program. Ignoring this for a moment, the results themselves do not give high confidence that the system proposed can meet the technical targets.
- The project has not been running very long since reinitiating. The attempt to project a membrane lifetime of 23,000 hours based on a few hundred hours of testing is not credible. The feasibility study seems to have led to dubious possible system designs (recompression of low-pressure steam).

Question 4: Collaboration and coordination with other institutions

This project was rated **2.0** for its collaboration and coordination.

- No partners were listed.
- There has been no collaboration reported.
- No collaborations or interactions are indicated.
- UTC Power might benefit from collaboration with partners that have expertise in compressors, but no collaborations are strictly needed for this project.
- As a system designer, UTC Power should be communicating and catering to its market. It is unclear who that is, and what the "voice of the customer" would ask for, in light of other UTC Power product offerings.

Question 5: Proposed future work

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

- The future work looks reasonable.
- The future work is appropriate, though it seems like a lot to get done between now and December 31, 2012.
- The work on desulfurization is valuable to the industry; however, the plan shown on slide 20 is unclear and lacks crisp, well-defined deliverables.
- There seems to be a lot more future work required than just developing an advanced fuel processing system. The project plan is not clearly defined; it looks like the researchers will need an extension.
- UTC Power would march forward and demonstrate the system; however, this reviewer's industrial experience indicates that UTC Power needs to step back and examine its target market and target system specifications.

Project strengths:

- This is a very technically and scientifically strong team.
- UTC Power has significant experience with the PEM fuel cell system.
- Developing a high-temperature PEM stationary system is a difficult challenge. A systematic approach to the various project elements is a good plan, although it is difficult to see this from the briefing.
- This project has no strengths.

Project weaknesses:

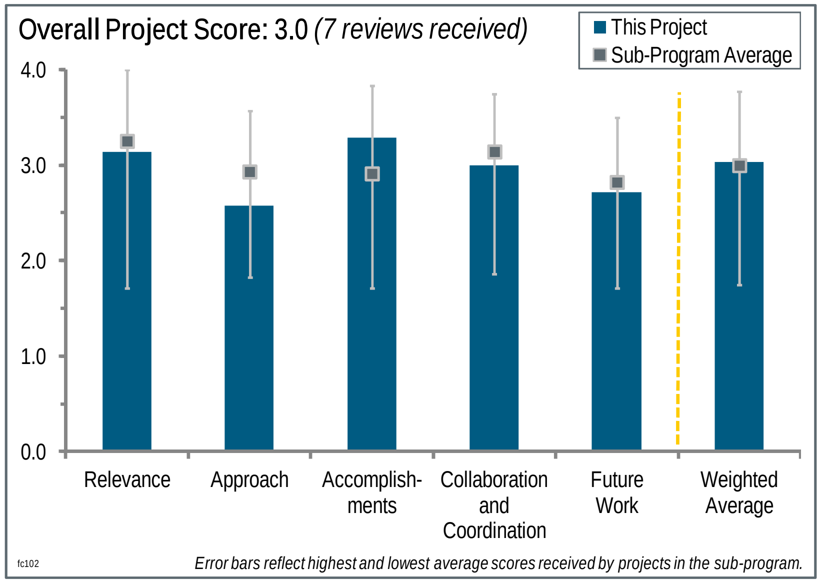
- This project needs market analysis (or needs to show existing market analysis).
- This project does not seem like it has accomplished a lot for the money that has been spent.
- This project has an unbalanced approach (too heavy on analysis, not enough testing), discontinuous history, unpromising results, and a low value-to-cost ratio.
- The poster did not convey this project's progress, so it is difficult to judge. The progress of this project appears poor, given the time and budget invested; the project started in 2004. It seems that technical feasibility is the only milestone achieved in eight years.
- This project is not exploring a new application space. PEM fuel cells have been around for so long, and it is unclear why UTC Power is deciding now to investigate this stationary application. It may have made more sense back in 2004 when the project originally started than it does now.

Recommendations for additions/deletions to project scope:

- Close this project as soon as practical.
- Because the project end date is December 31, 2012, it does not make sense to change the scope with so little time remaining. This project should not be extended beyond 2012.
- The researchers need to show what the market pull is for this type of system; how big the market is; and if the system makes sense, even if it accomplished $3,000/kW. It is unclear whether this system can offer something more than its big PAFC brother. If this system had a good market pull, then it seems unnecessary for DOE to provide funding for UTC Power to pursue it. UTC Power should spend more effort on this system with its own funding.

Project # FC-102: New High Performance Water Vapor Membranes to Improve Fuel Cell Balance of Plant Efficiency and Lower Costs

Earl Wagener; Tetramer Technologies, LLC



Brief Summary of Project:

Tetramer aims to design and develop high-performance, low-cost water vapor membranes for cathode humidification in fuel cells. Technical objectives are to: (1) demonstrate a water vapor transport membrane with >18,000 gas permeation units (GPUs), (2) develop a water vapor membrane with less than 20% loss in performance after the General Motors (GM) stress test, (3) limit the crossover leak rate to less than 150 GPU, (4) design temperature durability of 90°C to excursions of 100°C, and (5) limit the cost to less than $\$10/m^2$ at medium volumes.

Question 1: Relevance to overall U.S. Department of Energy (DOE) objectives

This project was rated **3.1** for its relevance to DOE objectives.

- Low-cost water vapor membranes to reduce the cost of the humidification system/fuel cell electric vehicle balance of plant fit well with the overall DOE objectives.
- The development of high-performance materials for humidifiers is a very important topic to be addressed to enable the introduction of fuel cells.
- The project is focused on one narrow aspect of fuel cells—namely, membranes to control hydration. While this is very important, it is not as broad as some of the other DOE-funded activities.
- The project will help to achieve better water stack management with existing materials. DOE is investing in new ionomers for hot and dry operation, so it is not clear that a humidifier will always be needed.
- Improved membranes for water recycling in polymer electrolyte membrane (PEM) fuel cells lead to cost and durability improvements. Current membranes exhibit adequate performance, but they are expensive. A lower-cost membrane for water management would provide a modest cost reduction.
- This work is pertinent to hydrogen (pure or reformate) and oxygen (from air) PEM fuel cells that need hydration, which is the PEM technology presently favored for automotive fuel cell power sources. This technology could help reduce the size and weight of this kind of fuel cell. This technology is not critical for high-temperature (HT) PEM fuel cells and solid oxide fuel cells.
- It is a bit unclear what the target truly is, and how it relates to DOE objectives. It is also unclear if the membrane is for HT application, humidifiers, or fuel cells. The project seems related to novel membranes with low cost.

Question 2: Approach to performing the work

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

- This work was a short, six-month Phase I, and under those constraints the principal investigator (PI) demonstrated a very efficient and focused effort to come up with a feasible membrane that shows great potential.

- It is difficult to evaluate the approach because it is proprietary. The reputation and experience of the researchers does foster a sense of confidence that they know what they are doing.
- It is hard to really describe the approach. Targets are clearly set, but how Tetramer Technologies is trying to achieve these targets is a mystery.
- It is not clear what criteria are used to drive material development. The approach seems to be exclusively experimental, based on trial and error. It is recommended to increase analytical support to experiments and to make it visible in the course of the next reporting period.
- This approach of using a water permeation membrane based on Henry's law and molecular void space is reasonable and desirable, as opposed to using inefficient and bulky mechanical humidifiers. It is hard to say that this approach is outstanding, because the authors could not share the chemistry of the membranes due to intellectual property issues. This work should be continued if the chemistry makes sense to "those in the know." The authors should file their patents or patent disclosures before next year so progress can be reviewed fairly and objectively.
- It is not clear whether allowing a huge amount of water through the membrane will help issues beyond water management. The materials presumably swell heavily and may have compromised mechanical integrity when wet.
- The approach seems fine, but without knowing anything about the membrane chemistry it is hard to judge the approach. The focus on gas permeation is a bit confusing with regard to whether it is a key metric or not. Also, it would be interesting to know about liquid water transport.

Question 3: Accomplishments and progress towards overall project and DOE goals

This project was rated **3.3** for its accomplishments and progress.

- The progress in the development of the work has been very good. Degradation rates need to be decreased; however, the candor of the PI was appreciated.
- These early results show achievement of critical flux rates and lifetime under accelerated protocols.
- Comparing the conductivity and IV performance of Tetramer Technologies membrane versus Nafion® 1000 is very competitive. The project is making steady progress toward gas permeation targets based on various generations. Durability, however, still has room for improvement.
- The team has made good progress in the area of performance. Significant challenges still have to be addressed in the area of durability. The presenter mentioned that seven different methods have been identified to reduce degradation, but no details were provided.
- This is new work, and the fact that the water permeation is better than in Nafion® for 200 hours is a good start. A key issue is to resolve the purification of materials, as the authors themselves stated, so a rational design of experiment can be done and discussed next time.
- The researchers have met the gas permeation goals. It is hard to comment on the accomplishments when the membrane chemistry/morphology is not revealed at all. The durability is very poor; it is good that Tetramer Technologies has identified several methods to mitigate the problem, but because these were not discussed, it is very hard to comment.
- The team accomplished good results in terms of permeation and conductivity. However, not all humidity ranges and temperatures were reported. The researchers seem to be reaching their milestones, although some more data about the experimental conditions is necessary.

Question 4: Collaboration and coordination with other institutions

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

- High-level partners are in place to build and evaluate prototype modules when the down-selected materials take place. At this time, no real interaction has occurred.
- The collaboration with GM is clear, and the roles of Ballard and Membrane Technology Research will become more evident in the following phases. In general, the team is well composed with two strong industry representatives.
- Most of the collaboration to date has been with GM. Phase II, if granted, would invoke additional partners. However, the GM collaboration has proved important in guiding the desired results.

- The collaboration appears to be validation testing from an original equipment manufacturer (OEM). Few details were provided.
- The authors are collaborating with GM and Ballard, which are leaders in this PEM technology and can guide the team and validate if the team is making significant impact.
- All collaborators are industrial—either component manufacturers or OEMs. It would be appropriate to include a national laboratory partner.
- For the size of the project, there is good collaboration, although independent testing of the membrane is expected.

Question 5: Proposed future work

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

- The proposed future work is hard to evaluate due to the proprietary nature of the project.
- Although more details on the future work would have been beneficial, the plan seems to address the major challenges. It is aimed at validating Phase I findings in real environments.
- The future work is dependent on a Phase II. Some issues to consider would include the ease of scaling up beyond the sheet casting that the PIs discussed, and developing a packing geometry appropriate for automotive applications.
- The authors are correct, and future work should include using a known structure to develop a new, better one, and scaling up for prototype testing. The researchers should include purification of a monomer for optimal synthesis (they said this in the body of the presentation), and also file patents so they can discuss the work in more technical detail.
- Scale-up of the films is highly desirable for testing at component suppliers and OEMs. It is still not clear whether the trade-off between performance and durability can be achieved when the films barely reach the performance target.
- The project plan seems good, but it is tough to say because gains in durability, etc. will depend on chemistry. No specifics were given about other characterization tasks.
- The proposed future work is: "Using structures developed in Phase I, fabricate membranes and optimize tradeoff in performance and durability. Scale up to provide partners with membranes for prototype testing." There is no way to gauge what that actually means.

Project strengths:

- The project's strength is the low cost of the membrane.
- Tetramer's strong partnership with industry members is an area of strength.
- The team's strong knowledge of separation membranes and familiarity with the chemistry allowed for quick progress to achieve the target water flux goals.
- The project's strength is the world-class polymer scientist leading the team.
- This is a strong academic and industrial team that has all of the resources to achieve its goals. The academic team is affiliated with Clemson University, a university with proven know-how for achieving new, high-quality membranes. The team should leverage the "Clemson connection" as much as possible.
- The novel polymer chemistry is an area of strength for this project.
- The supposed low-cost membrane that performs as well as Nafion® is a strength of this project.

Project weaknesses:

- It is unclear what the technology is about.
- The project lacks an analytical approach to accelerating the experimental findings. The roadmap to identify the trade-off between performance and durability is unclear.
- The highly proprietary nature of the project makes it very difficult to evaluate. There are no reports or publications to even assess the polymer chemistry used.

- It is not clear how to improve durability and optimize the membrane for permeability, because the composition and structure were not discussed. The researchers should file patent disclosures as soon as possible, or else they will be inhibited to discuss the work and will get limited help from the outside team.
- The approach is possibly too empirical and needs scientific input from a university or a national laboratory.
- The chemistry is not specified, so it is hard to say what is going to happen with durability, expected cost, etc. Some justifications are needed for the specific experiments chosen to be metrics for the membrane targets.

Recommendations for additions/deletions to project scope:

- The project scope is adequate.
- Humidifier housing can play a significant role in defining performance and durability. The team should investigate this aspect as part of the following phase.
- Recommendations include funding for Phase II and a focus on the packaging/scale-up.
- The researchers should include purification of a monomer for optimal synthesis (they said this in the body of the presentation), and also file patents so they can discuss the work in more technical detail.
- The team should make sure that all DOE membrane metrics are being met or examined, and test other transport properties under a whole range of conditions.