ERRATA SHEET

NREL REPORT/PROJECT NUMBER: BK-6A1-46600 DOE NUMBER: DOE/GO-102009-2920 TITLE: DOE Hydrogen Program: 2009 Annual Merit Review and Peer Evaluation Report AUTHOR(S): Satyapal, S. ORIGINAL PUBLICATION DATE: October 2009 DATE OF CORRECTIONS: May 2010

The following corrections were made to the Fuel Cells section of this report:

Page 386

The last paragraph was replaced.

Page 448

Christina Johnston was added to the list of authors.

Pages 597-598

Corrections to text in several bullet points were made, starting with Question 1. There were no changes to graph or project scores.

Fuel Cells Summary of Annual Merit Review Fuel Cells Subprogram

Summary of Reviewer Comments on Fuel Cells Subprogram:

Reviewers consider fuel cell development to be a critical enabling technology for the success of the Department's Hydrogen Program. Overall, the research and development portfolio was judged to be well managed, appropriately diverse, and focused on addressing technical barriers and meeting performance targets. Progress was considered remarkable. There was an undercurrent of concern about the announced program redirection and consequent effects on solid plans already in place and technical progress underway. As a result there was less confidence in future plans and prospects than in previous years. One portfolio gap appeared in several reviewer comments in the area of balance of plant (BOP) components, highlighted by the program's cost studies showing cost reduction in stack components making the BOP a significant contributor to system cost.

Fuel Cells Funding by Technology:

The Fuel Cells subprogram continues to concentrate on the critical path technology of stack components (membranes, catalysts/supports, bipolar plates/seals, water transport and management, and analysis/characterization). Additional projects address cross-cutting technologies and effects of impurities on fuel cell performance. Cost and durability of stack components continue to be a key focus of the subprogram.



Majority of Reviewer Comments and Recommendations:

This year 60 fuel cell projects were reviewed of the 63 projects presented. In general, the reviewer scores for the fuel cell projects span the range (1=poor, 2=fair, 3=good, 4=outstanding), with scores ranging from 3.6 to 1.6 for the highest and lowest scores, respectively. The average score of Fuel Cells subprogram projects was 2.9. The highest, lowest, and average scores were all lower than in the 2008

review values of 3.8, 1.9 and 3.0, respectively. The set of projects were reviewed by three to eight reviewers each, with an average over all 60 projects of six reviewers (posters had a lower average number of reviewers). Project scores reflect the technical progress made over the past year; relevance to the DOE Hydrogen Program; technical approach; extent of technology transfer; and proposed future plans. While reviewers tend to award those projects closer to commercial application with higher scores, their comments reveal that they also appreciate and support more fundamental work attacking key barriers to commercialization. Key recommendations and weaknesses are summarized below. DOE will respond to reviewer recommendations as appropriate for the scope and coherency of the overall fuel cell research effort.

Analysis/Characterization: The six projects in this category included two projects that tied for both the highest rank and second highest rank in the subprogram, with the overall category score well above average. These diverse projects were noted to strongly support the fuel cell program objectives and goals. The Oak Ridge National Laboratory microstructural characterization effort again ranked high, this year achieving the highest score in the subprogram tying with 3M and Delphi on other projects. Reviewers commend the advances in techniques and resolution, and continue to comment that correlating the microstructure of membrane electrode assemblies revealed in these images with performance data would increase the value of the effort. The National Institute of Standards and Technology neutron imaging project received the second highest score in the subprogram and demonstrated improvements in capability and resolution. The reviewers again encouraged the modelers in the fuel cell program to validate their models with real world data, and to work more closely with industry. Fuel cell manufacturers need to supply more experimental data to the modelers. The cost of an 80-kW automotive polymer electrolyte membrane (PEM) fuel cell system operating on direct hydrogen and projected to a manufacturing volume of 500,000 units per year continues to fall, and the cost studies help illuminate opportunities for cost improvement. The technical assistance to developers' activity at Los Alamos National Laboratory (LANL) is thought to provide value to the fuel cell community and was particularly noted for its collaborative approach.

Water Transport: Three water transport projects were reviewed with ratings from average to well above average. The LANL project was rated the highest, and the reviewers appreciated the scientific approach but felt that the project may be trying to cover too many phenomena, which limits depth. Reviewers recommended that both the modeling and materials effort of CFD Research Corporation and the visualization project at Rochester Institute of Technology work toward providing developers with models, recommendations, or insights that help advance fuel cell stack design and operation.

Impurities: The three impurities projects received above average scores, with the LANL effort achieving the second highest score of all the fuel cell projects. Reviewers consider these studies, on both the fuel and air sides of the fuel cell, to be very important to program success. The LANL project was lauded for improving understanding of the underlying degradation mechanisms and for suggesting/exploring mitigation strategies. The university projects were encouraged to quickly move to relevant contaminant levels and catalyst loadings, to increase modeling, and to address mitigation. Reviewers note the scope of the projects exceeds what can be reasonably accomplished with resources available. Although the researchers are sharing information and working on coordination, several reviewers recommended increased coordination to avoid duplication of effort.

Membranes: The sixteen membrane projects reviewed were ranked from well below average to well above average, with scores ranging from 1.8 to 3.4 and an average score of 2.7 against a subprogram overall average of 2.9. Giner's dimensionally stable membrane project and Vanderbilt's nanocapillary network proton conducting membrane projects received the second highest scores in the Fuel Cells subprogram, with FuelCell Energy's membrane project using humidification-independent cluster structures and 3M's project ranked just behind. The Vanderbilt project this year moved away from

sulfonated poly (arylene ether sulfone) nanocapillaries to perfluorosulfonic acid materials. The first three projects listed above met the DOE interim proton conductivity milestone at the 120°C, 50%RH conditions. As six projects pass the go/no-go milestone, some reviewers look forward to effort on development of compatible electrodes and fuel cell testing, while other reviewers find such work premature. In several projects, reviewers commented that membrane principal investigators would benefit from closer collaboration with fuel cell researchers and developers. A cost of production study of the most promising membranes was recommended for many of the projects. Reviewers provided specific recommendations to improve lower rated projects.

Catalysts/Supports: All but one of the five catalysts/supports projects reviewed received an overall rating above average, with two of the projects receiving the highest and second highest scores in the subprogram. The top-ranked 3M nanostructured thin film electrode project was particularly notable, exceeding three key DOE stack-level targets for 2015 on the single-cell level, on a mechanically-stabilized 3M membrane. The required total platinum group metal (PGM) content continues to fall as a result of subprogram research, and non-precious metal approaches show progress toward mass-activity targets. Reviewers continue to express concern about approaches that replace platinum with other PGMs. Some durability results from the non-precious metal catalyst projects are promising, but performance generally needs to be an order-of-magnitude higher before this durability matters. The reviewers commented that these efforts in alternative electrocatalysts, though high risk, represent a potential high pay-off option and should be supported in the future. Reviewers had mixed views on the conflict between improving fundamental understanding of a broad array of approaches with the desire to down select to a few candidate materials and focus effort as soon as possible. The only project in the category focused exclusively on supports scored below average, and reviewers offered a broad array of recommendations to improve the likelihood of success.

Recycling: The one remaining recycling project was evaluated and received an overall rating at the top of the range, achieving the second highest score in the subprogram. Reviewers consider PGM recovery an important aspect of the overall fuel cell life cycle, because it addresses both environmental issues and cost issues that impact the cost of fuel cell systems. BASF Catalysts has successfully completed the project, identifying and demonstrating the most efficient processes to recycle both catalyst-coated membranes and membrane electrode assemblies. Reviewers questioned the future impact of advanced catalyst forms, such as core-shell structures, and the economic impact of ultra-low PGM loadings, but generally considered BASF ready to commercialize PGM recovery whenever the market is ready.

Bipolar Plates/Seals: Two bipolar plate projects and one project on seals were reviewed in the Annual Merit Review and Peer Evaluation. These projects received average scores. Reviewers suggest increasing collaboration with stack developers and investigating remaining issues of concern, including: metal plate formability and joining; further reductions of processing temperature and cost; and the permeability, durability, and minimum thickness/formability of expanded graphite/resin plates. Reviewers believe more information on the seal materials developed must be provided if the UTC seal project is to be of use to the broader community. They also recommend permeability testing and testing under fuel cell cyclic conditions.

Water Management: Two projects in water management were reviewed. The Nuvera project received an above average score, and notably demonstrated that DOE cold-start targets can be met at the stack level. However, reviewers questioned the benefit to developers from the design and testing of a proprietary stack with limited data provided to the community, and they felt that the question of long-term durability of the stack under the start/stop protocol has not been adequately addressed. The Honeywell project received a score well below average, and reviewers questioned the value of testing of commercially available components. Reviewers noted that even if successful, the project is unlikely to improve the overall technological readiness of PEM fuel cell systems.

Distributed Energy: The ten distributed energy projects reviewed were generally scored below average; however, the Materials and Systems Research Solid Oxide Fuel Cell (SOFC) hybrid plant for the coproduction of electricity and hydrogen, and the UTC stationary PEM power plant verification project, scored notably above average. Distributed energy fuel cell applications are growing in program emphasis, and this year present a very diverse set of projects at widely varying levels of technical maturity and system development. Effort ranges from the fundamental to field testing of pre-commercial systems. Across the projects, reviewers noted both significant progress and significant problems and provided recommendations for the improvement of individual projects in approach, collaborations, and future planning.

Auxiliary, Off-Road, and Portable Power: Three auxiliary power projects, one off-road application project, and one portable power project were reviewed this year. The Delphi SOFC auxiliary power unit (APU) for heavy-duty trucks tied for the highest ranked project in the subprogram. Reviewers found the progress commendable, including two new on-vehicle demonstrations since the last review, and judged the project generally on track for success. The Cummins Power Generation SOFC heavy truck APU project was also rated above average, showing good progress, though the current design misses targets in several areas. The remaining projects in this category were generally rated below average. The IdaTech methanol-fueled PEM golf course maintenance vehicle received mixed reviews, with a lack of test data collection and analysis noted. The remaining projects, a solid acid fuel cell stack and a silicon-based SOFC for portable electronics, both demonstrated progress. However, they were judged to be relatively immature, and technical detail in the presentations was lacking.

Cross Cutting: The five cross-cutting projects reviewed this year generally scored well below average, and this category had the year's lowest ranked project from Microcell Corporation for commercial scaleup of a microfiber fuel cell, which was scored at 1.6 (below a rank of 2.0, or "fair"). The remaining projects in this category are all university-led, most of which tend to collect a number of disparate tasks spread across a number of investigators under a project umbrella. Although reviewers generally agree that the individual tasks address important issues in fuel cell development, reviewer comments indicate a lowlevel and diffuse effort on a broad number of technical areas does not constitute a well-defined, coordinated project likely to make progress toward targets.

Project # FC-01: Lead Research and Development Activity for DOE's High Temperature, Low Relative Humidity Membrane Program

James Fenton; University of Central Florida

Brief Summary of Project

The objectives of this project are to 1) investigate new polymeric electrolyte/phosphotungstic acid membranes; 2) develop standardized characterization methodologies, including conductivity, mechanical, mass transport, and surface properties of membranes; 3) provide the High Temperature Membrane Working Group (HTMWG) members with standardized methodologies; and 4) organize HTMWG biannual meetings. Fuel cell performance will be evaluated and the durability of membranes will be predicted. Membrane electrode assemblies (MEA) will be fabricated and evaluated.



Question 1: Relevance to overall DOE objectives

This project earned a score of 3.5 for its relevance to DOE objectives.

- This program supports the DOE R&D objectives.
- It was not clear whether this membrane development program was geared towards stationary or transportation applications. At 120°C and low relative humidity (RH), heat rejection and operation of the stack will be favorable for transportation applications with pure hydrogen. However, for stationary applications, the stack must operate at 40°-45°C higher than 120°C to avoid CO sensitivity, assuming they use natural gas (NG) or propane and a low-cost fuel processor.
- The project is critical to the Hydrogen Program and its need for membranes that can operate at higher temperatures under low humidity conditions. However, listing the electrolyte/phosphotungstic acid membrane development first in the list of objectives suggests a questionable set of priorities. Working with the HTMWG members is surely the first priority, as well as helping with standardized measurements or otherwise. This project has not lived up to its promise with regards to providing support for the HTMWG members and has shown that standardized measurements and biannual meetings are not sufficient to generate the necessary collaborative and collegial spirit to maximize the benefits of the HTMWG members' efforts. The phosphotungstic acid work is worthwhile, but not enough to merit dividing the team's attention. It is recommended that the team returns to the original proposal and revisits some of those ideas to generate more collaboration.
- The project goals are aligned with DOE targets and goals, and the project is relevant to DOE objectives.
- Tasks 3-7 are critical to facilitating the common conductivity testing of all the membranes in the HTMWG. Without this testing, it would be impossible to compare performance from one project to the next; furthermore, the tasks have addressed issues in conductivity testing and are intended to enhance it.
- Tasks 1 and 2 are dedicated to membrane development for fuel cells, which like all other membrane projects, is at least relevant.

Question 2: Approach to performing the research and development

This project was rated 2.7 on its approach.

- This program should be focused on meeting conductivity, mechanical properties, and chemical stability targets. Doing electrode development and evaluating fuel cell performance is premature in a materials-development program such as this.
- Given the scope of the project, it appears very well designed.
- The project needs to improve interactivity with other HTMWG members. The approach is a little too split between the project team's ideas on membranes and other's ideas. More assistance should be offered to other group members on more issues than just conductivity and MEA manufacture. For example, there seems to be little on mechanical properties, swellability, or gas cross-over. How are these measurements to be done? More leadership by others is necessary so that it occurs more frequently than just twice a year.
- The project team's work in the function of HTMWG lead is good. The approach to obtaining high temperature membranes led to an improvement by an approximate factor of two over Nafion®.
- The approach of adding heteropoly acids (HPAs) to perfluorosulfonic acid (PFSA) is basically reasonable, though more attention should be given to leaching prevention strategies. However, so far HPAs have not provided substantial improvement, especially at low RH, and it is not clear that the investigators have a good approach to further attacking the problem.
- The membrane development work lacks novelty. The literature contains many attempts to combine HPAs with PFSAs.
- The project does not show a convincing effort to defend against two of the most probable failure modes associated with HPAs: swelling and leaching of the HPA, with exception of the use of a reinforcing material.
- The project exhibits nice consistent effort to generate conductivity data with partner membranes, despite the fact that in-plane conductivity will mask anisotropy.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.7 based on accomplishments.

- Several of the partners have made good progress in meeting targets.
- It appeared that significant improvements have been made. Conductivity of membranes from all but one of the high-temperature membrane projects was already higher than Nafion®, with two of them already exceeding the target, and a third one being close to the target.
- Progress towards the HTMWG barriers as a whole is fair, but not compelling. This project's progress towards 50% RH is fair, but reaching this goal entirely seems rather problematic. Can a thinner membrane be used? The area specific resistance (ASR) is a good measure only if the gas crossover numbers are known. No real mention of this in the presentation.
- A technique was developed to measure HPA in the membrane and ensure it remains there during processing. Conductivity results are less than desired, but still exhibited an improvement by an approximate factor of two over Nafion®. The project team has developed thru-plane conductivity/ASR measurement with collaborators.
- The project exhibits some improvement over Nafion®, but is still not very close to the 120°C target and has no clear path to get there.
- None of the data clearly show that the addition of HPA increases the conductivity of membrane samples.
- No swelling data was reported.
- Some slides were missing important information, such as temperature.
- By this stage in the project, we should know something about the degree to which the HPAs leach from the Florida Solar Energy Center (FSEC) series membranes.
- The project team gave a good effort to produce a multi-port system for *in situ* testing.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.2** for technology transfer and collaboration.

- The project team collaborated well; however, more interaction with industry partners such as GM, Ford, and 3M would be useful.
- For a complex project with various stake holders like this, collaboration and coordination appeared to be at a very healthy level.

- Collaboration appears strong in the presentation, but really needs to be dramatically increased if the project is to be the lead project charged with coordinating the efforts.
- Overall, collaborations with membrane working group members appear to be good.
- This project has done an excellent job at staying consistent to its original intentions, especially while working with a large number of partners. Some may criticize the use of the in-plane method, and some may criticize the use of PFSA ionomers for all *in situ* testing, but the project has still managed to produce the results that it was assigned to do.

<u>Question 5: Approach to and relevance of proposed future research</u>

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

- As mentioned above, emphasis should be on conductivity and other material properties. Fuel cell testing at this early stage will not provide useful information for most of these new materials.
- The project team should add some sort of accelerated endurance testing for the membrane, as well as testing of the membrane as an MEA, to its future work plan.
- Future work plans appear to be confined to MEA preparation, without much fundamental basis for the proposed methods. Each polymer delivered from another group will need to be treated differently. This is not addressed.
- Redirection to stop membrane fabrication seems appropriate when test results are compared to milestone targets and other results in the high-temperature membrane projects. It is appropriate to begin planning for durability testing and integrating the new membrane working group project's membranes into MEAs.
- It seems unlikely that proposed future work, which mostly seems to involve varying processing and casting parameters, will result in sufficient improvement.
- The test system shown for running multiple fuel cells at one time appears promising.
- Future work on partner membranes was not clearly outlined; however, one slide implies that both mechanical and chemical durability tests will be pursued, which is good.
- The project should offer specific definition on ASR based on a specific protocol, and then seek the research community's feedback, rather than allow the community to define it.

Strengths and weaknesses

Strengths

- Several of the partners have made good progress in meeting targets.
- The team took a methodical approach.
- Standard procedures for conductivity are good.
- There was good collaboration between BekkTech, Scribner, and the University of Central Florida (UCF).
- The project team exhibited a strong ability to gather samples from the research community, perform needed testing, and fabricate test devices.
- The project team attempted to use reinforcement in membrane fabrication.

Weaknesses

- This program should be focused on meeting conductivity, mechanical properties, and chemical stability targets. Doing electrode development and evaluating fuel cell performance is premature in a materials development program such as this.
- The end user, or customer, was vaguely defined.
- The MEA standard procedure was not necessarily good. We need more research on what is important about polymer structures.
- Collaboration with other DOE workers in the membrane area seems to be weaker than this project requires, and leadership is not provided.
- Membrane development work has not increased high-temperature/low RH conductivity as much as desired.
- There was a lack of novelty in new membrane fabrication.
- There was a lack of initiative in addressing known membrane failure modes, such as swelling and leaching, within the opening stages of the project.
- The project team needs to more clearly define some future protocols.

- The project team needs to work with partners to understand and address material shortcomings to push as many new materials as far as possible. This will increase the likelihood of new and improved commercially viable membranes being developed.
- The plans should include fuel cell level testing to ensure that there are no interface level issues when these membranes are used in an MEA.
- The decision for UCF to stop membrane development work and concentrate on testing is appropriate.
- The Florida Solar Energy Center should eliminate development of its own membrane materials and focus on its other roles in the HTMWG.
- The project team should eliminate new membrane fabrication (which has been done).
- The project team should add emphasis on *in situ* testing of partner membranes and focus on the three ideal functions of the membrane: proton conduction, gas separation, and prevention of electrical shorting. This would create a wide temperature range before and after a stress test (e.g., open circuit voltage (OCV) or RH cycling). In other words, create a simple grid of measurements by which every membrane can be evaluated under *in situ* conditions.

Project # FC-02: Dimensionally Stable Membranes

Cortney K. Mittelsteadt, William Braff, Shelly VanBlarcom, and Han Lie; Giner Electochemical, LLC Fred Johnson and Israel Cabasso; SUNY-ESF

Brief Summary of Project

The ultimate goal of the project is to meet performance targets with film that can be generated in roll at DOE cost targets. The Year 2 milestones were achieved and interim conductivity targets have been met. Improvements in fuel cell performance have been shown, including electrodes. A realistic pathway for meeting cost targets can be seen for both paths. To reach the ultimate DOE goals, Giner needs to incorporate the low equivalent weight (EW) materials that have been developed at the State University of New York-Syracuse (SUNY).



<u>Ouestion 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of **3.6** for its relevance to DOE objectives.

- Dimensionally stable, high-temperature membranes are vitally important for fuel cell vehicle (FCV) commercialization. Whether or not FCV commercialization is still important to the DOE is another matter.
- High membrane conductivity over the range of operating conditions is critical to achievement of DOE targets. This project is focused on high-temperature and low relative humidity (RH).
- An important and essential aspect of the objectives of this effort is the high-volume supported membrane production capability development. This effort, together with the objective of meeting or exceeding the performance requirements, fully supports the DOE hydrogen R&D objectives.
- The project actually addresses the three primary barriers of durability, cost, and performance.
- Work is in line with Task 1, Barriers A, B, and C, to develop membranes that meet all targets.
- Membrane swelling remains an issue; hence the relevance of this task is undeniable. The composite membrane approach is a viable path to perhaps preventing membrane thinning, which is also observed in some failure modes.
- Improved conductivity, high-temperature, and low RH membranes are included in Hydrogen Program and DOE RD&D objectives. These are not the most critical material needs for enabling fuel cell commercialization.

Question 2: Approach to performing the research and development

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

- Unfortunately, Giner's unique laser etched reinforcement layer technology is not economically feasible due to an intrinsically high cost for the laser etching. Therefore, Giner is now working on commercially available support materials with ionomer from collaborators. It is unclear, with the 3DSM[™] support material, what inhouse technology Giner is bringing to this work.
- Using a mechanical support structure to allow lower EW ionomer has shown merit. The support structure prevents macroscopic swelling in the x-y plane.
- The performance of the 3-D stable membranes is impressive, particularly at 50% humidity, in this internally well-integrated project. The connection to other projects in the Hydrogen Program is less clear.
- Work is based on a previously established concept. The approach suggests a good chance for success. The 2DSMTM is a clever way to use less of the expensive ionomer and the better performing low equivalent weight

materials. The rationale of their approach seems logical and solid, and it is pleasing to see a possible alternative to the laser drilling.

- The approach is excellent, and the use of insoluble highly proton-conductive ionomers in a commercial 3D support should achieve the project goals.
- The approach appears very methodical, and it appears that testing the membrane in fuel cell embodiment and considering/factoring in production cost early on is a thoughtful approach.
- The project team employed a very strong approach using high-acid content ionomers and supported materials in which the mechanical properties are provided by the support. Down-select to a single approach, such as 3DSM, which should allow for more significant advances. Addition of Millipore as a team member supplying 3D significantly strengthens project.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.4** based on accomplishments.

- Proton conductivity measurements are showing progressively better values approaching the DOE target; however, the key to achieving the goals will solely lie with the ionomer producer, as mechanical minimums are not required by the DOE. In the case of 3DSM, support is provided from outside the company.
- Giner has met the Year-3 go/no-go target for high-temperature (120°C) and low-RH conductivity (50%).
- Data show the possibility of meeting the final target at 25% RH.
- So far, this program is one of the very few that has exceeded the DOE goal for conductivity at low RH.
- Fuel cell performance gains with the 2DSM are quite good with the 700 EW ionomer. Humidity cycling passes for all approaches and is giving a very good result. The new casting approach for the 2DSM is clever and encouraging. The project team should resolve the issue between its measurements and those of BekkTech.
- This project exceeded the go/no-go decision point target of 0.1 S/cm at 120°C and less than 50% RH.
- Technical progress appears consistent with expectations at this phase. The project team should make an effort to get rid of water and find a new mechanism for transport of protons.
- The project's progress over three years has been strong. In comparing this year's presentation to last year's, advances achieved in the last year seem a little limited focusing mainly on fuel cell performance and 3DSM advances. The fuel cell performance was deemphasized, and rightly so. The 3DSM results are interesting, show promise, and likely suffered from "learning pains;" but they show limited progress. The team should make more significant advances along these lines in the coming year. The results of the overall project are strong and show materials with significant property improvements.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.4** for technology transfer and collaboration.

- Based on the results, it appears interaction is high. This type of close collaboration is highly necessary, as both the ionomer and 3DSM comes from someone other than Giner.
- Giner knows its technical limitations and has partnered with appropriate organizations to fill in the gaps. He is working closely with GM and other partners including a porous support developer (Millipore) and a monomer synthesis organization (SUNY).
- Testing of SUNY polymers provides evidence of collaboration; however, the relation to GM was not clarified in the presentation.
- The program could benefit from collaboration with a larger film manufacturing company with large scale experience in film casting. This would provide not only experience, but also an understanding of cost and quality control issues.
- There have been collaborations with polymer chemists at SUNY and an industrial original equipment manufacturer (OEM) in GM.
- The amount of collaboration appears healthy but exhibits a strong dependency on SUNY for low EW materials.
- It was demonstrated that collaboration exists with SUNY. GM was mentioned several times during talk, and the addition of Millipore, a leader in porous membrane technology, significantly enhances the project.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- This program should focus on 2DSM with the funding of program DE-FG02-05ER84322. Giner's unique technology is 2DSM/laser etching, and finding a cost effective manufacturing process is very important. If 3DSM technology is used with outside ionomer, the unique sales point of Giner is lost.
- Future *in situ* fuel cell results will be beneficial in understanding the true potential of the material.
- A down-select to 2DSM or 3DSM should be done soon.
- Working on cost and manufacturability is appropriate and important.
- More effort on mechanical testing and RH cycling is needed.
- Future work was appropriate. Efforts should be devoted to enhancing performance at lower RH levels.
- It is not clear what the criteria will be to down-select between the 2DSM and the 3DSM approaches. What advantage does the project team expect with the 3DSM over the Gore membrane?
- The down-select approach to go forward will demonstrate cost feasibility, fuel cell performance, and RH cycling durability.
- I agree with the proposed future work, and propose additional thoughts on eliminating water as a medium for proton migration. Also, along with fuel cell performance, some endurance run is also essential to prove the feasibility of the down-select solution.
- Down-selecting to a single support membrane structure was presented clearly and with good rationale. Focus on high-acid content polymers continues to be appropriate. Advancing to even higher acid contents (by shorter side chains) was mentioned but not discussed as a topic of further study.

Strengths and weaknesses

Strengths

- Strengths include: 2DSM, strong collaborations, and a strong team at Giner.
- Identification of highly conductive membrane at low RH is a major accomplishment.
- The project team shows novel approaches with logical, easy to understand potential benefits.
- They seem to have achieved a practical solution for the required membrane performance.
- The project team has taken a methodical approach.
- This is one of the best high-temperature membrane projects. It has shown clear advances using relevant materials and methods, as well as provided increased understanding/insight of high temperature, low RH conduction.

Weaknesses

- Using 3DSM support materials and outside ionomer, it remains unclear to the reviewer what technology Giner brings to the table. Giner must focus on 2DSM with key ionomer producers.
- The project team needs an independent projection of cost, including the cost of the synthetic processes.
- The project team shows a lack of large scale manufacturing experience.
- I am still not convinced that the 2D laser-drilled support will ever be cost competitive or able to be mass produced. More information is needed here.
- Perhaps not a weakness of the project, but coming down on the cost curve appears to be a challenge.

- DE-FG02-05ER84322 should become fully funded so that Giner can fully focus 2DSM materials. Fuel cell experiments should shed more light on the potential of this material.
- The SUNY synthetic effort needs to be enhanced, so that larger quantities can be available for membrane production and in-cell evaluations.
- Cast and cure of the 2DSM approach should continue and perhaps be expanded to include a broader array of processing methods.

- The material should be evaluated at all temperatures relevant to the automotive industry, including temperatures as low as -20°C with a concentration on 95°C. That is, temperature and RH cycling down to -20°C and from 20%-100% RH should be done.
- None.
- Acid strength arguments for pursuing perfluorosulfonic acids (PFSA) are supported by data, but not proven. For an increased understanding of these systems, other conductivity data as a function of hydration number for related compounds such as methane sulfonic acid, perfluoro ethyl sulfonic acid, perfluoro propyl sulfonic acid, and perfluoro benzyl sulfonic acid, would be extremely interesting and insightful from a fundamental understanding point of view, and would provide enhanced justification of acid choice for ionomers.

Project # FC-03: New Polymer/Inorganic Proton Conductive Composite Membranes for PEMFC

Serguei Lvov, Mike Chung, Sridhar Komarneni, Zhicheng Zhang, Elena Chalkova, Chunmei Wang, Jeong Lee, Wentian Lin, Young Dong Noh, Yunchul Cho, Kanchan Grover, Joo Young Kim, and Mark Fedkin; Pennsylvania State University

Brief Summary of Project

The overall objectives of this project are to 1) contribute to DOE efforts developing high-temperature proton exchange membranes for transportation applications; and 2) develop a new composite membrane material with hydrophilic inorganic particles and vinylidene fluoride / chlorotrifluoroethylene (VDF/CTFE) polymer matrix to be used in proton exchange membrane fuel cells (PEMFC) at -20°-120°C and relative humidity (RH) of 25-50%. The main project objectives of the last year were to 1) develop a proton conductive inorganic/polymeric membrane for a PEMFC operating at elevated temperatures up to 120°C and significantly reduced RH and 2) increase durability with



improved fuel cell performance. Tasks for the last year included the 1) synthesis and characterization of functionalized polymeric materials suitable for the desirable composite membranes; 2) synthesis and characterization of inorganic proton conductive materials suitable for the desirable composite membranes; and 3) fabrication and characterization of composite membranes and membrane electrode assemblies for PEMFCs in automotive applications.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.3** for its relevance to DOE objectives.

- The project is relevant to DOE's 2010 and 2015 low RH/high-temperature membrane R&D objectives.
- The tasks are aligned with the low RH/high-temperature project goals.
- The intent of the project is relevant to DOE's Hydrogen Program.
- The development of new membranes is aligned with DOE R&D objectives.
- The project's goals are limited to immediate achievement of shorter-term milestones with what appears to be a dead-end for the really desirable milestones. This is a pity, as the project held the potential for some basic investigation into polymer-surface interactions that could have been generally applicable and useful to the entire High Temperature Membrane Working Group (HTMWG) program.
- The project's goals and targets are aligned with DOE program goals and objectives.
- The project relevance is high because it involves fuel cell membranes with high conductivity at high temperature and low RH.
- In as much as the project attempted to fabricate proton-conducting membranes for PEMFCs, it was entirely relevant to the DOE objectives. If it had succeeded, it could have been critical.

Question 2: Approach to performing the research and development

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

• The approach of synthesizing crosslinked membranes with a crosslinked hydrocarbon backbone, which is known to be unstable to free radical condition, was not appropriate for the intended fuel cell application.

- The sulfonated polyethylene (PE) polymers have formed a crosslinking group with benzylic hydrogen, which is very well known to be unstable under free radical conditions. The synthesis of such crosslinkers with benzylic hydrogen was not appropriate for the intended fuel cell application.
- Although neat phosphosilicate has the potential to meet low RH/high-temperature conductivity, since none of its diluted composite showed better performance with Nafion® binder, this indicates limitation of phosphosilicate use under DOE conditions.
- It is not clear how holding water more tightly will contribute to membrane conductivity. The researchers should provide experimental or theoretical evidence that these inorganic additives can provide enhanced conductivity. Inorganic materials that bind water tightly often do not contribute much to enhancing conductivity.
- Aliphatic HCs typically do not have adequate chemical stability for use in PEMFCs. Stability experiments need to be done to show if these materials will survive.
- The presentation does not demonstrate how fundamental principles are used to design the materials to achieve the goals. This seems very empirical which is not helpful to the overall program.
- Chemical stability of polyolefin has not been demonstrated in fuel cell conditions; PE background should be susceptible to peroxide attack.
- At the low wt% inorganic additive (lower volume percent), the membrane can only be acting as a water storage material; otherwise, a template for organizing the Nafion® will not offer an alternative conduction pathway as postulated, because there is not enough inorganic additive to conduct over any appreciable length before the conduction pathway is interrupted by Nafion®.
- The approach was not particularly creative. The materials to be examined, as neither the polymers nor the inorganic additives, represent a step-jump forward in potential fuel cell performance and durability. The organic and inorganic materials were not highly conductive.
- Adding inorganic proton conductors is a poor approach when the inorganic components themselves have conductivity far below the 120°C target. Increasing water retention with hydrophilic dopants is a more credible approach.
- The original intents of the project were to fabricate membranes with functional groups that were known to not be stable for PEMFC environments.
- The switch to polyolefins still leaves the prospect of instability to oxidizing/reducing environments.
- Phosphosilicates chosen to enhance proton conductivity do not demonstrate sufficient proton conductivity on their own at 120°C and 25% RH.
- No strategy was given regarding the stabilization of inorganic additives in the membrane.
- All measurements focused on 120°C, although many fuel cell applications require lower temperatures during operation.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 1.9 based on accomplishments.

- A considerable amount of work had been conducted towards the sample preparation and testing, but, due to the less effective approach, no success was realized.
- Under DOE's condition of 120°C, all the test samples showed performance as being the same or below Nafion® membrane.
- No conductivity was realized by improving the approach of phosphosilicate-based Nafion® composite.
- No progress has been made towards meeting the high-temperature membrane conductivity objective of the project.
- The materials produced in the program met the 30°C conductivity target, but showed no improvement at 120°C. The fuel cell testing is interesting, but an explanation should be offered as to why there is this difference. The "re-cast Nafion®" control had an unusually low open circuit voltage (OCV).
- Some reasonable technical progress was made but there is no sense of great insight into membrane structures, proton conduction mechanisms, or how the next set of milestones will be achieved. The project seems to have very limited goals, and hence has very limited achievements. Much more extensive insights could easily have been found in this project, and it is disappointing that the participants were so unadventurous.
- The project team has only achieved limited improvements in conductivity compared to Nafion® with dopants. Progress on the HC membranes being investigated has been slow, and they are not developed enough yet to

make membranes comparable to Nafion® in performance, so the overall idea of a composite HC membrane with inorganic testing has not been tested.

- The technical accomplishments were not impressive. Conductivities were no better or only slightly better than commercial Nafion®. The addition of inorganic particles resulted in a lower membrane conductivity. Fuel cell current-voltage data were presented. The improvement in fuel cell performance with the PI's new membranes could not be explained.
- None of the tested materials have had significantly better performance than Nafion®, and most have had worse performance.
- Proton conductivity targets were not met at 120°C.
- Proton conductivity targets were not met at lower temperatures for materials still discussed in this year's presentation.
- The combination of best inorganic additives (P-Si) with updated functionalized polymer (PE-based) was not completed.
- The lowest swelling in the project was for Nafion®/P-Si composites, which were not the intended membranes.
- No demonstration was made in which materials had the prospect of durability.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated 1.7 for technology transfer and collaboration.

- There is no industry, national laboratory, or other university involved in this project.
- Most of the work was only conducted by Pennsylvania State University (Penn State) groups.
- There is no indication of any technology transfer prospect from the present project.
- There appears to be no outside collaboration.
- Not much interaction is taking place with other institutions apart from the obligatory one with BekkTech. This is very disappointing, particularly as interactions with other groups could have helped this project live up to its true potential.
- Collaborations outside of Penn State are limited and appear to be only for membrane testing.
- Collaborations/partners were only with researchers from Penn State.
- The only collaboration involved in the project was the one that was enforced on all membrane projects (BekkTech).

<u>Question 5: Approach to and relevance of proposed future research</u>

This project was rated 1.7 for proposed future work.

- The reason is not clear for proposing the crosslinkable polyolephynes, when it is clear that this approach is not showing any sign of membrane conductivity improvement.
- Sulfonic acid functionalized inorganic additives could be a potential route of making membranes with higher conductivity, if they help in enhancement of the ionomer conductivity. The present data suggest the inorganic additives do not help in increasing the conductivity of Nafion® ionomer.
- Cross-linking is a good approach.
- The chemical stability of the HC polymer needs to be addressed first.
- The project is at an end, but suggested future work is totally uninspired.
- Given the lack of progress with the polyolefin materials tested to date and results from block copolymer systems in other work, it appears unlikely that these systems will be able to achieve high enough ion-exchange capacity (IEC)/low enough equivalent weight to meet the conductivity targets.
- Nothing new here: The project has been completed, so future work is irrelevant.
- Proposed plans are either very similar to approaches already taken by several others in the case of increased sulfonic group concentrations (so there would be no point in duplicating this effort), or not a significantly different from what they have already tried in the case of increasing water retention (so unlikely to lead to significant conductivity improvement).
- The PI indicates that P-Si would be combined with PE-based polymers if more time was given, which is good.
- It is entirely speculative as to whether future efforts would be able to generate sufficient proton conductivity, lower swelling, prevent leaching of inorganics, and prevent brittleness.

Strengths and weaknesses

Strengths

- This is a good academic project for helping students get introduced to the field of fuel cells.
- The program provides hands-on membrane synthesis and fuel cell testing opportunities to students.
- The program also covers diverse areas of polymer synthesis, material development, and electrochemical testing in a single project.
- The project has the capability of providing real insight in the fields of polymer-particle interactions and solidstate proton conduction.
- The fuel cell results are promising, although they cannot be explained and, thus, it appears to be difficult to move forward with new-generation materials.
- The project allows for the ability to survey a wide variety of chemistry.
- The project has moved away from prior membrane systems where swelling was excessive.
- The project conducts systematic elimination of inorganic candidates based on the inability to conduct higher than Nafion® at varied weight percents.

Weaknesses

- The approaches to enhance membrane conductivity are not effective.
- The material selection of conductivity enhancement to the ionomeric membranes under dry condition was not adequate.
- Benefits of different ionomeric binders, such as HC-based ionomers with phosphosilicate inorganic salts, should have been explored.
- Aliphatic HCs typically do not have adequate chemical stability for use in PEMFCs. Stability experiments need to be done to show if these materials will survive.
- There appears to be no outside collaboration.
- The project exhibits timid objectives, poor collaborations, no sense of vision, and is limited to meeting short-term goals.
- The weaknesses are the absence of new materials with dramatically improved properties, and the resulting low/disappointing conductivity data at high temperature/low RH. There was no additional membrane property data, such as gas crossover, water uptake, or mechanical properties.
- None of the approaches tried so far have yielded significant improvement, and plans for future work are unlikely to change this.
- The project is unable to begin with a functionalized polymer system that either had a well-understood prospect for durability, or that brought some novel means of water retention or proton conduction.
- No effort was given towards understanding whether additives could be immobilized.
- The overall approach was more Edisonian than it should be. Ultimately, there needs to be well-established reasons for choosing the chemistry of the membrane beyond the overall goal of dispersing proton-conducting additives within a polymer backbone.
- The swelling issue was addressed too late.

- The crosslinkable polymer possessing aromatic crosslinkers with benzylic hydrogen should be deleted.
- The team should explore new inorganic proton-conducting materials capable of enhancing proton conductivity to the ionomers under dry operational conditions.
- The stability of the inorganics towards hot water needs to be shown.
- The team needs to be more adventurous and show better direction.
- The stability of the polyethylene-derived materials in the fuel cell environment is questionable. The reviewer recommends testing some of these materials in a fuel cell environment, or *ex situ* in Fenton-type reagent before continuing this line of work.
- There needs to be a new direction and focus for this project, in order to attain the DOE target of 0.1 S/cm at 120°C and 50% RH.

- If the project were to move on, collaboration with an industrial partner would be a necessity. Such a partner would provide guidance on what swells, leaches, enables conductivity, etc.
- An industrial partner might also be able to give advice on crosslinking or membrane reinforcement.

Project # FC-04: Poly(cyclohexadiene)-Based Polymer Electrolyte Membranes for Fuel Cell Applications

Jimmy Mays and Suxiang Deng; University of Tennessee Mohammad Hassan and Kenneth Mauritz; University of Southern Mississippi

Brief Summary of Project

The objective of the project is to synthesize and characterize novel neat and inorganically modified fuel cell membranes based on poly(1,3-cyclohexadiene) (PCHD). To achieve this objective, a range of materials incorporating PCHD will be synthesized, derivatized, and characterized. Successful completion of this project will result in the development of novel and potentially inexpensive polymer electrolyte membranes engineered to have high conductivity at elevated temperatures and low relative humidity (RH).



Question 1: Relevance to overall DOE objectives

This project earned a score of 2.8 for its relevance to DOE objectives.

- Low-cost and high-temperature membranes are a vital technology needed for fuel cell vehicle (FCV) commercialization.
- Membranes that operate at high temperature and low RH are in line with DOE targets and objectives.
- This program is scheduled to end. Stated objectives support DOE objectives, but the work appears to not meet the go/no-go milestone. Also, in its synthesis activities, it is not geared to address the synthesis of actual membranes, but rather membranes that are intrinsically unstable.
- The project addresses the three most critical barriers of cost, performance, and stability.
- The development of low-cost membranes is critical for FCV commercialization.
- Work is in line with Task 1, Barriers A and B, to develop membranes that meet all targets.
- The project goals of fabricating and characterizing a high-performance polymer electrolyte membrane for high-temperature/low RH fuel cell operation are highly relevant to the DOE.
- Improved conductivity, high-temperature, and low RH membranes are included in the Hydrogen Program and DOE RD&D objectives. These are not the most critical material needs for enabling fuel cell commercialization.

Question 2: Approach to performing the research and development

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

- The PI knows that disulphide bonds and remaining chlorine atoms on cyclohexadiene are highly susceptible to hydrolysis (disulphine bonds) and platinum poisoning (chlorine), but chose to ignore this for the sake of "synthetic ease." Therefore, the reviewer does not understand the purpose of this research as it has neither reached the DOE's proton conductivity requirements, nor will it have the necessary durability. Furthermore, if it were to have succeeded, it is not apparent how the knowledge obtained would have been transferred to a more robust chemistry.
- The program is focused on the technical targets of increasing membrane conductivity over a wide operating range.
- Adding titania is questionable based on data presented on water uptake.
- Some mechanical properties are measures, but interpretation and relevance need to be clarified.

- The work is focused on model systems that have insufficient stability. The easiest synthetic path was chosen at a loss of relevance. Although the "right" crosslinking was mentioned, no argument was presented that the current work would be relevant to this.
- One concern is with the expected stability of the HC membranes from peroxide and free radical attack.
- Another issue is catalyst membrane integration when an HC membrane is used.
- The project seems focused on only one HC monomer. Are there others nearly as low cost that might work better?
- The approach of copolymerization with poly(ethylene glycol) (PEG) is a good idea for improving conductivity.
- The PI is applying relevant diagnostics such as water adsorption/desorption and dynamic mechanical analysis (DMA).
- The concepts being generated have extremely high swelling, and no viable path was presented to reduce swelling.
- Novel chemistry was based on polymerizing cyclohexadiene, and it is not clear how this would ever make an oxidative stable film. What advantages does this approach have over other HC technologies?
- The PI focused on a low-cost polymer for his membranes. The use of derivative forms poly (cyclohexadiene), however, is suspect; S-S bonds for crosslinking are prone to hydrolysis, and PEG is oxidatively unstable.
- The pursuit of polycyclohexadiene as an ionomer material is limited because the backbone will have poor durability in fuel cell environments. The addition of PEG as a block component and disulfur as a crosslinker have stability issues, but was at least addressed in as far as perfluoro PEG and other crosslinkers could be investigated. Still, the backbone polycyclohexadiene is unstable and the pursuit of these materials is unlikely to help achieve DOE targets particularly in the area of durability.
- The high water uptakes and stiff mechanical properties are unlikely to meet RH cycling requirements.
- The project focused too much on physical characterization of materials.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 1.9 based on accomplishments.

- The membranes did not meet DOE proton conductivity targets.
- The membranes will not meet durability needs due to known chemical weaknesses.
- Nafion® conductivity was matched, but neither the interim conductivity milestone of 30°C and 80% RH, nor the Year 3 decision point of 120°C and 50% RH, were met.
- Work proceeded along the "easiest" lines with loss of relevance. Even with the introduction of less appropriate PEG moieties, the performance did not meet the conductivity goals.
- No direct swelling data vs. water uptake have been shown yet.
- The membrane conductivity at 120°C is comparable to many other approaches.
- There is a concern as to why the polarization curves with titania in the Nafion® were so bad. This has been tried by others many times.
- The project team should be utilizing fuel cell testing resources sooner and more often for functional performance feedback and guidance.
- The conductivity has been significantly improved.
- Why does the addition of titania not reduce the ion exchange capacity (IEC)?
- The combination of high-swell and high-dry modulus is the worst case scenario for mechanical durability under RH cycling.
- Oxidative stability is a concern.
- New polymers were produced, but none of these met the performance metrics, and many were similar at best to Nafion® in conductivity under some conditions.
- Membranes were made and some characterization data was collected. Water swelling in some membranes was very high. Some results could not be explained (e.g., same water absorption isotherms at 25°C and 80°C). Some polymer components may not have the requisite chemical stability. No membrane attained the DOE target of 0.1 S/cm at 120°C and 50% RH.
- The project was unable to meet conductivity targets or significantly improve on Nafion®, and it has therefore been chosen for a no-go decision. The addition of titania as an additive or PEG as a block showed modest changes in properties, but nothing suggesting high-conductivity goals could be met with further effort.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **2.3** for technology transfer and collaboration.

- Collaboration between the University of Tennessee (UT) and the University of Southern Mississippi (USM) is high (even the talk was split between the two). Other than that, there were no other partners with a role. Perhaps an industrial partner would have been beneficial to the program.
- The team does not include commercial stack developers/integrators.
- The participation of ORNL and USM has been helpful.
- Work seems to proceed with little evidence of direct collaboration.
- It is not clear what the role of ORNL has been. It seems the project could really benefit from more fuel cell testing expertise.
- Jimmy and Ken have a nice collaboration, but it would be nice to see some industry input.
- This project involves two universities and a national lab, but no industrial partners neither an automotive original equipment manufacturer (OEM) nor a membrane electrode assembly (MEA) manufacturer.
- The primary collaboration was with Ken Mauritz at USM, who performed some membrane characterization experiments.
- The project team comprises two teams, one for synthesis and the other for characterization. While this is a logical combination, the need for and value of characterization provided in this project is limited.

Question 5: Approach to and relevance of proposed future research

This project was rated **2.1** for proposed future work.

- A fundamental shift in chemistry is needed before the occurrence of further optimization of the reaction conditions or optimization of the composition.
- The PIs need to remove chemically weak points from membranes as soon as possible; otherwise the project is not relevant to the program and should be shifted to Basic Energy Sciences.
- The DOE project ends in August 2009.
- The project needs chemical durability data, such as Fenton's test, the open circuit voltage (OCV) hold test, and mechanical durability data, such as RH cycling.
- More attention to cost is needed.
- It is not clear what will be new in the synthesis routes that will improve progress towards the DOE goals for high temperature.
- The top priority should be to reduce extremely high water uptake.
- The project team should focus on making thinner membranes (~25 um) to approach area specific resistance targets.
- The project will be terminated, and wrap-up activities look reasonable.
- Future work lacked specifics as to how the PI will reach the DOE conductivity target. The project is concluding, so future work is not relevant.
- The project is 99% complete, making this category largely irrelevant. The future work proposed suggests pursuing similar electrolytes for Li batteries. This makes some sense as similar materials like PEG or polyethylene oxide are used commonly in these applications.

Strengths and weaknesses

Strengths

- A strong synthetic PI and an excellent membrane morphology PI are working on this project.
- The project team is using relevant diagnostics such as water uptake and desorption as a function of temperature and RH.
- This is a novel polymer system.
- The polymer will probably be low cost, but it might not have the required properties and durability.
- The project team is also using chemistry that allows reasonable control of blocks.

Weaknesses

- The chemistry chosen at the beginning of this project makes it a dead-end, which will be of little use to the fuel cell community unless it is modified shortly.
- The project shows a lack of relevance.
- There has been no fuel cell testing expertise on the project to provide valuable functional performance properties.
- The membranes have an extremely poor combination of high swell and high modulus, with no significant plans given for reducing swell. Adding titania doesn't appear to help much.
- It never looked like it would work, and it didn't after the first three years.
- The chemical durability of the PI's membranes is questionable. The inability to fabricate a membrane with high proton conductivity is a project weakness.
- The primary weakness is the use of a backbone not inherently stable in fuel cell operation and membranes of conductivity properties that are unremarkable.

- The project is complete at this time. For future funding opportunities, chemistry building blocks must be changed and industrial partners should be brought in.
- The decision to terminate is appropriate.
- This reviewer concurs that the project will be terminated.
- This project is ending, but more fundamental membrane synthesis/fabrication work is recommended, with less emphasis on complicated/sophisticated characterizations (e.g., dielectric spectroscopy).
- For materials with poor chemical stability and questionable mechanical properties, these do not merit further investigation. This conclusion is also based on the observed properties reported and have been properly down-selected.

Project # FC-05: Advanced Materials for Proton Exchange Membranes

James E. McGrath and Donald G. Baird; Virginia Polytechnic Institute and State University

Brief Summary of Project

The objectives of this project are to 1) design, identify, and develop the knowledge base to enable proton exchange membrane films and related materials to be utilized in fuel cell applications, particularly for H₂/air systems at 100°-120°C/low relative humidity (RH); 2) nanophase separated hydrophilic-hydrophobic thermally stable multi-block copolymers; 3) correlate water diffusion coefficients with proton conductivity under partially hydrated conditions; and 4) relate thermodynamics of nanophase formation to ordered morphology and to conductivity, diffusivity, and novelmembrane self-assembly.



<u>**Ouestion 1: Relevance to overall DOE**</u> <u>objectives</u>

This project earned a score of **3.6** for its relevance to DOE objectives.

- The project is relevant to the objectives of DOE's Multi-Year R&D Plan.
- The initial activities were very much aligned to DOE's goal, but in the later part of the project, the focus became a bit distracted from the goal.
- Improvement of low RH membrane conductivity is critical to the success of DOE's Hydrogen research initiatives.
- This program supports the DOE R&D objectives.
- Work is in line with Task 1, Barriers A, B, and C, to develop membranes that meet all targets.
- The project goals align with DOE objectives, but the switch to polybenzimidazole (PBI)-phos acid block copolymers is not in line with objectives to be able to use these membranes in the presence of condensed water.
- The project goals of fabricating and characterizing a high-performance polymer electrolyte membrane (PEM) for high-temperature/low RH fuel cell operation are highly relevant to the DOE.
- The project aims to produce membranes that meet conductivity targets for proton exchange membrane fuel cell (PEMFC) commercialization. For that reason, every task of the project fits the DOE R&D objectives, and if successful, could be critical to the Hydrogen Program.

Question 2: Approach to performing the research and development

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

- The approach of making low equivalent weight (EW) HC based membranes with a high concentration of sulfonic sites is a good strategy for retaining membrane conductivity at low RH and high-temperature operational conditions.
- The approach of making nanophase separated sulfonic acid membranes to control membrane swelling and retain better conductivity is the right approach.
- The approach of surface fluorination of HC membranes to improve membrane compatibility with perfluorosulfonic acid (PFSA)-based electrodes is good.
- However, the approach of making PBI-biphenyl sulfone (BPS) copolymer does not fit with the goal of this project. Phosphoric acid-doped PBI can meet DOE's goal of low RH/high-temperature conductivity. So why bother making PBI-BPS copolymer, when it still needs acid doping? This approach is not good.

- The block co-polymer approach is very systematic and holds promise for meeting DOE goals.
- Obviously, applying the knowledge of the McGrath group to this problem allows one of the greatest minds in polymer science to systematically improve the science. Proton conductivity can be understood in terms of morphology and the other desirable properties of the film. Unfortunately, the target was only based on proton conductivity, and perhaps too much time was spent making the perfect PEM in the first three years.
- The approach has led to membranes that have good mechanical properties and durability, but less than desirable conductivity. The choice to pursue phosphoric acid doped membranes that will likely not be stable in contact with liquid water is problematic for automotive applications. Polymer processing work is a bit premature since polymers do not have desired conductivity.
- The approach of using biphenyl sulfone H form (BPSH)-type polymers is moderately innovative, but much of the project tasks appear to be logical extensions of the PI's prior research. There was no truly creative tasks and/or materials described in the PI's approach.
- Not enough attention has been paid to improving membrane conductivity, which should be focused on at this stage more than other membrane attributes.
- The approach has generally been the highlight of this project, since, in principle, the project should be able to generate a wealth of fundamental knowledge. A well-defined, multi-block copolymer with variable lengths of hydrophobic and hydrophilic blocks is an ideal system for realizing trends for conductivity and mechanical properties vs. variations in the two block lengths.
- The project begins with a polymer system where some rationale for expected durability does exist.
- In the absence of success with the original approach, other approaches were added (addition of PBI and fluorination processes) with dubious implications for cost and commercial acceptance.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.3 based on accomplishments.

- In terms of conductivity improvement, not much progress has been made towards the goal since last year.
- Except for Hydroquinone-Based Poly(arylene ether sulfone) (HQSH)-BPS, none of the membranes showed better membrane conductivity than Nafion®.
- However, under the DOE goal of 120°C and 50% RH, the conductivity of HQSH-BPS is very close to the Nafion® membrane, and significant conductivity improvement is needed to meet DOE's goal.
- This work gives insight into structure-property relationships for block copolymer ionomers. The demonstration of low XY swelling is important for durability in fuel cells. Conductivity targets were not met, but a systematic study of these systems should provide improved conductivity and valuable insight for future development work.
- Adequate proton conductivity was not demonstrated. Many other useful PEM attributes were achieved.
- Progress in increasing conductivity at low RH has been slow. Conductivity has not improved much over the course of the project. In general, these materials have conductivity which exhibits a higher dependence on RH than Nafion® conductivity. The team has not looked into the effect of the anisotropy of their membranes on conductivity.
- Many of the technical accomplishments were not connected to the overall goal of fabricating a fuel cell membrane with a conductivity of 0.1 S/cm at 120°C and 50% RH. Direct methanol fuel cell DMFC crossover and fuel cell performance data are not relevant. Similarly, it is not clear why acid doped BPSH-PBI blends and surface fluorination were studied. The effort was much too broad, without an organized plan to achieve the DOE conductivity target.
- Membranes have been developed that have reasonably good properties for fuel cell applications, as demonstrated by the fuel cell testing and durability testing; however, the conductivity just isn't where it needs to be.
- The ability to adjust both conductivity and swelling with block length was shown for multiblock copolymers.
- Multiblock copolymers did not meet conductivity targets at any temperature.
- A PBI-based system that demonstrates 80 mS/cm at 80°C without phosphoric acid (PA) is intriguing, but RH needs to be specified.
- Some idea of the cost implications for fluorination must be understood before conductivity/dimensional stability enhancements can be considered promising. As a fundamental study, however, this does offer some useful insight.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.1** for technology transfer and collaboration.

- There is good interaction with different industry, academics, and national lab groups.
- The demonstration of large quantity polymer synthesis at Akron Polymer Synthesis (APS) is relevant to the focus of the project.
- The overall team has done well with the intended research on high-temperature membrane development.
- Work with an industrial membrane/membrane electrode assembly (MEA) developer would benefit this program.
- The project team is collaborating with national labs and industry.
- Collaborations were listed, but the actual collaborative work was not clearly identified in the project presentation.
- Effective use of technical abilities of partners has been made, particularly with LANL.
- Akron appeared to be successfully leveraged for membrane fabrication.
- The difference between Arkema and Giner contributions was not entirely established in the slide materials.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.7 for proposed future work.

- Post-fluorination of random and block copolymers will not provide low RH conductivity of the membrane, and so they are not relevant to the goal of this project.
- High ion-exchange capacity (IEC) crosslinked polymers are known to be unstable under a free radical environment: therefore, crosslinking homo and multi-block copolymers may not be the right strategy to achieve a low RH/high-temperature membrane.
- The biphenyl system has potential for further gains in conductivity. The fluorination of polymers is an interesting approach for membrane stabilization.
- The wrap-up activities seem adequate.
- Future work covers a variety of areas. The reviewer suggests that future work focus on high IEC crosslinked materials.
- The listed future work was very general and non-specific. This project is ending, so "future work" may not be relevant.
- Future work plans are reasonable, and there is slight hope of reaching targets, but there is still a long way to go.
- Lowering EW of the multi-block copolymers in order to return to the original goals of the project is a worthwhile exercise.
- In general, the question of what controls conductivity at low RH (in particular for sulfonic acid PEMs) is not only good to address for this project, but could have been a great project unto itself if the DOE had begun three years ago. This is one of the fundamental questions that the DOE Hydrogen Program should answer.
- It would be interesting to see if future work would include non-PA PBI-based membranes.

Strengths and weaknesses

Strengths

- This is a new approach to solve the problem of low RH membrane conductivity.
- The synthetic method is novel, and it is a valuable contribution to the development of ionomer chemistry.
- The concept of nano-phase separation and correlating this fact to the membrane conductivity at low RH is a well-thought-out concept.
- This work gives insight into structure property relationships for block copolymer ionomers. Demonstration of low XY swelling is important for durability in fuel cells. Conductivity targets were not met, but systematic study of these systems should provide improved conductivity and valuable insight for future development work.
- This is one of the few projects to try and optimize all PEM properties.
- BPSH materials represent a class of polymers with some potentially useful/interesting properties.
- The PI exhibited enormous skill and expertise.

- The original approach allowed levers of control for membrane conductivity, dimensional stability, and mechanical properties.
- The original membrane concept had a previously identified rationale for stability.
- This incorporation of collaborations can competently produce membrane samples and perform testing.

Weaknesses

- The BPSH type HC ionomer seems to have limitation in membrane conductivity under low RH conditions.
- Anchoring BPSH to other base polymers, such as PBI, will not add any conductivity effect to the copolymer, unless the copolymer is doped with acid. This is not a viable approach and doesn't meet the goal of the project.
- Surface fluorination may help to improve the MEA interface, but it is not going to improve the membrane conductivity to the order of magnitude presently needed for BPSH-type polymers.
- It is not clear that this approach would ever have met the target, but a valiant effort was made nonetheless.
- Progress in increasing conductivity toward the target was low. The project lacked a focus on increasing the conductivity at high temperature and low RH.
- The obvious weakness is the PI's inability to attain the DOE's go/no-go conductivity target of 0.1 S/cm at 120°C and 50% RH.
- The project exhibits insufficient focus on strategies for improved conductivity.
- The original concept is unable to meet conductivity targets.
- The ideas generated after not reaching the conductivity targets might not be commercially accepted.
- The opportunity to fundamentally address low-RH conductivity for -SO₃H membranes was not taken.

- It may be necessary to incorporate proton-conductive and water-retaining inorganic salts, such as hetropolyacid or phosphosilicic acid, to make the BPSH composite a more viable approach to improve membrane conductivity under low RH.
- Further development of crosslinked materials may not help in attaining the goal of the project.
- This project should be considered for continuation if possible.
- If the focus remains dual-use (for automotive and other applications), it is recommended that the project team delete the work on phos-acid doped membranes. If stationary systems become more of a focus, the phos acid doped PBI-block copolymer systems may be worth investigating.
- N/A since this project is ending.
- The project has ended, but hopefully the investigators can continue with other funding sources. With a tighter focus on strategies to improve conductivity, there is promise for development of high-performance materials.
- If this project were to continue, instead of scrambling for "what works," it would be interesting to see further progress framed in terms of fundamentals. For example:
 - What is responsible for conductivity at low RH? adjust such elements as EW, block lengths, processing solvent, and morphological parameters;
 - If fluorination works, why? Then an industrial partner could attempt to answer whether a low cost means exists to realize it; and
 - Further efforts to demonstrate stability are always good.

Project # FC-06: Protic Salt Polymer Membranes: High-Temperature Water-Free Proton-Conducting Membranes

D. Gervasio, C.A. Angell, R. Marzke, and J. Yarger; Arizona State University W. Youngs; University of Akron

Brief Summary of Project

The objective of this project is to make proton-conducting solid polymer electrolyte membrane (PEM) materials having 1) high proton conductance at high temperature (up to 120°C); 2) effectively no co-transport of molecular species with proton; 3) reduction of fuel cell overvoltage; and 4) good mechanical strength and chemical stability. PEMs are being made based on "solvent free" protic ionic liquid concepts, which can be used to model membrane factors such as stability and conductivity, and to act as plasticizers in membranes. Acid and base moieties and polymers are varied to optimize properties in two kinds of PEMs. Proton conductivity will be characterized by electrochemical impedance spectroscopy from -20° to 120° C.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.1** for its relevance to DOE objectives.

- High-temperature membranes, which do not rely on water and function at all temperatures, would be an amazing breakthrough for the fuel cell community especially for vehicles.
- The relevance of this project is high. Conductivity at low relative humidity (RH) and high temperature has been one of DOE's key technical targets.
- The project is highly exploratory in nature, and although the objectives are stated in accordance with DOE objectives, the actual work is not proceeding along those lines.
- The project addresses two of the major barriers: durability and performance.
- Developing a PEM that does not rely on water for proton transport would enable higher temperature operation and a potentially simpler water management system.
- This project is very relevant. It works on removing water as the limitation of membrane performance. This is a critical barrier for use of fuel cells in vehicles. The observation of better catalytic activity is also important. The project is aimed at very relevant objectives; however, in the execution, the relevance sometimes seems to be lost. It would be best to simply focus on the method of proton conductivity first, and then make the appropriate membranes. Some of the materials chosen are unlikely to be stable, so the relevance of choice of materials might be questionable. This does not matter if the basic mechanism of proton conduction is elucidated.
- The reviewer has some doubts about the project relevance. The reviewer thought the project goals were to fabricate and characterize a high-performance polymer electrolyte membrane for high-temperature/low RH fuel cell operation. The PI wrote that his goal was to fabricate an anhydrous proton-conducting electrolyte membrane. Such a membrane would operate at a very high temperature. It is unclear whether this is what DOE wants. The reviewer questions this goal because a fuel cell operating temperature of 275°C is outside the targets set in the DOE Multi-Year R&D Plan.
- Membranes with improved conductivity that function under high-temperature/low-RH conditions are included in the Hydrogen Program's and DOE's RD&D objectives. These are not the most critical material needs for enabling fuel cell commercialization.

Question 2: Approach to performing the research and development

This project was rated **1.9** on its approach.

- While the concept is an excellent one, the PI may not be pursuing this in the best way. The first step should have been to screen and pick the best protonic salt based on proton conductivity. The second step should have been immobilizing the best salt to prevent water solubility, while maintaining the proton conductivity. The third step then should have been the fuel cell experiments. Too much time was spent on the third step and not enough on the first two.
- The original ionic liquid formulations did not meet DOE targets.
- The new indium tin phosphate (ITP) approach shows high conductivity, but crossover and mechanical strength of the pure ITP may be issues. Blending with poly-vinyl pyridinium phosphate (PVPP) may reduce crossover, but conductivity may also be reduced.
- It is unclear what membrane/electrode interface issues will arise with an ITP membrane, or how they will be addressed.
- It is interesting to look at ITP materials and composites, but the conductivity of these compounds appears to be substantially below what is needed. There appears to be no clear path for improvement, so the team should, instead, switch to some other as yet unidentified compounds.
- The concept of a non-aqueous proton conductor is good, but the approach seems to be too diverse and unfocused. At such an early conceptual stage, it might be more productive to take a model system and study it in depth to better understand the fundamental prospects and limitations. This should be combined with bonafide material property and even fuel cell functional property evaluation to prove feasibility and identify the most significant problem areas.
- Protic ionic liquids (pILs) have demonstrated the ability to conduct protons.
- There has been too much focus on fuel cell testing, and not enough focus on proving proton conduction in actual films.
- ITP is not recommended because of H₃PO₄ release and likely severe performance loss over time.
- The approach is okay, but seems to be unfocused in many places and trying to do too much at once. The ITP work seems like a diversion. Showing such apparently poor polarization curves is not very helpful at this time. The project team should concentrate on the proton conduction mechanism first, and then add the other issues.
- The use of pILs is an interesting approach. Unfortunately, the PI was not able to achieve the DOE conductivity target of 0.1 S/cm at 120°C and 50% RH with such materials. It was not clear why the PI switched to the use of ITP. While a purely inorganic film (very thick) met the DOE conductivity target, an ITP/polymer composite exhibited a low conductivity (0.01 S/cm). The reported fuel cell performance of the ITP membrane electrode assembly (MEA) was very poor. The focus of looking for membranes that operate without water at 275°C does not seem to be appropriate for this DOE program.
- The project has significant weaknesses, but may have some impact on overcoming barriers. The investigation of pILs is of interest from a fundamental proton transport standpoint, but is unlikely to yield materials suitable for commercial applications. The addition of ITP adds a material that shows significantly more promise, but these studies do not build on previous studies, and seem like a subset of results already reported by Hibino and coworkers.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.0 based on accomplishments.

- Decent progress was made in finding a protonic salt and immobilizing it with PVPP; however, more work should have gone into further optimization of this system with *ex situ* testing such as PC and water soak testing, and not fuel cell testing.
- IV Curves with an OCV of 0.8V and 30 mA/cm² at 0.4V does not help the fuel cell community, nor does it help develop this material from a basic membrane understanding perspective.
- The original membrane technology did not meet the DOE targets.
- Target conductivity has been realized with pure ITP, but ITP issues are not insignificant.
- It would appear that the current emphasis on doped tin metaphosphates may not be effective at room temperatures. It is also not clear how ionic liquids could lead to an appropriate fuel cell membrane. This work

appears to be an extension of some other work by some of the investigators on ionic liquids. The relevance to the Hydrogen Program is unclear.

- There is a fundamental problem of some kind preventing their oxygen reduction reaction (ORR) with the protic salt-based electrolyte. The cyclic voltammogram on slide 13 may imply a fundamental issue with catalyst poisoning.
- Fuel cell performance examples are low by up to two orders of magnitude. Something is very wrong, and the PI should get help to understand if this is just their inexperience or a fundamental limitation with the technology.
- The claim on slide 6 to have made a neat ITP membrane with conductivity that exceeds the DOE targets by such a large amount, at 120°C and totally dry, is remarkable. The presentation should have focused on this, and then tried to explain why fuel cell performance on slide 29 is so terribly bad. Is the water generated in the fuel cell hurting the electrolyte?
- The fuel cell tests showed extremely poor results.
- The liquid electrolyte conductivities do not meet DOE targets, bringing to question why immobilized ILs have any chance to meet targets.
- ITP results must be verified and proved stable.
- Good progress has been made in making polymers. There are concerns about stability, particularly with regards to the pyridinium ions which are known to be easily oxidized; however, the structural effects on proton conduction mechanisms should be useful.
- The DOE conductivity target at 120°C and 50% RH was not met. Too much of the work focused on nonmembrane work, e.g., cyclic voltammetry studies of ORR, hydrogen pump experiments, and studies of liquid electrolytes. For non-leachable membranes, the proton conductivity was quite low (<10⁻² S/cm, as shown in slide 24).
- There are several scientifically interesting results presented. In particular, the high open circuit voltage (OCV) demonstrated for some liquid studies and voltammetry showing currents at high potentials, although these are not represented in any of the polarization curves presented.
- The results for liquid systems show poor performance and no clear path for stable, realizable systems.
- The ITP-based systems show similar results compared to those shown by Hibino with no clear insight/rationale as to how or why further improvements to this system should be expected by this team's work.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **2.0** for technology transfer and collaboration.

- Some collaboration exists with the University of Akron (UA), but the reviewer does not fully understand their role in the project.
- Collaborations do not include stack developers or integrators.
- Collaboration was not addressed in this presentation, but may be presumed to have occurred.
- It is not clear what the contributions of the other, limited collaborators were.
- There is a collaboration with UA, but the project could benefit with industry collaboration and support from someone with MEA preparation experience.
- The collaborations are okay, but could be better. It is not clear who is doing what or what kind of feedback is coming from the Florida Solar Energy Center (FSEC) and partners.
- The collaborative work appears to be minimal.
- There is some collaboration. This topic is not particularly relevant for grading this project.

Question 5: Approach to and relevance of proposed future research

This project was rated 1.5 for proposed future work.

- Future work should focus on the fundamentals of this material and really understanding the proton mechanism and its potential in an idealized form. An outside modeling group could potentially be involved to find out.
- This group should not conduct fuel cell testing until acceptable proton conductivity and water non-solubility are achieved.
- Future work should focus on ITP and blends.

- Much of the future work seems to continue study of the failed ionic liquid systems.
- The work is highly and broadly explorative. Program reformulation is required to focus on materials that can actually serve as fuel cell membranes. In view of the poor cell performance of just the ionic liquids, it is not clear why the work merits further effort. A better case needs to be made.
- The project should focus more on why the electrode kinetics are so bad. It is a much bigger issue than the membrane conductivity, and it needs to be understood before more effort is put into the membrane alone. The membrane has to be integrated with the catalyst and other components to work effectively, but when the combined system is one to two orders of magnitude below expected performance, it is not simply a matter of tweaking the MEA making. Something more fundamental may be at play that needs to be understood.
- There has been too much focus on fuel cell testing.
- The decision to focus on non-leachable PEMs is good.
- Before too much work is done on ITP, the PI should be absolutely sure that it is not releasing phosphoric acid.
- The future plans are very unclear. It is unclear where this project is going. The ITP work is most confusing.
- The future work is suspect. pILs do not seem to work. ITP is not a viable option when mixed with a polymer binder (conductivity is too low). The PI should not focus on anhydrous fuel cell operation at a very high temperature. It seems that the PI has abandoned the DOE target of 0.1 S/cm at 120°C and 50% RH.
- The proposed future work seems to hinge critically on polyphosphazene development, poly vinyl pyridine (PVP) and PVPP as binders with ITP. These aspects were not well explained during the presentation, and are somewhat confusing from the presentation text. It seems that the motivation is based on an assumed interaction between ITP and these polymers with no presented basis. ITP is not an ionic liquid it is a solid, which is key in the way it behaves in terms of its conduction process. If the polymer was co-synthesized, or the ITP particles dissolved, the positive properties observed would likely be destroyed. If the ITP particles remained intact, the polymer would serve only as a binder and would not be expected to result in positive effects.

Strengths and weaknesses

Strengths

- The concept of immobilized protonic salts is a great concept and should be funded by DOE.
- The project team took an imaginative approach.
- The concept is high risk, but with a potentially high payoff.
- It is one of the few projects that focus on anhydrous proton conductors.
- The project exhibits very relevant goals and objectives. The project addresses issues that are of fundamental importance to the program.
- The PI seems to be well versed with a variety of analytical electrochemical techniques.
- The team is investigating non-typical materials with interesting properties.

Weaknesses

- The approach is weak, with too much emphasis on fuel cell data.
- Poor materials performance suggests that the approach is not going to lead to the desired results.
- The team does not have enough experience with whole MEA performance requirements.
- No fundamental work has been done to suggest that pILs have the potential to meet DOE proton transport resistance targets.
- The project team is too scattered; it is trying to do too much. It does not help the cause to show polarization curves that are not good without adequate explanation of why they are so poor.
- The PI appears to be focusing on H₂/O₂ fuel cell operation under anhydrous conditions at 275°C, but the DOE fuel cell operating conditions were 120°C and 50% RH.
- A lack of scientific basis for the guidance of future work is a major weakness. The poor OCV of the ITP systems and the liquid nature of the pIL systems result in materials that are further weaknesses.

- The project team should add a modeling group to understand the full potential of this material.
- The project team should spend more time on the fundamental side and material development.
- The project team should stop doing fuel cell experiments.

- So far, the results do not support continued work on the ITP blend materials.
- The project team should focus on establishing a solid, credible benchmark performance with the ITP system, and go from there.
- There should not be any more fuel cell testing until stable membranes are prepared with conductivity within 1/10 of DOE target conductivity.
- The project team should concentrate on making solid polymer electrolytes that have the required conductivity and forget about the rest for the moment. Any changes in the structures should be designed to elucidate the mechanisms of conduction.
- The PI did not present any evidence that the use of tethered acid and base groups will yield a membrane with the requisite proton conductivity at 120°C and 50% RH. The PI needs to provide a clear and convincing path to achieve high conductivity. At the present time, it is questionable if this project will yield useful fuel cell membranes.
- The addition of ITP adds a relevant material to the study; however, these results are essentially completely independent of prior reported pIL studies and do not show signs of advancing beyond other reported studies on similar systems (Hibino). Arguments can be made for this project as meeting DOE conductivity targets based on conductivities of phosphoric acid, but it reflects a poor metric for the go/no-go decision on these projects as this project should have been held to a higher standard.

Project # FC-07: Fluoroalkyl-Phosphonic-Acid-Based Proton Conductors

Stephen Creager and Darryl DesMarteau; Clemson University Oleg Borodin, Grant Smith, and Gregory Voth; University of Utah

Brief Summary of Project

The overall objective for this project is to provide new electrolyte materials for use in next-generation hydrogen fuel cell-powered sources, especially for automotive transportation applications. The specific objectives of this project are to 1) synthesize and characterize new proton-conducting electrolytes based on the fluoroalkylphosphonic acid (FPA) functional group; and 2) create and apply new computer models to study proton conduction in FPAbased electrolytes.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.4** for its relevance to DOE objectives.



- The project supports DOE's Hydrogen Program.
- All the aspects of this project are aligned to the hydrogen vision and DOE RD&D objectives.
- This program supports the DOE R&D objectives.
- The relevance of the project is undeniable, but it may be time to do a risk assessment.
- The program addresses important questions regarding proton-conduction mechanisms and polymer structures. The combination of synthesis and modeling is very powerful and appropriate for addressing the objectives of the Hydrogen Program.
- The project addresses appropriate DOE barriers. Membranes that operate under hotter and dryer conditions are critical to fuel cell vehicle commercialization.
- The project intends to develop a new family of proton-conducting membranes for proton exchange membrane fuel cell (PEMFC) applications. As such, the project is entirely relevant to DOE R&D objectives and maintains the possibility of becoming important.

Question 2: Approach to performing the research and development

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

- The approach of this project is very valid.
- The project is related to the common belief that the phosphonic acid polymers may have better low relative humidity (RH) conductivity than sulfonic counterpart as proton transport occurs without any assistance of water.
- The approach of studying the model compound is a good way to predict the behavior of phosphonic acid fluoropolymers.
- The use of phosphonic acids in fuel cell membranes has been proposed many times. This is a well designed, systematic study which can show if this is a promising path.
- The approach appears systematic, but conductivity targets are not being met. There may be too much emphasis on the computer models. It is unclear why the PBI approach is not being pursued.
- The approach is okay, but perhaps light on the synthesis and measurement part. The modelers seem to be far ahead of the ability to make and measure the materials. The lack of investigation of bases other than the amphoteric phospohorus groups and water is curious. One would think it probable that the modelers could

easily throw in some heterocylic bases to the mix to see what effect would be expected. The program seems a bit inflexible.

- The approach is focused on trying to obtain conductivity under hotter and drier conditions. Modeling is useful, but not developed enough to provide results to guide the synthesis effort yet.
- The general approach of employing perfluorophosphonic acid membranes, supported by computational studies and examination of model compounds, is sound.
- The beginning approach of this project was simple and strong, i.e. replace the sulfonic acid group in a perfluorosulfonic acid (PFSA) membrane with a phosphonic acid at the termination point, to see whether there are advantages for conductivity, particularly as they relate to enhanced proton hopping.
- The project has sought to verify directions for enhanced conductivity through both molecular dynamics modeling and model compound studies.
- While the model compound studies do compare phosphonic/phosphinic species to sulfonic, the molecular dynamics studies unfortunately do not.

<u>Ouestion 3: Technical accomplishments and progress toward project and DOE goals</u></u>

This project was rated **2.3** based on accomplishments.

- So far, the progress made has not been focused towards the DOE goal of low RH membrane.
- Very little progress was made for the development of synthetic pathways for phophonic acid bearing fluoropolymers.
- The conductivities of the polymers are the same as the Nafion® membrane. So far, no improvement of the phosphonic acid approach has been demonstrated.
- While conductivity targets were not met, progress was made in preparing lower equivalent weight (EW) samples and understanding the ionomer morphology.
- It appears to be a high-risk project, as the conductivity target was not met. The membrane became brittle and failed during conductivity testing at BekkTech.
- Much progress has been made in synthesis and modeling. The results are useful in the study of proton transfer mechanisms. There seems to have been some disconnect with the Topic 2 program at the Florida Solar Energy Center (FSEC).
- The team has prepared new phosphinic and phosphonic acid derived membranes. Conductivity of these materials has not been as high as desired, and has been substantially lower than Nafion®. Improvements in morphology have not yet led to the improvements in conductivity deemed necessary. Decreased EW from the two acid groups on the phosphonic acid does not seem to be as beneficial as would be expected; a more detailed comparison of phosphinic and phosponic acid membranes may shed some light on this.
- Some progress has been made, but conductivity is still far below targets.
- Molecular dynamics were able to agree that certain model compounds would show proton hopping (CF₃PO(OH)₂), but not for all model compounds that showed proton hopping.
- Desired membrane structures appear to have been produced, but further chemical identification would be helpful to confirm this.
- The project failed to achieve conductivity targets. It is clear that further optimization in terms of casting solvent, EW, means of crosslinking, etc. need to be pursued aggressively. In some cases, this is necessary just to be able to do a conductivity measurement.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **2.7** for technology transfer and collaboration.

- Good collaborations were carried out between different research groups.
- Good coordination has been carried out between industry, national labs and the University of Utah.
- The collaboration with Giner is a plus. Collaboration with a PFSA manufacturer will help a lot in moving this program forward more quickly by allowing the controlled preparation of low EW membranes and other samples for conductivity optimization.

- This is a good fundamental study; however, more consideration is perhaps necessary with keeping the real customer in mind. The project team should go back and make the same recommendation as was made last year, to consider collaboration with an industrial partner.
- Good collaborations have been made with lots of good groups, but the collaboration with FSEC and its subcontractors seems not to have been good. It should be expected that materials like these will behave differently from Nafion®, and there should have been much communication to avoid the problems that occurred.
- Collaboration between team members has been good. Collaboration with High Temperature Membrane Working Group and others is apparent.
- An industrial partner could be extremely useful to this membrane fabrication aspect of this project. The services provided by Giner and JEOL were not applicable towards membrane fabrication.
- Perhaps greater use of nuclear magnetic resonance (NMR) expertise could have been applied towards the membrane samples.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- The combination of sulfonic and phosphonic acid in the same polymer will result in lowering the effective EW of the ionomer. Instead, why not to use low EW sulfonic acid polymer?
- Crosslinked polymers are not stable in a free radical environment. Why pursue the crosslinked polymer route?
- The researchers seem to be on the right path to optimize this system. Greater focus should be placed on lower EWs and methods to increase phase separation (domain structure).
- The risk level for the proposed future work is unclear.
- Future plans are not well laid out and do not overcome the limitations of the system.
- Plans for work with higher acidity fluoro-sulfonimides are intriguing.
- Incorporating superacid moieties into perfluorophosphonic acid membranes is a logical path to improving membrane conductivity.
- Introducing the sulfonimide chemistry would likely prove to be a distraction. This might be better contemplated after 3M has finished its project on similar chemistry.
- It is agreed that crosslinking needs to be a future activity, but particular modes of crosslinking should be targeted.

Strengths and weaknesses

Strengths

- Phosphonic acid based fluoropolymer is a novel route to address low RH membrane conductivity challenges.
- There is a good combination of synthetic and modeling groups to address the wide scope of this project.
- The use of phosphonic acids in fuel cell membranes has been proposed many times. This is a well designed, systematic study which can show if this is a promising path.
- The project uses an approach of fundamental study supported by modeling and experiments.
- The project exhibits good fundamental studies on important issues, with good synthetic skills and strong modeling.
- The project has a strong team of theorists and experimentalists.
- It seems to be a well-thought out and logical approach to achieving high conductivity at low RH.
- The project has a simple and strong premise.
- The project has the ability to predict conductivity using molecular dynamics and model compound studies.
- The project also has the ability to still produce desired membrane chemistry despite earlier setbacks related to keeping monomers in solution.

Weaknesses

- As expected, phosphonic acid fluoropolymers did not show high membrane conductivity under low RH conditions.
- The addition of sulfonic acid groups to phosphonic acid polymers is not expected to enhance proton conductivity of the membrane to the same extent as low EW sulfonic acid polymers.
- This program needs to show greater progress towards meeting conductivity goals.
- The project requires improved gap analysis and consideration for practical use.
- It is perhaps weak on measurements and definitely somewhat inflexible in trying to overcome limitations.
- The relative knowledge of perfluorophosphonic acid materials is not as developed as that for sulfonic acid derivatives.
- Despite a promising approach, a lack of positive results so far makes the technical viability questionable.
- The lack of industrial collaboration made it tough on the project to be more nimble about optimizing membrane fabrication such as casting solvent and EW.
- There is a lack of molecular dynamics comparison to sulfonic acid groups.
- More information about the membranes should be reported, including swelling, NMR identification, and perhaps mechanical/thermal characteristics.

- The crosslinked membrane approach may not give free radical stability to the membrane, so there is no point in carrying out this task.
- It will be interesting to see whether acid-containing inorganic composites with phosphonic acid-based fluoropolymers will help in enhancing the membrane conductivity.
- I would like to see a greater focus on lower EWs and methods to increase phase separation (domain structure)
- The team should consider a membrane with PBI.
- While performance of perfluorophosponic acid derivatives is of interest, the poor conductivity seen to date suggests that this may be more appropriate for Basic Energy Sciences-type studies rather than the more near-term applications oriented EERE funded projects.
- Temptations to dive into other forms of chemistry beyond the original project premise should be avoided.
- An industrial membrane collaborator should be added to optimize the membrane faster.

Project # FC-08: Rigid Rod Polyelectrolytes: Effect on Physical Properties Frozen-in Free Volume: High Conductivity at Low RH

Morton Litt; Case Western Reserve University

Brief Summary of Project

The objectives of this project are to 1) synthesize polyelectrolytes that reach or exceed DOE low humidity conductivity requirements; 2) use materials and synthetic methods that could lead to cheap proton exchange membranes; 3) understand structure/property relationships in order to improve properties; and 4) develop methods to make these materials water insoluble and dimensionally stable with good mechanical properties. Case Western Reserve University has decided to work with poly(pphenylenes) with one and two sulfonic acids per ring. These have lower equivalent weights (EWs) and cannot hydrolyze.



Question 1: Relevance to overall DOE objectives

This project earned a score of 3.6 for its relevance to DOE objectives.

- With a stable and low relative humidity (RH)/high temperature, the membrane supports the Fuel Cell Program at DOE.
- The relevance to obtaining low RH/high-temperature membranes is high.
- The team took a well-formulated and innovative approach and fully supports DOE objectives.
- Developing membranes with high conductivity at low RH is critical to automotive proton exchange membrane fuel cell (PEMFC) commercialization.
- This program supports the DOE R&D objectives.
- This work is in line with Task 1, Barriers A, B, and C to develop membranes that meet all targets.
- It is a relevant project with an out-of-the-box concept.
- Membranes with improved conductivity that function under high temperature / low RH are included in the Hydrogen Program and DOE's RD&D objectives. These are not the most critical material needs for enabling fuel cell commercialization.

<u>Ouestion 2: Approach to performing the research and development</u>

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

- The project is another DOE funded HC membrane using poly(biphenyl [di]sulfonic acid) membranes. While the concept is worthwhile, the reviewer does not see this as a long term solution to fuel cell membrane commercialization.
- The performance is too low, the cost is too high, and the membrane lacks the required durability (the membrane is water soluble or disintegrates in water).
- The rigid rod structure is intended to provide permanent volume for water, even at low RH. Conductivity results are very good.
- Mechanical and chemical durability have not been sufficiently addressed.
- The rod polymers are very promising. The project team needs to construct some fuel cell-type membranes to verify how these would function in an actual fuel cell configuration.
- Trying to improve mechanical stability is an excellent approach.

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- The team has taken a very novel approach towards producing a highly conductive polymer.
- This is probably one of the few legitimate hydrocarbon films that has a shot at achieving program objectives. The team is currently concentrating on making water insoluble films.
- This project has an interesting concept; however, there are uncertainties about the risk level for this approach.
- The high conductivities of the novel ionomers presented are promising for developing materials with decreased conduction losses. Developing ultra-high acid loaded polymers is interesting and has largely been shown viable through the approach presented. The focus needs to shift further to producing materials with improved mechanical properties.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.8 based on accomplishments.

- Proton conductivity targets were met with a water soluble membrane or a membrane which disintegrates in water. Benchmark material used by the PI is Nafion® 117. For the amount of money spent by DOE, the reviewer does not see anything in this presentation that is unique or has not already been attempted.
- Excellent conductivity has been achieved.
- Mechanical and chemical durability have not been sufficiently demonstrated.
- Perpendicular conductivity is presumed to be higher than the in-plane conductivity, based on the rod orientations, but this assertion needs to be demonstrated experimentally.
- There were a few new ideas for mechanical stabilization via copolymer development, but there was little new data, and little progress toward making a membrane that is mechanically stable in water.
- The conductivity results are very impressive. The work towards producing materials that can be tested in fuel cells should be accelerated. The oxidative stability of the pendent aliphatic groups should be studied.
- Very high proton conductivities were demonstrated, and the project comfortably met the go/no-go criteria.
- The team performed a good analysis and material characterization; however, physical properties are major limitations now.
- There were a few more "new results" in this year's presentation compared to the prior year's presentation, but there is still less progress advancing this promising area of science than would be expected at this funding level. The graft copolymer approach shows some promise at improving properties, but further advances are necessary in the area of mechanical properties/durability.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **1.6** for technology transfer and collaboration.

- Collaboration with outside companies remains non-existent.
- There were no industrial collaborators because the membrane is at a very low level.
- The program should have had more collaborators from the beginning.
- There seems to be very little outside collaboration, although "interest from GM and 3M" is encouraging.
- Collaborations were not addressed in the presentation, and cannot be judged from the material provided.
- Professor Litt could benefit significantly by collaborating with experts in block polymer synthesis.
- It would be beneficial to this team to work with an industrial partner who can do a more in-depth evaluation of these materials
- Interest from 3M and GM is not the same as working with 3M or GM. The PI must involve industrial collaborators who can give relevant advice and aid in the commercialization of the material.
- The project may benefit from collaboration with any of the major industry partners.
- There have been relatively few interactions; however, they are not particularly needed at this point in time.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.8 for proposed future work.

- Future work is as expected for this type of material, e.g. increase molecular weight (MW) and graft copolymers. One highlight would be to use these ionomers with Professor Pintauro's electrospinning technique.
- More work on the mechanical and chemical properties of the materials should be done. Approaches to solving these issues are not compelling.
- Future work is well planned and builds on the present results.
- The proposed focus of copolymerizing sulfonated poly(p-phenylenes) with non-soluble copolymers is warranted.
- The focus on making higher MW polymers is also directionally correct.
- The grafting work does not seem to show much promise.
- The project shows very good direction towards improving mechanical properties, although chemical stability (oxidative) should be evaluated. More emphasis on fuel cell testing would be appropriate at this stage.
- It really looks like a serious effort is being made to make these materials practical.
- The risk of the proposed approaches for improving mechanical properties is not known.
- Work should move beyond the current studies suggested involving an increasing degree of sulfonation. The past year showed advances in this area without major impact on performance. The suggested approaches that involve copolymer synthesis by Suzuki reactions are quite reasonable. No direction is shown as to the approaches that will be followed, but rather a list of many possible studies is presented.

Strengths and weaknesses

Strengths

- There is a potential for collaboration with Professor Pintauro; the electrospinning technique is very unique and could be of high value.
- It is an innovative approach that actually works.
- Professor Litt has demonstrated the best conductivity of High Temperature Membrane Working Group projects.
- The conductivity results are very impressive.
- There was good emphasis on understanding structure-property relationships.
- The hydrocarbon material was very highly proton conductive.
- The project team took an out-of-the-box approach, and exhibits a fundamental understanding of the mechanism.
- Extremely high conductivities were demonstrated.

Weaknesses

- The chemical backbone is not unique; sulfonated polyphenyl has been extensively studied and does appear to have the inherent properties needed for fuel cell vehicle membrane requirements.
- This project desperately needs partners for synthesis and durability issues.
- The project shows no obvious weaknesses.
- Poor polymer synthesis expertise to make high MW copolymers.
- There are still issues with making water insoluble materials.
- This project could perhaps use a more holistic approach, where all requirements are addressed relatively simultaneously.
- This project is very slow in advancing the technology. The particularly impressive conductivity results are now more than two years old, and relatively little has happened in advancing these materials since their creation.

- At this funding level, more output is expected. The focus should be on creating a membrane with a relatively high proton conductivity and decent mechanicals. This would speed up collaboration with Professor Pintauro.
- This work is yielding excellent results. Fuel cell testing should be planned, and equipment made available to do this.
- The project team should obtain through-plane conductivity data because membranes are not isotropic.
- The project team should to add more polymer synthesis expertise to the project.
- Work with an industrial partner who can do more in-depth evaluation of these materials would be beneficial to this program.

- More morphological studies and RH and temperatures cycling are necessary.
- These synthesis studies are often slow and laborious, but the focus could be modified to focus on more processable systems as mechanical properties seem to be a limitation of these materials. These are extremely promising materials, and it is suggested that the project team introduces a separate mechanical component (either through support incorporation or producing blocks) and/or some backbone flexibility without diluting acid concentration substantially (at least within the conducting domain).

Project # FC-09: NanoCapillary Network Proton Conducting Membranes for High Temperature Hydrogen/Air Fuel Cells

Peter Pintauro; Vanderbilt University Patrick T. Mather; Syracuse University

Brief Summary of Project

The objective of this project is to fabricate and characterize a new class of NanoCapillary Network proton conducting membranes for hydrogen/air fuel cells that operate under high-temperature, low humidity conditions. The 2008-2009 project goal was to fabricate membranes with a proton conductivity of 0.10 S/cm at 120°C and 50% relative humidity (RH) in preparation for the Year 3 DOE go/no-go decision.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.5** for its relevance to DOE objectives.



- The project is aligned with DOE's RD&D objectives.
- The focus of this project is to develop a low RH membrane, which is aligned with the hydrogen vision.
- All the tasks in this project are focused towards the low RH, high-temperature membrane initiative.
- This project is very relevant to DOE's objectives, and on track to meeting all of them.
- The program investigates novel ways of making membranes with controlled channel structures of perfluorosulfonic acid (PFSA). This method provides interesting ways of making membrane structures; however, it is not clear how this will work in the membrane electrode assemblies (MEA) and the Norland embedding material must be suspect if it is a polyurethane. Although the program is not very profound in its goals and objectives, it is of considerable benefit to the Hydrogen program.
- The project is aligned with DOE goals and targets and addresses appropriate barriers.
- This project fully supports DOE RD&D objectives by attempting to create a novel membrane for proton exchange membrane fuel cells (PEMFC). If successful, it will be critical to the Hydrogen Program.

Question 2: Approach to performing the research and development

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

- The approach of transporting protons through proton conductive fibers is interesting.
- The concept seems to be an inverse concept of reinforced membrane, and the non-conductive NOA63 seems to be a good mechanical reinforcement material.
- The limitation of proton conductivity in 3M 733 equivalent weight (EW) material indicates that densification of the fiber is needed.
- The use of sulfonated polyhedral oligomeric silsesquioxanes (SPOSS) seems to be boosting the proton conductivity.
- This is an interesting, almost systematic approach, by decoupling conductivity and mechanical strength. The project team needs to incorporate fuel cell level testing and understand the impact of trace impurities, thermal cycling, and robustness of the membrane over longer periods of operations.
- The approach is slightly pedestrian beyond the initial premise. One would like to see more work on variation of the channel dimensions, EW, and other additives besides SPOSS. There is a suspiciously empirical feel to this

that is troubling. One would like to see a bit more fundamental objectives, such as the mechanism of proton transport. Also, there really is no mention of how to make these work with electrodes.

- The project team changed course away from polysulfones when it was seen that they would not meet the 120°C target and moved to PFSA materials. The approach is flexible and may be useful for other ionomers as well as PFSAs.
- The team took a very well-organized approach to addressing technical barriers, and it was logical throughout.
- While some doubts still exist about whether the volume consumed by the non-conducting resin will prevent commercialization, the approach still remains simple and novel: electrospun PFSAs provide conductivity while the embedded resin provides mechanical support.
- This year's approach was excellent; the electrospinning of PFSAs was well worthwhile.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.3** based on accomplishments.

- Good progress has been made during the last year.
- Achievement of the DOE goal by a sample 4 mm thick NFN-02 is impressive.
- The spinning method seems to be very versatile and capable of making fiber with many different ionomers.
- The team has made impressive progress and is on track to meeting all its objectives.
- The team has made good technical progress in achieving the set objectives, but it needs to set some more ambitious goals for the future otherwise, this will just be another membrane that is not quite as good as Nafion[®]. The mechanical properties are described as good or better without any discussion of what is wanted. Some better thought into this would be good.
- The project team met the very aggressive DOE milestone of 0.1S/cm at 120°C and 50% RH. Flexibility of the approach increases chances of meeting all the DOE membrane targets simultaneously.
- So far, the project is meeting all its milestones, and the membranes also have good mechanical properties.
- This project made quite a leap forward this year with the electrospinning of PFSAs. Conductivity measurements showed that the DOE 120°C target was achieved. It should be noted, however, that a more realistic 120°C target (25% RH) would not have been met, and that the target at 80°C was not met.
- Swelling and gas crossover data were not reported in the slides. In the future, the project should be sure to present a "scorecard" of membrane attributes to make sure everything is covered.
- The sample thicknesses are still too large.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.3** for technology transfer and collaboration.

- The inclusion of two industrial partners, 3M and Nissan, is good for the progress of the project.
- It would have been nice to have a national lab and university involved in the project.
- There appears to be strong collaboration with 3M.
- The program has good collaborations with industry, but not many with other academics that might increase the curiosity into the fundamental mechanisms. This should be emphasized in the future.
- There have been collaborations with an MEA manufacturer and an original equipment manufacturer (OEM). The project team initiated work with Nissan this year. The team is working with 3M, and with Professor Litt to try to incorporate his high-conductivity material.
- The project team has exhibited good collaboration with 3M.
- The project team has very effectively worked with 3M as a collaborator. It was 3M who provided the PFSAs that helped greatly in achieving the DOE 120°C conductivity target.
- Collaboration from Nissan does not yet appear to have generated much data.

Question 5: Approach to and relevance of proposed future research

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

- The proposed future work is aligned with the present issues faced in the project.
- The exploration of insoluble SPOSS material will be helpful in controlling the conductivity of the membrane.
- The development of core-shell fibers and use of inorganic fillers are good strategies.
- Proposed future work should help the project achieve all its technical milestones.
- Future work will no doubt be useful, but it could be improved with more fundamental work on proton conduction mechanisms and how such elements as channel size and EW affect it.
- Future work is focused in the right areas looking at the durability of SPOSS and Norland Optical Adhesive (NOA), and options to replace these, and looking at further options in PFSA materials.
- A number of good ideas were presented for further development. It is good that there are multiple pathways under consideration for further improvement of membrane performance, and also that stability is being addressed.
- The future work section correctly recognizes that the use of SPOSS is likely a liability, and that it should ideally be replaced.
- The future work section also correctly regards the resin with suspicion.
- A baseline of the chemical and mechanical stability of present materials (in DOE Accelerated Stress Tests, ASTs) would be good. Or, if speculation is strong enough, it would be good to present some commentary on why these ASTs would not work with materials as they are (e.g., swelling is too high so the mechanical test would fail quickly).

Strengths and weaknesses

Strengths

- Developing wired matrix for proton conduction is a very interesting and novel concept.
- The feasibility of the concept has been demonstrated well.
- Improvement of the membrane's mechanical strength by using NOA63 polymer matrix is a good concept.
- The project team has had early success with decoupling membrane conductivity and stability functions and working on areas to build on this success.
- It is an excellent concept and rapidly produces new ways of affecting morphology.
- The flexibility of the electrospinning approach allows the benefits of a composite membrane to be seen with materials with poor mechanical properties.
- It is a novel concept unlike other projects.
- This is a teachable project that has embraced feedback from reviewers to create even better materials than before.
- This is also an evolving project that realizes the drawbacks of particular components, but has a solid plan to address them.
- The project has shown the use of quick technical decisions to make challenging tasks (e.g., electrospinning of PFSAs) work out.

Weaknesses

- The project team needs to find a non-leachable SPOSS type proton conductor to have a practical membrane.
- The chemical stability of NOA63 is still an issue.
- SPOSS is expected to deform the fibers upon prolonged RH cycling.
- The team needs to improve the NCM surface to enhance the membrane electrode integration.
- The reviewer didn't get a good feel for cost.
- There has not been enough fundamental thought to what is going on. This has the feel of an engineering project where not too much inquiry into the basics is going on.
- The durability of SPOSS and the inert membrane filler NOA have not been demonstrated.
- There is a lack of reporting certain attributes that reviewers need to know about, e.g., swelling and gas crossover.
- The membrane thickness is a weakness.
- It is still unknown whether a chemically stable embedding resin will be found.

- The project team needs to focus on cost and ability to manufacture in large volumes, along with performance in fuel cell environment.
- The team needs to add aq modeling components and force the program to look into basic mechanisms, such as how the channel dimensions affect the behavior.
- The team also needs to work with lower EW material being made at 3M, and material being made for Giner at Syracuse if possible. Look at polyPOM materials as additives.
- Once it is known that a fairly durable PEM has been fabricated (which has not yet necessarily happened), it would be good to have *in situ* DOE accelerated stress tests performed.
- While it is good that 3M has contributed the PFSA for electrospinning, it might also be wise to add their services in order to figure out how to decrease the thickness of the membrane. It might also be interesting to see how lower EW PFSAs perform in electrospinning trials.

Project # FC-10: High Temperature Membrane with Humidification-Independent Cluster Structure *Ludwig Lipp; FuelCell Energy, Inc.*

Brief Summary of Project

The objectives of this project are to: 1) develop polymer membranes with improved conductivity at up to 120°C; 2) develop membrane additives with high water retention and proton conductivity; 3) fabricate composite membranes; and 4) characterize polymer and composite membranes (in-plane conductivity). FuelCell Energy, Inc. has 1) fabricated three polymer iterations, six nanoadditive batches and more than 10 composite membrane batches; 2) improved mC^2 uniformity and conductivity with concurrent process simplification; 3) integrated additive functionalization and composite membrane fabrication; and 4) demonstrated >2ximproved conductivity at 120°C over 2008 (>3x higher than NRE-212[®]).



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.3** for its relevance to DOE objectives.

- A high-temperature membrane that operates at low relative humidity (RH) is one of the key technologies needed for the commercialization of fuel cells.
- High conductivity at all operating conditions is critical to achieving overall DOE targets of cost and performance.
- This project is fully in line with DOE objectives.
- It appears to address only one critical barrier of one membrane electrode assembly (MEA) component.
- High-performance membranes are critical to automotive commercialization, but lack of specifics doesn't allow others to benefit from fundamental knowledge gained from the work done.
- There appears to be good alignment with DOE's current objectives. A broader consideration should include the ability to operate at higher temperatures to have low CO sensitivity required for stationary fuel cell systems operating on infrastructure fuels.
- Membranes with improved conductivity that function under high temperature/low RH are included in the Hydrogen Program and DOE's RD&D objectives. These are not the most critical material needs for enabling fuel cell commercialization.

<u>Ouestion 2: Approach to performing the research and development</u>

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

- While the PI does not divulge the details of their approach, the concept of adding a variety of technologies, e.g., co-polymer, support polymer, water retention additive, and proton conductivity enhancer, to obtain a viable fuel cell membrane is good approach.
- Unfortunately, the reviewer has no idea what these various technologies really are.
- The necessary functionalities such as water retention, conductivity, and strength, are being provided by additives to form a composite membrane.
- Complicated systems usually lead to high cost.

- The project has overcome significant technical barriers. The disclosed material was, however, short on details because of the proprietary nature of the additives. Some of these additives are known to be very expensive, so new synthetic routes may also need to be developed.
- The approach appears to be a truly "engineered" membrane physical versus just chemical. It appears to be quite successful in breaking down the overall functional performance properties into separate characteristics.
- The approach is hard to judge without knowing anything about the materials, other than it seems to work for getting good conductivity at low to moderate RH. The approach may not be compatible with durability or cost targets. There is no way to know without knowing what the materials are or having any durability data.
- The project team has taken an excellent problem solving approach. Considering all required functions of strength, water retention and protonic conductivity early on allows for narrowing down to a comprehensive solution.
- The approach involves a multicomponent system containing a support, low equivalent weight (EW) ionomer, water retaining additive and proton conduction enhancing additive. The multicomponent approach seems to provide additive or synergistic effects based on conductivities reported. Evaluation of the approach is limited as it is unclear what additives are being investigated and how they work.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.6 based on accomplishments.

- Based upon the PI's results and the results at BekkTech, the research obviously meets or exceeds the DOE's targets; however, the PI did not discuss the mechanical nature/swelling of the membrane, which could be the potential weak point.
- All project targets, goals, and decision points have been met to date.
- The amounts of additive have not yet been optimized.
- Swelling may need to be addressed.
- The performance of the composite membranes, particularly at 50% humidity, is spectacular.
- The project shows very encouraging results and is meeting all program milestones, but it still needs to demonstrate that fuel cell performance will not have any surprises. Also, other membrane durability tests have not been carried out yet.
- The conductivity results are very good as verified by BekkTech. If the materials turn out to be durable, I'd say progress was outstanding.
- The project team has made excellent progress. The reviewer is curious to see the actual fuel cell performance and endurance data.
- Of the conductivity results reported to date, the recent results are impressively high. Other data regarding the materials is lacking in the presentation. In particular, water uptake and mechanical properties would be useful as only conductivity is reported anywhere without any information on durability/leaching.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated 2.7 for technology transfer and collaboration.

- Collaboration with various institutions appears to be very high; however, as everything is highly confidential, the reviewer does not know who they are: a polymer company, nano-materials company, or university? Are they receiving DOE funding?
- Although impressive progress implies impressive partners, the partners are not named and, therefore, cannot be rated.
- Collaboration was not addressed and cannot be judged.
- It appears they brought together the key technology experts to methodically address their membrane component/functional requirements, but why aren't the collaborators fully identified?
- The unnamed collaborators seem to work together to provide a product with good conductivity. It's impossible to know the value of partners as no specifics were given.
- There was excellent collaboration among vendors, material experts, academia, and test entity probably the best of any team.

• There were relatively little interactions; however, they are not particularly needed at this point in time. A partner is mentioned as contributing to the project, but no further information regarding this partner is given. This makes an assessment of the value of this teaming arrangement difficult.

Question 5: Approach to and relevance of proposed future research

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

- The proposed work is based upon existing/established membranes and is well thought out.
- Fuel cell experiments with performance and durability experiments are on the agenda, which is excellent at this stage of development.
- Future work is right on the mark, addressing durability/stability, cell performance, cost, scale-up, and synthesis simplification.
- The accomplishments to date are extraordinary, and definitely warrant continued and intensified development efforts.
- The project team needs to demonstrate compatibility with catalyst electrodes to identify any future issues.
- The two areas of recommended future work are durability testing and high-temperature fuel cell testing.
- Further polymer optimizations should only be made while taking durability under consideration. Performance is already satisfactory.
- I agree with proposed future work. The ability to manufacture in high volume and low cost needs to be a consideration.
- It is not certain how the project has resulted in the conductivity reported, and this makes future advances uncertain. Further optimization is the first bullet noted in future work, but conductivity does not seem to need significant advances. Durability is noted but not discussed, and at the present time it is uncertain what the durability concerns are.

Strengths and weaknesses

Strengths

- The approach is based upon what the reviewer can see is very sound and logical, and results support this fact.
- The additive approach works.
- The PI's company has a strong background of practical understanding of fuel cells and what is required for commercialization.
- The project team has made a membrane with good conductivity.
- The project team has taken a highly methodical and holistic problem-solving approach. They are staying focused on project goals and optimizing around meeting objectives.
- The team has taken a combined approach that seems to be yielding enhanced benefits from additive approach, particularly high conductivity.

Weaknesses

- The only weakness (and I'm not even sure if it a weakness) is the secrecy of the project. Does this work help the overall industry or only the company itself? Does that matter?
- The lack of identification of partners and chemical/structural constituents prevents full analysis of the project.
- Cost issues may need to be addressed.
- There is a lack of disclosure of materials.
- There is a lack of mechanical and swelling data on membranes.
- There is a lack of durability data.
- There is also a lack of cost analysis.
- I can't think of any. This is probably the best project!
- There is a lack of technical data, other than conductivity and a lack of information regarding materials and scientific basis for advances. This makes the project more difficult to evaluate and is frustrating, as three years into the project they still haven't discussed any materials from even initial studies.

- The team should continue as planned; *in situ* fuel cell experiments are the planned next step to look at performance and durability. It will be very important to learn from these experiments and cycle back to the beginning if the results do not meet expectations.
- The reviewer recommends continued evaluation of longer term tests, and possibly stability testing with Fenton's reagent.
- The team should run DOE specified durability tests. Further recommendations will fully depend on the durability data.
- The team should provide a cost analysis or, better yet, have an independent party such as TIAX or DTI do it.
- The project team should move beyond conductivity to other properties.

Project # FC-11: Novel Approaches to Immobilized Heteropoly Acid (HPA) Systems for High Temperature, Low Relative Humidity Polymer-Type Membranes

Andrew M. Herring; Colorado School of Mines(CSM) Mathew H. Frey; 3M Corporate Research Materials Laboratory

Brief Summary of Project

The overall objective of this project is to fabricate a hybrid heteropoly acid (HPA) polymer from an HPA functionalized monomer with >0.1 S/cm at 120°C and 25% relative humidity (RH). The objective for 2007 was the synthesis and optimization of hybrid HPA polymers for conductivity from room temperature to 120°C. The 2008 objective was the synthesis and optimization of hybrid HPA polymers for conductivity from room temperature to 120°C with an understanding of chemistry/morphology conductivity relationships using a model system. The 2009 objective was to optimize hybrid polymers in more practical systems for proton conductivity and mechanical properties.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.5** for its relevance to DOE objectives.

- The project is very relevant to the Hydrogen initiative and supports DOE RD&D objectives.
- This project is focused on meeting DOE objectives on low RH membrane conductivity.
- The project addresses important issues of particular relevance to the Hydrogen Program. It attempts to study the factors that are important in controlling proton transport under low RH conditions, as well as simply trying to meet milestones. This is important, as meeting the 25% RH goal looks difficult for this system, and fundamental studies will be necessary to meet the goal.
- The project is aligned with DOE goals and milestones, and it addresses relevant barriers.
- The project goal of fabricating and characterizing a high-performance polymer electrolyte membrane for high-temperature/low RH fuel cell operation is highly relevant to the DOE.
- The project, like many other membrane projects, is attempting to produce a membrane for polymer electrolyte membrane fuel cells (PEMFC). The production of membranes that can operate at high temperature and low RH is central to DOE Hydrogen Program goals.

Question 2: Approach to performing the research and development

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

- The approach taken for achieving the goal is unique.
- The incorporation of HPAs in PFSA-type materials to enhance low RH conductivity is a good approach.
- However, the incorporation of HPAs into other types of host polymers, which are unstable under free radical environments, may not yield a practical membrane.
- Ultra-violet (UV) curable polymer host materials may contain leftover free radicals, which may degrade the polymer under fuel cell conditions.
- The approach seems fair. Use of the polyacrylate polymers is okay and justified by the need to make membranes and get on with the polymer properties. The design space is rather mysterious and not well defined.

The axes are not understandable; however, at some time the durability needs to be addressed. It is also unclear on how the membranes will function with the electrodes. Some ideas for this might be useful.

- Initial work has been done and has resulted in easier-to-produce, quicker-to-make polymer linkers, which allowed quicker progress and enabled the team to meet the conductivity target with a totally new polymeric ionomer system. With this newer system, this was the appropriate approach, and allowed them to demonstrate that they can approach the targets with this new type of ionomer system. This is a good mix of synthetic work with advanced characterization techniques, which allows the team to understand its system, as well as other systems that have been studied for much longer.
- The use of HPAs is interesting, but there are many potential pitfalls, including membrane brittleness, leaching of HPAs from the membrane, the thermal stability of such HPAs, and the method of mixing such inorganic materials with a polymer.
- The approach is good novel, well-designed, and feasible although it would have been better if a stable comonomer had been used from the start.
- While the approach recognizes that a water soluble group such as an HPA would be best situated as an immobilized part of a polymer chain, issues of swelling and morphologically derived conduction inefficiencies still remain as hazards.
- The project shows that its approach is changing; however, positioning the HPAs as pendant groups off a main chain might return HPAs to a vulnerable position.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.7 based on accomplishments.

- The project has achieved the conductivity objectives, but has not addressed the mechanical properties of the membrane.
- The HPA co-polymer (slide-14) did not show good conductivity above 100°C.
- It seems that the HPA polymer hybrid material is behaving similar to perfluorosulfonic acid (PFSA)-type material and losing its conductivity above 100°C.
- Many measurements have been taken, and some reasonable progress on how morphology affects conductivity has been made. The low RH results are a bit worrying, and some more work on the polyoxometalate (POM) mechanisms might be useful.
- The project team has achieved the very aggressive DOE milestone of 0.1 S/cm at 120°C and 50% RH, and developed a totally new class of polymeric protonic conducting materials.
- The data presented by the PI is confusing. No BekkTech conductivity data were presented and some data were confusing: PolyPOM75v membrane loses all its water at 56% RH for all temperatures (slide 12), and thus has no conductivity at this RH, but PolyPOM80v has a very high-proton conductivity at 120°C and 50% RH. It is unclear why this is the case. The two membranes are similar. In addition, the use of acrylate polymers because of their poor chemical stability is questionable. It is unclear why the team has not used 3M's perfluorosulfonic acid polymer since 3M is a collaborator on the project.
- The project is meeting conductivity milestones, which is good, but it needs to accelerate efforts to provide good chemical and mechanical stability.
- Numerous drawbacks are found with the original strategy, including membrane brittleness and low conductivity at low RH.
- Some promise is found with certain samples achieving 0.1 S/cm at 120°C/50% RH, and with the use of UV polymerization.
- Although it can be said that certain conductivity measurements achieved their goal, not enough is known about the membranes, particularly with respect to swelling and leaching. Furthermore, chemical identification (NMR, IR/Raman) within a fairly complex system would be useful.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.0** for technology transfer and collaboration.

- The project involves good industry and academic collaborations.
- Involvement of some national labs could have been better.

- Partners are well coordinated, but collaboration with the Topic 2 group is not well presented. Outside collaborations are not prominent, and could be increased in order to attack the low RH problem which looks to be big.
- The project team is collaborating with 3M.
- There seems to be a good collaboration with 3M.
- Interaction between CSM and 3M has been fruitful.
- 3M has played a consultant role for a while, but it appears a more earnest role is just beginning, as indicated by the future work.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.5 for proposed future work.

- The proposed research is very vague.
- No clear direction in the future work on improving the membrane morphology is given.
- There is no clear indication on the types of monomer to be used. It is understandable that the monomer structure could be proprietary, but the nature of materials could have been mentioned.
- Future work is a bit mysterious and not easy to judge. How is the morphology to be improved? The details on this are fuzzy.
- Future work with 3M offers the potential for a link to fluorinated linkers and fuel cell testing. More information on monomer systems to be used would be helpful. Whatever information can be provided without divulging intellectual property (IP) would help (are they using hydrocarbons, fluorocarbons?).
- The future work slide was very general and lacked specifics. There were no mechanical property measurements in the 2009 AMR presentation and no plans to make such measurements next year. Mechanical tests on the membrane (both wet and dry) should be carried out next year. Also, the water swelling and chemical/thermal stability of the membranes should be evaluated.
- Future work plans are logical and sound, but the lack of detail makes this one a bit hard to judge.
- The prospect of positioning an HPA as a pendant group off a main group may not be appealing. The HPA may be more susceptible to leaching, and the channel size may have to be increased, in which case perhaps conductivity would require greater RH. This is all just speculation, but there is no assurance that the new possible structure will work.
- Thankfully, the old concept has not been entirely tossed aside. It would be interesting to see if 3M could still find a way to keep the old system from being mechanically unstable.

Strengths and weaknesses

Strengths

- The team has taken a good approach by harvesting the low RH conductivity property of HPA.
- The team shows good collaboration with an industrial partner.
- The project team has taken an interesting approach of making HPA hybrid material using UV curable polymers.
- The team has a good combination of industrial and academic workers.
- Polymer chemistry and HPA chemistry give the prospect of a genuine breakthrough in proton conduction mechanisms that could solve the low RH issue.
- The team has a good mix of synthetic work and characterization which allows them to have a better understanding of their system. The presence of 3M provides the polymer experience necessary to advance the project, and the material offers the potential for very low RH conductivity.
- The team has taken an interesting approach that appears to be working.
- The team has strong collaboration with 3M.
- The team has attained high-conductivity measurements despite complex chemistry and numerous materials drawbacks.
- The team is collaborating with 3M.
- Certain prospects still exist for improving membranes, e.g., UV polymerization.

Weaknesses

- There are no clear directives towards the future work.
- Using UV-curable host polymers to make the HPA hybrid membrane is not very practical.
- The second generation hybrid material (slide 18) may result in poor mechanical properties of PFSA membrane due to the bulky size of HPA incorporated between the polymer chains of the backbone polymer.
- The team should move on from polyesters. Future plans are veering into secrecy at an inopportune time. More progress needs to be made on the basic HPA polymer interactions before drawing a secrecy veil around the work. The team did not show where the membrane electrode assembly (MEA) work is planned. Durability issues need to be more explicitly addressed.
- There are many unanswered questions, such as why there are no BekkTech third-party proton conductivities, and seemingly confusing data, e.g., the difference in properties of polyPOM75V and polyPOM80v.
- The PI has not yet addressed any critical issues regarding the chemical stability of his polymer and the mechanical properties of his polymer-HPA materials.
- The project has exhibited sample brittleness and the possibility of other material disadvantages.
- The project has also exhibited low conductivity at low RH.
- The general concept of using HPAs is risky due to swelling, leaching, etc.

- How is the MEA to be prepared with these materials?
- The group should begin to address mechanical properties and durability issues.
- The group should clarify the results obtained to date, and ensure that reproducible membranes can be made. It is unclear why the PI is planning to look at morphology. He already has met the DOE conductivity target, and should focus on chemical/thermal and mechanical stability.
- Work focused on the original concept should continue, but with more intense collaboration from 3M to resolve brittleness, low conductivity at low RH, uniformity of the material, and other problems.
- Perhaps some theoretical work previously applied to other membrane projects could be used here to help predict the degree of proton hopping to be expected for distinct membrane structures.

Project # FC-12: Improved, Low-Cost, Durable Fuel Cell Membranes

James Goldbach, David Mountz, Tao Zhang, Wensheng He, and Michel Foure; Arkema

Brief Summary of Project

The objectives of this project are to 1) develop a membrane capable of operating at 80°C at low relative humidity (RH) (25-50%; 2) develop a membrane capable of operating at temperatures up to 120°C and ultra-low RH of inlet gases (<1.5 kPa); 3) use commercially available matrix materials as a low-cost approach; and 4) elucidate ionomer and membrane failure and degradation mechanisms via ex situ and in situ accelerated testing. Mitigation strategies will be developed for any identified degradation mechanism. M41 was shown to have superior durability in accelerated in situ testing and shown to operate down to 65% RH. Initial M70 membranes show an order-of-magnitude increase in conductivity versus M43.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.1** for its relevance to DOE objectives.

- Low-cost and durable membranes are critical to achieving DOE targets.
- The project does not clearly address DOE objectives, as the project targets are for performance at 80°C rather than 120°C.
- The project focuses on just one fuel cell membrane electrode assembly (MEA) component the membrane. This may not be adequate to show ultimate feasibility.
- Mechanically robust and low-cost membranes are critical for automotive fuel cell commercialization.
- Work is in line with Task 1, Barriers A, B, and C, to develop membranes that meet all targets.
- The project goals of fabricating and characterizing a high-performance polymer electrolyte membrane for high-temperature/low RH fuel cell operation are highly relevant to the DOE.
- Membranes with improved conductivity that function under high temperature/low (RH) are included in the Hydrogen Program and DOE's RD&D objectives. These are not the most critical material needs for enabling fuel cell commercialization.

Question 2: Approach to performing the research and development

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

- It is not clear how membrane conductivity can be maintained when a substantial fraction of the material is composed of an insulating material such as Kynar®.
- However, the Kynar does help with mechanical and chemical durability.
- Biphenyl sulfone H form (BPSH) probably does not have the necessary conductivity to achieve DOE targets.
- Large volume production of the BPSH-Kynar blends appears to be feasible, but performance of the membranes is significantly below target.
- Focusing on commodity Kynar is a good cost strategy, but the downside is that it may limit options for meeting the performance and durability goals.
- Blending polyelectrolytes with polyvinylidene fluoride (PVDF) is a solid approach to make a durable, potentially high performing, and low-cost membrane.

- Working with highly functionalized polyelectrolytes is also a good approach.
- The choice of ionomers so far has been poor. Ionomers with a chance of meeting the targets should be used.
- The use of polymer blends is a sound approach, although not particularly novel. A focus on low-cost approaches is important and worthwhile.
- Investigating Kynar (PVDF) as a potential cheap/durable polymer is sensible for a company with PVDF experience. The use of polymer blends is worthy of investigation, but testing to-date has not shown significant promise, as current low RH conductivity is poor.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 1.7 based on accomplishments.

- Arkema has increased the conductivity an order of magnitude over the early formulations, but it is not clear how continued optimization of blends of Kynar with M7x or BPSH can realize another order of magnitude to reach the DOE high-temperature/low RH targets.
- Conductivities are way below what is needed for these blends.
- Given where the project was with the M4X series, the M70 is showing much improvement, but it is only just matching the old Nafion® standard.
- It would be good to have already demonstrated some fuel cell performance with the M70 membranes. If they had membranes for the conductivity measurements, then why were the fuel cell measurements not done as well?
- The rate of progress of the program is low.
- While they have tested a lot of membranes, none approach the low RH resistance targets.
- I commend the PIs for running RH cycling and open circuit voltage (OCV) hold tests, and showing very good durability in both tests.
- It was also nice to see some results on the sensitivity to Kynar content.
- Proton conductivity is still very low, and a lot of time is being wasted testing the materials for properties that can be optimized after the conductivity targets have been met.
- Membranes that have been made to-date lack the requisite low humidity conductivity. A conductivity of two mS/cm at 50% RH is unacceptably low. New membranes have a conductivity less than that of Nafion® for all values of RH. BPSH/Kynar blends do not appear to be promising; a highly charged water soluble BPSH must be crosslinked to prevent solubilization, and this will undoubtedly lower proton conductivity and make the polymer highly brittle. It is unclear why durability tests were performed on the M41 membranes when they have insufficient proton conductivity.
- The approach presented has not met low RH conductivity of even Nafion®, and remains far from DOE targets. Performance of membrane materials in MEAs is not presented particularly for durability studies. The comparably funded project by 3M shows greatly improved conductivity and extremely long durability. Membrane development on the materials presented by this lead organization has been funded by DOE for several years, and the current level of advancement is disappointing for the significant investment.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.0** for technology transfer and collaboration.

- A stack manufacturer would add to the project team capabilities.
- Collaboration was not addressed.
- It is not clear what, if any, contributions were made by the University of Hawaii.
- They could benefit from much more fuel cell functional property testing.
- The collaborations provide access to many ionomer and catalyst materials for membrane and MEA development.
- The transmission electron microscope (TEM) work at Oak Ridge National Laboratory provides useful insight into blend morphology and miscibility.
- Hawaii's role is unclear.
- The project team exhibited a good mix of university, national lab, and industrial partners.

- There are quite a few collaborators on this project. Most appear to be peripherally involved with the work, and do not directly assist in formulating new membranes and new membrane morphologies (e.g., they provide polymer catalyst samples or they do TEM studies; membrane fabrication and morphology control studies are done solely by Arkema).
- The project has strong partners, such as Johnson Matthey of Oak Ridge and Jim McGrath; however, it is not clear how Johnson Matthey interacts.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.0 for proposed future work.

- For the time left on the project, the future plans are appropriate. The latest formulations will be optimized and more completely characterized.
- There is a lack of clarity of what to do next, or lack of intention to come closer to the DOE targets.
- The team seems to be satisfied with the commercial viability of current status.
- Testing should be done extensively now on their best membrane option in fuel cells as a function of membrane thickness as well as polymer composition and morphology, and fuel cell testing conditions (temperature and RH) to really establish a benchmark of this technology approach and reveal both operating windows of opportunity and material issues and gaps.
- There was nothing specific in the future work plans to suggest they have a good idea for how to improve low RH performance.
- Emphasis should be put on using highly conducting ionomers, and understanding proton conductivity from -20° to 120°C at dry conditions. A detailed understanding of morphology, chemistry, and transport is needed so that the conductivity in these materials can be understood and optimized.
- The PI could not explain why the proton conductivity of Arkema membranes with the low EW polymer was much lower than that of Nafion®. Without a firm understanding of why the Arkema membranes do not perform up to expectations, it is difficult to see how improvements can be made.
- Future work focuses on many of the same ("continued") studies. The specific approaches proposed are vague and seem to hinge on the hope that crosslinkable end groups could allow higher conductivity and reduced water uptake by using BPSH-100. It seems highly unlikely that this change alone would result in the significant advances necessary to yield new materials with competitive properties.

Strengths and weaknesses

Strengths

- Large quantities appear to be easily produced.
- The project team shows a strong technological understanding of polymer properties and processing, particularly with their core technology, Kynar.
- The project team performs durability testing.
- The project team has access to McGrath ionomers.
- A low cost potential exists.
- PVDF blending capability exists.
- Arkema and the PI are experienced in using PVDF in various types of membranes. PVDF is an attractive material for blended fuel cell membranes.
- Cheap/chemical resistant polymer systems could lower the cost of fuel cell ionomers.

Weaknesses

- There are large quantities of membranes of questionable utility.
- There has not been enough functional property and durability testing in full MEA configurations to understand water management properties.
- There has also been very little progress towards main objective (low RH conductivity) with little demonstrated fundamental understanding as to why conductivity is so low.
- The project team made a poor choice of ionomers for blending.

- Proton conductivity on new membranes is very low (much lower than Nafion® 212). A credible strategy for overcoming this weakness was not presented.
- Fuel cell testing has been too limited, and the approach taken has not addressed a key issue of low conductivity at low RH.

- It is not advisable to continue this project in view of poor results and the lack of a clear path to improvement.
- The project team should work with McGrath's partially fluorinated block copolymers.
- The project team should do more fuel cell testing (H₂/Air) with these materials as conductivity can be misleading.
- The project team should also concentrate on meeting proton conductivity targets first. A go/no-go point that requires a material substantially improved versus Nafion® 212 should be developed.
- No additions/deletions to the project scope are recommended.
- The team should focus on higher acid content polymers and crosslinkable end groups, and it should stop studies with ionomers with lower acid content than BPSH-100.

Project # FC-13: Membranes and MEAs for Dry, Hot Operating Conditions Steven Hamrock; Fuel Cell Components

Brief Summary of Project

The objective of this project is to develop a new proton exchange membrane (PEM) with higher proton conductivity and improved durability under hotter and dryer conditions compared to current membranes. Fuel Cell Components is developing new membrane additives for both increased conductivity and improved stability/durability under dry conditions. Experimental and theoretical studies will be conducted of factors controlling proton transport both within the membrane and mechanisms of polymer degradation and membrane durability in a membrane electrode assembly (MEA).



<u>**Ouestion 1: Relevance to overall DOE**</u> <u>objectives</u>

This project earned a score of 3.8 for its relevance to DOE objectives.

- Membranes that can operate stably with high conductivity over the entire range of expected temperature and humidity are critical to DOE goals.
- The project is in alignment with DOE's objectives.
- The project addresses all the relevant DOE targets of looking at membrane performance at high temperature/low RH and durability.
- Improved membranes with wider operating window, higher performance, and better durability are critical to the Hydrogen Program.
- This project is highly relevant to the Hydrogen Program. It systematically seeks to attack the issue of conductivity at low humidity by lowering membrane equivalent weight (EW), while still maintaining a mechanically stable structure.

Question 2: Approach to performing the research and development

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

- 3M is paying a lot of attention to durability under cycling conditions.
- 3M is addressing composition, additives, synthesis, and manufacturing methods, and theoretical analysis to support the project. It is not clear how or if these approaches will be down-selected or combined.
- The approach to nailing down and optimizing a system appears to be too broad. The number of options should be narrowed down after performing appropriate risk analysis.
- The project team is looking at stability, durability, and performance. It builds on strengths of the current 3M ionomer, while investigating the limits of that chemistry. The combination of theory, synthetic work, and characterization help understand effects of crystallinity, EW, and acidity on conductivity and physical properties.
- The approach covers all critical areas and provides good balance between new materials development, fundamental understanding, scalability, and stack performance.
- The approach is very ambitious in terms of the broad range of materials and additives being investigated.
- The approach appears complex, but is very simple: reduce EW while maintaining mechanical stability, which is the essence of most of the decent membrane projects.

- The approach makes room for all the major techniques for achieving the above: blends, polymer modification, reinforcement, and crosslinking. Not all these methods receive equal treatment, however.
- The polymer modification part is the best conceived. As groups are added as chemical handles (increasing EW), more conductive groups are added (returning to low EW).
- Other efforts (SSC, tethered HPAs) are interesting, but not yet integrated into the mainstream of the project.

<u>Ouestion 3: Technical accomplishments and progress toward project and DOE goals</u></u>

This project was rated 2.8 based on accomplishments.

- Membranes have been fabricated that approach 0.1 S/cm at 50% RH and 80°C.
- Mechanical stability under RH cycling is a serious issue.
- Reasonable progress has been made. The team needs to narrow down the scope to start hitting project goals.
- The team has demonstrated membranes in an MEA with durability exceeding 10,000 h. They have determined EW-conductivity relationships for the baseline 3M ionomer. They have prepared derivatives of the 3M ionomer for bis-acids with increased conductivity compared to the base ionomer with the same backbone.
- The project has shown conductivity meeting DOE 2010 targets with low EW polymers.
- The project has given impressive fuel cell durability data.
- The team is meeting or approaching conductivity targets, although mechanical and chemical stability are still a concern.
- The synthesis progress in its own right has been good, although bottom-line conductivity results have not been delivered. For example, the ortho/meta bis-acid membranes have been fabricated, but conductivity measured at 80°C/50% RH is still 40-60 mS/cm.
- An urgent need still exists to show that the ortho/meta bis-acid membranes have not become low enough EW that they are on the "wrong side" of the solubility threshold.
- Electron spin resonance (ESR) shows that the base 3M 825 EW membrane is more resistant to radical attack, but it is not entirely understood yet why.
- Additive A appears to extend lifetime, but the test used for lifetime is still more of a load cycle than a drive cycle.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated 3.7 for technology transfer and collaboration.

- The team includes an auto original equipment manufacturer (OEM) and all other disciplines necessary.
- The project appears to have strong collaboration, but the scope of collaboration appears too broad for the given scope.
- The project team has collaborated with several top-name people.
- The collaborators have provided important input and feedback.
- 3M has assembled a wide range of collaboration, some of which is more useful than others.
- GM, BekkTech, and the University of Detroit-Mercy all have clear and necessary tasks.
- Colorado School of Mines (CSM) and the University of Tennessee (UT) are providing solid contributions modeling morphology, but it still remains up to the project leadership to integrate this better into the rest of the project.
- Case Western Reserve University (CWRU) appeared to be used early on to look at proton diffusion at low humidity, but this has not been repeated yet for the newer membranes.

Question 5: Approach to and relevance of proposed future research

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

• The team is addressing mechanical durability and water solubility by several approaches including theoretical analysis.

- The scope of future work is too broad, and requires a more holistic approach to meet all requirements as opposed to targeting individual product attributes.
- The team needs a comprehensive plan to address the barriers by characterizing various important underlying aspects of performance and durability.
- The team clearly needs a lot of planning for future work. Their strategy is very ambitious essentially to examine every possible route to improve performance and stability they can think of, which is a lot; if they are able to achieve this level of effort, then good results are likely.
- There are a lot of tasks in the future work, split amongst a number of parties. The challenge will be to create some coherent sense out of all the disparate parts.
- The emphasis needs to remain on making a stable, high-conductivity, low-EW membrane. Towards this end, the morphological studies need to be getting used to better direct the polymer modifications. Reinforcement may be important at some point if the chemical stability offered by additives is not sufficient. The project's success will depend on keeping about 7-8 "plates spinning."

Strengths and weaknesses

Strengths

- The project team has taken a diverse approach.
- The team has used a good combination of synthetic work, characterization, and theory.
- The team has taken several parallel and complimentary approaches to increasing performance and durability.
- They have used interesting and systematic work to develop new polymers.
- Investigators have lots of ideas, and are effective in the examination of multiple strategies for improved performance and durability.
- The team has come up with creative, well-conceived ideas for polymer modifications.
- The team has exhibited a wide range of collaboration that includes modeling, *in situ* testing, and analytical expertise.
- Background demonstrates deep understanding of what the problems are.

Weaknesses

- The cost discussion is weak.
- The scope is too broad.
- The project has not yet delivered a higher conductivity membrane with lower EW that has been shown to not be soluble.
- The project contains a lot of separate parts that have not all delivered as much as can be expected.
- There could still be a number of issues that are, as of yet, untouched in the material presented (swelling, leaching, catalyst poisoning, etc.).

- The team needs to ensure consideration for high-volume manufacturing and cost, while staying focused on solving technical problems.
- The blends made so far do not look promising; the team should consider establishing a go/no-go milestone for this approach in 2010.
- The project should remain focused on polymer modifications and morphological refinements until conductivity goals are met, with a secondary focus on the solubility/RH cycling of the highest conductivity/lowest EW materials made to date. All activities are details in comparison.

Project # FC-14: New Polyelectrolyte Materials for High Temperature Fuel Cells

John B. Kerr; Lawrence Berkeley National Laboratory

Brief Summary of Project

The objectives of this project are to 1) develop knowledge that leads to materials that can meet the performance needs at acceptable cost; 2) create fuel cells that operate efficiently without external humidification at operating temperatures between -40°C and 120°C; 3) develop durable water-free membrane materials that meet the 2015 DOE targets as set out in the Multi-Year R&D Plan; 4) achieve conductivity of 0.1 S/cm at operating temperatures ($\leq 120^{\circ}$ C) and inlet water vapor partial pressures <1.5 kPa; 5) demonstrate durability with cycling >5,000 hours at over 80°C; and 6) achieve oxygen and hydrogen crossover currents ≤ 2 mA/cm^{2} .



Question 1: Relevance to overall DOE objectives

This project earned a score of 3.7 for its relevance to DOE objectives.

- A non-water dependent proton conducting membrane, which works well from -20°C up to 120°C, would be an amazing breakthrough for the fuel cell community.
- The project is relevant to the high-temperature membrane development requirement to meet overall DOE objectives.
- The goals and objectives of the project are designed satisfactorily.
- The multi-year plan is in line with DOE RD&D objectives.
- Anhydrous proton-conducting membranes can enable higher temperature fuel cell operation enabling potentially simpler and less expensive systems.
- The work is in line with task 1, barriers A, B, and C, to develop membranes that meet all targets.
- Membranes that can operate in a wide range of conditions, including 120°C and low relative humidity (RH), are critical to meeting the performance and system cost targets for stationary and automotive fuel cells.
- The objective of this project, to fabricate durable membranes that operate in a fuel cell without external humidification at moderate temperatures (at most 120°C), is highly relevant to DOE.

Question 2: Approach to performing the research and development

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

- The approach is sound with a strong synthetic approach to prepare tethered imidazole membranes. The PI understands the strengths and shortcomings of imidazole and is making the necessary adjustments.
- The blending of imidazole with perfluorinated sulfonic acid (PFSA) type materials, however, seems rather banal compared to the rest of the program, and it is unclear how much value it is in both the near term and long term.
- The project has been designed appropriately to address the technical barriers of low RH conductivity.
- The approach of the project is good and its outcome will address some of the key technical barriers.
- The technical feasibility of the synthetic route to an oxidatively stable ionomeric polymer is doubtful.
- The oxidative stability of the new polymer should have been considered.
- The grafted imidazole polymer possesses benzylic hydrogen, which may affect the durability of the membrane.
- Developing a material that can use Grotthus-hopping within a polymer is a unique approach.

- This is a truly novel approach borrowing heavily from technology used in the battery industry. The team could potentially develop a truly anhydrous proton conductor.
- The water-free membrane is a very ambitious goal, and the primary focus on synthesis and conductivity is appropriate.
- The use of membrane-bound (grafted) heterocycle acids and bases, ionic liquids, and doped polymers is not a new approach; other researchers have investigated similar approaches, with limited success. It appears that the PI is mostly working on polymer synthesis.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **2.3** based on accomplishments.

- For the amount of funding this project has received, it appears that the rate of progress is quite low. An undergraduate summer student performed the largest amount of work. While the reviewer understands the difficulty of synthesis for this type of chemistry, the work thus far is lackluster.
- The conductivity efficiencies of proposed new polyelectrolyte material are not known.
- If the new polyelectrolyte functions, as proposed by the team, then the benefit would be immense.
- The blend of Nafion® and grafted imidazole polymer may not possess phase compatibility.
- The technical progress had been good, and the development plan is satisfactory.
- Polymers were made to enable conductivity measurements, which is a major step forward from last year.
- Conductivities with free-acid electrolytes are below DOE target conductivity at 120°C, and significantly lower at lower temperatures. No rationale is given to lead one to expect that the systems with polymer electrolytes would have higher performance. Nafion®-containing membrane conductivities are still at least an order of magnitude too low.
- Modeling results do not clearly establish that DOE target conductivities are possible with this approach.
- Oxidative stability to Fenton's test was done, but not sufficient to prove oxidative stability.
- As would be expected from this team, there is a large amount of good science. Unfortunately, the conductivities achieved are still rather low.
- The project team has made good progress, but membrane conductivity needs significant improvement to reach targets.
- The PI's results are not particularly encouraging. The measured proton conductivities are generally low (too low for the membranes to be used in a fuel cell).

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.3** for technology transfer and collaboration.

- Collaboration can be seen with LANL for the membrane electrode assembly (MEA) fabrication, and 3M for their PFSA material; however, the overall level of effort seems to be quite minimal.
- The project has good collaborative effort for the proposed development work.
- There has been good technical interface and information-sharing among the concerned groups.
- The project has an industrial partner as well as other national laboratories.
- Overall, the project possesses good teamwork.
- The team with 3M, LANL, and Berkeley provides a comprehensive skill set including world class materials, synthesis, modeling, and characterization capabilities.
- It appears as though this team of national lab/university/industry is working very well together.
- There has been good interaction with partners and analytical facilities.
- There is some collaboration with other national labs, such as ORNL and NIST. Michael Guiver of the National Research Center in Canada provided polymers for testing. More collaborations are needed. Why not collaborate with other high-temperature membrane researchers?

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- Proposed future work is in line with the project. The addition of molecular dynamics modeling to understand the potential proton conductivity limits of this material would be highly useful for the go/no-go decision.
- The future research with imidazole end group modification of PFSA polymer would not give a stable membrane.
- The siloxane modified route to new polyelectrolyte is not expected to give an oxidatively stable polyelectrolyte.
- Working with 3M, low EW PFSA and functionalized block copolymers to blend/attach/copolymerize with imidizoles is a good path forward.
- Fenton's test is not recommended. The team should wait until stable membranes can be made and then run DOE open circuit voltage test.
- Approaches towards practical proton conductivities should be emphasized. There needs to be more studies involving water and fuel cells that make water, because it will not do any good if an anhydrous proton conductor is developed that is sensitive to moisture in a bad way.
- It is a good plan with go/no-go decision criteria in place.
- It is not clear if the proposed future work will produce a usable fuel cell membrane. It is doubtful that a conductivity of 0.1 S/cm at 120°C and <25% RH can be attained by continuing to attach imidazolium groups to polymers. The PI did not explain the connection between the future work on membrane fabrication and the future work on the oxidative stability of polymers. Why work on the latter when the membrane properties are not yet at DOE targets?

Strengths and weaknesses

Strengths

- The PI is a world leader and this project is a great concept that, if successful, will be very valuable to the fuel cell community.
- It is a good group with a solid knowledge base of fuel cell fundamentals.
- There is good interaction between the collaborative groups.
- The team has taken a good fundamental approach in understanding the major technical barriers of low RH proton conductivity.
- The team has exhibited excellent collaboration.
- The team has improved and reduced focus from last year to imidizole/sulfonic acid systems.
- The team's very high level of scientific understanding should lead to a truly unique material that could be a practical proton conductor.
- The team is using novel polymer chemistry combined with known materials and techniques.
- The PI has a good reputation in the field of membrane fabrication. The PI has pursued many different approaches.

Weaknesses

- Progress to date with this level of funding is quite low.
- No measures have been in place to evaluate the oxidative stability of the proposed electrolyte materials.
- The new polyelectrolyte material may be interesting to study, but it is not expected to give the long-term stability under fuel cell conditions.
- The oxidative stability of the polymers developed in this project is doubtful.
- The materials-to-date are still relatively poor conductors.
- The materials do not run well when wet.
- There is not enough thought towards practical proton conductivity.
- The proton conductivity results to-date are not encouraging. The PI's primary approach (tethering imidazoles to polymers with acidic functional groups) has not produced interesting materials with the potential for use in a fuel cell.

- The work should continue as planned. The hiring of motivated/well-qualified scientists for this project is highly important for the overall success. To-date, the work done is below where it should be.
- The PI should consider evaluating the oxidative stability of any new polyelectrolyte material.
- The backbone change to manipulate the polymer morphology should be reconsidered, and the approach should be changed to synthesize oxidatively stable polyelectrolyte.
- The team should include mechanical and swelling characterizations of membranes.
- The team should study conductivity versus RH, not just temperature. The materials might actually work okay at 50% RH, but there is no data.
- Modeling efforts should be increased to understand fundamental limits of the approach.
- The project team should look at the effect of moisture.
- It is not clear why the PI is looking at the oxidative stability of membranes with imidazolium groups when such membranes do not possess the requisite proton conductivity for use in a fuel cell. Similarly, MEA fabrication studies with hydrocarbon polymers appear to be premature. The PI should focus on new membranes with properties (e.g., proton conductivity) that are much improved as compared to what has been achieved so far on this project.

Project # FC-15: Development of Novel PEM Membrane and Multiphase CFD Modeling of PEM Fuel Cell Susanta K. Das and K. Joel Berry; Kettering University

Brief Summary of Project

The overall objectives are to develop a 1) novel proton exchange membrane (PEM) for fuel cells; and 2) multiphase computational fluid dynamics (CFD) model of a PEM fuel cell for improved water and thermal management. Objectives for 2008-2009 were to 1) perform real-time membrane testing for a single cell, and stack and real-time testing for stability and materials properties for a low-cost, highperformance membrane; and 2) conduct a performance evaluation for different parametric conditions for the integrated multiphase CFD model for PEM fuel cells.



Question 1: Relevance to overall DOE objectives

This project earned a score of 2.3 for its relevance to DOE objectives.

- This project partially supports the Hydrogen initiative and the DOE RD&D objectives.
- The major focus of the project should have been on the membrane conductivity at low RH, high temperature (120°C, 50% RH). There is no consideration of this goal.
- Membranes are not tailored for automotive (dry) conditions.
- CFD tools are not novel.
- The membrane development work addresses the DOE R&D objectives. The researchers have not made it clear how the work on the control strategy for the stack addresses the barriers listed.
- The project looks at new membranes that need water for conductivity. CFD modeling was used to predict water flow in cell to optimize performance. This project does not try to make a membrane that works at reduced RH.
- Membranes for high-temperature operation and water and thermal management are important issues and are aligned with DOE goals and objectives.
- The project goals of developing a novel PEM for fuel cells and developing a multiphase CFD fuel cell model for improved water and thermal management are highly relevant to DOE. These two project tasks are very different, and one does not support the other.
- The project title and intent was definitely relevant, but the execution was not because the membrane work was too early stage and the CFD modeling was simplistic.

Question 2: Approach to performing the research and development

This project was rated 1.7 on its approach.

- The nature of the membrane material and its synthetic approach is not clear.
- The CFD approach to provide a better gas diffusion layer (GDL) and flow channel designs is good.
- More information on the membrane material would have helped in understanding the approach to solve the membrane conductivity barriers.
- Membrane materials were not disclosed, so synthesis approach cannot be effectively judged.
- The conductivity method is questionable and, after three years, has still not been benchmarked against accepted direct current (DC) or impedance-based methods.
- The results were not reported in units that allow comparison to literature data or those from other DOE projects.
- Conductivity model fits data too well to be meaningful and provides no fundamental basis.

- It is not very clear from the information presented if these materials are suitable for use in PEM fuel cells.
- The measurements are unusual and probably not useful. The induction time just shows that the membranes are very swellable and take up lots of water. The approach is not designed to solve DOE's stated goals.
- Non-standard approaches are used for proton conductivity presented in nonstandard units. This makes it difficult to compare to other materials. Unusual dependence of resistance on temperature observed for Nafion® suggests there is more being measured with their technique than membrane resistance; others do not see this unusual dependence. The membrane properties desired are higher conductivity under hotter-drier conditions. Their technique does not allow one to determine conductivity under drier conditions.
- Chemical and mechanical durability of membranes are also important issues. They are not addressed at all here. Without any knowledge of the chemistry of the membrane begin presented, durability is of concern.
- The modeling appears to lack validation with experimental data.
- It was very difficult to assess the PI's approach. Very few details were presented regarding the composition of the PEM. A standard AC impedance method was not used to characterize the membrane, for reasons not well understood. It was not clear how the PI's CFD modeling work differs from that of other investigators.
- The first half of the presentation was on membrane development, but membrane chemistries were not revealed nor were the membranes compared to others (except for a simplistic and erroneous comparison to Nafion® 212). The error was a comparison based on electrolysis (for conductivity and resistance) rather than direct fuel cell comparisons. What about comparisons to other polymer systems, too? And why not use accepted fuel cell characterizations?
- CFD's work, as explained during the presentation, was simplistic and the questions from the audience were not answered satisfactorily.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 1.7 based on accomplishments.

- The conductivity of styrene-acrylonitrile-vinylsulfate (SAS)-type membranes should have been measured in true fuel cell conditions.
- The measurement of proton diffusion is not the correct way of measuring the proton conductivity of the membrane.
- The proton transport can happen if there is a pinhole in the membrane leading to leakage of HCl acid to the water chamber.
- No temperature or RH cycling work was done.
- No new membrane data was shown since last year's review except one set of conductivity curves from BekkTech.
- No new dynamic model simulation data was shown either.
- Based on the data presented in the "conductivity test" developed by Kettering, they may be measuring the permeability of the membrane to chloride. The conductivity data from BekkTech was far below the target.
- Measurements seem to be inappropriate and most unusual. Conclusions drawn are not straightforward and probably incorrect. Membranes need to have conductivity measured by standard methods. The PI seems to pay no heed to other groups working in the field.
- Measurements suggest they may have very good conductivity under high (100%) humidity conditions. This is not the area of interest. There also appears to be some other factors being measured, since the temperature dependence of the Nafion® resistance shows an unusual temperature dependence not observed in other studies. It is not clear that they have isolated protonic mobility from other mobile species (Cl-) in their measurement. Initial tests from BekkTech show less than stellar conductivity under lower humidity conditions.
- Some results (e.g., the dependence of temperature on the maximum in the proton transfer capacity) could not be explained. It was not clear how water uptake was measured, or if the PI's membrane is a satisfactory proton conductor.
- Solid work, if we were focused on developing HCl electrolysis membranes, but our focus should be on fuel cell membranes. The speaker revealed that no fuel cell tests were conducted on these membranes and no fuel cell data (of substance) was presented.
- Unusual and unexpected "humps" in the Nafion® 212 resistance measurements were not explained when the question was posed. There may be experimental errors in the resistance curves were presented.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **1.3** for technology transfer and collaboration.

- There is no collaboration with any of the leading national labs.
- There is no interaction with any other academic institutions.
- Bei-Tech provides polymer membranes. No other collaborations were evidenced.
- The researchers need to work with someone who has expertise in evaluating fuel cell membranes.
- Collaboration is non-existent.
- BekkTech was mentioned as a collaborator, but the team was having difficulty getting conductivity measurements from them, so collaboration appears to be ineffective. They do not appear to have other collaborators. Collaboration with others in the field would be useful in validating their measurement technique or in allowing them to measure their membrane's conductivity using standard techniques/protocols.
- There was essentially no collaboration. The slides list Bei-Tech as a collaborator for polymer membranes, but their role on the project is not clear.
- The only collaboration involved the Bei-Tech membranes, but beyond the supply of the membranes, no useful data (to build further work on) was presented.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.1 for proposed future work.

- The crosslinked membrane stability in the fuel cell environment may be a problem.
- The team needs to do temperature and RH cycling to understand membrane stability.
- The conductivity must be reported in S/cm or resistance as ohm-cm² and compared to literature data.
- Membrane durability should be considered and evaluated (mechanical and chemical).
- CFD work needs a stack partner for relevance and validation.
- Future plans are not well developed. No viable follow-on is proposed.
- The project is near completion and plans are to wrap-up.
- There was no connection between present results and future work. For example, the PI lists as future work: polymer crosslinking to make the membrane chemically inert towards reactant gases; however, no data were presented showing polymer degradation. Also, it was not clear why the PI wants to map water history.
- The approach was good because the team was finally going to move to fuel cell tests.

Strengths and weaknesses

Strengths

- The project team conducted good CFD modeling work.
- There was industry engagement in membrane fabrication.
- The CFD modeling work appears to be sound, although the reviewer is not an expert in this area.
- The team had a solid defense of their system as an electrolysis membrane based on a single-cell test.

Weaknesses

- The team lacked the use of proper membrane evaluation methods.
- There was no fuel cell performance data for SAS membranes.
- There is a need to get true conductivity data of SAS membranes.
- Based on the data presented, the "conductivity test" developed here may be measuring the permeability of the membrane to chloride. The conductivity data from BekkTech was far below the target.
- The approach is wrong. The execution appears to be done in the absence of checking with the literature. The nature of the membrane is not revealed. It appears to be a mystery. If this is patented, then why is there not a proper description of the membrane materials? The project does not try to address DOE goals.
- There was a lack of information on the proprietary membrane composition. Unusual non-standard techniques were used to evaluate membranes under conditions that we are not very interested in. These techniques cannot obtain information on proton conduction in the region we are most interested in.

- The membrane fabrication and characterization work is weak. Critical details were missing from the presentation. Standard methods for evaluating the membrane were not used. Membrane performance that was presented was confusing and could not be explained adequately.
- Apparently, very early-stage work was done with no cross-references to an established body of literature on the many polymer chemistries explored by other researchers. A defense of why they chose their membranes (with a revelation of the chemistry) would have been a necessary first step.
- Work displayed shows relative "immaturity" of the team in this arena.

- The team should include real fuel cell and membrane conductivity measurement methods in this project.
- The project is to end next month. Please do not renew.
- This project should be discontinued.
- There is a disconnect between the membrane fabrication work and the CFD modeling. My recommendation is that the membrane work be dropped from this project.
- Drop funding for this effort, unless further basic background work is conducted or presented. Include a body of past literature research and why they chose the membrane for scale-up relative to the many membrane developments already underway. The team should also include fuel cell test data.

Project # FC-16: Applied Science for Electrode Cost, Performance, and Durability

Christina Johnston, Eric Brosha, Fernando Garzon, Andrea Labouriau, Marilyn Hawley, Rex Hjelm, Yu Seung Kim, Kwan-Soo Lee, Nate Mack, Bruce Orler, Tommy Rockward, Dennis Torraco, and Cindy Welch; Los Alamos National Laboratory

Piotr Zelenay and Karren More; Oak Ridge National Laboratory Bryan Pivovar; National Renewable Energy Laboratory Hui Xu; FuelCell Energy

Brief Summary of Project

The overall objective of this project is to assist the DOE Hydrogen, Fuel Cells & Infrastructure Technologies Program in meeting cost, durability, and performance targets by addressing issues directly associated with electrodes. For 2009, LANL will 1) explore the impact of solvent choice in catalyst ink on fuel cell performance; 2) relate the structural and chemical properties of the ionomer in different inks to electrode performance and structure; and 3) initiate use of bilayer/gradient structures in electrodes.



<u>**Ouestion 1: Relevance to overall DOE**</u> <u>objectives</u>

This project earned a score of **3.3** for its relevance to DOE objectives.

- The objectives of this project align well with the DOE program. Membrane electrode assembly (MEA) optimization is often an overlooked area of research, which can provide substantial performance and durability improvements without changing the basic components, such as membranes, electrodes, ionomers, and gas diffusion layers (GDL).
- The project directly addresses a fundamental understanding to correlate electrode processing with performance to provide insight for improved performance and durability, and to a lesser degree, potentially reduced cost.
- A fundamental basis for optimizing electrode structure based on physical properties of the components is critical for developing new electrode materials.
- Work is in line with task 2, barriers A, B, and C, to develop electrodes that meet all targets.
- This program is indeed relevant to the development of polymer electrolyte membrane fuel cell technology, and thus to the overall DOE Hydrogen Program. Understanding the catalysts' ink composition and the required catalyst structure to achieve the highest platinum utilization is one of the key components in designing the MEA. Most project aspects are aligned with the Hydrogen Program and DOE RD&D objectives.
- The project seeks to address the cost and durability of proton exchange membrane fuel cells (PEMFC) by examining parameters associated with catalyst inks. Since catalyst inks have not been dismissed as irrelevant to commercial PEMFCs, this project is still relevant. However, DOE should carefully think about the extent to which a catalyst ink processing project will achieve results that meet commercialization targets. The results of this project will be incremental at best.

Question 2: Approach to performing the research and development

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

• The overall approach is very strong, and the area researched is very interesting with a lot of valuable information for the fuel cell community.

- The biggest weakness of this approach is the focus solely on decal LANL-type MEAs. Many companies are now also working on spray, die-coating type, and screen-printing type electrodes, and the reviewer wonders how transferable the findings here are.
- The team has taken a well-defined approach that provides good baseline systems for understanding correlations at the particular length scale of each technique, while enabling integration of the knowledge across broader length scales. The approach includes both baselining activities and experiments designed to test the theories that emerge from the initial studies. The approach does a great job of isolating individual effects (e.g., ionomer structure, catalysts, gradient structures) and their impacts to develop a complete understanding of the complex electrode structure. Electrode processing materials and methods are relevant for practical fuel cell systems, so that results from this fundamental research project should be readily transferred to more applied projects.
- Studying the effect of ionomer and solvent on performance is a valuable approach.
- Characterizing the ionomer properties is also valuable.
- More focus should be applied to studying the electrode pore structure and the structure of the ionomer within the electrode.
- Looking at degradation and changes in electrode properties over time/cycling is a positive approach.
- The industrial manufacturers have almost certainly optimized their electrode assemblies using the tools available to them. This project needs to emphasize the advanced instrumentation that can only be used at national labs, rather than repeating anything empirically.
- The approach developed for probing structural properties of MEAs is multifaceted and, if utilized effectively, could contribute significantly to overcoming technical barriers related to structural properties of ionomers. The authors used a variety of spectroscopic and structural techniques to probe the effect of solvents on physico-chemical properties of Nafion[®]. Hopefully in the future, equally successful probes will be developed to probe the stability of the catalyst layer itself. Therefore, the approach could be improved.
- The conclusions derived (especially about mobility and solvent interactions) could have been derived equally as well without resorting to the sophisticated methods employed.
- It is not clear as to what has prompted the choice of solvents.
- Many in the industry will expect that this project is too applied of an effort for government funding; in other words, it should be expected that MEA manufacturers will understand performance and durability differences that stem from processing. Polarizations and transmission electron microscopy (TEM) images that show very subtle differences based on solvent choice, confirm the suspicion that ink processing studies will result only in incremental improvements that ultimately do not address the commercial PEMFC targets.
- Despite the implications towards long-term targets, the researchers themselves excellently crafted a hypothesis and the means by which to test it. The hypothesis was that ionomer history would be preserved on a molecular scale, and that the effect of this would be realized in fuel cell testing. In this sense, the approach was simple and elegant. Perhaps more importantly, the approach was universal and can be applied for all ink processing.
- Results from the bilayer task were somewhat expected. More interesting results might be produced by looking at lower ionomer content in different parts of the catalyst layer, rather than higher.
- In my opinion, this is a weak part of the program. The success will be measured based on understanding the properties of the catalysts layer, rather than the ionomer itself. The authors should focus more on degradation of platinum catalysts including platinum bi/multi-metallic surfaces.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.0** based on accomplishments.

- A lot of high-quality work has been done at this time with some very useful data and findings. However, more temperatures and relative humidities should be investigated to confirm/understand their effect.
- The project team has made significant progress in correlating processing solvent with electrode microstructure and validating emerging theories about impact of ionomer structure and mobility. Their experiments are well defined with appropriate interpretation at the relevant length scale. The team is beginning to combine information from several analytical techniques to develop a complete suite of ideal electrode features and the necessary processing materials and methods to achieve improved electrodes. This is a very challenging project that has already demonstrated that there should be approaches to improving both mass transport and kinetic performance.
- Significant progress has been made since last year.

- Performance results have been correlated with ionomer properties based on nuclear magnetic resonance, small angle X-ray scattering, and TEM/atomic force microscopy.
- The solvent type has been shown to affect both performance and durability.
- The value here would be to emphasize the application of advanced instrumentation; solvent effects are well known by the industry. Industry needs to be given detailed mechanisms here for this project to be more useful.
- The results obtained are ambiguous; however, more time should be provided to the team to work through the research objectives and tasks.
- Despite the concerns that the approach of this project is not suitable for government funding, it must be acknowledged that the actual project execution has improved tremendously from the past year.
- The researchers have done an excellent job in attempting to rationalize why certain solvents represent the changes that they see in different regions of the polarization curve, by exploiting NMR, small angle neutron scattering, and AFM. However, where inconsistencies exist (e.g., water), it would be interesting to find some deeper explanations. Are mobility and phase separation truly enough to explain all the data?

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated 2.9 for technology transfer and collaboration.

- Collaboration at this time is limited but fruitful. Excellent characterization of MEAs is ongoing with one of their partners. The team may want to bring on-board an MEA fabricator to help the PI understand in more depth the industrial realities.
- The team leverages other national lab expertise in analytical characterization methods to interrogate relevant length scales over a broad range. The project goals could not be achieved without these interactions.
- Collaboration with ORNL provides opportunity for structural characterization.
- The project could benefit from collaboration with an MEA supplier.
- There needs to be much more industrial collaboration.
- The program is multifaceted and the PI successfully built a team of experts capable to deal with the projected challenges.
- Collaborations exist and are being leveraged.
- ORNL has been used in the past by this project, but this year's work does not obviously show where they contributed.
- The role of NIST is not entirely clear.
- The use of an industrial partner would seem to be an obvious step for this project.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.7 for proposed future work.

- Proposed future work is okay, but it would be nice to see the expansion to different types of MEA fabrication techniques and more in-depth fuel cell studies such as temperature effects, RH effects, and durability.
- The team will build on recent work understanding ionomer structure, mobility relationships, and gradient electrodes by expanding the range of solvent systems, incorporating catalysts into the systems, and adding that layer of understanding.
- The proposed work on Nafion® interactions with catalysts will be very important.
- I would recommend not focusing on multi-layer/gradient structures in favor of the ionomer/carbon/catalyst interactions.
- The plan to look at different carbon dimensions should be interesting. I would like to see different carbon types also investigated.
- Again, emphasize the light sources, neutron sources, and the microscopy at ORNL.
- The proposed work is good, and if executed, may significantly improve the understanding of MEA interfaces.
- The proposed work seems to branch out into numerous directions, and a more focused approach using a single model system would perhaps yield more tractable results.
- There is no end to the parameters to consider in ink processing, but considering the effects of catalyst and temperature is a good start. Stir rate, fluidization dynamics, temperature ramps, and time involved in

temperature hold and drying are just a few more parameters that could be studied (or have been studied, but perhaps not with the rigor demonstrated by this past year's project performance). Unfortunately, the project is boxed into a situation where either a large number of parameters must be studied, or else the results are only relevant to a very particular ink processing scheme.

• It would be significantly better (if an ink processing project must be done) to take an industrial partner's experience and direction and combine that with LANL's analytical prowess (both in terms of instruments and scientific expertise) to begin blocking off whole regions of parameters that are undesirable. Then, begin attacking what the true issues are for processing. Is solvent choice truly something that Gore or DuPont is still agonizing over? It would be interesting to bring them into this and find out, but without allowing the industrial partner to take away the universality of the project.

Strengths and weaknesses

Strengths

- World-class fuel cell team investigating a very important fuel cell topic.
- Great fundamental research designed to provide relevant practical results/impact.
- There is a strong combination of thorough material characterization with fuel cell testing.
- The fundamental approach to understand the dynamic and structural properties over the range of relevant length scales is a strength.
- Builds on LANL's fuel cell expertise.
- This is a well-organized project with a clear goal to understand the MEA interface. However, the question of how much this will help in improving fuel cell performance still remains. The approach used in this work could contribute significantly to overcoming technical barriers related to structural properties of ionomers.
- This is a strong team.
- Analytical techniques have been used to draw simple and elegant conclusions regarding set hypotheses.
- There has been experimental execution.
- The results are interesting and thought-provoking.

Weaknesses

- Due to the inherent strength of the team, the reviewer wonders if the PI looks outside enough to see the changes in the fuel cell community.
- None.
- The focus has been on bulk ionomer properties. The ionomer properties of very thin films in an electrode are likely to be very different, and it would be great if the PI could make progress in better understanding the properties of the ionomer in the electrode.
- The team has spent too much time reinventing the wheel; there needs to be a higher level of science.
- The program should focus more on testing catalyst layers. Furthermore, the PI should focus more on science and less on testing; the latter should be done in collaboration with an R&D program of industrial partners.
- There is a lack of focus.
- The overall premise that this is the right work for government funding is questionable.
- Collaboration with parties who would have a lot to say on the topic is missing.
- There is a lack of confirmation that the parameters being studied are the ones that would have the most impact on cost and durability of PEMFCs.

Specific recommendations and additions or deletions to the work scope

- The reviewer would like to see the expansion to different types of MEA fabrication techniques, such as spray, die-coating, and screen-printing.
- More in-depth fuel cell studies need to be done, such as temperature effects, RH effects, durability, etc.
- More focus is necessary on the impact of process conditions such as drying temperature and rate.
- More focus is necessary on bulk electrode properties such as pore volume and sheet resistance.
- The team should correlate performance losses during voltage cycling with electrochemical surface area loss.
- More characterization is necessary (both ex situ & fuel cell testing) at lower RH.
- The team should emphasize using advanced instrumentation to really study this problem.

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- This is a very good team of people who should be permitted more time to study the issue; however, a more focused approach with fewer variables in terms of materials should be recommended.
- An industrial partner must be added.
- With recommendations from an industrial partner, the team should focus the project on the true issues in ink processing and figure out what parameters could make the greatest impact on cost and durability of PEMFCs.
- Perhaps the project could be changed entirely to look at new electrode structures for ultra-thin catalyst layers. Is it always true that a catalyst layer of 1 µm or less must be plagued by flooding at lower temperatures? That would be more interesting in a world where catalyst loading has significantly decreased. To what extent do large and small hydrophilic pores help or hurt water transport? Are hydrophobic pores necessary to avoid flooding?

Project # FC-17: Advanced Cathode Catalysts and Supports for PEM Fuel Cells *Mark K. Debe; 3M Company*

Brief Summary of Project

The overall objective is to develop a durable, low-cost, high-performance cathode electrode (catalyst and support), that is fully integrated into a fuel cell membrane electrode assembly with gas diffusion media, fabricated by high-volume capable processes and is able to meet the 2015 DOE target. The objectives of this project for the past year were to 1) define and implement multiple strategies for increasing nanostructured thin film (NSTF) support surface area, catalyst activity, and durability, with total loadings of <0.25 mg-Pt/cm²/membrane electrode assembly (MEA); 2) work closely with subcontractors to fabricate and screen new electrocatalysts using high throughput characterization



methods for activity and durability gains; 3) conduct fundamental studies of the NSTF catalyst activities for oxidation reduction reactions; 4) apply more severe accelerated tests to benchmark the NSTF/MEA durability; 5) define and implement multiple strategies to optimize the MEA water management; 6) advance the high-volume roll-good NSTF catalyst/membrane integration; and 7) work closely with system integrator to validate NSTF functional properties/issues.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.7** for its relevance to DOE objectives.

- This project is critical to the Hydrogen Program. The research addresses all major objectives of the program, and results are above the DOE targets on several technical barriers.
- The project is critical to the Hydrogen Program as it focuses on implementing DOE goals on development of new durable supports and highly active catalysts. It is highly relevant because it offers a new approach to MEA development and manufacturing.
- This project is extremely relevant to the DOE goals for reducing the cost and improving the performance and durability of polymer electrolyte fuel cell (PEFC) power systems. These are the critical barriers to implementation of PEFC power systems for a variety of applications.
- This program is forging ahead toward the metrics needed for fuel cell catalysts needed to transition an affordable and durable fuel cell vehicle. They have made outstanding progress on all fronts. The main problem with the program is that the DOE metrics might be somewhat irrelevant for this class of catalysts where the water management at high power is the major problem for transition. That is, they might be able to achieve the activity and durability metrics, but the catalyst morphology may not be inherently useful for vehicle applications. Mark Debe said, though, that they are moving toward more focus on water management issues and explained that they needed to work on the scale-up first before they could answer this problem meaningfully. With more time, hopefully the water management problem will be addressed.
- The 3M project is dead-on target to address the necessary targets that make fuel cells commercial, low cost, high performance, and of extended durability.
- Thanks to making improvements to MEA performance and especially durability its primary target, this project is very relevant to DOE objectives.
Question 2: Approach to performing the research and development

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

- The research is focused on technical barriers; my reservation pertains to the combinatorial design of the catalysts. Stability and surface segregation in these mixtures usually invalidate initial activity. Scientific support for selecting the compositions is desirable, in particular for binary catalysts.
- The project is well designed and addresses multiple aspects of the development of practical fuel cells.
- The team has taken a good approach, with considerable coordination with other research groups. It would be nice to see more corroboration with stack integrators.
- The approach leverages the higher durability and activity of bulk-like Pt catalysts, but achieves low Pt loadings by dispersing this catalyst structure as a thin film on a high surface area structured support.
- This 3M team has the intellectual and experimental horsepower to address the multiple complexities associated with the scale-up and commercialization of the 3M MEAs. They have a complex approach, which is needed to solve the problems.
- Dr. Debe has shown consistent excellent planning, thoughtful evaluation, and focused direction on a very complex, multidimensional technical problem.
- 3M has pursued the NSTF approach for many years by now. It remains promising, but significant challenges remain; they need to be addressed.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.9** based on accomplishments.

- With the exception of Pt mass activity, results are better than the DOE targets on several technical barriers.
- The project already met almost all DOE targets for 2015.
- The team has shown impressive effort, with combination of fundamentals and in-cell demonstration. The team is asking and answering some key questions about the correlation between ideal catalyst systems and practical MEAs.
- It is important to understand the value of exchange-current density at varying oxide states, allowing correlation between *ex situ* rotating disc electrodes and in-cell MEAs.
- The team exceeded DOE's 2015 inverse power density target for MEA.
- The team met the DOE activity target of 0.44 A/mg platinum group metal (PGM) in rotating disk electrode (RDE) experiments, but not in MEA.
- The team has made excellent progress in developing and improving the activity of MEA, and in meeting the durability targets.
- Tremendous progress has been made within the goals of the DOE metrics.
- Too many reviewers thought the DOE milestones were perhaps too aggressive, but they have been addressed and met, way ahead of schedule and under budget. This is just a success story!
- There was very good high-current response in fuel cell testing, but not so good at low currents, in which range there has been no improvement since 2007 (possibly caused by the lower catalyst loading though). Would higher cathode catalyst loading result in needed better low-current performance?
- It is not clear whether performance improvements distribute equally between the two fuel cell electrodes. Lowering catalyst loading on one electrode rather than both at the same time would provide better insight into the sources of observed improvement.
- What was the reason for using very high catalyst loadings in RDE testing? While obviously helping the RDE data appearance, this seems to be causing lots of confusion in the electrocatalysis community. Testing at lower loadings would be desirable.
- What was the outcome of GM testing? The "real-world gaps and issues" should have been spelled out in this year's presentation, rather than merely mentioned in the summary.
- In spite of previous declarations to the contrary (see responses to 2008 reviewers' comments), there has been little data and no emphasis on water management in the past year.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.4** for technology transfer and collaboration.

- The collaboration with excellent national laboratories and university teams seems quite successful.
- There is close collaboration with universities, national labs, and industrial partners.
- There is very good corroboration and collaboration.
- This is an excellent team with varied and complementary expertise.
- It appears that the choice of catalyst composition is not based on RDE experiments, but on MEA tests, and also that the catalysts are treated differently for RDE and MEA tests (intrinsically acid-washed vs. not acid-washed), so what is the value of RDE experiments? It was also not clear if the post-fabrication processing of catalysts performed at ANL was just for the RDE experiments, or if it is being implemented in the catalysts that are fabricated into MEAs.
- It is not clear if the Dalhousie and Jet Propulsion Laboratory (JPL) work led to the identification of the best of class catalyst (PtM), or if it has guided the choice of catalyst compositions used in the present MEAs or in previously studied MEAs. In particular, the JPL results were not included in presented material.
- The 3M team is very big, but seems to be managed appropriately. The program recognizes how many problems need to be solved to commercialize the 3M membranes, and has the right team to solve the problems. The role of JPL was a bit unclear.
- The collaboration is outstanding, especially because the partners have very clearly defined and supporting roles. One understands that there is one project, and several excellent groups all singing on the same page, but not the same notes. This is another result of Dr. Debe's management style, building a well-functioning team.
- There has been very good collaboration between 3M and Argonne, but the roles of two other partners in the project are not well defined.
- The impact of Dalhousie's University combinatorial testing is unclear. In spite of a significant amount of the Dalhousie data in the presentation, the University seems to have played a rather limited role in the progress achieved.
- There has been little contribution from JPL to the project in the past year. The Laboratory's role should be either enhanced, or its participation re-considered.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- The plans address some remaining obstacles to prototypes construction, including stack testing, durability, Pt mass activity, and water management.
- Future work is carefully planned and built on past progress.
- In light of program success, it is suggested that the team scale back or redirect to go after the remaining problems more aggressively, rather than staying on the current path.
- The biggest challenge with the NSTF-based MEAs appears to be water management; therefore, the future proposed work focused on gas diffusion layer development/optimization is on target.
- The 3M team should put less emphasis on the global DOE metrics for catalysts, and focus on the metrics that are specifically a problem for the 3M MEAs.
- Dr. Debe discussed the need to rethink the tail end of the project, being in the unique position of having pretty much crossed the finish line, well ahead of the pack. The next phases need some careful thinking.
- Focus on water management, including testing under "wet" conditions, is necessary. Results need to be summarized in next year's presentation, which has been long overdue.
- The effect of impurities on NSTF-based catalyst performance, especially at the cathode, should be studied.

Strengths and weaknesses

Strengths

- There are excellent teams at 3M, national laboratories, and the university.
- There are possibilities for large-scale catalyst production.

- The team executed good planning of critical research tasks.
- The team showed creativity in development of new catalysts.
- The team took a unique technical approach with good effort on fundamentals. Explaining the difference between model systems and MEAs is of primary value.
- The greatest strength of the project is the unique approach that combines the stability and area-specific activity of bulk catalysts with a unique support and dispersion technique that allows high dispersions (cm²/g catalyst). Another strength of the project is the rolled-goods approach to manufacturing.
- This is a very good team solving a complex problem.
- Dr. Debe has this wonderful combination of profound theory mixed with a very good pair of highly capable hands.
- The theory suggested new, complex nanostructures, and the hands worked to make those visions real. It was a team effort, and required the technical excellence of a company that made its fortune by coating material rolls of highly controlled uniformity. This success would have been very unlikely outside of the 3M corporate excellence, the people who began with sand paper, "Scotch Tape", and now NSTF electrodes.
- The team showed excellent MEA durability in 3M fuel cell testing.

Weaknesses

- There was a lack of characterization with atomic resolution.
- There are no weaknesses.
- I would like to see more third-party stack verification and characterization of mass-transport issues.
- The unique architecture that allows high dispersions on chemically and electrochemically stable supports also presents challenges in terms of removal of water from the electrode layer. 3M believes they can overcome this challenge with the correct design of the microporous layer (MPL) and gas diffusion layer (GDL). This next year will be critical to see if they can address these challenges.
- It's not clear if the water management problems associated with this class of MEAs can be solved. More research in the next few years should show whether this is possible.
- It is hard to find any weaknesses.
- There is an apparent further delay in addressing water management issues.

- See above.
- The project should focus more on durability issues.
- The reviewer recommends sharper focus on understanding what happens between *ex situ* and *in situ* testing.
- The project should more clearly present how the subcontractors are guiding the development of the catalysts and processing of the catalysts that are actually used in the MEA. For example, it appeared from the presentation that ANL is developing a catalyst treatment to improve the activity of the catalysts in the RDE experiments, but is this implemented in the preparation of catalysts incorporated into the MEAs? Why are the catalysts in the MEAs not acid-treated prior to incorporation into the MEA, when it was stated during the presentation that the majority of the non-noble metals easily leach from the catalysts in an acidic environment? If the catalysts are not acid-leached prior to incorporation into the MEA, then the non-noble metals are most likely being leached during MEA testing and may be poisoning the membrane.
- Funds need to be redirected for the next project phase. This reviewer suggests that tasks that support commercialization of the existing formulations, perhaps leading to simpler or less costly manufacture, higher durability, or other goals comprise the next phase. Some additions to the project scope are in order.
- After many years of development and continued DOE support, it's time for the NSTF-based MEAs to be independently verified in a broad range of operating conditions.
- The effect of common anode and cathode impurities needs to be addressed. This is especially important for NSTF-supported catalysts, not as well dispersed as other catalysts.
- The "rotating electrolyte" case needs theoretical treatment that would relate it to rotating disk geometry and help interpretation of oxygen reduction reaction (ORR) kinetic data from JPL.

Project # FC-18: Highly Dispersed Alloy Catalyst for Durability

Vivek S. Murthi; UTC Power

Brief Summary of Project

The objective of this project is to develop a structurally and compositionally advanced cathode catalyst that will meet the DOE 2010 targets for performance and durability. The impact of oxygen on Pt dissolution and structural stability for various core/shell systems has been qualified. A number of elemental and alloy cores have been evaluated; Pd₃Co and Ir cores lead to the highest improvement in oxygen reduction reaction (ORR). Various PtIrX alloys have been synthesized and tested to understand activity and durability trade-off.



<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of **3.5** for its relevance to DOE objectives.

- The cathode catalysts development is of key importance for successful deployment of fuel cells. Therefore, this program is indeed relevant to the development of polymer electrolyte membrane fuel cell technology, and thus to the overall DOE Hydrogen Program.
- The project team took a very highly coordinated approach to lowering catalyst loading in fuel cells.
- The development of more active, less costly, and more durable cathode catalysts is very relevant to the DOE polymer electrolyte fuel cell (PEFC) objectives.
- High performance, excellent durability, and low cost in proton exchange membrane (PEM) electrocatalysts are primary DOE goals, and therefore appropriate.
- This project is relevant to DOE objectives, directly focusing on the cathode electrocatalysis challenge.

Question 2: Approach to performing the research and development

This project was rated 2.7 on its approach.

- The approach developed for probing activity and stability of catalysts for the ORR is, in general, *ex situ* which, by definition, is not sufficient to provide information required to understand surface processes at an atomic scale. The latter is the key for tailoring catalytic properties of catalysts.
- This project encompasses two approaches, Pt alloy catalysts and Pt shell-Pd alloy core catalysts. UTC Power is pursuing the Pt alloy approach, whereas BNL and Johnson Matthey Fuel Cells (JM) are pursuing the core-shell approach. It is not clear how the UTC Power and BNL approaches are integrated. It is also not clear how the core-shell approach pursued in this project differs from the core-shell approach pursued by BNL in the LANL-led catalyst project. Unfortunately, the UTC Power approach involves replacing a portion of the platinum with an even less abundant metal, iridium. It is also not clear how the modeling activity is impacting the choice of materials or explaining the experimental data for the Pt alloy activity. Are there any characterization results that show the calculated structures are those that are obtained experimentally?
- Feedback between modeling and experimentation could be more clearly reflected.
- Restructuring/dissolution of ternary catalyst during potential cycling/fuel cell operation remains a concern, and hence, it is not clear if alloying with less noble metals is the way to go.
- The materials science aspects of the program are fine, and there is a focus on barriers.

- The approach is not entirely clear. There is some "theory," but much more trial and error. The partners appear more as vendors. Indeed, there was no description of team interactions and assignments, although one assumes that JM makes the catalysts, etc.
- Comments were made that others had superior supports, although there was no description of how such supports would be built into the program.
- The project's heavy reliance on iridium, a metal approximately ten times less abundant than platinum, is a serious drawback. In reality, there is no path forward for the use of Ir in practical fuel cell catalysts.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.8 based on accomplishments.

- The team has created structures with high-catalytic activity for the O-O bond making and bond breaking events. For the former, much more fundamental experimental work is required in order to demonstrate a true activity and stability of ternary alloys. For the latter, appropriate testing in the rotating disk electrode (RDE) configuration would be desirable in order to be able to confirm the observed increase in catalytic activity of the oxygen reduction reaction (ORR) on the Pt monolayer (shell) structure. Overall, significant progress has been made in overcoming barriers.
- The team has made steady progress, with good fundamental understanding.
- Progress was shown in replacing a significant portion of the iridium in the alloys with less expensive, more abundant metals (Co and Cr). The modeling activity shows that the "non-segregated" alloys are more stable against dissolution than pure Pt; however, are the Ir and Cr stable against dissolution if they are in the surface layer of the alloy?
- It is stated that the purpose of the iridium is to stabilize the Cr or Co against leaching from the alloy. The activity and stability of the PtCrIr and PtCoIr ternaries should be compared with their binary PtCr and PtCo counterparts rather than to Pt, as these binaries have been shown to have improved activity and durability compared to Pt in past studies (i.e., show the necessity/advantage of adding an expensive platinum group metal (PGM) to the binary).
- There have been issues with maintaining activity when scaling up core-shell catalysts for Pt on Pd₃Co.
- There has been good progress in terms of reproducible preparing and testing of the catalysts.
- I am skeptical if using chromium is a good idea given potential impact on membrane degradation. However, applicants should be allowed to demonstrate through long term tests that membrane degradation rates are not significantly altered.
- The data were not convincing, and results showed a large loss of performance which appears far from state-ofthe-art. Many would claim the lack of durability is disappointing.
- Improved performance of Ir-containing catalysts is interesting for better understanding of the ORR on platinum group metal (PGM) alloys, but of limited practical consequence (the use of iridium).
- Johnson Matthey's scale-up of the synthesis of two catalysts is a notable accomplishment.
- High angle annular dark field-scanning transmission electron microscopy (HAADF-STEM) data indicating the presence of a core-shell structure in as-synthesized Pd₃Co is convincing. The post-cycling data are somewhat ambiguous. Is there core-shell structure preserved after catalyst operation in the cathode?

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.2** for technology transfer and collaboration.

- The program is multifaceted and the PI successfully built a team which is well coordinated.
- This is a very strong team that works well between fundamental studies/modeling, catalyst synthesis, and cell testing.
- There is good coordination between the project participants in the core-shell activities. It is not evident that there is very extensive coordination in the alloys part of the project.
- The project team exhibits nice collaborations.
- Some roles seem poorly defined. There seemed to be more competition with BNL than cooperation.
- There has been good collaboration between four partner organizations, but no collaboration mentioned with partners outside the project.

<u>Question 5: Approach to and relevance of proposed future research</u>

This project was rated 2.7 for proposed future work.

- The proposed work should be more challenging. It seems that the authors are focusing on routine experiments, and unfortunately, with this approach, no progress will be accomplished.
- Proposed future work should focus on demonstrating the effect and necessity of Ir by benchmarking against binary alloys (ternary alloys minus the Ir component).
- The future work was not described clearly.
- Prospects for increasing mass activity of both dispersed and core-shell catalysts remain uncertain. Ir-containing catalysts should be abandoned.

Strengths and weaknesses

Strengths

- This is a team of highly experienced and very good scientists, especially in the field of fundamental understanding of the ORR. The team is multifaceted, and with appropriate focus may bring us closer in our quest to design better catalysts for the ORR.
- The project team had very good partners.
- There was a good mix of practical and fundamental work.
- There were attempts to scale-up BNL core-shell catalysts by a commercial catalyst manufacturer. There was also testing of core-shell catalysts in an MEA environment.
- It is a strong team that had reasonable focus in terms of materials studied.
- The team utilizes an excellent supply of chemical tools, and seems to have strong materials science excellence.
- Activity and stability modeling is interesting and represents this project's strength.

Weaknesses

- The correlation between theoretical and experimental results is defocused. There does not appear to be any relevance between the d-bend center position and catalytic activity and stability. The theoretical approach in resolving segregation of Pt alloys is not clear and does not contribute to understanding this important phenomenon. For the experimental part, the differences in catalytic activity between Pt and alloy surfaces is rather small (factor of 2) and, thus, very careful analysis is required in ordered to claim catalytic improvement, especially in the case of ternary alloys. The core-shell structures stability of surface atoms should be addressed more rigorously.
- It is still unclear as to what will be happening as core-shell structures age.
- There is a lack of coordination between core-shell and alloy activities. There is a lack of atomic level characterization of Pt alloys and benchmarking against binary analogs of Ir-containing ternaries.
- The dissolution and restructuring of less noble components over the long term may render the proposed system impractical.
- Although the "core shell" results from BNL were mentioned, the main consideration seemed to be to demonstrate that the "highly dispersed" materials were superior. Even so, there was no mention of experimental error, and the sets of data demonstrated disappointing durability. The UTC Power materials were somewhat more stable, but hardly good enough. Even so, the results presented were not convincing that there was any advantage.
- The presentation was poorly done. The graphics were difficult to read, especially the legends. This is probably some Microsoft problem; even so, from the second row while squinting, it was not easy to read.
- The use of Ir is a major weakness.
- Catalyst performance should refer to the total PGM content, not only to Pt, as used in most slides.

- From a practical viewpoint, there should be more focus on durability, since both catalyst and membrane as a resultant of catalyst dissolution is needed.
- The roles and tasks of the team partners need to be defined.

- There is no indication of error in measurements and repeatability, thus making it difficult to evaluate the utility of the results and conclusion.
- Although "fundamental" (in regards to approach) is thought to be an attribute, the need now is on commercial development.
- Testing of core-shell catalysts' durability in fuel cells is needed, not only in the RDE.
- Ir should be eliminated.

Project # FC-19: Development of Alternative and Durable High Performance Cathode Supports for PEM Fuel Cells

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Brief Summary of Project

The overall object is to develop and evaluate new classes of alternative and durable high-performance cathode supports. The objectives for 2009 were to identify compositions with 2x better stability than carbon black-supported catalyst for cell demonstration and demonstrate durability under accelerated test protocols that meet DOE lifetime criteria.

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of **3.4** for its relevance to DOE objectives.



- The project addresses DOE targets for fuel cell durability and cathode supported catalyst performance.
- The project addresses DOE goals on development of more durable supports.
- The project supports goals identified as key to durability success for both automotive and stationary applications.
- This program is indeed relevant to the development of polymer electrolyte membrane fuel cell technology, and most project aspects are aligned with the Hydrogen Program and DOE RD&D objectives.
- The nature of the catalyst/support interaction is very important and critical to DOE's long-term goals.
- This is a good project that tries to characterize the fundamental nature of what happens at the catalyst/support interface. It is appropriate to have both modified carbons and metal supports.
- Catalyst support corrosion has been one of the problems identified as a barrier to proton exchange membrane fuel cell (PEMFC) durability. This is the only project that directly addresses catalyst supports. It is most certainly relevant to the DOE Hydrogen Program goals.

Question 2: Approach to performing the research and development

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

- There is a good mix of fundamental theory and experiment to guide approach. Significant emphasis on model systems should provide fundamental insight into cathode catalyst and support effects.
- The consistency and clarity of the approach are not obvious.
- The 2009 objective stated as "2x better stability than carbon black" is not specific enough.
- The approach is focused on durability only, and underestimates the influence of metal-support interaction on platinum electrocatalytic activity.
- Using modeling to guide the search is a good basis for approach, especially with experimental follow up.
- It is an interesting approach to decouple carbon from Pt corrosion mechanisms.
- How is electrode manufacturing related to this approach? Are there limitations to catalyst selection due to ink making problems?

- In general, combining experimental and theoretical methods in resolving catalytic and stability properties of electrochemical interfaces is the right way to go. Based on this, the authors are on the good track and they should proceed with the approach they developed. However, in order to overcome important barriers, the author should develop methodology on how to test the activity of catalysts.
- The team has taken a good approach that handles problems from a variety of perspectives, such as modeling, characterization and cell performance.
- Given the problems with carbon, it is at the least deflating to find that the project intention is to support the alternative supports on some form of carbon. Since this is the only support project in the portfolio, it would be preferred for this project to have begun by laying out support requirements, and the entire range of materials that could be considered.
- Some of the materials involved (ITO, SnO₂, TiO₂) have conventionally low surface area. It would be more interesting to see what could be done to make these materials at high surface area, both in terms of synthesis protocols and tradeoffs with conductivity. Then, if you have to support on carbon, you would make that decision after exploring every avenue to avoid it.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.4 based on accomplishments.

- Many different experiments looked at individual aspects of the issue, and each approach made good progress in a niche area. The team has made piecemeal progress toward meeting the DOE targets. The path to bringing all the individual pieces of information together to develop a cathode catalyst to achieve the DOE targets is not clear. They have incorporated carbon nanotubes (CNT) into the mix, but no case has been made that this will be a cost-effective approach, and there has been no direct performance comparison to the other support materials under consideration.
- Reference points for improved durability are ill-defined.
- Published data on intrinsic properties of the substrate materials considered in the project are not taken into account.
- Progress has been made towards the identification of new stable supports.
- It is unclear whether the wt% of oxide on carbon support has been optimized.
- The graphene work is encouraging.
- The identification of potential supports looks good, but the PI should be able to reference DOE's catalyst kinetic targets too, since while durability is a key target, kinetics are ultimately important, too.
- Unfortunately, the approach resulted in very little useful information regarding relationships between stability of support and activity of catalysts. In particular, the presented activity of Pt is unacceptably low (a couple of orders of magnitude from the benchmarking values); therefore, before the PIs start focusing on the stability of the substrate, they must provide the evidence that activity of Pt on different supports is sufficiently high that any improvement in stability will be beneficial.
- There has been some good progress towards the goal, and towards understanding the nature.
- I would like to see a more relentless root-cause analysis to understand specifics/mechanisms of differences between the various support/catalyst combinations.
- Rotating disc electrode (RDE) evaluations appear to have been completed in sulfuric acid, which means that anionic interactions with the Pt surface may distort results.
- Experimental data that shows Pt/WC was more stable than WC was already established in prior years. Modeling that shows Pt/WC and Pt/VC should be more stable and active is interesting. Higher activity is shown for Pt/WC/ordered mesoporous carbon (OMC) with RDE, but what is the mass/specific activity at 0.9 V?
- The low degradation shown by TiO₂ is promising, but more material characteristics should be shown for TiO₂. Is it rutile or anatase? What is the conductivity? What is the surface area and particle size? Is the stability a function of any of these properties?
- It is strange that following a high voltage hold test on an MEA, TEMs would be shown of Pt particle size. Would SEMs of MEA cross-sections be preferable to observe layer thinning with support corrosion?

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.1** for technology transfer and collaboration.

- Strong coordination existed with partners that provide complimentary expertise and capabilities.
- It is not clear how interactive the team was.
- There was good collaboration with industry and national labs.
- They assembled a good team. It was not clear which of the team members contributed to specific work packages.
- Collaboration between different institutions is close and partners are fully participating.
- These are very good technical teams, with appropriate alignment.
- Some collaboration exists, but it is not entirely clear that ORNL and the University of Delaware have contributed anything more than delivering carbons, which is not the most interesting part of the project.
- Fuel cell testing at the Automotive Fuel Cell Cooperation has not yet yielded the results that are crucial to showing progress; those results (activity of Pt/WC/OMC and durability of Pt/TiO₂/Vulcan) have been shown with RDE.

Question 5: Approach to and relevance of proposed future research

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

- These really are two parallel projects, the model Pt/carbon system and the broad survey of other potential catalysts/supports. The PIs need to tie these together more closely and/or conduct some of the same fundamental studies on the newer systems to develop a broader understanding more quickly.
- There are quite a few reported studies on the topic of "Study C Degradation."
- Plans are to build in order to meet the 2x durability improvement goal.
- The team should incorporate electrode-making, i.e., ability to make an ink and electrode of good porosity should be made part of the process.
- Future work would require both more RDE measurements for the ORR on different supports, as well as a deeper understanding of relationships between the nature of support and catalytic activity of Pt.
- The reviewer would like to see more characterization to understand the nature of degradation, and whether relative importance of various failure modes changes among the supports.
- The future plans include further durability testing and material development on OMC and CNT, which is to be expected.
- Evaluating Pt/carbon (whether OMC, CNT, or other carbons) may be of little importance since these carbons' instability or cost makes their prospects of enabling PEMFC commercialization questionable.
- Future work should emphasize material properties much more so than it does. There is not just one form of TiO₂ or WC; we need to know more about the surface area and phase of these materials to relate these properties to activity and stability.

Strengths and weaknesses

Strengths

- The team has stable supports, but alternatives to carbon are needed.
- It is a model approach to development of new support.
- The PIs have performed careful microscopic characterization.
- The team has made good use of modeling and experimental verification to base studies.
- The team has identified some good potential candidates for stability.
- I like the idea of developing a new type of supports and the approach used to tackle this issue.
- There was a good focus on fundamentals of catalyst/support interactions.
- The research includes both ORR and catalyst stability.
- The PIs showed some progress with WC and TiO₂.
- The team has the ability to back up experimental results with some modeling capability.
- The team also has the ability to use a large array of analytical techniques.

Weaknesses

- The team has done lots of individual niche experiments without a clear path to developing the ideal cathode material. There are two separate projects for model systems and new catalysts/supports, the latter of which is highly exploratory without adequate scientific support for the broad range of materials (thus a lot of niche experiments that are not linked).
- The project is lacking a clear focus, and much is needed as it enters the final period.
- The quality of RDE data is poor and does not allow reaching a conclusion about higher activity of Pt/WC/C.
- The quality of polarization curves in membrane electrode assemblies (MEAs) is poor and does not meet current standards.
- The team should also include cost considerations for the various supports, for example, current cost at low volume and projected cost at high volume.
- The team should start to include kinetic data in selections, or show how you would improve kinetics.
- The activity of Pt shown is below the benchmark activity of Pt and this must be corrected.
- The authors must understand why Pt supported on oxides is more active than Pt supported on carbon.
- I see no reason for using the density functional theory calculation without trying to understand how this method may help in designing better supports.
- Scanning tunneling microscope micrographs would be more helpful if the Pt images are compared between different supports.
- A plot of the percent of degradation vs. the percent of carbon loss seems to have no mechanistic basis, and doesn't have a particularly good fit.
- The role of ionomers in mediating performance and degradation, and the role of catalyst ink formulation should be explained.
- The team needs to use proper RDE conditions and show validation of the technique.
- The overall premise of simply supporting materials on carbons is questionable towards meeting cost and durability targets.
- The execution has been slow in materials synthesis.
- There is a lack of materials characterization reporting, but more importantly, a lack of relating materials properties to activity and stability.

- The team should carefully characterize platinum-oxide interactions.
- The PIs should either include a kinetic/activity task, or a plan to increase this if a highly durable support is identified, but initial kinetic rates are lower than state-of-the-art or reference material.
- The project team needs to include relative cost of materials, near term (low volume) and longer term (high volume), using Vulcan XC-72 as a benchmark.
- The sensitivity of model results to assumptions about an electrode/electrolyte interface should be addressed.
- The PI should comment on whether catalyst degradation is occurring more on the carbon or platinum side of the interface.
- The contact resistances/electronic-phase resistance of MEA should be characterized.
- The project would be more useful with an intense focus on the novel support materials (ITO, SnO₂, TiO₂, SiO₂, WC). Information on carbons usually appears similar to what has already been studied. While actual performance would certainly be lower, disposing of the carbons might force this project to explore the novel materials even further, and then create more useful materials for later application.
- The project would benefit from laying out requirements for catalyst supports, even if it is only as a function of active species parameters.

Project # FC-20: Non-Platinum Bimetallic Cathode Electrocatalysts

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J. Regalbuto, S. Ekambram, and H.-R. Cho; University of Illinois at Chicago

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K. More; Oak Ridge National Laboratory

P. Zelenay, F. Garzon, H. Chung, and G. Wu; Los Alamos National Laboratory

Brief Summary of Project

The overall objective is to develop a nonplatinum cathode electrocatalyst for polymer electrolyte fuel cells to meet DOE targets that 1) promotes the direct fourelectron oxygen reduction reaction (ORR) with high electrocatalytic activity; 2) is chemically compatible with the acidic electrolyte and resistant to dissolution; and 3) is low cost. The objectives for the past year were to 1) optimize ORR activity and stability of Pd-Cu nanoparticles, study the correlation between Pd-Cu electronic structure and activity, and perform membrane electrode assembly (MEA) tests; and 2) synthesize and evaluate the oxygen reduction activity and stability of nanoparticles of one palladium alloy system



and two rhodium alloy systems (Pd-Co, Rh-Co, and Rh-Fe).

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.4** for its relevance to DOE objectives.

- The project addresses fuel cell targets for electrode performance, durability, and cost. Specific quantifiable targets for the program are listed in the brief, indicating a clear understanding of the basis for progress and success.
- Expanding the option for catalysts beyond platinum, to include other platinum group metals (PGMs), is okay.
- The project addresses three of the most critical barriers for MEAs, but is limited to only the cathode catalyst and cost reduction by reducing the PGM content.
- The project addresses durability, kinetics, and reduced cost via the core-shell approach and is highly relevant to achieving goals.
- Reducing catalyst PGM-loading is the highest priority for enabling automotive fuel cell commercialization.
- It is unclear if the proposed systems are practical from either the performance or (more importantly) the durability viewpoint.
- This project takes a rigorous and systematic analysis of non-platinum catalysts. The PIs focus on Pd-based materials that may be critical to next generation catalysts for proton exchange membrane fuel cells (PEMFC).

Question 2: Approach to performing the research and development

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

• There is a strong fundamental scientific basis for this approach and for bimetallic systems selected for initial studies. The team has used a strong iterative process to develop the theoretical methods to determine the DOS for various systems and to identify the most promising materials, experiments to validate the theory, and further theory and experiments to refine the approach.

- It is not clear how the theoretical/modeling work is guiding the experimental effort. Perhaps it should be vice versa.
- The approach is very theory-based, and is actually led by theory rather than the other way around. This is good, but may not be adequate to reveal the right path due to the myriad of factors that material processing can introduce that affect the real catalyst synthesis and performance.
- The team used a strong theoretical basis to making these materials.
- There was good coordination and explanation of team members' activities and contributions.
- The combination of modeling and experimental work will be helpful to enable optimal alloy and core-shell structures to be developed. The missing piece is the characterization aspect so the PIs know what their particle surface and near-surface atomic structures are.
- Also, the PI should be careful of replacing Pt with other PGMs (Pd and Rh).
- The team took a very good approach with combined theory and analysis. The only drawback might be that the theory is not able to predict the actual materials that will be stable at the cathodes of PEMFCs, after Pd and some of the other metals oxidize.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.9 based on accomplishments.

- The team addressed several key concerns mentioned in the reviewer comments from the 2008 AMR review, specifically the basis for the overall approach and the validity of the aqueous treatment approach used to screen the Pd-Cu samples. The attempt to modify existing colloidal particles with Pd was not successful, but the PIs will continue to pursue other methods to fabricate the bimetallic systems. The team demonstrated good correlation between theory and experiments for Pd-Cu density of states. The team is making good progress on demonstrating the benefits of this combined theoretical/experimental approach.
- A large number of samples were prepared (and tested).
- The work done to-date is very impressive for its fundamental and theoretical contributions. However, the technical accomplishments so far have not demonstrated any catalyst systems that can approach the required activities or cost reductions needed to overcome the target barriers.
- The team has made good progress; the synthesis of core-shell catalysts presents unique challenges that may consume resources.
- The "best" candidate is still below the current state-of-the-art.
- ORR mass activities per PGM still have not shown improvement over Pt.
- Progress has been made by optimizing alloy content, and has a fundamental basis from d-band and X-ray photoelectron spectroscopy (XPS) studies.
- The molecular model was developed at California Institute of Technology, but still needs validation with experimental studies.
- It was difficult to decipher data as presented, and very few results are meaningful from an applications viewpoint; only one MEA test was seen and was not compared to one with a traditional Pt-based catalyst.
- Much more MEA testing is needed.
- The team has made a lot of progress toward probing the properties of Pd-based catalysts.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.6** for technology transfer and collaboration.

- This is a strong team with complementary expertise. There is good interaction between the team activities.
- Too many participants for effectively steering the program toward a common/unified goal.
- The project shows very good team strength compatibilities.
- The project has a strong government and university team. Perhaps one commercial partner or qualifier would help, especially with catalyst production.
- There is very good coordination and collaboration throughout a complex project covering theory and practice.
- This is an outstanding team with expertise in synthesis, characterization, modeling, and testing capabilities.
- This is a very broad, highly capable team.

<u>Question 5: Approach to and relevance of proposed future research</u>

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

- Proposed future work builds on prior results. It is highly dependent on the outcome of purely theoretical calculations, and would benefit in the screening process from including some additional literature research on the catalyst materials that are identified to see if they are really suitable for a practical fuel cell environment.
- The classes of sample catalysts have to be reduced.
- I suggest that the theoretical strengths be put more on improving the world's best Pt alloys, since they are much better than the pure Pt target of the current project already, and they probably have a better chance of reducing total Pt loading without a loss of performance.
- No d-band center alloy with Pd₃M approaches the Pt₃Co alloy's d-band center of 2.76 eV. The target of trying to just reach that of pure Pt, 2.47 eV, may not be adequate and, therefore, no Pd system can ever reach the performance or mass activity of the best Pt alloys.
- The future plan is acceptable, but the synthetic component should be increased.
- More focus should be put on making core-shell structures to increase mass activity.
- The plan should be expanded to include stability to voltage cycling and advanced characterization of alloy surface structures.
- More focus is needed, and more time needs to be spent, on meaningful test systems such as a single cell.
- Durability will remain an issue and must be aggressively addressed.

Strengths and weaknesses

Strengths

- The project team is collaborating with a lot of high-power institutions and researchers.
- There is a very strong fundamental understanding among the collaborators.
- There is a very strong theoretical basis to guide research, validated by extensive literature support.
- They have a strong team assembled.
- The team has modeling capabilities.
- The team also has a wide range of materials made and tested.
- This is a very good team with a multi-pronged approach to implementation of non-Pt catalysts.

Weaknesses

- There are too many high-power institutions and researchers.
- Sub-par reference RDE data is reported.
- This project could benefit from more materials synthesis and screening to help guide the modeling to demonstrate trends that the modeling can then try to explain.
- Synthetic approaches to build the designs dictated by theory may ultimately limit progress.
- There is a lack of evidence of alloy structure to validate models.
- The best mass activity is still below Pt.
- There is a lack of MEA test data.
- The team needs a more focused approach with respect to practically relevant goals.
- They may not be modeling the materials that are electrochemically stable at the cathode.

- At this stage, the project needs a very narrow focus.
- The team should apply the modeling and first principles to improving the activity of the very best Pt-based alloys known to be in practice or in the literature. This approach could have better payback in reducing the Pt or PGM loading without a loss of performance.
- Try to let more experimental measurements drive the modeling as well as the other way around.

- A combinatorial style component to sort through some of the finer details of what is trying to get accomplished (such as changing ratios of the metals, sintering temperature, sintering gas such as hydrogen vs. Ar) may be needed. The team should use the theory to determine the boundary conditions for the combinational approach.
- More modeling in the presence of water and oxygen is needed in the computational studies to determine susceptibility to dissolution/oxidation.
- The PIs need to develop a better understanding of atomic layering on the core shell.
- The project should focus on appropriately characterizing the alloys to be able to validate the modeling work.
- More work on voltage cycling is recommended.

Project # FC-21: Advanced Cathode Catalysts

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Brief Summary of Project

The overall objective is to develop an oxygen reduction reaction (ORR) catalyst, alternative to pure platinum, capable of fulfilling cost, performance, and durability requirements established by the DOE for the polymer electrolyte fuel cell cathode. In the past year the project team 1) developed a number of catalysts with much reduced platinum content, ORR activity exceeding the DOE target, and very good rotating disk electrode cycling durability; 2) accomplished industrial scale-up of the first core-shell catalyst; and 3) demonstrated non-platinum group metal catalysts with volumetric ORR activity on track to meeting the DOE 2010 target.



Question 1: Relevance to overall DOE objectives

This project earned a score of 3.7 for its relevance to DOE objectives.

- The project addresses DOE targets for electrode performance, durability, and cost. Inclusion of the quantifiable targets in the brief indicates a clear understanding of the metrics for progress and success.
- Low Pt/platinum group metal (PGM) or PGM-free catalysts are critical.
- The project supports DOE goals on reduction of Pt loading and fabrication of non-precious metal catalysts.
- All aspects align with the Hydrogen Program objectives but, to this reviewer, not all are critical, because there are other more promising approaches already demonstrated. The nanostructured polymeric materials (NPM) catalysts are simply not necessary to enable a viable commercialized fuel cell business, given that existing internal combustion engine vehicles have the equivalent of 10 g of Pt in them.
- The inclusion of electrode structures places this program above typical catalyst-only projects.
- There are many excellent aspects to this program, for instance, the scale-up of the BNL materials. Some of the work seems somewhat irrelevant, but LANL is doing a pretty good job of ending approaches that do not work, thus keeping the program relevant.
- The most crucial area of study for meeting the DOE Hydrogen Program goals and objectives is novel oxygen reduction catalysts. This project studies exactly that, and is therefore extremely relevant to the objectives.

Question 2: Approach to performing the research and development

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

• There is a good balance of catalyst composition breadth while still conducting key experiments to evaluate viability of each approach (Pt-containing and non-Pt-containing), and specific materials sets within each approach.

- The approach is well designed.
- Very tough challenges are addressed by this project to meet the Pt equivalency (or better Pt alloys) with the non-precious metal approach. There is a very small chance of success if not fundamentally impossible for NPM catalysts to meet the required performance targets, despite the significant progress demonstrated by this project.
- The ultra-low Pt shell-core catalyst work is much more promising, but still suffers from the fundamental specific activity limitations of nanoparticles.
- They have made good use of the team's strengths to accomplish goals of project.
- The team has made great use of analysis to support and guide results.
- The approach is very good in that LANL is using multiple approaches to solve the complex problems associated with the development of new ORR catalysts. They are making good use of their team. The only downfall is that they are looking at too many catalyst systems, and they might make more progress if they narrowed their scope. They are down-selecting their catalyst systems, but probably not fast enough.
- The approach has been considerably improved this year. Catalysts that were not performing (chalcogenides) have been removed, while the remainder of the portfolio has been sharpened.
- Improvement could be found on the crucial aspects of the non-PGM catalyst work: narrowing the mechanistic possibilities for ORR, and identifying why durability does not yet exist.
- More direction should be given as to the intent with PtAu catalysts. Last year, high activity was shown in the context of a Ni core, while this year, the stability of PtAu (no Ni) was touted. The project needs to consistently show progression of activity and stability with the same series of PtAu catalysts and/or note when formulations are changed and why.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.6** based on accomplishments.

- Performance of industrial scale-up of Pt/Pd/C systems was impressive. The team has made good progress in understanding fundamental science that contributes to the observed catalyst behavior, while making significant progress toward meeting the DOE quantifiable metrics.
- Excellent activities have been reported by BNL, LANL (non-PGM), and Cabot (scale-up).
- The project demonstrates steady progress towards DOE goals.
- Relative to past work in each technology approach, this team has made very good and impressive progress. However, that progress still falls short of demonstrating the potential to overcome the target barriers.
- The group provided an excellent demonstration of the potential of the Pt monolayer on the Pd/C system.
- The peroxide sensor in polymer electrolyte is a significant accomplishment.
- The stabilization of the Pd-based catalyst is exceptional.
- Scale-up to 5 g, while a benchmark, should really go to the 20-50 g range as first step, unless there is a precedent that 5 g in process can be consistently scaled to 500 g, for example.
- The LANL team is making a lot of progress on multiple catalyst systems.
- Progress this past year has been excellent, although not outstanding since the prospect of stability with higher activity core-shells is still questionable, and the prospect of durability with higher activity non-PGMs is perhaps 10x more questionable. That said, however, the higher activity is impressive.
- The 0.75 A/mg Pt on multiple-wall carbon nanotubes (MWCNT) is interesting. It would also be interesting to see if this can be done on shorter MWCNTs and how reproducible the result is.
- The silica-derived electrode structures appear to be making progress with smaller pores, but it is unclear how this will impact either PGM or non-PGM electrode design and why.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u></u>

This project was rated **3.6** for technology transfer and collaboration.

- This is a strong team that includes collaborators outside of the DOE program.
- As with most/all national lab projects, participants work (here successfully) on their own.
- Cabot scale-up does not seem to have much in common process-wise with the BNL effort.
- The project demonstrates close collaboration with universities, national labs, and industrial partners.

- The project team exhibits strong and broad expertise.
- This is a massive effort with a large team. It is good to see an industrial partner, which has been deemed critical.
- The LANL team is large but talented, and seems to be well managed, as evidenced by their high productivity.
- What was formerly a weakness of this project has now turned into a strength. It is now possible to look at each partner in the project and point to how they are contributing (BNL: core-shell; ANL: non-PGM characterization; University of New Mexico (UNM): silica-derived materials and XPS; University of California, Riverside (UCR): nanotubes; Cabot: scale-up; ORNL: STEM).
- Praise must be given for exercising a no-go option on a collaborator that was not appreciably contributing University of Illinois at Urbana-Champaign (UIUC).
- There is still some work left in incorporating UNM's silica-derived materials better into the remainder of the project.

Question 5: Approach to and relevance of proposed future research

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

- Proposed future work builds on prior work for the most part. Expansion to include nanorod and nanowire structures was not explained. The project continues to include a very broad range of materials and approaches and should add to the fundamental understanding of these systems.
- Future work is built to overcome current technical barriers.
- The catalysts should be tested under more rigorous durability conditions, such as scanning from 0.6 to 1.2 V in actual fuel cells at elevated temperatures, to really judge their tolerance for Pt dissolution.
- The team should stop the nanostructured polymeric materials work and focus more on the core-shell catalyst.
- The majority of work focused on the catalyst; it would be nice to see more electrode work as part of the title objectives.
- The team has proposed very good future work. The team would benefit from selecting their top catalyst systems and focusing their resources on fewer systems.
- There are certain points in the future work that need further justification. For example, if smaller nanoparticles are less stable (a hypothesis that may need further validation), why make smaller nanoparticles? It is not clear that smaller nanoparticles will maintain activity.
- Will nanostructures for polyaniline (PANI)-based catalysts eliminate carbon and decrease layer thickness? If so, this approach makes sense, but the rationale is not clear.
- It would be good to take a look at Pt monolayer stability using conventional accelerated stress tests before investing a lot in fabricating more catalysts with Pt monolayers.
- At some point in the near future, the scaled-up batches of core-shells should be attacked with durability testing.

Strengths and weaknesses

Strengths

- The individual participants have good accomplishments.
- The team has made fast progress in the development of new materials.
- The team has made careful in situ characterization with a variety of spectroscopic techniques.
- The team members are excellent researchers.
- The collaborators have extensive experience.
- This is a highly productive team with a wide assortment of offshoots giving a greater chance of success.
- The team has a great portfolio of potential solutions to meet DOE goals.
- This complex team is very well managed.
- This is a large effective team trying to sort out the advantages of different fuel cell catalysts systems.
- The team has demonstrated higher activity for both PGM- and non-PGM-based catalysts.
- There is a considerable breadth of novelty.
- There has been better collaboration than in the past.
- There is a powerful range of analytical techniques available.

Weaknesses

- This is still a very broad project, and it is not clear what the ultimate goal/output of this project is at the end of the four year performance period. Will there be a further catalyst down-select recommendation to focus future efforts, or would the full scope of fundamentals to industrial scale-up continue in a follow-on effort? It is not clear that this effort wrapsup at the end of the performance period.
- The project has too many parts that are not related to each other.
- There are too many resources on approaches that appear to be fundamentally limited to ever overcome the target barriers, e.g., the NPM approaches.
- Scale-up level (5 g) seems still under a level that would be a significant first step to show feasibility.
- The team is spread too thin looking at too many catalyst systems.
- There is a tendency to drive towards a finished product before entirely exploring fundamentals (e.g., why PtAu is durable, why non-PGMs are not durable, whether or not scaled-up catalysts are durable).
- PGM catalysts still do not meet mass activity targets on a per mass of PGM basis.
- There are still some aspects of the project that need to tie-in better to the remainder.

- The UNM effort does not seem to be related to the project, and it is not clear what kind of contribution can be made for the duration of the project.
- The team needs to extend scale-up synthesis to quantities higher than grams.
- They need to refocus the NPM work on the best core-shell and non-carbon support combinations that can exceed the DOE barriers in order to facilitate moving the fuel cell technology into reality in the nearer term.
- The team needs to boost up electrode structure work.
- The PIs should scale-up the synthesis task to 20-50 g, minimum.
- There needs to be greater emphasis on non-PGM durability.
- There should also be more emphasis on the durability and uniformity of scaled-up core-shell nanoparticles.
- Fundamental studies on the durability of Pt/Pd nanotubes from UCR are needed.

Project # FC-22: Effects of Fuel and Air Impurities on PEM Fuel Cell Performance

Fernando Garzon, Rod Borup, Eric Brosha, John Davey, Fernando Garzon, Mark Nelson, Tommy Rockward, Josemari Sansiñena, Tom Springer, and Francisco Uribe; Los Alamos National Laboratory Brian Kienitz and Thomas Zawodzinski; Case Western Reserve University Idoia Urdampilleta; Cidetec, Spain Thiago Lopes; University of So Paulo, Brazil

Brief Summary of Project

The overall objective of this project is to contribute to the scientific understanding of the effects of fuel and air impurities on fuel cell performance and how it affects DOE fuel cell cost and performance targets. The specific objectives are to 1) understand the effects of fuel cell operation with less than pure fuel and air, i.e. simulate "real world" operation; 2) understand how impurities affect DOE fuel cell cost and performance targets; 3) contribute to the scientific understanding of impurity-fuel cell component interactions and performance inhibition mechanisms; 4) develop sciencebased models of impurity interactions upon fuel cell performance; 5) perform experimental validation of models; and 6) develop mitigation strategies and methods.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.8** for its relevance to DOE objectives.

- Understanding fuel cell degradation mechanisms due to fuel and air impurities is one of the key remaining barriers for commercial applications of fuel cells. Understanding degradation mechanisms will allow fuel cell developers to design more reliable systems at reduced cost since impurities affect some of the most costly components of the system (e.g., Pt).
- Membrane electrode assembly (MEA) materials' robustness with real world usage profile is technically critical for fuel cell variability.
- The effects of impurities in air and fuel are important for developing fuel standards and determining lifetime. The team needs to concentrate on fuel impurities for the fuel standards timeline.
- Air side contamination work is needed. It would make far more sense to test the air side with representative air quality of, say, Los Angeles.
- This project is excellent and very well delivered. It is of real-world relevance and critical to RD&D objectives. The project is science-based, methodical, and useful.
- Much work has been done using ultra-pure fuels. The understanding of fuel impurities for real application is of utmost importance.
- The most relevant impurities were chosen for the investigations.
- Long-term tests of up to 100 hrs were addressed.
- DOE cost and performance targets are addressed through understanding of contamination mechanisms. Incipient mitigation strategies are developed.
- At this stage, the investigations meet the critical issues and DOE targets fully. A broad range of impurities is considered. In the next stage (for future projects), it is recommended to include impurities originating from pipeline systems.

Question 2: Approach to performing the research and development

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

- The PI investigated the biggest contributors to fuel cell poisoning; thorough testing allowed the PI to develop models of the contamination effects.
- Model development/validation is necessary for H₂S and NO_x degradation mechanisms. Molecular dynamics can be applied. Metal cation analysis properly uses modeling and empirical approaches.
- The project is well designed and focuses on important impurities. Testing appears to be all on Pt commercial fuel cells/vehicles that are likely to utilize Pt alloys to get needed activity. Work on these higher activity alloys, modeling, or other activities to relate Pt results to Pt alloys is needed.
- There is pressure to develop a fuel quality standard. Fuel impurity data is needed for this, and a focus on fuel impurities versus air impurities would help this effort.
- This approach would make more sense if gas mixtures were used. Single constituents have been tested by a number of institutions.
- Again, there were very clear explanations of the methodical approach to single-gas effects and testing protocols. The team gave solid explanations of plausible mechanisms of degradation and possible reversal of effects, in some cases.
- Elucidation of mixed-gas effects (if any) was not presented. Effects of aromatic contamination were not discussed.
- The approach is scientifically solid and appropriately considers the technical challenges, like long-term stability and air contaminations like SO₂; however, more work will be needed in the future. The current program is very well focused on the technical challenges, and the effort fully justifies the budget assigned to it.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.5** based on accomplishments.

- The PI addressed the biggest original equipment manufacturer (OEM) suggestion from the 2008 AMR, that the PI should begin developing degradation models.
- It was not clear if the model was validated with real world operation and experience. The model is clearly based on testing (important), but will it correlate well to future designs? Time will tell.
- Coefficients/constants should be provided in the model to guide developers.
- The project had well-identified H₂S, NO_x, and cation poisoning.
- Measurements on H₂S, along with methods to measure S in the C support, and drive cycle testing effects of 10 ppb H₂S are important work. Ammonia and hydrocarbon work is useful for hydrogen fuel quality standards.
- Testing mixtures would be more relevant as the effects are not additive.
- There is good progress shown, and the team gave excellent explanations of cation effects.
- An H₂S crossover measurement method was developed, and crossover rates were measured. NO_x was investigated as a poison, and the interaction of humidification and NO_x poisoning was noticed, though it is not fully understood. Ammonia and cations were addressed as well. The investigations were backed by modeling in case of the cations, to identify the blocking mechanism. These investigations are ongoing activities.
- The results will help to overcome barriers.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.2** for technology transfer and collaboration.

- LANL is a well-recognized team in fuel cell research; they collaborate well with the industry and other academic institutions.
- For fuel contaminations, it is recommended that they work with the hydrogen storage technology team to identify possible NH₃ contamination levels.
- The team exhibited good collaboration with other national labs and universities.
- A wide range of partners was selected for the modeling effort.

- The team demonstrated good collaborations, but the role of The Canadian National Research Council (NRC) Institute for Fuel Cell Innovation is unclear.
- The work was shared between LANL and Case Western Reserve University for modeling, Berkeley National Laboratory for modeling, and NRC for fuel cell impurities, as well as two further partners. The partners mentioned are internationally renowned. The work share is based on competencies, and the partners share vital parts of the activity, thus, they are full participants and well coordinated.

Question 5: Approach to and relevance of proposed future research

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

- The team should continue with model development; results should be published with general guidance provided for constants and general behavior.
- Knowledge transfer to other/novel gas diffusion layer designs in various operating conditions should be addressed.
- This project is ending.
- Future work should include some work to determine effects on alloys that are likely to be used in commercial fuel cells.
- Future work as outlined was quite shallow and could use more depth.
- The team had good conclusions on possible non-uniformity challenges, but specific experiments (or analytical methods) to investigate such inhomogeneities should be addressed.
- The team has good future plans for cationic effects.
- This is a very good and relevant project that is very well conducted.

Strengths and weaknesses

Strengths

- The PI is very receptive to OEM feedback.
- The PIs have performed well-controlled testing to evaluate poisoning effectiveness.
- There is much analytical expertise present in this project and understanding of the fuel cell catalysts.
- Air side contaminant effects have been needing study for some time. This particular project seems to have a path, rather than the infinite loop of past investigations.
- This was a clear presentation that was very methodical with mechanistic/modeling explanations. They took solid single-gas approaches.
- This team has conducted rock-solid scientific investigations on a broad range of contaminants.

Weaknesses

- There is not much modeling capability of H_2S and NO_x poisoning.
- The team needs more mixed gas experiments. They need to look more at chlorides (and their associated cations).

- The PIs need publications to make valuable data available.
- I recommend some initial work with high activity alloys such as PtMnCo (3M) and core-shell catalysts to see if effects are significantly different.
- The team should explore more cyclic effects with simulated diesel or gasoline exhaust as inputs on the air-side.
- Contaminants of the hydrogen distribution chain should be identified, and their impact should be investigated using the same approach. This is not meant to be added to the project, but would represent a new project. The same holds true for further gases, particularly exhaust gases of internal combustion engines.
- A way to seek out mal-distribution effects, i.e., some impurities cause areal inhomogeneities over surface leading to current density variations and hence, spotty degradations over a membrane surface could be explored.

Project # FC-23: Effects of Impurities on Fuel Cell Performance and Durability

J.G. Goodwin, Jr., Jack Zhang, L. Hongsirikarn, and Michael Martinez; Clemson University Hector Colon-Mercado and Scott Greenway; Savannah River National Laboratory Peter Finamoore; John Deere, Advanced Energy Systems Division

Brief Summary of Project

The overall objectives of this project are to 1) investigate in detail the effects of impurities in the hydrogen fuel and oxygen streams on the operation and durability of fuel cells (CO, CO₂, NH₃, H₂O, HCs, O₂, inert gases, and H₂S); 2) determine mechanisms of impurity effects; and 3) suggest ways to overcome impurity effects. Objectives for 2008-2009 were to: 1) complete the investigation in detail of carbon monoxide poisoning of platinum as well as the effect of NH_3 poisoning; 2) complete the study of NH₃ poisoning of the Nafion® membrane as a function of impurity level, relative humidity (RH), and temperature; 3) finalize the study of the minimal effect of carbon monoxide on



Nafion® conductivity; 4) investigate the effect of a Cl-containing hydrocarbon (PCE); 5) carry out long term study of effect of NH₃ poisoning per DOE request; and 6) start correlation of fundamental and fuel cell performance results for NH₃ poisoning.

Question 1: Relevance to overall DOE objectives

This project earned a score of 3.6 for its relevance to DOE objectives.

- Understanding effects of impurities is important to the objectives of the DOE Fuel Cell Technical Team (FCTT) with respect to improving fuel cell durability and reducing cost due to Pt requirements and catalyst poisoning. Some of the impurities the PI has selected are questionable in the sense that they have already been extensively tested (CO) or may not be a significant poison in real world operation (chlorinated HCs).
- The project directly responds to the need of understanding the effects of contaminants on fuel cell performance and durability, and aims at developing mitigation strategies for these effects. The project targets a significant amount of individual contaminants.
- The effect of impurities is critical for real work usage profile. Fuel impurities can affect hydrogen fuel cost and availability.
- Effects of impurities are a key aspect of fuel cell durability.
- The project is tracking more closely to the industry needs for data to support the International Organization for Standardization (ISO) fuel specification activity. Impurities effects studies are divided among working group members to provide input to the ISO hydrogen fuel quality specification process. Clemson is studying the effects on fuel cell performance of perchloroethylene, CO₂, ethane, and ethylene.
- Determining hydrogen fuel impurity effects is a high priority to guide not only fuel cell development, but also production and storage development.
- The study of impurities and their effect on the performance of proton exchange membrane fuel cells (PEMFC) is very important and the project supports DOE RD&D objectives. The two major impurities discussed, CO and NH₃, are important impurities to understand their impact on performance.

Question 2: Approach to performing the research and development

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

- The PI continues D₂-H₂ testing despite the recommendations of the FCTT to use simpler, more cost effective techniques.
- The project combines experimental studies to understand results. Modeling would additionally be beneficial to understand the poisoning mechanisms. Modeling results should be used in addition to experimental results to develop mitigation strategies.
- Milestones address the study of a large number of impurities.
- Milestones do not address the development of mitigation strategies proposed in the technical approach.
- If the modeling approach is combined with a phenomenological measurement approach, it would be more effective to determine mechanisms of impurity effects.
- It is necessary to clarify how to distinguish multiple mechanisms of impurity effects in fuel cell performance analysis.
- The combination of phenomenological and fuel cell experiments is an excellent approach.
- The Clemson project is focusing on contaminants of interest such as NH₃ and perchloroethylene. Longer-term testing is underway, and lower impurity levels are being tested that are more aligned with the proposed ISO fuel standard.
- The team has matured this year, and their technical approach has improved. However, impurities studied to date are replowing old ground.
- The approach to CO poisoning confirmed information that was in the literature, but it did not necessarily add a new level of understanding. The use of H₂-D₂ is interesting, but it was not clear how this would improve our understanding of CO poisoning.

<u>Ouestion 3: Technical accomplishments and progress toward project and DOE goals</u></u>

This project was rated **3.2** based on accomplishments.

- PI has thoroughly researched these materials.
- Contaminant concentrations that are closer to the suggested values for fuel quality guidelines should be investigated. The CO concentration tested was 50x above the 0.2 ppm SAE value, and the NH₃ concentration was 20 times above the comparable 0.1 ppm value.
- Little progress was reported on durability aspects of fuel cell contamination.
- No progress was reported on the development of mitigation strategies.
- There was interesting phenomena of hydrogen spillover. It was good to quantifiably identify CO poisoning on Pt/C.
- Much progress has been made in characterizing effects of CO, NH₃, and PCE.
- Progress appears to be good, and the Clemson results can be compared with those from other organizations as the round robin testing has been completed and results are comparable at any of the locations.
- There was a concern raised last year about the reliability of the impedance spectroscopy measurements. The presentation did not address this concern. The study of hydrogen spillover from the Pt catalyst onto the carbon support is not really germane to the impurities discussion. Further work in this area should be de-emphasized, as automakers have devised operating strategies to deal with CO poisoning.
- Progress continues to be slow, though the fuel cell testing seems to be coming up to speed. This project is halfway through in time and money, with little added to date to overall understanding.
- The claim of surface restructuring of the platinum catalyst did not appear to be corroborated by atomic force microscopy or some other method. It is not clear how the authors justified this claim. Did the electrochemical surface area (ECSA) change as a result of addition and removal of CO? The ammonia data did not appear to provide any new information. "A higher impurity tolerance and PEMFC performance would be expected at higher humidity." Is this true for all impurities, or just for impurities in the membrane? This statement needs justification. The mechanism for perchloroethylene poisoning should be addressed.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.1** for technology transfer and collaboration.

- The PI does not seem to be collaborating with LANL impurity research efforts. LANL studies are generally regarded as practical and useful to the FCTT. The FCTT recommended PI establish a relationship with LANL in the 2008 Annual Merit Review.
- The type of collaboration between project contributors and ANL for modeling efforts is unknown. Such an exchange may significantly support the project's goal to "suggest ways to overcome impurity effects."
- It would be good to work with the Hydrogen Storage Technology Team to identify possible levels of NH₃ in the hydrogen fuel from the hydrogen storage system.
- There has been excellent collaboration between Clemson and SRNL.
- Clemson participates in the DOE Hydrogen Fuel Quality Working Group activities and collaborates with other organizations to avoid duplication of effort. The results are shared amongst the members and with the representatives to the ISO technical standards committee.
- Reported collaboration and coordination is good, though this set of projects has a DOE funded activity to ensure coordination, so it is not clear how much of this is due to this PI and team.
- They are good collaboration partners. It is not clear what the contribution of John Deere is to the program. The advice of John Deere should be provided.

<u>Question 5: Approach to and relevance of proposed future research</u>

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

- I would recommend that the PI drop the CO work (plenty of literature exists for this).
- The PI should really investigate what the real world exposure of a fuel cell system (FCS) to chlorinated hydrocarbons is.
- The future work does not include the development of mitigation strategies.
- The future work is addressed at the key issues.
- For the 2009-2010 timeframe, Clemson plans to study the effects on fuel cell performance of perchloroethylene, CO₂, ethane, and ethylene. Lower levels of impurities are consistent with recommendations of the DOE Hydrogen Fuel Quality Working Group.
- Some of the future work appears to duplicate effort already or nearly completed elsewhere, e.g., H₂S, and coordination activity needs to work hard to make this set of projects accomplish as much as possible.
- The project team is finishing up program, but with no explanation of deliverables. Will models and/or mechanisms be defined or developed for effects of PCE, CO, ethane, or ethylene impurities? After testing 1,000 hours with 0.1 NH₃, what will be done with the data? Most future work is studies, but the deliverable should be described.

Strengths and weaknesses

Strengths

- New techniques were successfully applied to the study of the effects of contaminants in fuel cells.
- The project addresses a significant number of relevant fuel cell impurities.
- The project demonstrates that it successfully contributes to the general understanding of the effects of fuel cell impurities.
- The team has conducted well-controlled testing to measure the effectiveness of impurities.
- The PIs have taken an excellent approach for evaluating, understanding, and mitigating effects of impurities on fuel cell degradation.
- The collaborations are good.
- The PIs have used a lot of equipment.

Weaknesses

- Up-to-date experiments addressed contaminant concentrations that were significantly higher than the expected concentrations in hydrogen fuel.
- It is unclear if and how modeling is contributing to reaching the project's goal.
- Up-to-date development of mitigation strategies is not addressed.
- There was no mention of the impedance spectroscopy issue raised in last year's review or its resolution. Neither was there a lot of evidence shown that Clemson has a thorough understanding of the relatively vast literature on impurities effects in fuel cells.
- The team is still learning fuelcell-relevant conditions. Progress has been slow (but appears to be improving).
- For a university and national laboratory, the RD&D did not appear to be very focused, and seemed to be a little undergraduate in nature. Everyone knows CO and NH₃ poison PEM fuel cells. What are the mechanisms? What are the catalyst crystal faces that are poisoned? Is one face more selective than others? What if the membrane was poisoned with NH₂D? Would this provide some fundamental understanding of the poisoning mechanism? What happens to anode catalysts when oxidizing PCE off the surface? The researchers do not appear to have a structured mechanistic approach and are conducting Edisonian R&D.

- More emphasis should be placed on mitigation strategies. Less emphasis on CO and on the H₂-D₂ reaction is recommended; they should use cyclic voltammetry instead. These activities do not relate directly to the task at hand, that is to provide data that can be used to set fuel quality specifications. Other membrane materials should be considered.
- The set of impurities to be studied in the remainder of project should be revisited, in coordination with related projects and DOE.
- The team should refocus this project. The DOE should take a strong hand to guide the program and assure that new and pertinent information will be obtained.

Project # FC-24: The Effects of Impurities on Fuel Cell Performance and Durability

Trent M. Molter, Ph.D.; University of Connecticut

Brief Summary of Project

The overall objective of this project is to develop an understanding of the effects of various contaminants on fuel cell performance and durability. The specific focus for the past year has included 1) the screening of hydrocarbon impurities per standard test protocols to identify impurities of concern; 2) the quantification of the effects on fuel cell performance; 3) the effects of cations on membrane properties; and 4) the development of fundamental models based on experimental findings.

<u>**Ouestion 1: Relevance to overall DOE**</u> <u>**objectives**</u>



This project earned a score of **3.4** for its relevance to DOE objectives.

- Understanding fuel cell poisoning effects is critical towards designing optimal fuel cell systems and meeting DOE targets.
- The project directly responds to the need of identifying fuel cell contaminants and identifying fuels purity standards for hydrogen fuel. The project is in contact to groups that define International Organization for Standardization (ISO) standards for hydrogen fuels purity for fuel cell vehicles. These standards are essential in moving forward towards hydrogen-based transportation.
- In general, the effects of impurities are critical for the viability of fuel cell systems. Hydrocarbon-related contamination is more relevant to stationary applications and less important for automotive applications.
- Understanding impurity effects on membrane electrode assemblies (MEAs) is a very important aspect of fuel cell system durability.
- The University of Connecticut (UC) is a member of the Hydrogen Fuel Quality Working Group and regularly participates in group meetings and teleconferences. The group is dividing the impurities of interest according to each member's interest and capabilities. The selected impurities and overall objectives are relevant to the DOE Fuel Cell Technologies Program.
- Effects of impurities in the hydrogen fuel and air are of high relevance to the department's program. Understanding will help guide not only fuel cell development, but also production and storage development.
- The determination of the effects of fuel impurities on the performance of proton exchange membrane fuel cells (PEMFC) is of critical importance to the advancement of fuel cell systems. Foreign cation impurities can significantly reduce the efficiency of PEMFCs.

Question 2: Approach to performing the research and development

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

- Prioritize the species to be tested: do the experiments in order of what is most contaminating to fuel cells.
- The team has taken a very systematic and straight-forward approach that focuses on the objectives of the project, and has thoroughly considered the details of the approach to ensure valid results.
- In addition to an empirical modeling approach, a first principle modeling approach would be effective.
- The PIs took a well-structured and thorough approach.

- The approach is very good and includes a literature search, experimental studies, and contaminant modeling. UConn and the other impurities projects should consider using state-of-the-art MEAs with lower catalyst loadings.
- Test cell break-in on different test stands must be done carefully to avoid introducing experimental variables other than the impurity. Cleaning of "wetted" components between tests is a commendable practice, and shows the team strives for quality testing. They need to resolve MEA performance issues and move to lower Pt loadings.
- The approach focuses on technical barriers identified by DOE and builds on existing databases. This approach should minimize repetitive research reported by others. The development of empirical models should assist others in predicting the effects of impurities on PEM fuel cell performance.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **2.9** based on accomplishments.

- The PI prioritized (as per OEM request) halogenated hydrocarbons as a more important contaminant to study.
- The project team did consider material sets; it is hard to tell if there was an effect or not.
- The project team completed a study of a known fuel cell poison so the project can show it can measure contaminant effects in line with others' data.
- Some difficulties in experimental equipment were successfully overcome.
- Researchers may consider using the existing gas analysis equipment to test for reactions of the contaminants in the cell. This information would add significant value to the results.
- The testing conditions (impurities level) should be explained.
- The team needs to clarify the rationale of impurities assumptions.
- Formic acid has been identified as an impurity with a potential deleterious effect on MEA performance.
- Among other accomplishments, UConn demonstrated the importance of fully characterizing the initial state of the MEA. This is a necessary step toward specifying a commercial MEA that is stable under these test conditions.
- Characterization data showed variation within and between MEA lots. UConn is working with the manufacturer and other labs to identify lots that show more consistent performance and has spawned an interlab effort to specify generic, stable MEAs. This effort is critical to ensure the reliability of the impurities effects data.
- No work has been done yet on halogenated compounds in spite of slide titles. The PIs have not yet started looking at effect mitigation. The sharing of raw data with modelers is an improvement this year.
- The concentrations used in the experiments were too high to provide critical information regarding those impurities that affect performance, e.g., 50 ppm formic acid. It is recommended that after initially observing impurity poisoning, the concentrations be reduced to levels consistent with fuel cell experience, e.g., <5 ppm. The data on ammonia impurities is beneficial; however, ammonia is a well-known impurity and testing at lower concentrations is necessary. Testing at 25 ppm does not appear to be productive. The recovery from ammonia poisoning is also well documented. Something new is needed, and other laboratories need to be involved.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.3** for technology transfer and collaboration.

- There is a significant amount of collaboration and exchange among the work groups and other institutions.
- There is effective collaboration.
- Collaborations are in place to address issues of testing standardization and MEA variability.
- Collaboration is very good within the Hydrogen Quality Working Group. Data is being fed to the modeling efforts in an attempt to provide a solid argument for impurity limits in the ISO fuel specification effort.
- Addition of Fuel Cell Energy to the team is an improvement. ORNL helped train a student in MEA sectioning for transmission electron microscopy. It is not clear that Hamilton Sundstrand (electrolysis background) adds much. DOE funds a coordination activity for this group of projects.
- Collaboration is high and should productively help the program.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- While the fuel specifications are critical in the near term, there are also impurities that could come from fuel cell system materials, e.g., plasticizers. Someone should eventually account for this in the overall impurities project.
- There should be a continuation of the systematic approach for future work.
- This experimental procedure was expected to be tuned for impurities that show performance effects.
- A systematic approach for development of mitigation strategy for contamination was not presented.
- There should be more specifics as to what hydrocarbons and cations will be examined in the future.
- Contaminant studies should be extended to ensure that all relevant species are studied and models validated against the experimental data. The need for a commercial source of stable MEAs with state-of-the-art material sets has become an important issue in establishing the reliability of the data output from this project, as well as for the other impurities projects.
- The project team needs to work on halogenated compounds typical of those found in operating environments.
- There has been good experimental activity, but the deliverable from the future work should be explained.

Strengths and weaknesses

Strengths

- The project team uses a very systematic approach.
- This project is highly focused to respond to immediate needs.
- This is a strong team with versatile capabilities.
- There is high exchange with other institutions and organizations.
- The PIs have a well-controlled experiment capability.
- The project is relevant and well structured. Significant results have been obtained in a timely fashion.
- The PIs have taken a good approach with literature search, experimental effort, modeling, and model validation.
- There is a good mix of experimental science and modeling effort. The team seems to have matured this year and is positioned to make more of a contribution.
- There is good collaboration in the program and an important collection of experimental methods and equipment.

Weaknesses

- The request for peer-reviewed journal publications has not yet addressed.
- Possible reactions/conversions of impurity in operating cells have not been considered.
- The MEA variability is a concern. Perhaps a "standard" MEA should be employed.
- Results to-date may be with MEAs configuration that may distort the true behavior that might be expected with state-of-the-art material sets.
- Impurities tested to-date are of minor interest. Issues with their commercial MEAs need to be resolved and the PIs need to move quickly to lower Pt loadings.
- The project should focus on the relevant concentration of impurities.

- Of the remaining species, the halogenated hydrocarbons may be most important.
- The project team should consider material sets; it is hard to tell if there was an effect or not.
- Do a study of a known fuel cell poison so the project can show that it can measure contaminant effects in line with others' data.
- With respect to the project's cation level studies:
 - The conductivity tests are being conducted at extremely high ion-exchange levels (which are conditions under which a fuel cell would not perform well anyway).
 - \circ Perform the tests at lower concentrations and show conductivity data.
 - Consider developing a standard impurity testing protocol.
- The PIs should continue the project, with careful coordination with similar projects to maximize learning from available effort.

Project # FC-25: Development and Demonstration of a New-Generation High Efficiency 1-10kW Stationary PEM Fuel Cell System

Durai Swamy, Ph.D.; Intelligent Energy

Brief Summary of Project

The overall objective of this project is to develop and demonstrate a proton exchange membrane fuel cell (PEMFC)-based stationary power system that provides a foundation for commercial, mass produced units that address identified technical barriers. The technical objectives are 1) 40% electrical efficiency (fuel-to-electric energy conversion); 2) 70% overall efficiency (fuel-to-electric energy plus usable waste heat energy conversion); 3) the potential for 40,000-hour life; and 4) the potential for \$450/kW. Intelligent Energy (IE) will engage international partners and demonstrate phase in an International Partnership for the Hydrogen Economy (IPHE) country other than the United States.



Question 1: Relevance to overall DOE objectives

This project earned a score of 2.8 for its relevance to DOE objectives.

- Design and demonstration of a small adsorption-enhanced fuel-processor-based combined heat and power (CHP) unit is consistent with DOE broad stationary fuel cell system goals.
- The project supports the DOE fuel cell program objectives to develop, demonstrate, and introduce fuel cell systems into the stationary marketplace.
- The aspect of international cooperation is appropriate. The technical and economic targets are laudable, but what markets are truly served at this power level? Has there been a detailed market analysis or is this simply a technology push?
- Refocusing of the DOE program to nearer-term applications makes this project even more relevant. CHP is a challenging market for a low-temperature polymer electrolyte membrane fuel cell, but it is important to determine how applicable the technology can be.
- The project addresses the construction of a fuel cell system that includes a reformer system. The focus of the project is continuous operation. Critical fuel cell data, e.g., decay, were not presented. The efficiency target was not adequately discussed since it was not identified if the target was beginning-of-life or end-of-life. The end-of-life target should lose about 5%. The presentation was too high-level for the maturity of the audience.

Question 2: Approach to performing the research and development

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

- The approach is methodical and follows normal steps of design analysis, sub-scale testing, and scale-up.
- Actually, answers given during the Q&A session led me to the conclusion that the overall system concept is not finally developed. For instance, questions about efficiency were answered, from my point-of-view, very optimistically.
- Developing a low-temperature PEM CHP system may not yield a system that has the potential to meet the project and DOE technical targets, because the temperature of the waste-heat stream is quite low. Absorption-enhanced reforming has been proposed by others and does not appear to have been widely adopted because of the process complexity it introduces. In fact, from the presentation, it is not clear if the reforming options

mentioned in last year's presentation have been abandoned in favor of the current concept. The approach to increasing the fuel cell, reformer, and power conditioning efficiency, and reducing parasitic power losses is generally sound.

- Having a flow chart as an approach chart is novel and elegant (given that, in my view, this is a system engineering demonstration). Such system-level demonstrations are truly appropriate, especially if DOE is moving toward technology transition programs (as opposed to its traditional role of simply demonstrating science frontiers).
- The integrated systems approach, where thermal sources are appropriately utilized to support parasitic loads, is good; however, caution must be exercised because component optimization may often lead to system sub-optimization see comment in the next section.
- The team continues to work on absorption enhanced reforming, but has shifted the project baseline to steam methane reforming with QuestAir pressure swing absorption for clean-up, which seems appropriate for this project. The team passed the August 2008 go/no-go point. They are now pursuing coated stainless steel (SS) plates (coating may be gold).
- The approach appears to be to assemble the components. No discussion was had on the design, construction, and performance parameters of the system. The team achieved >69% thermal efficiency, but they did not discuss how they achieved this efficiency. Variation in heat recovery was discussed and ranges given for cathode off gas, etc; however the electrical output was shown as a constant. The range for electrical efficiency would be 33% to 52% and the high value is probably incorrect.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.8 based on accomplishments.

- The project seems to be on schedule.
- On slide 18, it is impossible to gauge fuel cell performance without cell size, number of cells, temperature, pressure, fuel composition, catalyst loading, etc. It is difficult to assess fuel cell performance improvement without a baseline.
- The absorption enhanced reformer (AER) may need more R&D to improve natural gas conversion and thermal efficiency.
- More or less state-of-the-art components are going to be integrated together inside the final application.
- Progress during the last year appears substantial; however, the paucity of data in the presentation makes it difficult to assess the project's true technical progress. The hydrogen production data shown does not indicate the fuel type. Hydrogen composition into the fuel cell is not shown. The short run times for the major system components do not inspire confidence that the project durability target can be met. No cost projections are presented. Balance of plant issues appear to be surfacing, which will negatively affect system durability.
- AER systems are best used in continuous operations. So, what happens to overall system efficiency when the system is turned down? The team needs to better explore transitions and turn-down ratio effects on overall system efficiency.
- Sub-system results are clearly explained, but need more experiments at different output levels.
- They have completed the system design. They have achieved more than 2,000 h of testing on stack, 180,000 cycles, and 150,000 cumulative on cells during development. System efficiency projected was high, but not demonstrated; depends in part on using cathode exhaust for absorber purge (unproven). They claim very high efficiency on the stack (60% at 60% power). The cost was reduced 30%. The stack has 192 cells, 200 cm², and operates at 68°C.
- Progress toward operating an integrated system was demonstrated. The discussion on the CHP System Design was more of an advertisement listing properties or future properties. There was no information regarding decay in performance or stability of balance-of-plant (BOP) subsystem. The failures were all associated with BOP failures. Data showing system performance and stack performance as a function of time would have been helpful.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **2.2** for technology transfer and collaboration.

- Participation of BOP vendors is not explicit in terms of minimizing cost, weight, or volume.
- Collaborators are indicated in the presentation, but there is no indication of the extent or significance of their contribution.
- The use of CE standards at the component level was great, and so is the certification strategy.
- However, the project team needs better, clearer definition of work related to grid-interconnections (beyond the single slide.)
- Collaboration seems limited to working with subcomponent developers. Coordination with the utility to host the integrated test seems adequate.
- Partners are identified; but no information on contributions from the University of South Carolina (USC), California State Polytechnic University, Pomona, or SNL.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- IE is marching along the schedule and following the logical sequence of tasks.
- Fuel processor start time of 1 hour needs to be shortened.
- The future work plan is consistent with the project objectives to demonstrate a PEM-based CHP system.
- More details are needed on future work and the team needs a clearer definition or identification of critical barriers/road blocks.
- Necessary steps for an integrated consumer appliance and representative integrated testing all appear to be appropriate and underway.
- Future activities are identified. Is Phase 4 the same as Task 4? If not, there was no discussion on Task 4. If they are the same, then change the terminology to be consistent with the Gantt chart.

Strengths and weaknesses

Strengths

- Fuel cell stack performance appears good. Operating on high-purity hydrogen should enhance durability.
- This is a well-articulated approach and systematic testing of components/sub-systems.
- The team took a solid approach to system design, development, and testing, which may well facilitate early commercial entry and market transformation.
- The main project strength is that they got the system to work for a brief period of time.

Weaknesses

- Seeking certification for an as-yet-to-be-tested complete CHP system may be premature.
- They need to work on turn-down challenges with comparisons to incumbent/legacy systems used in this market space.
- The efficiency projected may be hard to achieve.
- There was very poor discussion of the technology and benefits of fuel cell systems. The following were unclear: the durability of the pressure swing adsorption unit (PSA); the concentrations of impurities exiting the PSA; the life of the adsorbants in the PSA; and the durability of the fuel cell. The project does not give adequate information; even information that would be very general in nature is not provided.

- Emphasis going forward should focus on durability issues and confirming performance projections.
- The project team needs to elaborate on future plans.
- They need to continue to completion.
- The reviewer recommends that the program be terminated unless more information is provided by IE. The information provided was more of a press release. The presentation was too high-level for a review.

Project # FC-26: Stationary PEM Fuel Cell Power Plant Verification

Eric Strayer; UTC Power

Brief Summary of Project

The objectives of this project are to 1) evaluate the operation of a 150 kW natural gas-fueled proton exchange membrane fuel cell (PEMFC); 2) assess the market and opportunity for utilization of waste heat from a PEM fuel cell; 3) verify the durability and reliability of low-cost PEM fuel cell stack components; 4) design and validate an advanced 5 kW PEM system; 5) conduct demonstrations of PEM technology with various fueling scenarios; and 6) evaluate the interconnection of the demonstration 5 kW power plants with the electric grid.



Question 1: Relevance to overall DOE objectives

This project earned a score of 3.3 for its relevance to DOE objectives.

- Small stationary fuel cell units are consistent with DOE's broad goals. Durability and cost are being addressed.
- This project focuses on system and performance optimization of a practical stationary PEM fuel cell system, and is very relevant to the goals and objectives of the Hydrogen and Fuel Cell Program.
- The project clearly meets and fully supports DOE objectives. The project advances the technology and prepares the technology to eventually enter marketplace.
- The project addresses major barriers for commercialization of cost and reliability.
- The project is relevant to the DOE Fuel Cell Program objectives. If the system appears viable, it could be selected for early deployments.
- The project serves objectives A, B, and field robustness.
- The coverage of this project is very broad and comprehensive. UTC fulfills the expectations fully. The integrated approach from basics to engineering is considered to be contributing notably to the success.
- However, there is one point which might be considered a weakness: as of now there is no PEM membrane material available which has been proven to fulfill the lifetime requirements of 40,000 hrs. Existing materials are good for about 10% to 30% of the required lifetime. A strategy should be outlined for how to overcome this potential barrier. Long-term testing of different materials may be necessary. It is not required to demonstrate the ultimately required longevity, yet the potential of a material should be validated.
- With DOE program refocusing to nearer-term applications, the project has increased in relevance this year.

Question 2: Approach to performing the research and development

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

- UTC is applying accepted design and engineering methods to this project.
- The durability and reliability of cell/stack components are being verified in parallel, as well as cost.
- The approach is based on implementing incremental improvements in system design and performance for stationary applications. The approach is clearly structured and appropriate.
- The project is sound and it's easily believable that UTC will perform well, but there should have been a greater discussion of barriers and challenges. The presentation discussed water management issues, but what other barriers and challenges make this a difficult project?

- The UTC project is very effective and contributes to overcoming boundaries, but an expansion of the challenges would have made this project stand out. What is truly difficult that others are not facing?
- Durability testing including start-stops is important. It is not clear what load cycle was utilized for the durability testing or how it relates to expected real world operation. Including multiple units in the tests provides some confidence in field durability data. It is not clear that the current platform (5 kW-33 cell stack) is an appropriate size to utilize for lab tests for an eventual 150 kW system.
- UTC displays an understanding of requirements for commercial viability in the back-up power market. Reduced cost is absolutely required, as is operability and durability that matches or exceeds conventional technology.
- The technical approach is professional and scientifically sound. The system's approach, which has been taken, is strictly oriented to overcome the barrier of commercialization through addressing power density, stack cost, and degradation. See the potential barrier above.
- The approach is solid, as expected from a long-term successful commercial system developer. Stack cell components are evolving from demonstrated UTC PEMFC stacks. The project appears on track for meeting their internal metrics and advancing progress toward DOE targets.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.6 based on accomplishments.

- Power density has been quadrupled.
- Grid connection issues are being addressed.
- Cost has been reduced by 30%.
- Several early units are installed in the field and operating very well.
- Significant progress has been reported in improving system design and performance, while lowering cost based on a total system perspective (both fuel cell stack and balance of plant).
- The project is clearly working towards goals and yields a great deal of product and data. Program metrics indicate that work is completed on those areas where significant barriers could exist, and that remaining work most likely has limited barrier threats.
- The team has increased the packing factor by ~45% and increased power density to ~28 kW/m³ for a 5 kW system. It has developed water management strategies that allow for <30 second start-up time for fuel cells; the team achieved >3,500 h operation and >450 starts on the system. The team achieved good system efficiency (45%), exceeding its target. It appears to be behind schedule; 60% complete after 4½ years on a 5 year project.
- Considerable progress has been made in the past year. The UTC advanced system is 30% less costly than the baseline system. The power density matches that of engine generator sets. Costs, as has been the case throughout this project, are not presented, and neither are cost targets.
- A low-cost stack has successfully been developed. Degradation was investigated, yet the time of 100 hrs is not sufficient to address the required longevity for a stationary application. The power density of the UTC system performed well when benchmarked against other commercial fuel cell systems. The 5 kW systems performed well with 42% efficiency and 99.6% availability.
- The program is very focused, straight-forward, and designed to overcome the main barriers. As mentioned above, the barrier of limited longevity of PEM stacks is not addressed appropriately. The rating is kept on outstanding, since in all other aspects the program falls into this category.
- It appears that considerable progress was made this year on durability, power system volumetric size/packaging, cost reduction, and system integration.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **2.6** for technology transfer and collaboration.

- The team is well-rounded, including testing services and power electronics/grid entities.
- Appropriate collaborations are reported that augment the expertise of UTC Power. Clearly the collaborators have made significant contributions to the impressive results reported.
- Partners' roles exist, but this project seems close to being solely a UTC affair, especially as Houston Advanced Research Center (HARC), U.S. Hybrid, TDI, and Avalence are non-cost share suppliers rather than equal collaborators and partners.

- The project appears to most closely resemble the statement that most of the work is done at UTC with little outside collaboration, especially given the large scope of the project, almost \$11.4 million from DOE.
- The presentation makes clear that the role of partners is non-cost share, but there remains little definition of their roles. Their roles seem miniscule compared to the effort of UTC, almost as if their greatest and sole role is to be listed as partners for the project.
- The project team is collaborating with HARC, and others.
- The development efforts are concentrated at UTC and its suppliers. Numerous locations have been identified as demonstration sites which could provide the information necessary to verify commercial viability.
- There is a limited role of cooperation partners. Since UTC has a broad in-house base in fuel cell technology and systems technology, collaboration is more critical and this might not be regarded negatively.
- System-development collaboration seems limited to working with component developers and testing at the HARC. Coordination with a number of test locations seems on track.

Question 5: Approach to and relevance of proposed future research

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

- Future work is a logical progression of the project. It looks as if the project will need to be extended.
- Adequate plans for completion of this project were described.
- Work to date appears to have been significant and supports future completion of the project as well as possible transitioning of the product to market.
- Future work is carrying out field demonstrations. Bringing the field test results back into the reliability modeling to get feedback in to the loop would be very beneficial.
- Not much time is left in the project. It may be too short to provide sufficient information to determine commercial viability.
- The project appears to be a well-planned, commercially focused effort with multiple test locations and fuels. It should result in a near-commercial system suitable to a variety of special applications.

Strengths and weaknesses

Strengths

- There are strong collaborators.
- UTC is a capable company that clearly is a leader in advancing the hydrogen economy.
- The main strength is the high and comprehensive competency of UTC. The project is designed to that strength and is well-conducted.
- The team is an experienced developer executing and planning well. Internal metrics add valuable commercial focus to DOE system-level targets.

Weaknesses

- There is very little collaboration with other organizations.
- There is a lack of cost targets and any indication of how close this system is to meeting those targets.
- There is a limited longevity of PEM membranes.

- The project team should consider making the system freeze-capable.
- None are noted at this time.
- Extending the demonstration period should be considered so that sufficient data can be gathered to assess the commercial viability of the system.
- The project team should include investigations as to whether, with novel membranes, the 40 khr goal can potentially be achieved.
- Continue to conclusion.

Project # FC-27: Intergovernmental Stationary Fuel Cell System Demonstration *Richard Chartrand; Plug Power*

Brief Summary of Project

The objective of this project is to design and produce an advanced prototype proton exchange membrane fuel cell (PEMFC) system with the following features: 1) 5 kW net electrical output; 2) flex-fuel capable (liquefied petroleum gas, natural gas, ethanol); 3) reduce material and production cost and increase durability; 4) increase electrical efficiency over the current alpha design; and 5) increase total efficiency by incorporating combined heat and power (CHP). Plug Power will also show a path to meet long-term DOE objectives, including 1) 40% system electrical efficiency: 2) 40,000-hour system/fuel cell stack life; 3) \$750/kW integrated system cost (with reformer); and 4) \$400/kW fuel cell stack cost (direct hydrogen).



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.3** for its relevance to DOE objectives.

• Relevance would have gotten an outstanding rating if one of the fuels for this system was hydrogen. The team claims to show a path to \$400/kW fuel cell stack cost operating on direct hydrogen. The PI does not test direct hydrogen based on a relevance chart, not to mention that this was a high-temperature system until chart 13. Is it a high-temperature system, and what is Ballard's function with a high-temperature system?

Question 2: Approach to performing the research and development

This project was rated 2.9 on its approach.

- The approach is to follow accepted engineering principles to design, build, and install the prototype demonstration unit.
- The unit will be installed at Construction Engineering Research Laboratory (CERL) to provide CHP for an existing building.
- The team has conducted demonstrations of a low temperature unit at CERL and a high-temperature unit at Union college, both reformate-based and providing both power and heat. The low temperature unit will have 32% efficiency.
- Great identification of tough barriers and challenges including system efficiency, system and fuel cell stack direct material cost, and system and fuel cell stack durability. These are barriers that if only partial progress was achieved, it would still be a significant accomplishment.
- The work is highly relevant towards meeting DOE goals and could yield benefits throughout industry.
- The approach is sound. Plug Power is focused on cost reduction and system simplification. The decision was made to purchase fuel cell stacks from Ballard for integration into the Plug Power system. This decision conserves Plug Power resources and provides a developed fuel cell stack for integration into the system.
- The project seems to be more system design and integration rather than component development or improvement, with a focus on grid interconnection testing. They selected a Ballard stack and have developed GenSys targets to supplement DOE stationary targets.
- The approach chart was a list of the activities from a Gantt chart and did not provide information on what the technical problem to be solved was and how the approach would lead to a solution.
Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.7 based on accomplishments.

- The project appears to be on schedule and making tangible progress. Site installation is imminent.
- Concept development and system definition is 100% complete. The system integration part of the project is 40% complete, and the field demo task is 10%. Have passed design review with DOE. System integration began for both steady state and transient operation. The team claimed to achieve system material cost reduction of 53% (in production quantity).
- There was great identification of tough barriers and challenges including system efficiency, system and fuel cell stack direct material cost, and system and fuel cell stack durability. These are barriers that if only partial progress was achieved, it would still be a significant accomplishment.
- The team has made clear progress compared to last year's presentation.
- The prototype system has been designed. Plug Power indicated that they had achieved material cost reductions of about 53% compared to the system as defined in the previous year. The reduction assumes production quantities of 10,000 units per year; however, the presentation did not indicate how close Plug Power had approached the DOE cost target.
- The prototype system achieved an electrical efficiency of 32%. Achieving the 40% target could be difficult.
- The team passed DOE go/no-go decisions for concept development and system definition. Efficiency is projected at 32%, far below the DOE target of 40%. They have a creative definition of "appropriate performance," that being components that demonstrate their "specified performance."
- The technical accomplishments provide information that progress is being made, but do not discuss the benefits of the accomplishments. There was no information on performance and durability of the system. Was this proven previously? The criteria for passing a go/no-go decision point were not fully identified. It is not clear if this is polybenzimidazole (PBI) or a perfluorinated sulfonic acid system. These are two very different fuel cell systems with different performance targets.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.0** for technology transfer and collaboration.

- CERL has extensive experience with fuel cell installations.
- Ballard is a respected fuel cell supplier.
- National Grid (utility) provides grid connection expertise.
- Both CERL and Union College are just demonstration sites. They have done good work with National Grid to get installed at Union College.
- Plug Power appeared to avoid resume-building that listed team members whose sole role was to be listed as a partner on a presentation. Instead, Plug Power appeared to have teamed with strong active partners. Though few in number (three partners), the partners appeared to be true partners in the project.
- They were well-coordinated with CERL in the role of customer activity. CERL is providing requirements that Plug Power is working to meet. This aspect will be crucial to making the product saleable in the marketplace.
- Strong partners are involved!
- The major collaboration is with Ballard Power Systems that brings world-class expertise in fuel cell technology to the project. The system will be demonstrated at the U.S. Army CERL site. A high-temperature system will be demonstrated at Union College. National Grid will participate in defining and specifying the interconnections between that system and the existing electricity grid.
- Ballard is supplying stacks. There is coordination with U.S. Army's CERL and National Grid for testing.
- It is not clear why collaboration with Ballard on system integration of a high-temperature stack is beneficial. Maybe a definition of high temperature is needed. Is this a PBI-phosphoric acid system, a silicon carbidephosphoric acid system, or a perfluorinated sulfonic acid system?

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- Future work is straightforward for this demonstration project.
- The team will install a unit at CERL in the 3rd quarter of 2009 for winter heat. Union College is awaiting installation. They will complete field tests and post test analysis and close out in 2010.
- Work plans are clearly built on past success and when work plans are completed, the work path will lead to well-thought next steps. Current efforts appear to be part of a coordinated plan so that current work can be built on with future work just as current work was built on past work.
- The project plans are to complete installation and shakedown testing of the units in 2009 and commence longerterm testing in 2010.
- No path to improve efficiency was suggested. This project will demonstrate grid-connected operation (which has been demonstrated before), and does not seem to advance the state-of-the-art.
- What is the prototype system to be commissioned at CERL? Is it the same high-temperature system for Union College? What are the criteria for the commissioning of the system at CERL? Not enough information was provided.

Strengths and weaknesses

Strengths

- The project team put on a grid at Union College; a good example of working with a utility.
- This is a well-coordinated project in all aspects.
- The team took a good approach and good collaboration was apparent from the presentation.
- The team has done well with system integration and packaging.
- Plug Power has extensive 5-kW system building and testing of PEM.

Weaknesses

- There is some question that it will be possible to complete the project on time. Only two units will not prove out durability and show potential for cost reduction, as is claimed. Very short term testing is to be done (less than one year).
- The presentation did not convey any information concerning the cost of the systems. The presentation did not provide an indication of the overall efficiency expected from these systems, or any details concerning the technology employed in the high-temperature system.
- Nothing in the project seems to advance technology toward DOE targets.
- It was not clear what the fuel cell system was, because data were not made available.

- The team should add in more units to prove out durability and cost reduction.
- No recommendations are noted.
- Future presentations need to be more informative and provide a better description of the technology.
- The remaining project is primarily system testing, which should probably be completed at this point.
- The team needed to define the system to the reviewer.

Project # FC-28: Development of a Low Cost 3-10kW Tubular SOFC Power System

Norman Bessette; Acumentrics Corporation

Brief Summary of Project

Acumentrics creates battery base uninterruptible power supplies for harsh environments. The overall objectives for this project are to 1) improve cell power and stability; 2) cost reduce cell manufacturing; 3) increase stack and system efficiency, 4) prototype test meeting system efficiency and stability goals; and 5) integrate to a micro combined heat and power (mCHP) platform.

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>



This project earned a score of **2.9** for its relevance to DOE objectives.

- The project is ultimately aimed at mCHP units, consistent with DOE goals.
- This project is very relevant to the Fuel Cells subprogram since responsibility for solid oxide fuel cell (SOFC) stationary systems were added to the subprogram portfolio.
- Project goals are to reduce SOFC cost, increase power density, increase efficiency to >40% lower heating value (LHV), increase durability to meet a 40,000 hr lifetime for stationary power applications, and CHP.
- There was some description of relevance to DOE goals, but not very clear.
- The mCHP appliance could be an important early market for high-temperature fuel cells. With the DOE refocus to nearer term applications, this project is very relevant.
- The project does not fully address the program goals of the Hydrogen Program. The 1-kW home appliance is better suited for Europe or Japan than North America. The program does not appear to address the trade-off of electric power which is costly, and thermal energy which is inexpensive.

Question 2: Approach to performing the research and development

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

- The list of necessary improvements is listed, but details of how the improvements will be achieved are vague.
- The proposed approach to systematically improve system design and performance is appropriate and sound.
- This work started as a Solid State Energy Conversion Alliance (SECA) project and has been funded through this program since April of 2008. The approach is to perfect the system components, then integrate them into a system. The system will be a mCHP prototype with 90% energy efficiency. The mCHP will have 10%-25% lower primary energy consumption, 10%-25% lower CO₂ emissions, and 10%-25% lower cost. It is intended to use line natural gas for fuel; a 1 kW system for residential.
- The barriers being addressed are key to improvements.
- There is good focus on overcoming relevant barriers and a good approach to attacking each problem.
- The project demonstrates a systematic approach to improving required performance, cost, and durability. Internal targets have been developed, and progress toward those targets assessed and used to guide further development activities. They have an understanding of real-world heat to electricity ratios (residential maybe 3:1) and implications for system design and operation are apparent.
- The "Acumentrics Approach" slide states that they were going to make things better, but does not provide information on the pathway to improve the power and stability or stack integrity. Better explanations are needed.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.1** based on accomplishments.

- Several of the progress graphics only show data through 2007. What progress has been made in the last 1.5 years?
- Most of the data are for single cells. Stack improvements can only be inferred from the single-cell data and from the diminishing stack volume.
- There are insufficient data on the uniformity of fabricated cells in terms of initial performance and degradation rate.
- Very significant progress was reported in system design and performance. Advances in size, weight, and volume were also reported.
- The project is 25% complete. Because the project has been funded for many years through SECA funding, it is difficult to determine what has been accomplished explicitly using Energy Efficiency and Renewable Energy (EERE) funding. Apparent progress has been made in manufacturing and moving to isostatic pressing; elimination of one braze joint; decrease in tube wall thickness by 45%; improved (decreased) cell degradation rate to approximately 1% per 1,000 hr. They have also apparently been able to run the system without a purge. Progress has also been made in comparing catalytic partial oxidation (CPOX) with steam reforming. Steam reforming gives better thermal balance. Also, decrease cells in the stack from 126 to 36, although this probably started under SECA funding.
- Good improvements in cell degradation rate have been made.
- Manufacturing process improvements contribute to cost reduction.
- The project team has made good durability progress.
- CPOX and steam methane reforming capabilities are good for multiple applications.
- Heat recuperator weight and volume reductions are excellent.
- Good progress was demonstrated on improving performance and durability. More attention to projected costs would be good.
- Some (much?) of the progress reported seems to have been under the SECA program, but technology progress is nonetheless impressive. Progress that can be specifically tied to this project, such as lower-cost, higher performance tube manufacturing is also impressive. Electrical efficiency is high-(46%-48%); heat utilization further improves fuel conversion to useful products.
- Acumentrics data shows improvement in the cell power and the degradation process. How were these improvements achieved? In Chart 11, what is Method 1? Progress is shown, but explanations are not given. If "equivalent performance" was achieved with a reduction in wall thickness, what was the benefit? Was Acumentrics seeking improved performance with a reduction in wall thickness? What is the average layer voltage?

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **2.6** for technology transfer and collaboration.

- The list of strategic partners is impressive, but their roles are not elucidated.
- There was no discussion of collaborations. It is assumed that all of the work was done in-house. Some interactions with European energy providers were discussed. It is suggested that collaborations with U.S. energy companies be sought in order to maximize the projected benefits to U.S. taxpayers.
- There were no real collaborations, but they were demonstrating a residential system in Europe with utilities.
- The team is complimentary.
- European members have good experience
- The collaboration seems excellent. Coordination of European tests (which must utilize "waste" heat) seems good.
- The "Strategic Partners" list does not mean collaboration and coordination with other institutions. A better explanation is needed.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

This project was rated 2.1 for proposed future work.

- There are no "Future Work" slides.
- The future plan appears to be adequate to complete this project in an orderly and timely fashion.
- They need to finish units and test home units and get data. Future plans are not well-defined.
- The plan seems to be adequate.
- Additional pathways to improve current density will be more productive.
- Additional information would be helpful.
- Future plans have not been clearly addressed. Apparently, future plans are to continue the current work.
- This project seems on track to advance the state-of-the-art, perhaps significantly.
- There is no proposed "Future Work" chart.

Strengths and weaknesses

Strengths

- The CHP application is positive. They have demonstration of residential units in a willing market (Europe).
- The technology based on an anode-supported design has shown maximum advantages.
- The team has a good technology base.
- The team has an experienced developer and a strong approach, with measurement of metrics and redirection as required.
- Acumentrics is experienced in the development of SOFC tubular stacks and cell components. They have made improvements, but it is not clear they are reaching their business targets.

Weaknesses

- The project shows very low power density for cells, <0.3W/cm². This will make it difficult to commercialize. They only will demonstrate in Europe.
- SOFC technology is not as mature as other fuel cell technologies, and will likely not be market-ready soon.
- Data on cell stack life was not presented. Does the cell stack last five years in order to fulfill the needs for CHP application? Little or no information was provided on methods of improvement. The application is not consistent with the Hydrogen Program.

- The project team should seek U.S. collaborations to define system requirements applicable to U.S. stationary power deployments.
- The team needs to provide system cost analysis, and work to demonstrate in the U.S.
- Efficiency data should be based on electrical AC output.
- Continue to conclusion.
- Acumentrics should define their targets better so a judgment on progress can be made.

Project # FC-29: Fuel Cell Systems Analysis

R.K. Ahluwalia, X. Wang, K. Tajiri, and R. Kumar; Argonne National Laboratory

Brief Summary of Project

The objective of this project is to develop a validated system model and use it to assess design-point, part-load and dynamic performance of automotive fuel cell systems. This project includes supporting the DOE in setting technical targets and directing component development as well as establishing metrics for gauging progress of research and development projects. Argonne National Laboratory will 1) develop, document, and make available versatile system design and analysis tools; 2) validate the models against data obtained in the laboratory and at Argonne's Fuel Cell Test Facility; and 3) apply the model to issues of current interest.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.6** for its relevance to DOE objectives.

- The project to develop and validate fuel cell component and system level models is critical to the success of the DOE Hydrogen Program and addresses many of the key targets for performance, system thermal and water management, air management, start-up and shut-down time, transient operation, and cost.
- This project is developing a sophisticated fuel cell system model that provides very relevant input to the Fuel Cells subprogram.
- Having a model to provide fuel cell system analysis is highly desirable; however, so much of the technology that really drives major portions of the system definition is still on fairly steep development curves. Furthermore, the details of system architectures are generally among the most tightly guarded secrets with the original equipment manufacturers (OEMs), since these are generally the areas where competitive advantage is key. It is important that this effort be focused in areas that are more generalized whenever possible.
- The project could extend systems expertise to all fuel cell applications.
- There is reference to this work in the Directed Technologies, Inc. (DTI) cost analysis work presentation, but no linkage explicitly explained in this talk; integration of efforts should be highlighted.
- This project is fully relevant and supports program targets and goals. It provides necessary input to cost studies, but the program is not absolutely critical to the success and progress of the technology, i.e., the results of this program do not directly "raise the bar."
- The technical program supports the DOE program and FreedomCAR technical teams by providing critical modeling and analysis for the DOE of industry and university performance/durability data. The project evaluates the complete system and provides baseline analysis of balance-of-plant (BOP) components. It developed a baseline design for fuel cell systems and established interaction of BOP components in the system.

Question 2: Approach to performing the research and development

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

• The approach is a good balance between component- and systems-level considerations in optimizing fuel cell systems. The clearly identified baseline fuel cell system design and specific components provides good starting points against which to compare other components and system trades. The approach provides significant

flexibility to consider other components and designs. The project took a strong approach to provide a widely available modeling capability, rather than an overly proprietary model that cannot be broadly built upon.

- The approach involves continuous improvement of the comprehensive, complex analytical model and validation of the model with the latest fuel cell experimental performance data. The results of this model couples with a companion cost model to provide valuable planning for the Fuel Cells subprogram.
- The project is too limited on air machinery and thermal management system. It is not clear that stack model is realistic enough; there was no collaboration with an appropriate organization like an OEM.
- The project is very focused, maybe a little too much so; three major factors are highlighted.
- The approach could be broadened.
- Evidence of more parties from the industry tuning into this work would be valuable.
- The approach is appropriate for the stated goals.
- The approach is well-organized and based on experimental data. A potential weakness is the dependence on data from industry without the ability to crosscheck the data.

<u>Ouestion 3: Technical accomplishments and progress toward project and DOE goals</u></u>

This project was rated **3.0** based on accomplishments.

- The project has been very flexible and agile in evaluating key issues during the past performance year.
- Substantial progress has been made during the past year in model development and improvement. The model has been used to explore system design configuration and allows the subprogram to make informed and validated decisions regarding programmatic directions, performance target definition, and research investment opportunities.
- Subfreezing start-up simulations are not likely to be very helpful since they lack specific details of the stack and membrane electrode assemblies (MEA). It is not clear that the issue of ice formation in the electrodes is accounted for.
- The work on system pressurization and freeze start is excellent.
- Attention to duty cycles and integrated efficiencies seems lacking.
- What about the battery or energy buffer? It is missing! This needs attention as a key (potential) powerplant component, especially hybridization, electrical issues, and battery performance decay.
- There was little consideration of aging effects overall.
- For a year's worth of effort dedicated to analysis, results seem a little light.
- Modeling expertise is very solid but needs to be applied to more critical areas, or at least where the effort can contribute to advancing the state-of-the-art (impurity, freeze, water management modeling, and validation).
- The technical accomplishments working with 3M and Honeywell provide the background validation of the two emerging technologies. The team identified efficiency issues for the compressor/expander module. The discrepancies in the Hawaii Natural Energy Institute (HNEI) CO data and the simulation were presented in additional data, but it was not fully discussed why the discrepancies occurred. The data analysis on the 3M materials needs a much better explanation than was presented at the AMR. The test appears to be at constant efficiency (50%) with a strong variation in the Pt content with temperature and relative humidity (RH). A 5°C temperature difference (90°-95°C) greatly changed the Pt content, e.g., at 60% RH. A 5°C change is very common within a cell, so this would suggest the system is working on a knife edge. There was no discussion or explanation which was disappointing.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.4** for technology transfer and collaboration.

- Results indicate very close collaboration/interaction with the full spectrum of component and system developers to develop and validate the models. These interactions provide confidence in the model outputs.
- This project has extensive collaborations ranging from those providing performance inputs for model validation to those who use the tool for guidance and direction.
- The level of collaboration was good, given the collaborators chosen.
- The project could be extended to a broader field of stakeholders, e.g., Vairex should have been consulted regarding the compressor motor expending unit, in addition to Honeywell.

- Interactions outside the U.S outside the US should be included if they exist.
- The team has a good working relationship with 3M, the Hydrogen Fuel Quality Workgroup, and Honeywell.
- The team needs more collaboration with other DOE impurity/contaminant programs and freeze and water management programs. These areas can greatly benefit from detailed modeling support.
- Efforts with 3M, Honeywell, HNEI, etc. are all very good. The Hydrogen Program establishes ANL as the lab to develop models and simulations for much of the R&D in other projects. It would be important to validate the quality of the ANL simulations and modeling with the input from 3M, HNEI, etc.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.7 for proposed future work.

- The approach to future work builds on prior results. It would be nice to see when this project is anticipated to move beyond model validation and begin to impact some of the technical projects.
- The proposed future work is logical and sensible and continues to meet the analytical needs of the Fuel Cells subprogram.
- The project team needs to extend the model to non-automotive applications.
- I would like to see a high-temperature case exploiting high power density (allowing efficiency relaxation) across a range of driving cycles to really understand heat rejection issues and possible thermal management strategies; the radiator question needs to be revisited.
- A fundamental analysis of the trade-off of stack pressure drops and cell pitch and aspect ratios should be added to the pressure analysis.
- The project team should validate freeze modeling with Nuvera's freeze program.
- The team needs to increase effort on impurity and contaminant modeling (cathode air borne impurities as well as anode).
- They should consider a joint effort with LBNL, LANL, and NIST to elucidate water transport in the MEA.
- The future work is a shopping list of activities with no real statement of what would be delivered.

Strengths and weaknesses

Strengths

- The team has a solid technical staff generating good system models.
- The system tool is VERY powerful and broadly applicable.
- The PI is dynamic and has component modeling expertise.
- The project has access to a large amount of data generated by other research groups in the Hydrogen Program, and has applied simulation models to increase the understanding of the fuel cell processes. ANL is a collection point for many of the BOP data and simulations, which is a necessary function for the Hydrogen Program.

Weaknesses

- The team could do a better job clarifying what is purely modeling, what is modeling using experimentally provided parameters, and what has been validated experimentally. This project covers a lot of things, and each requires a somewhat different modeling approach depending on the type of data available and what is possible from a purely theoretical standpoint. They could do a better job making the iterative process more clear (i.e., whether they started with a model, then saw how it fit the data, whether the model has been modified based on newer data, and whether new experiments were done to validate the model outcomes).
- The relevance and impact of this modeling effort is hampered by the lack of direct interaction with stack and system designers.
- The results seem a little lean for the funding level and timeframe.
- Duty cycle considerations are needed.
- There is too much emphasis on system optimization (this is the realm of the OEMs).
- The plots of the stack data appear to have "data points" and simulation curves. If a stack was operated, the plots should show its size/rating. If not, the project team should take the data points out of the plots since it gives the impression of real data. The PI needs to identify firm deliverables, e.g., how they will validate JARI and U.S. data and whether there is some absolute model that validates the data.

- It would perhaps make more sense to place less emphasis on these system-level activities in the future, because of the inherent issues associated with a national lab being able to get the most relevant technical input.
- There should be less focus on system optimization, and less effort on well-understood thermal management.
- The team needs to increase the focus on impurity modeling, freeze validation, and water transport through all layers of the MEA. They should validate their models using resources from the other DOE Hydrogen Program projects.
- There should be more effort on validating other DOE Hydrogen Program materials and claims.
- Is the Argonne reference 2009 data consistent with all vehicle fuel cell manufacturers or just one? With the program moving toward pre-automotive fuel cell systems, changes to simulation models and fuel cell system design data should be explained.
- It would be good to see more evaluation of systems used in other DOE-sponsored programs. It would also be good to see inclusion of other possible components for the air machinery and thermal management subsystems.

Project # FC-30: Mass-Production Cost Estimation of Automotive Fuel Cell Systems

Brian D. James and Jeffrey A. Kalinoski; Directed Technologies, Inc.

Brief Summary of Project

The objectives of this project are to 1) identify the lowest system design and manufacturing methods for an 80-kWe direct H₂ automotive proton exchange membrane fuel cell (PEMFC) system based on three technology levels (2008 status technology, 2010 projected technology, 2015 projected technology); 2) determine costs for these three technology level systems at five production rates (1,000; 30,000; 80,000; 130,000; and 500,000 vehicles per year); and 3) analyze, quantify, and document the impact of system performance on cost. Some costs were not included (warranty, building costs, sales tax, and non-recurring engineering costs).



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.6** for its relevance to DOE objectives.

- A reliable, detailed manufacturing cost estimate is essential to gauging progress and determining the relative importance of technology areas for the allocation of resources. Manufacturing cost is an important metric for original equipment manufacturers (OEMs) to make a go/no-go decision for the hydrogen fuel cell vehicle production decision.
- Cost analysis is imperative to measure technology status and important input for R&D direction.
- The program is relevant and in support of the objectives of the Hydrogen Program; however, assumptions regarding vehicle volume ramp-up rate are probably over optimistic.
- The analysis establishes a baseline for the cost of materials and manufacturing of proton exchange membrane fuel cell (PEMFC) systems at production rates approaching automotive needs.
- This project has an important role for the program to ensure that fuel cell vehicles are economically feasible at future high-volume production rates, ensuring there are no show-stoppers.

Question 2: Approach to performing the research and development

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

- The extensive use of detailed Design for Manufacturing and Assembly (DFMA®) analysis for stack and system components provides a solid basis for the system costing. Representative component designs were used where actual configurations were not available. The system model is derived from input derived from individual component suppliers and not thermodynamically integrated.
- The stack technology assumption is good, except 2010 Pt loading. Why it is higher than 2008 status?
- Fuel cell system architecture and its evolution should be analyzed to lower the total fuel cell system cost before technology assumption is fixed. There should be some trade-offs between operating conditions and cost sensitivities of each component, e.g., if humidification cost sensitivity is small, the system doesn't have to eliminate external humidifiers.
- The barriers as described will not be significantly impacted by the approach. Manufacturing costs will be driven by vehicle volumes, and this assumption has already been addressed as over optimistic. Platinum group metal (PGM) costs will be out of the control of any manufacturer and projecting these costs is very difficult.

- The approach uses DFMA, innovation, and practicality, and grounds the results in realistic components and costs.
- The team took a solid approach, building on previous successful years of cost projections.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.4 based on accomplishments.

- Since the Directed Technologies, Inc. (DTI) 2007 cost analysis report, nearly every component cost has been updated or refined (some costs increased and some decreased) from both a design and analysis basis, which resulted in a net substantial system cost reduction. Further, new component technology and designs were analyzed and the cost was estimated for various system components.
- Progress in system cost analyses of this type is difficult, as many of the subsystems under consideration are in a constant state of flux. Without close involvement of OEMs and Tier 1 suppliers, true costs are difficult to pinpoint.
- Analyses and results are systematically developed and demonstrate a firm technology/cost base for projecting the cost of an automotive PEM fuel cell system. Some of the costs developed by DTI appear to be aggressive based on present industry practice. It was not clear there was adequate technical justification for the Treadstone coating work.
- The team gave an outstanding explanation of what has changed (specifically) since the 2007 analysis, and what impact that had on the overall fuel cell system cost (\$/kW net).
- The team made good use of detailed process analysis for things like stamping metal bipolar plates and membrane electrode assembly (MEA) frame gaskets.
- It is fortunate that Pt cost dropped precipitously due to the recession (by a factor of 2x), or the "current" evaluated costs of the fuel cell system could be significantly off.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.6** for technology transfer and collaboration.

- The detailed analysis of various new and improved components indicates substantial collaboration with component developers and suppliers.
- The team did not provide enough detail regarding collaborations.
- DTI works closely with industry to meet the goals of the cost analysis and provide a practical base for the data.
- Good collaborations are apparent with companies like Honeywell.
- The project team should discuss whether interactions have occurred with TIAX cost study, or are they totally independent?
- Would be useful for both TIAX and DTI to have one slide highlighting the major differences between the two studies.

Question 5: Approach to and relevance of proposed future research

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

- A plan and time schedule is presented in detail that includes NSTF, and low pressure fuel cell system analyses that are among the most fruitful areas for cost reduction.
- As mentioned at the "Approach" section, the cost analysis approach should be improved.
- Proposed future work plows no new ground and refines only what those analysis already performed.
- The future work discussion was lacking in detail, and appeared to be more of a schedule than a statement of deliverables. Future work should include the polybenzimidazole (PBI)-phosphoric acid (or equivalent fuel cell system) analysis.
- The 2009 updates look interesting.
- How will range-extended EVs (such as the Volt) be factored into potential sizing reductions for the system? Specifically, how much more (\$/kW) would a 40 kW fuel cell system cost than the 80 kW system?

Strengths and weaknesses

Strengths

- The team conducted fair detailed work with a good synergetic view of actually all cost influencing parameters/technologies.
- All major and some minor stack and system components have had detailed DFMA analysis performed, including updates for recent advances. The number of components included is extensive and representative of a complete Bill of Materials (BOM) assuring that the calculated cost is realistic.
- Cost modeling development was good.
- The team is well-grounded in the principles for conducting cost analyses. The program is well-organized and structured.
- The team gave an excellent presentation with clear explanation at each step of the process.
- They provided detailed results of what changed in the updates and what the impact on costs were.
- Reduction of labor cost was appropriate given the current labor market conditions.

Weaknesses

- The system model was based on component vendor input rather than thermodynamics, which can lead to incorrect performance/cost trade-offs and erroneous system efficiency.
- The vehicle volume projections are over optimistic.
- A broader range of information sources including European and Asian fuel cell manufacturers could improve the data.
- The PIs need to include the impact of handling freeze on cost.
- While the Monte Carlo analysis was performed, it would be nice to see the results presented.

- The presentation is kind of lost in details, and the slides are by far too busy and almost impossible to read/understand.
- A comprehensive summary with the most important findings is recommended.
- In addition to the extant plan, the team should increase the 2009 scope so that DTI can incorporate a thermodynamically correct model equivalent to that developed by ANL and use it to make the cost/performance trade-offs required to attain the lowest system costs. Further, multi-variable sensitivity cost studies should be added.
- The team should expand the effort to include PBI-phosphoric acid, silicon-carbide-phosphoric-acid, and other high-temperature (HT) membrane fuel cell systems. Do a comparison of these HT fuel cell systems to PEM.
- With such a high sensitivity to Pt cost, they need to have a plan of how to have year-to-year comparisons (and yet still a relevant projection) if Pt cost goes back up to where it was at its peak and stays there.
- Analysis of lower pressure systems (which was mentioned) is critical because of multiple OEMs heading in that direction.

Project # FC-31: Direct Hydrogen PEMFC Manufacturing Cost Estimation for Automotive Applications Jayanti Sinha, Stephen Lasher, and Yong Yang; TIAX LLC

Brief Summary of Project

The overall objective of this project is a bottom-up manufacturing cost assessment of an 80 kW direct H₂ proton exchange membrane fuel cell (PEMFC) system for automotive applications. The objectives for 2008 were to perform 1) a high-volume (500,000 units/year) cost projection of the Argonne National Laboratory 2008 PEMFC system configuration assuming a nanostructured thin film catalyst-based membrane electrode assembly (MEA) and a 30 µm perfluorosulfonic acid membrane; 2) bottom-up manufacturing cost analysis of both stack and balance of plant components; 3) sensitivity analyses on stack and system parameters; and 4) independent peer review of cost analysis methodology and results.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.6** for its relevance to DOE objectives.

- A reliable detailed manufacturing cost estimate is essential to gauging progress and determining relative importance of technology areas for allocation of resources. Manufacturing cost is an important metric for original equipment manufacturers (OEMs) to make a "go/no-go" decision for the Hydrogen Fuel Cell Program.
- An independent assessment of fuel cell system cost is directly relevant to the DOE Hydrogen Program objectives, and thus supports and is a necessary component of the program.
- None.
- The projection of cost based on present technology, present manufacturing capability, and future production provides a metric to judge the progress of the Hydrogen Program.
- Projecting fuel cell system cost is very important to show the glide pathway toward commercial viability in light-duty automotive applications.

Question 2: Approach to performing the research and development

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

- The utilization of the integrated system model developed by ANL gives additional credibility to the selection and sizing of components. This selection is critical to the determination of overall system efficiency, which in turn impacts system cost.
- Various types of fuel cell system architecture should be studied to lower the total system cost. Trade-offs between operating conditions and components cost sensitivities should be studied. Fuel cell systems for this cost study are not optimized to lower the cost.
- Overall model refinement should include some form of risk analysis in the approach description.
- The assumption of complete OEM control of the stack is unrealistic and should be changed to include some estimation of markup from tier 1 suppliers.
- A systematic approach was used that is based on inputs from the DOE HFCIT Program management, the Fuel Cell Technology Team, and the ANL system model. Critical Bill-of-Material was identified and used to specify manufacturing equipment and materials. The bottom-up approach provides practicality and realism to cost analysis. Interaction with industry builds confidence in the approach.

- The approach is solid, using a bottom-up approach to determining the high-volume manufacturing cost.
- There was good involvement of industry for vetting the assumptions and results; it keeps projects bounded by the inputs of experts.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.9 based on accomplishments.

- Major progress has been made in stack cost reduction as a result of lower catalyst loading and increased power density of the nanostructured thin film catalyst (NSTFC)-based MEA. The balance-of-plant (BOP) components do not appear to be updated and have not had any cost reductions compared to the 2007 cost study report.
- Cost analysis on 3M's NSTF catalyst MEA is valuable. It is a new catalyst concept and different from Pt/C.
- The program has accomplished their stated objectives in a competent manner, and overall results are reasonable and believable.
- I do not give it a higher rating since the program cannot, by its objectives, actually move the technology forward as it just assesses this progress.
- The whole project seems to have a seat-of-the-pants feel.
- The project systematically forecasts the cost of the cell components, cell stack, and BOP. The values are consistent, but in some cases optimistic, with industry inputs. The project maintains checks and balances through its interaction with DOE, the fuel cell technical team, and industry.
- This year's update looks like it has made incremental improvements to respond to any weaknesses from previous years.
- Excellent detail provided of the key costs of each of the major parts of the systems.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.0** for technology transfer and collaboration.

- Collaboration with ANL has been important in establishing a valid system model. Only limited collaboration with non-stack component suppliers is indicated even though these components contribute 50% of the cost.
- Interactions with the fuel cell technical team and an independent review panel are key steps that have been taken.
- However, it may benefit from earlier screenings of the system and assumptions with system developers.
- It is not clear as to the degree of collaboration between investigators.
- Collaboration with industry, ANL, and DOE is high and should be maintained. Fuel cell system models other than the ANL model should be considered.
- It is not clear how coordinated this is with the work of Directed Technologies, Inc. (DTI) (perhaps it is not supposed to be so that they are two totally independent/objective evaluations).
- I'm not sure if redundancies with DTI work are intended or accidental.

<u>Question 5: Approach to and relevance of proposed future research</u>

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

- Generic discussion is provided for proposed future work, but specific plans and focus areas are not available for review. It is anticipated that the substantial differences in the ANL 2009 system configuration would require a major rework in the costing.
- The cost study approach of the fuel cell system architecture assumption should be modified and include a system optimization process to lower the total cost.
- The modest goal of updating the stack and fuel cell system cost to align with current performance status, in this option year, are in line with the modest funding and appropriate for what is required at this time.
- The reviewer recommends removing the anode humidifier as developers have demonstrated they can do without (for example, see mechanization in SAE 2008-01-0420).
- I recommend using thinner membrane for improved performance and reduced cost (no greater than 25 microns).

- None.
- The future work will build on 2008 results for a final report.
- I look forward to knowing whether the preliminary 2009 estimates hold at \$24-33/kW.
- The team also needs to be evaluating smaller fuel cell systems to account for likely hybrid scenarios, including the much smaller size that might be used in a range-extended electric vehicle, such as the Volt.
- Back-up slides provide important information; see if some of those details can be worked into future review presentations.

Strengths and weaknesses

Strengths

- The presentation had clear graphics and introductory illustrative flow sheets in the presentation.
- Literally all aspects for cost were covered.
- The Design for Manufacturing and Assembly (DFMA®) bottoms-up analysis for unique fuel cell systems, internal technology-based cost models, and off-the-shelf costing for commercially available components is a good use of resources. Reliance on ANL system models provides a sound technical basis. Single and multi-variable cost sensitivity studies are a plus.
- The team performed a risk analysis of overall cost.
- They have an experienced team at TIAX and ANL. The bottom-up approach is believed to be the proper approach. Monte-Carlo methods are beneficial but need better explanation.
- Really like the Monte Carlo analysis to show the sensitivity of cost to changes in input parameters; it makes the result more believable than a simple single-point analysis.
- This project builds on a strong baseline from several years ago, with future updates providing meaningful and timely insights.

Weaknesses

- Not enough effort was devoted to non-stack system/component costs and the trade-off between the performance and cost of individual components.
- The whole analysis has a seat-of-the-pants feel, meaning that the level or depth of analysis seems rather shallow.
- The cost of membrane materials may be underestimated since high volume (for chemical industry) will not be achieved at 500,000 vehicles per year.
- The project team needs to clarify the role relative to DTI; compare methodologies, whether the same industry experts are consulted, etc.

- A comprehensive summary would improve the presentation.
- The team should incorporate the latest system/component performance and design results from the 2009 AMR into the 2009 cost analysis study. They need to perform more DFMA studies of unique fuel cell components, and evaluate the trade-off between system performance and cost to develop the lowest system cost.
- The team should consider replacing the 30 micron membrane with a thinner membrane, likely 25 microns or less for commercial automotive fuel cell systems.
- The cost of \$16/m² for the membrane seems a bit too high. The team needs to compare this cost to projections from GM (Mathias et al., "Two Fuel Cells in Every Garage," The Electrochemical Society Interface, Fall 2005).
- An area utilization of 85% of the plates seems too high. If feasible, the project team should confirm with the Graftech DOE plate project.
- The project needs more depth and increased involvement of collaborators.
- The team should continue effort and expand industry contacts to Europe and Asia.
- No major changes required; project is on a good course.

Project # FC-32: Microstructural Characterization of PEM Fuel Cell Materials

Karren L. More, Larry Allard, Harry Meyer, and Shawn Reeves; Oak Ridge National Laboratory

Brief Summary of Project

The overall objectives of this project are to 1) identify high resolution imaging and compositional/chemical analysis techniques for the characterization of the material constituents comprising proton exchange membrane fuel cell (PEMFC) membrane electrode assemblies (MEAs); 2) apply these analytical and imaging techniques for the evaluation of microstructural and microchemical changes that determine fuel cell stability; and 3) elucidate microstructure-related degradation mechanisms contributing to PEM fuel cell performance loss. Collaboration with industry, academia, and national laboratories will be conducted to make these techniques (and expertise) available to



correlate structure and composition with MEA processing and/or life-testing studies.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.9** for its relevance to DOE objectives.

- These techniques are tremendously valuable to various aspects of materials development and failure analysis. It is important that enough time/effort can be effectively channeled to application of these methods to a wide array of materials.
- It is an outstanding and unique tool for understanding electrode structures before and after testing.
- Understanding the catalyst distribution and morphology, and the chemical composition of the MEA with time is key to understanding the degradation mechanisms of the fuel cell and is then key to improving the durability.
- This project has great possibilities to investigate catalyst particles with various methods.
- This project provides a unique and very useful capability to the fuel cell research community for materials characterization and development.
- The PI has developed several novel techniques for MEA and other fuel cell materials analyses.

Question 2: Approach to performing the research and development

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

- The potential for these techniques to provide valuable insight is clear. The ability to image electrode materials at conditions more appropriate to fuel cell operating conditions is extremely valuable. My only concern with the approach is that Dr. More's lab seems to get inundated with many samples from a wide range of studies at other labs. For microscopy analysis of this caliber to be truly successful in identifying fundamental information about electrode materials structures and durability, it is essential that efforts can be focused on a well-thought-out set of samples. Typically, this requires more time than perhaps the collaborating parties are willing to invest.
- It is not clear if DOE is footing the whole bill for this work. The assumption is that this capability would have application in a range of nanotech areas. My only comment is whether there could be synergies with other applications to accelerate progress in developing techniques.

- This effort focuses on development of a user facility providing state-of-the-art facilities for users to study the composition of MEAs at various stages of degradation. This excellent/optimal approach leverages the expertise of the microscopy experts with the fuel cell MEA expertise of the users/collaborators of the facility.
- Established know-how is used and adapted to current topics of interest.
- The project applies state-of-the-art electron microscopy techniques (including sampling preparation, mounting, and handling) to very high resolution (to Å scales) for materials imaging and compositional analyses.
- To address special issues and user needs, the project develops specialized approaches, such as microtomy to obtain depth profiles, and low-angle sectioning to determine variations in even very thin films and samples.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.4 based on accomplishments.

- There are many fronts on which excellent progress has been made including the Å-level resolution of imaging and chemical analysis (with the Si-drift detector), *in situ* analysis of catalysts in liquid, 3-D tomography, and the ultra-low-angle microtomy method to perform through-plane chemical analysis via X-ray photoelectron spectroscopy.
- There are a lot of delays, which is understandable with such specialized equipment, but the presentation did a great job of highlighting progress.
- New tools have been added and existing capabilities have been improved.
 - Can study the chemical composition of MEAs to understand degradation;
 - Can look at the 3D distribution of the catalyst nanoparticle;
 - Can look in detail at the Å-level at the composition of the catalyst nanoparticle; and
 - Can study *in situ* the behavior of catalysts in a liquid electrolyte environment.

All of these are important accomplishments to help guide the community towards an understanding of the fundamental degradation mechanisms in the fuel cell.

- Large progress was made compared the last year report.
- This project is continuing to develop innovative techniques to provide useful data on MEAs and other fuel cell components.
- They are now able to provide Å-scale image resolution by aberration-corrected scanning transmission electron microscopy (STEM), which can be used to study core-shell nanoparticles, for example.
- They have built a liquid-flow cell for *in situ* electron microscopy at the nanometer scale.
- They are using electron tomography for 3-D analyses and imaging.
- They have developed ultra low angle microtomy to obtain as many as 19 30-µm "spots" across a 100-µm-thick MEA to characterize composition across the MEA.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.4** for technology transfer and collaboration.

- Clearly, this resource is being broadly used by many other institutions.
- Lots of collaborators; however, there is a lack of connectivity to any specific problem being addressed or solved. The audience is left to imagine utilization of the techniques for their own purposes. For example, there have been a lot of questions about electrode optimization:
 - o How uniformly dispersed is the catalyst in commercial products?
 - Why can 50% electrochemical surface area (be lost and performance unchanged?
- It would be good to show beginning-of-life vs. end-of-life with technical interpretation to fully bring home the value of these techniques.
- As an open-user facility, this project provides the best opportunities for technology transfer. The project has a good set of collaborations; still it seems there could be a significantly greater base of collaborations/users that could be developed.
- This project collaborates extensively with many of the organizations conducting fuel cell R&D.
- ORNL is also collaborating with several universities under the Shared Research Equipment Program, which provides access by other organizations to this project's specialized equipment and techniques.

<u>Question 5: Approach to and relevance of proposed future research</u>

This project was rated **3.6** for proposed future work.

- Continued refinement and application of these powerful methods will be greatly beneficial to fundamental understanding of electrode materials.
- Development of the techniques is core.
- My suggestion is to place more emphasis on application of the techniques to solving specific user/customer questions/problems. This approach will help a bigger audience appreciate the value, and undoubtedly induce more solicitations for use.
- The impact of methods on temperature (heating) and/or electrification of samples may need further characterization.
- Future work is guided towards providing the state-of-the-art capabilities for fuel cell microscopy in support of the DOE program. The team must continue to develop collaborations to enhance the work performed at the facility.
- Due to good cooperation with different partners, the established methods are further developed in the right direction.
- Their plans call for developing the various techniques still further by adding other capabilities, such as electrical contacts to the samples in the liquid flow cell for *in situ* analyses.

Strengths and weaknesses

Strengths

- The project exhibits the excellent technical skills of the PI and world-class instruments. There are seemingly strong support staff and facilities at ORNL. The "user" nature of the lab to allow ready access to these techniques by other organizations is good.
- They are a talented research team with state-of-the-art equipment, and the user facility gives everyone access.
- The instrumentation and techniques developed in this project provide unique insights into MEAs, which gives insight of great value in understanding materials degradation over time and as a function of the operating environment and operating history of the MEA.

Weaknesses

- The team has the potential to get bogged down in too much technique development and somewhat diffuse analysis of a wide range of samples. It is important that these tools are focused on a couple of carefully planned sets of samples to get at a more systematic analysis of electrode materials properties and degradation modes.
- The team is a little biased toward the techniques and the "wow factor" (which IS definitely impressive!) than bona fide problem solving.

- A thought on a study that could be broadly beneficial: Analyze several commercial catalysts in powder form, as a new electrode film, and as an aged electrode (run for a few hundred hours) to identify general features, such as:
 - carbon and catalyst particle size/morphology;
 - interaction of catalyst-support among these 3 types (how well each adheres);
 - carbon-ionomer interaction in the electrodes.
- Balance heavy lab/academia portfolio of collaborators with commercial/industrial collaborators, especially stack users with durability issues arising in real system applications.
- The planned future work described in the presentation should be executed.

Project # FC-33: Platinum Group Metal Recycling Technology Development

Lawrence Shore; BASF Catalysts LLC

Brief Summary of Project

The objectives of this project are to 1) achieve a platinum recovery rate of 98% to lower the effective cost of platinum used in a fuel cell membrane electrode assembly (MEA); 2) simplify the process so that platinum recovery is achieved using a single leach to reduce the cost of platinum recovery by increasing throughput; and 3) determine chemistry and reaction conditions to optimize platinum leaching to reduce reagent usage, reducing process cost, and to reduce cost of construction for the reactor vessel by identifying the most appropriate reactor liner. This project has achieved the objective of 98% platinum recovery, identified room-temperature alternatives, and reduced reagent usage for platinum leaching.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.6** for its relevance to DOE objectives.

- This project to evaluate Pt group recycling technology directly addresses DOE targets for overall cost and for stack material and manufacturing cost.
- Platinum is the single largest material cost component of the stack and system. Any reduction in this cost through recycling would move fuel cell stack costs closer to reaching the system cost targets.
- This is a great program to recover platinum and therefore reduce its cost as well as the cost of downstream products.
- Outstanding platinum recovery rates offer a method to minimize platinum costs and improve re-utilization of older systems.
- Due the fact that Pt loading is one of the main cost drivers in 2010 and 2015 stack cost analysis, the recycling becomes more and more relevant; therefore, this project has a high relevance.
- This project is developing simple, low-cost processes for recycling Pt. There is a significant concern, however, that this technology is not yet mature, and may change substantially between now and the time of commercialization.
- Pt recycling is a significant positive factor in stack life-cycle cost and PM supply, thus is of significant importance to the DOE Hydrogen Program.
- Recovery of the platinum from fuel cell electrodes is very important to the viability of fuel cell systems. This BASF Catalysts LLC project shows a relatively straightforward and cost-effective means to do so.

Question 2: Approach to performing the research and development

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

• This is a strong approach that is integrating prior work to develop a final system-level capability and analysis and focus on remaining primary challenges. The approach addresses both current and emerging polybenzimidazole (PBI) membrane MEA technologies. There is a good range of Pt recovery approaches evaluated, while keeping adequate focus on the overall goals/targets.

- The overall approach of exploration, validation, and process flow development has yielded positive results and attainment of project objectives of 98% or greater Pt recovery.
- Solid, steady progress has been made on a variety of well-identified challenges.
- The team has done very good work to overcome technical barriers and reduce costs of platinum recovery. The project was well-designed, feasible given the payback received, and fits well into overall DOE goals to make fuel cells affordable to the consumer.
- Their approach has been to select processes that are not specific to the platinum matrix, verify that the processes work with MEAs and other fuel cell components, develop a process flowsheet, and validate and seek process improvements.
- The PI has taken a methodical and comprehensive approach to the program and appears to have successfully met objectives, though it is unclear as to the potential impact of ultra low loaded and alternative electrodes which may be required to meet program commercialization (cost) objectives.
- The approach for this program is "outstanding" because they effectively accomplished their goals, and even expanded their effort to the BASF Celtec membranes. They made an effort to look at spent membranes that would be similar to what would be returned for platinum reclamation. This research is not very exciting, but is very good.

<u>Ouestion 3: Technical accomplishments and progress toward project and DOE goals</u></u>

This project was rated 3.6 based on accomplishments.

- Significant progress has been made in achieving the DOE targets and in identifying key issues/solutions for practical systems ranging from the Pt recovery processes to processing equipment corrosion impacts, and approaches to mitigate those issues. The team demonstrated a method for leach process quality control/analysis and made significant progress in simplifying the Pt recovery process (i.e., single vessel leaching). The team provided a good estimate of cost impacts for increasing Pt recovery by 1% from 98% to 99% based on their recovery experimental results.
- Process parameters appear to have been optimized and only details remain to be wrapped-up in a technology commercialization phase. The basic oxidizing acidic solution approach with aqua regia, HCl, H₂O₂, etc., had been identified in earlier patents by others.
- I wouldn't mind awarding BASF a score "5" for this area.
- Recovery processes offer real promise to lower platinum costs through the reduction of a number of in-use materials.
- It appears that BASF was willing to try, change, and adjust processes that meet goals, rather than finding a goal that meets the existing process.
- By developing a simplified leaching process, good progress was made since the last DOE report.
- A key accomplishment has been to achieve 98% Pt recovery without forming and releasing any hydrofluoric acid (HF).
- Another significant accomplishment has been the development of a one-step, one-vessel process (including vessel material validation).
- An added benefit of the process is that it avoids cryogenic milling using liquefied nitrogen cooling.
- The performance of the process has been verified with new and aged MEAs.
- The PI has taken a methodical and comprehensive approach to the program and appears to have successfully met objectives.
- It looks like they finished the job and are just waiting on 10,000 fuel cell cars now.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.1** for technology transfer and collaboration.

- The team had strong collaborations with partners that provide complementary expertise and facilities/capabilities.
- Collaboration has occurred with potential suppliers of processing equipment and chemicals.

- I recognize that BASF is only working with one collaborator now, which shows that the project has progressed from requiring a number of partners and collaborators to a project near its end-state. It is not worth punishing success, as BASF worked itself into a position to close-out collaborator support.
- Several important partners are involved in these activities.
- They have worked with several organizations from industry, suppliers, and consultants.
- The team may benefit from interactions with developers of novel electrodes (NSTF, core-shell) unless the PI can state assurance that the process will be applicable to these.
- BASF did a good job getting MEAs from different vendors; however, they might have expanded their effort more to some of the alternative catalysts being developed by other collaborators.

Question 5: Approach to and relevance of proposed future research

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

- The project is wrapping up. Proposed future work will complete key outstanding experiments and compile the results and analysis into a final report.
- Further process optimization/simplification is planned, but specific plans are not available for review.
- It is very nice to have a project completed as planned.
- This project has demonstrated outstanding year-to-year progress during the life of the project.
- The project has well-thought plans for the future work site floor plan.
- They will primarily be wrapping up some final points (effects of sonication to promote leaching, optimize process temperature and pressure, etc.) in the project that is already 98% complete.
- The program is near completion, but I'd like to see the following:
 - An estimate of yield and cost benefit/impact of lower Pt loaded electrodes (0.025-0.05 mg/cm² anode, 0.05-0.1-0.2 mg/cm² cathode) as well as alternate structured electrodes (NSTF, core-shell); and
 - An estimate of overall cost benefit (Pt yield vs. process cost) to assist DOE in assessing life-cycle Pt cost.
- The program ends this year. BASF has the opportunity now to build a plant once fuel cell vehicles become commercialized.

Strengths and weaknesses

Strengths

- The project clearly demonstrated that >98% recovery could be achieved with a simplified process using lower acid concentrations with reduced environmental load.
- The team did an outstanding overall job.
- They have an industrially strong team of collaborators.
- The team has the expertise of the PI in precious metal recycling, and took a solid, methodical approach.
- The project team solved the problem.

Weaknesses

• The project was relatively low-risk compared to others in the fuel cell portfolio since it was basic process optimization/simplification and did not move the Pt reclamation technology forward dramatically.

- The project team should wrap-up the program with a detailed report so that industry can benefit from results.
- The project is already 98% complete. There are no significant recommendations to offer.

Project # FC-34: Neutron Imaging Study of the Water Transport in Operating Fuel Cells

David Jacobson, Daniel Hussey, Eli Baltic, and Muhammad Arif; National Institute of Standards and Technology

Brief Summary of Project

This project aims to develop and employ an effective neutron imaging-based, nondestructive diagnostics tool to characterize water transport in proton exchange membrane fuel cells (PEMFCs). The objectives of this project are to 1) form collaborations with industry, national laboratories, and academic researchers; 2) provide research and testing infrastructure to enable the fuel cell/hydrogen storage industry to design, test, and optimize prototype to commercial grade fuel cells and hydrogen storage devices; 3) make research data available for beneficial use by the fuel cell community; 4) provide a secure facility for proprietary research by industry; 5) transfer data interpretation and analysis



algorithms techniques to industry to enable them to use research information more effectively and independently; and 6) continually develop methods and technology to accommodate rapidly changing industry/academia need.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.6** for its relevance to DOE objectives.

- Strong *in situ* analytical tools for measuring water conditions in active/running fuel cells are of paramount importance to developers in understanding and designing their systems. Water management is one of the most important aspects of fuel cell performance and durability. Any tool to help developers with water management will play a significant roll in helping DOE meet objectives of the program.
- One of the very few ways of seeing inside the "black box," this project is tremendously valuable for reconciling models and empirical results with respect to water management.
- The project helps to address fuel cell performance issues.
- It is not clear how any of this addresses the #1 key issue of fuel cell cost.

Question 2: Approach to performing the research and development

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

- The PI is using one of the inherent advantages of neutron images (having a larger cross section of H compared to other methods like X-ray that don't see H well) to help the objectives of the team. Recently, the PI has worked to improve the resolution of the equipment to visualize through plane water profiles, which is very important. The PI has also worked to compliment the water imaging by correlating with current and temperature profiles of the same systems. This combination of data (*in situ*) is very useful for developers.
- The project team has shown good focus on utility and application of the method to real problems.
- The capabilities are well-deployed, but it is not clearly presented what the exact challenges are to achieving higher resolutions and scan rates. The progress with LANL is excellent, although future membranes may be as thin as ~8-10 μ m; suggesting an ultimate resolution of ~2 μ m (4x higher than now) may be desirable. Is it possible and what will it take to get there?
- It is not clear if scanning slit option is available. A detector with a high aspect ratio (instead of square) over which a cell is moved, may provide more economy.
- I see a need for a high-resolution detection scheme that has a height equal to an actual fuel cell.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.4 based on accomplishments.

- The PI has heeded the recommendations of the Fuel Cell Technical Team and original equipment manufacturers (OEMs) by completing the upgrade to higher resolution systems, correlating work with temperature and current density profiles. He has also worked to develop an improved understanding of the model discrepancies with real world testing.
- High-resolution work is excellent; keep going!
- Experiments at low temperature are extremely impressive!
- The work on H₂ storage was interesting, but could have been more succinct.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.6** for technology transfer and collaboration.

- NIST is teamed with the appropriate end users of the technology such as GM and Ford, and some of the best fuel cell research labs such as LANL. The combination of these partners provides NIST with the expertise and resources needed to build a successful program.
- The project has a very impressive portfolio of users!
- The technique is well-established, and technical publicizing has been extremely effective.

<u>Question 5: Approach to and relevance of proposed future research</u>

This project was rated 3.4 for proposed future work.

- Any further increase in spatial resolution (up to 1 µm) to visualize through-plane behavior of membranes would be appreciated, since membranes are becoming increasingly thinner as the technology progresses.
- In addition to listing collaborators, it would be considerably more compelling to have a matrix showing what type of study was done with that user; this activity is strongly encouraged for next year.
- The focus seems to be on developing new instrumentation and better capabilities, rather than working with major stack developers and helping them solve their problems.

Strengths and weaknesses

Strengths

- NIST is one of the few labs in the world that can conduct this work.
- They make a very expensive tool available to the research community at a cost-effective price.
- NIST is a unique facility with world-class staff and equipment; the users are obviously deriving high value.

Weaknesses

- Evidence of a user-voiced specification for what detection is necessary.
- The team needs high resolution over more realistic dimensions (~0.25 µm height).

Project # FC-35: Water Transport Exploratory Studies

Rod Borup; Los Alamos National Laboratory

Brief Summary of Project

The overall objective of this project is to develop an understanding of water transport in proton exchange membrane fuel cells (PEMFCs). The specific objectives are to 1) evaluate structural and surface properties of materials affecting water transport and performance; 2) develop (enable) new components and operating methods; 3) accurately model water transport within the fuel cell; 4) develop a better understanding of the effects of freeze/thaw cycles and operation; 5) develop models which accurately predict cell water content and water distributions; 6) work with developers to better the state-of-art; and 7) present and publish results.



Question 1: Relevance to overall DOE objectives

This project earned a score of 3.3 for its relevance to DOE objectives.

- Understanding water management is critical to optimal design of fuel cell systems and thus to meeting DOE objectives.
- This study developed experimental methods to look at water transport. Of particular interest (to me) were the neutron scattering pictures. Presumably the sort of techniques presented here will prove important to analyzing fuel cells for many years into the future.
- The focus of project work could be better.
- The issue of water management in PEMFCs is clearly important to understanding and addressing the key technical barriers for fuel cells, including optimizing performance and cost. The project approach based on detailed experimental characterization and modeling of water transport seems very relevant to the overall objectives, provided the approach continues to move toward clear recommendations. The non design-specific approach of the program makes it a little more difficult to move to specific recommendations.
- Water transport impacts numerous areas of major importance to the success of the hydrogen fuel cell effort: performance, durability, freeze survivability, and start. Thus, it is an important area for continued DOE funding.
- Serves objectives B, C, start-up energy consumption, and freeze start operation.
- The project team has designed an independent approach focusing on water concentration in MEAs. The project addresses cost and performance targets of the DOE, and involves the relevant industry. Water transport is of utmost importance for PEM technology. The program is critical to the Hydrogen Program of the DOE, and fully supports the objectives.

Question 2: Approach to performing the research and development

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

- The direction of effort is not clear.
- The project timelines, deliverables and go/no-go points need to be addressed. How will the data feed into a model?
- The PI generally did a better job this year of clearly specifying fuel cell operation conditions.

- Some methods seemed good, but others less-so. There are too many groups; they each do okay work, but each seemed to be going its own way.
- The "Characterization Approach" seems to be well thought-out and scientifically sound, taking into account industry approaches and current materials, while keeping the approach non design-specific as much as possible. The project's approach needs to continue towards making clear recommendations in the area of developing or enabling new materials and operating methods.
- The project team has taken a good approach to integrate the diagnostic effort with LBNL's modeling expertise.
- However, the program appears to be too wide-ranging and lacking focus. What are the primary objectives and expected outcomes? Perhaps it would be better to pick two or three areas. For example, developing a quantitative understanding of water transport through the electrode, membrane, and gas diffusion layers (GDL) by combining diagnostics such as NIST neutron imaging experiments with modeling efforts.
- Cutting edge *in situ* measurements of membrane water are being carried out. Water profiles for different membrane geometries were investigated, and the results were published. In the presentation, specific reference to published results was given. The water content vs. time was measured for wetting and drying modes. This is a case in point for the technical focus of the program. The same holds true for investigations on freezing. The x-ray tomography delivered impressive results. The modeling activities support the investigations. Modeling might gain additional importance as the project proceeds.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.2** based on accomplishments.

- Experimental methods shown seemed good. Other work seemed less well developed. It was hard to pin down exactly what was accomplished beyond visualization.
- The technical accomplishments of understanding in the water transport and behavior are good, and the project needs to continue towards making recommendations.
- The project team has done good diagnostic work with many interesting observations, but little in the way of conclusions.
- The research raises a lot of questions (such as the actual lambda of membrane vs. relative humidity (RH) and temperature slide 7), but again, it is not clear as to the answers. Is membrane lambda vs. RH and temperature considered still controversial? Is this real or an artifact of signal/noise ratio?
- The high-resolution measurement quotes lambda to one decimal, but the measurement of thickness in a water-filled channel varies by over 30%; error seems significant (slide 7, center figure).
- On slides 10 and 11 (wetting/drying transients), is time scale actually due to GDL properties as suggested, or just due to airflow saturation (i.e., was the longer dry out time scale after down-transient due to reduced capacity of reduced airflow to hold water, or did I misunderstand and airflow was held constant)? It is not clear how wetting transient analysis is being used in the overall project.
- What size channels are being used for freeze durability experiments? Is the cell purged at shut-down? The degradation shown is not observed in application hardware with paper GDLs.
- The technical results are outstanding in that freeze properties can now be monitored. Fundamental characterization of the GDL was performed, including the introduction of the 3D x-ray tomography.
- A table of many accomplishments is included in the presentation.
- The progress made is outstanding in terms of quantity and quality.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.5** for technology transfer and collaboration.

- The project team has demonstrated good collaboration with industry, national labs, and academia.
- If the PI is being used primarily as a data resource for other groups, then he should indicate it. The PI is conducting a tremendous amount of work on many fields, but has no specific end goal in mind.
- There are too many groups. They each do okay work, but each seemed to be going its own way.
- The project has an extensive list of collaborators, which appear to be actively involved and contributing to the program.
- The team demonstrated excellent interaction with NIST and the modeling effort.

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- It is less clear what the Nuvera interactions were. Can LANL provide insight into the observations and results from the Nuvera freeze study?
- There are six institutional partners involved and four companies. All of them are renowned, or at least well known. The program looks very well-organized.

Question 5: Approach to and relevance of proposed future research

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

- The PI proposes a large amount of work. The PI should reduce his scope and work on topics that may not overlap with other projects.
- Future goals, methods, and milestones seemed vague.
- The plans address key areas of further understanding of the behavior and changes in the GDL. It is important to focus this understanding on improvements and recommendations to address the key barriers of performance optimization.
- I support the modeling proposals.
- I would like to see more interaction with the Nuvera freeze project.
- The plan for how all these experiments are going to tie together is not clear. It seems that many fundamental issues have been highlighted, but does LANL have the resources to dig into each one?
- The focus on the 3D tomography is appreciated, as well as the increasing importance of modeling.

Strengths and weaknesses

Strengths

- Nice visualization methods are shown.
- The project brings together an excellent team with clear focus on understanding the fundamental behavior of water transport in the PEM. The project makes use of state-of-the-art scientific methods to develop an understanding of the fundamental characteristics that affect some key performance barriers in PEM fuel cells.
- The project has generally solid, broad-based experimental studies with lots of key fuel cell partners.
- The team has an excellent analytical technique portfolio and analysis capabilities.
- The broad scientific input through institutes and the technical input from the industrial cooperation partners are strengths of the project team.

Weaknesses

- There is a tremendous amount of data presented in this presentation. The PI does not do a good job of tying it together or describing the relevance to the overall objectives. It is unclear whether the PI is focusing on, cold performance, modeling, durability effects, or material effects.
- Beyond visualization, the project seemed to wander, and has a lack of focus, with no clear goals for future work.
- The project has a non design-specific approach, which may be a weakness in moving to clear recommendations to address the barriers. The project should try to make clear the recommendations for materials and operating profiles to address these issues, at least within the generalizations of a non design-specific cell configuration.
- It would be nice to come to recommendations from all these excellent measurements on how to come to improved performance via operating parameters and/or novel materials.
- There is some lack of program focus.
- There are no notable weaknesses.

- The PI does not tell me why his results are important, or how they relate to the bigger picture. The objectives are extremely vague and do not help tell the story either.
- The team needs to weed out collaborators. The PIs should focus work on targeted, defined goals.
- The team needs to continue to look at materials and operating profiles to move to recommendations for cell performance optimization.

- I would like to see more interaction with the Nuvera freeze program.
- I am concerned about the use of serpentine flow fields in material degradation studies, as these flow fields are well-known to accumulate water in the U-bends. During freezing, water "slugs" can expand and damage the unitized electrode assembly (UEA). A non-serpentine flow field is recommended. Without U-bends, such a rig is more relevant to application hardware used by most or all automotive OEMs.

Project # FC-36: Water Transport in PEM Fuel Cells: Advanced Modeling, Material Selection, Testing, and Design Optimization

J. Vernon Cole and Ashok Gidwani; CFD Research Corporation

Brief Summary of Project

The overall objectives of this project are to 1) develop advanced physical models and conduct material and cell characterization experiments; 2) improve understanding of the effect of various cell component properties and structure on the gas and water transport in a proton exchange membrane fuel cell (PEMFC); 3) demonstrate improvement in water management in cells and short stacks; and 4) encapsulate the developed models in a commercial modeling and analysis tool. The fiscal year 2008 and 2009 objectives were to 1) complete baseline characterization for gas diffusion layer (GDL) materials; 2) gather experimental data under controlled conditions, test and apply models for water



transport in GDLs, channels, and across interfaces; and 3) evaluate performance and water management sensitivity in operational cells, evaluate cell-scale water transport models on a component level, and integrate with electrochemistry and test.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.4** for its relevance to DOE objectives.

- This project aims to improve the understanding of water transport in automotive fuel cells, issues of freeze/thaw cycle tolerance and cold start-up.
- Understanding water transport within PEM fuel cells is important for optimizing fuel cell performance, cost and durability
- Start-up of a fuel-cell car requires that frozen water in the FC can be purged; operation requires that water can be removed in operation. They found that these effects are strongly temperature dependent, but do most of their work at 35C, and study mostly the GDL. 35C operation may be a good average value to study, but it hardly constitutes the major operation conditions. Also, is water transport in the GDL the main water transport issue?
- Project addresses barriers D, E, and G primarily under task 4, develop gas diffusion layers.
- The project uses modeling and experimental characterization with the ultimate goal of improved components and fuel cell operating concepts. Current devices have power density limitations and durability concerns due to water transport. While this project addresses power density limitations, it has not as yet addressed any durability concerns. Water transport, while important, is not the most critical aspect limiting fuel cell commercialization.

Question 2: Approach to performing the research and development

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

- The project combines theoretical and experimental investigations of water transport in various components of PEM fuel cells.
- Characterization of materials feeding properties to modeling is a good approach to future developments.
- They do a good job of visualizing the flows, and have put them into multi-physics form. I'm sorry to say, but these models hardly include the effect of temperature, and seem to be based on constant low-temperature operation. My sense is that a fuel cell that operates at 65°C or 110°C will have very different behaviors than

shown here. My sense, also, is that higher temperatures are more relevant to the auto industry, so that there should not be as much plug formation as shown here. There seemed to be no formal way to integrate experiments with models.

- The team uses extensive characterization and modeling to understand water transport. The results are used for subsequent optimization.
- Combining modeling and experimental characterization is a reasonable approach to addressing key issues relevant to water transport in fuel cell systems. The project suggests the ultimate goal is improved components or operating concepts, but has shown no example how these will be achieved, or examples of how they are being currently addressed (at best this is not clear from the presentation). It is unclear how many of the experimental results are being tied to modeling studies.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **2.6** based on accomplishments.

- At the midway point, the project is still in the phase of collecting data and formulating models. To date, the project has not produced results to demonstrate a clear path to improved understanding. Hopefully, this will happen soon.
- The team provided good materials characterization of GDLs.
- There seems to be minimal performance measurements in actual operating fuel cells.
- Laser scans of GDL structure and variation with polytetrafluoroethylene (PTFE) loading are interesting.
- They do a good job visualizing the flows, and have put them into multi-physics form. I'm sorry to say, but these models hardly include the effect of temperature. It is not clear how hard it would be to add temperature better. Whetting angle, viscosity, and vapor-liquid equilibrium will be much different at 80°C, for example. Similarly, ice behaves differently than water. We need models that help with the important issues of membrane electrode assembly (MEA) operation. There was some experimental data, but there seemed to be no formal way to integrate experiments with models.
- The team extensively characterized GDL properties. They had a model developed, although comparison to experimental data presented to validate the model was unconvincing. It also appears that the model is limited by the coarseness of the grid used, and is not yet applicable to varying flow field channel designs.
- The project has presented significant experimental studies including base materials characterization. The models being applied are not presented in full enough detail to assess shortcoming/limitations, and show poor agreement with experimental data under many conditions (much worse than other reported studies). The value of these models and the ability to be further refined for increased value is unclear. To date, it is not clear how the experimental studies are helping material design or operating strategies. It is also unclear how relevant self-humidified studies are and how applicable the reported results are to long-term operation where wetting characteristics of materials are known to change.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.0** for technology transfer and collaboration.

- The project has assembled a credible team.
- The team demonstrated good interactions with an appropriate set of partners.
- The team had a good selection of partners, but the presentation did not differentiate what work was done by which partners, or what benefits were gained from distributing the project. To the extent that experiment and theory work was done in different locations, there seemed to be poor integration.
- There was a good mix of partners with complimentary skills. If Ballard is involved, why does the team need to initiate technology transfer through the University of South Carolina?
- There are several strong partners with relevant skills. The level of coordination between partners is unclear in several areas, and it is unclear how experimental studies are feeding modeling work (slide 19 on). There was very little from the project publicly presented, with only a "Transactions" paper listed in the current literature. Although a paper and second "Transaction" paper are listed as accepted, this lack of data in the literature makes supplemental information about the project harder to evaluate.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

This project was rated 2.4 for proposed future work.

- The future plans appear logical, but there is no evidence that the project will produce models that can actually be useful in optimizing the structure of fuel cell components or cell performance.
- The objective of the project is to demonstrate improvements in water management in cells and short stacks; however, this seems to be missing from the future work plans.
- There is no particularly strong plan to cover the weaknesses presented. Future work goals and milestones were fuzzy.
- The team will complete the characterization and modeling of the GDL. It is essential that the effect of temperature gradients and electrochemistry be explored on both.
- It is not possible to evaluate whether or not the proposed future work will lead to enough modeling improvements to make the models useful in a predictive fashion. Current models show relatively poor agreement with experimental results and adding aspects heat transfer and phase change may help, but it's unclear they will be enough to resolve all issues. Without a validated model, exploring optimization strategies doesn't make sense.

Strengths and weaknesses

Strengths

- This is a good team.
- They team had good combined modeling and experimental studies.
- Coupling of material characterization and modeling is important for good developments.
- The team had nice pictures (multi-physics can be powerful).
- The team was sharply focused on the real problems in GDLs, and aiming for optimization of manufacturable complements.
- There was a good mix of team strengths, with modelers and experimentalists well-represented. There was significant materials characterization at least within a specific window (slide 6, for example).

Weaknesses

- There were no breakthrough results.
- There were no plans to produce data, or models that will address issues of start-up and shut-down, start-up from subfreezing temperatures, or freeze/thaw tolerance.
- The use of numerically generated GDL microstructure is not an accurate representation of GDL materials.
- The data presented look-like analysis, and was at unlikely operation temperatures; temperature effects remain largely unstudied. There was also poor integration between experiment and theory. At this point, water transport analysis seems unlikely to aid in fuel cell design and operation.
- There was a lack of integration of electrochemistry and the effect of temperature.
- Results to-date are not compelling, and there is no reason to believe future efforts will necessarily lead to significant improvements. The wide team seems to be taking on multiple aspects of water transport (at different levels) without addressing any of them particularly well.

- The team needs major correction by looking at other (higher) operation temperatures. They need to integrate theory with experiment in terms of design/operation models. What happens when flow is in "up" direction?
- More electrochemical engineering and characterization of temperature is needed.
- The team should focus on a single aspect of water transport; for example, flow in the GDL or in the flow channel, rather than several aspects such as flow in the GDL, in the channel, and the transition between the GDL and flow channel. It is better to have a reasonably good picture of one of these aspects, than inaccurate representations of many.

Project # FC-37: Visualization of Fuel Cell Water Transport and Performance Characterization Under Freezing Conditions

Satish Kandlikar and Zijie Lu; Rochester Institute of Technology Thomas Trabold, Jerry Gagliardo, and Jon Owejan; General Motors Jeffrey Allen and Reza Shahbazian-Yassar; Michigan Technological University

Brief Summary of Project

The overall objectives of this project are to 1) gain a fundamental understanding of the water transport processes in the proton exchange membrane fuel cell (PEMFC) stack components; 2) minimize fuel cell water accumulation while suppressing regions of dehumidification by an optimized combination of new gas diffusion layer (GDL) material and design, new bipolar plate design and surface treatment, and anode/cathode flow conditions; and 3) meet U.S. DOE targets for 80 kWe transportation stacks. The goal for 2008 was to implement changes to the baseline system and assess the performance of 1) ex situ combinatorial performance; 2) in situ combinatorial performance: 3) water distribution and



current density distribution; and 4) microscopic study and models for water transport in GDLs and parallel channels.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.2** for its relevance to DOE objectives.

- Understanding water flow distribution is essential to building better fuel cells. Understanding misdistribution effects will help significantly reduce water purge energy, etc.
- The start-up of a fuel-cell car requires that frozen water in the fuel cell can be purged; operation requires that water can be removed in operation. They found that these effects are strongly temperature dependent, but do most of their work at 35°C, and study mostly the GDL. 35°C operation may be a good average value to study, but it hardly constitutes the major operation conditions. Also, is water transport in the GDL the main water transport issue?
- The project's approach to understand water management and address the effects under normal and freezing conditions clearly addresses some of the key technical barriers to fuel cell performance and adoption.
- This is a very effective visualization tool to qualitatively analyze what happens in the course of a purge.
- However, this doesn't seem to be set-up specifically to close a technical gap. This falls more under the purview of basic research and development, rather than a means to close the gap related to energy expenditure under a cold start.
- The project serves objectives C, D, and E.
- Relevant materials are investigated.

Question 2: Approach to performing the research and development

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

• The PI did a good job understanding how undesirable plug flow develops - high contact angles caused by hydrophobic GDL materials. This will present developers with a tradeoff situation of designing GDLs that don't induce plugs (hydrophilic materials) yet need to prevent flooding at high current density (hydrophobic

materials). Work is thoroughly well correlated to fuel cell operating conditions (temperature, humidity, stoichiometry, etc.).

- Results should lead to some kind of model development.
- The project team does a good job visualizing the flows, and taking the important step of making relevant models that look like they'd be easy to use and relevant. Unfortunately, these models are all based on data at 35°C. A fuel cell that operates at 65°C or 110°C will probably have very different behaviors. These higher temperatures seem more relevant to the auto industry than 35°C. Interestingly, the power data shown was taken at 80°C. The majority of data should have been taken at about this temperature, not at 35°C. Also, since transport in the GDL was very pore-structure dependent, a variety of pores should have been studied, or the effort should have been made to pick an optimal pore.
- The overall approach seems to address characterizing the water movement and accumulation in the fuel cell both by developing a clear understanding of the baseline conditions and a parametric study of the components. The approach to optimize the GDL and bipolar plate designs, as well as the operating conditions to provide improvements and recommendations, is important to addressing the key barriers of water management and control especially for freezing conditions.
- There is significant technical content here, as well as Parametric Studies followed by *ex situ* studies, *in situ* studies, and *in situ* performance in combinatorial studies.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.0 based on accomplishments.

- The PI addressed many of the original equipment manufacturer (OEM) concerns by investigating effects of GDL intrusion. The results were somewhat intuitive though: increased intrusion equals increased pressure drop. What effect does that have on water formation?
- The technical collaboration to date is impressive.
- GDL properties are broadly covered. A capillary flow model was developed.
- The presentation is very descriptive and draws on the work done. Little information is given about lessons learned, rendering a lower score.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.0** for technology transfer and collaboration.

- The collaboration has generally been good. The project team could be working with LANL as well (LANL is working heavily in this field).
- The project team made a good selection of partners, and the partners have done good work. Given that RIT has developed the high-temperature fuel cells, they should be expected to contribute such membranes not the traditional low-temperature membranes.
- The project has sufficient technical collaboration and it seems to be well coordinated for the program.
- The degree of collaboration and the comprehensive approach towards characterization are impressive.
- Through GM, there is an industrial partner with a strong fuel cell background in the consortium.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.5 for proposed future work.

- The project team should continue the study on effects of thermal conductivity and the important yet under appreciated characteristic of GDLs.
- The project team should plan is to vary flow channels and do more observations. The project team did not present any particularly strong plan to cover the weaknesses. The problem may be the lack of future funding and the impending bankruptcy of GM.
- The planned activities address the materials and measurements to target overcoming the barriers with materials and the operating parameters to address the purge and freezing behavior.
- The project team should provide a future plan with quantifiable goals.

FY 2009 Merit Review and Peer Evaluation Report

Strengths and weaknesses

Strengths

- This project provides a thorough study of material characteristics on GDL water management behavior.
- The project team produced usable analysis of water transport.
- The project has an appropriate team and is addressing both the characterization and the optimization materials to address the key barriers associated with the water management and shut-down behavior for freezing situations.
- The project team has good focus on the fundamentals of an interesting mix of characterization, modeling, and analysis. This will help to develop a comprehensive understanding of water content and water removal rates.
- There is a strong focus on GDL, combinatorial studies, and two phase flow studies.

Weaknesses

- The project team has a lot of topics to work on in a condensed time frame. The PI should try to aggregate results into a meaningful model or recommendations for GDL design.
- Testing conditions seem more relevant to the ease of testing than to actual operating conditions. The team should expect different water channel behaviors at 60°C to 100°C.

- The project team should perform work at higher temperatures. The team should try polybenzimidazole (PBI) membranes instead of Nafion[®]. On start-up, the project team should assume shut-down from idle (low power draw) at a low temperature. This tends to produce a worst-case water-plugging scenario.
- The project team has presented interesting and important findings; however, the translation of these findings into real improvements of performance is not clear.

Project # FC-38: Subfreezing Start/Stop Protocol for an Advanced Metallic Open-Flowfield Fuel Cell Stack *Amedeo Conti; Nuvera Fuel Cells, Inc.*

Brief Summary of Project

The overall objective of this project is to demonstrate a proton exchange membrane fuel cell (PEMFC) stack meeting the DOE 2010 cold start targets. The goals for fiscal year 2008 were to 1) achieve the -20°C cold start target respecting the energy budget; and 2) identify electrochemical material freeze cycle aging modes. The goals for FY 2009 were to: 1) provide reliability and durability of the -20°C start-up procedure, and 2) achieve the -40°C cold start target, enabled by new stack technology.

<u>**Ouestion 1: Relevance to overall DOE**</u> <u>**objectives**</u>



This project earned a score of **3.4** for its relevance to DOE objectives.

- The project addresses the issues of start-up and shut-down time and energy for a unique flow field design.
- The ability of a fuel cell to withstand freezing is absolutely critical to it being usable in the sorts of remote and mobile applications where it adds customer value.
- The program is focused on the key barriers of water management and the effect/optimization of start and stop protocols for fuel cells. Their program seems to maintain focus both on the energy requirement effects of the start and stop cycles, as well as the implications for the durability and reliability of the materials.
- The project addresses barriers D and G through Task 11, and has developed innovative concepts for fuel cell systems.
- The team has made a product development effort, which most of industry is addressing.
- Subfreezing effects, principally start-up and shut-down are critical to RD&D objectives, and are the primary focus of this task. The project itself is very Nuvera design-specific, and many of the details regarding how targets can be met or how the community in general can benefit from the lessons learned in this project limit the relevance of this project.

Question 2: Approach to performing the research and development

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

- The project has taken an experimental approach to determine the important parameters that affect the start-up time and energy and material durability.
- The approach seemed to be to develop new Nuvera fuel cells that start well from low temperatures, down to -40°C, and then to take data on these cells. There were no particularly general findings. For example, the effect of different start-up processes is not studied in a formal way.
- The program's approach seems to be following an overall sound scientific and product-development approach to understanding and addressing the barriers. Improvements in the approach to the measurements and testing of the effects of freezing conditions are good for ensuring relevance to the real world tests, as well as the increased collaboration to ensure the effects of freeze/thaw conditions on durability are taken into account fully.
- The set-up this year using an environmental chamber is a huge improvement on last year's freezer approach. There was good use of partners' skills in being able to diagnose failures due to freeze/thaw cycles.
- The project team needs to identify failure modes, root causes, and corrective actions to mitigate failures.

• The approach of the project has been effective at resulting in designs and data that meet DOE targets. The focus on lowering thermal mass is clear and open flow fields as part of the approach is mentioned for the andromeda stack design. Still, it is not clear whether or not the design/materials used and the lessons learned from this project are broadly applicable due to the lack of specific data that is likely considered proprietary. Information relating to the specific lessons learned, such as where and how thermal mass are cut from the stack without impacting other critical factors, would be beneficial.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.1 based on accomplishments.

- The project has met the time and energy target and is now concentrating on failure mechanisms.
- The project team made a low thermal mass Nuvera fuel cell and used a high-pressure purge that they believe is good for start-up from freezing. Is this the best method? We really don't know. It is not clear that we learned much about what one does generally to deal with freezing conditions. Perhaps a resistive heating cycle based on a brief short-circuit operation period would have worked better. It is also not clear that this method would have worked at all if more water were frozen in the cell.
- The team has demonstrated good energy management for the shut-down and start-up. It was not clear how aging affected this from the presentation. This project needs further work, understanding, and validation of the durability and energy behavior over time.
- The project demonstrated DOE's goal of 50% of rated power in <18 s, and identified the main failure mode.
- The start-up energy and time from -20°C to 50% rated power for the system are impressive and comfortably meet DOE targets. The improvements in start-up energy from last year to this year also suggest significant progress in the past year. Specific changes and their potential impact on durability or longevity of the stack and performance over time are not clear, and therefore it is not certain that the changes made don't also have negative consequences.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.1** for technology transfer and collaboration.

- The project team includes an integrator, membrane electrode assembly (MEA) manufacturer, gas diffusion layer (GDL) supplier, and a university.
- Though two university groups participated, there seemed to be little theoretical input.
- This was a good project team to address the effect of the materials and water management on the stack behavior. The projects involved a larger group of collaborators to address and understand the issues with the freeze/thaw and the materials durability and aging.
- This was an excellent team comprising an MEA manufacturer, a carbon supplier, and a university. The team also held a freeze workshop.
- The workshop is a good way to show collaboration/coordination with other institutions. The project has strong partners, but it is unclear how the partners have worked together to provide the improvements in performance presented. There is little in terms of publication from this group based on this project (for example, the PI this year, Conti, and last year, Cross, are not authors on the few publications listed). This further reflects the low level of information released from this largely publicly funded project that limits its value to the fuel cell community.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.8 for proposed future work.

- The future plans are to incorporate durability consideration into the start-up protocol, and to evaluate the feasibility of an H₂ purge.
- The next step follows from the previous. It is not clear that any general conclusions will be derived, but it will not be worthless. The team has taken a very practical approach.

- Future work should continue to loot the validation of the energy management, as well as the stack design, to examine the effects of durability/aging on these parameters.
- There is insufficient detail to really evaluate this; however, it appears that now that test protocols have been identified, that integration of next generation fuel cell components will not be a problem.
- The future work proposed is vague and only inferred from two bullets on the summary slide or by interpolation from the overall approach slide. The plans may lead to improvements, but it is not certain whether future advances will be possible. Again, this is due to a general lack of detail throughout the presentation.

Strengths and weaknesses

Strengths

- This is a good team.
- The project has identified the key issues of thermal mass and its relationship to power density (current density at rated power).
- Though two university groups participated, there seemed to be little theoretical input.
- The team took a good overall approach to modifying the stack, MEA component, and operating profiles to address the two barriers of energy requirement.
- The team demonstrated realistic testing protocols, and collaborated with a materials and components supplier that can supply state-of-the-art and improved components.
- The project meets or exceeds a number of key DOE targets.
- The project produced a working model. It's nice when a project gets to a state where there is an actual product on the horizon.
- The project continues to focus on the effects of aging and durability on the materials, and the effects of these aging changes on the energy performance and operating profiles.

Weaknesses

- The project has taken an engineering option and is not structured to developing a fundamental understanding of start-up/shut-down or material durability/compatibility.
- The project has not systematically looked at the effect of the shut-down protocol.
- The project needs more consideration of what is happening to the ionomer during freeze/thaw.
- The rationale, specific materials, and choices for this project are not presented in enough detail to understand whether or not there are critical shortcomings to the approach, or provide value for the general fuel cell community to apply similar approaches. This should be able to be done to a greater extent without meaningfully compromising proprietary information (or be done in the absence of public support).

- The project team should develop a method to characterize the MEA at the end of the purge cycle (beginning of the start-up cycle).
- The next step follows okay from the previous step.
- The team needs more polymer science.
Project # FC-39: Development of Thermal and Water Management System for PEM Fuel Cells *Zia Mirza; Honeywell*

Brief Summary of Project

The overall objectives of this project are to 1) improve proton exchange membrane fuel cell (PEMFC) performance and life by maintaining the humidity of the inlet air stream at a high level (>60%); 2) eliminate the need for an external water source by transferring water from the stack exit air stream to the inlet stream; and 3) design, build, and test high-performance full-size radiators to meet the 80 kW fuel cell stack cooling requirements. Specific objectives are to 1) validate the performance of fullscale humidification devices sized for an 80 kW PEMFC by testing the Emprise enthalpy wheel and the Perma Pure membrane module; 2) evaluate planer membrane humidification devices; 3)



perform performance testing at sub-ambient conditions; 4) increase the performance required to dissipate lowquality heat; and 5) optimize the weight, size, and cost.

Question 1: Relevance to overall DOE objectives

This project earned a score of **2.4** for its relevance to DOE objectives.

- Advanced humidification systems are important.
- Balance-of-plant is not a technically critical area for fuel cell research. It is an engineering area to the design and development of fuel cell systems, and it is a competitive area.
- System water and thermal management are important aspects of automotive fuel cell development, although they are of secondary importance to catalyst performance/cost, membrane development, and durability.
- This doesn't seem to address the major issues of fuel cell development: cost, durability, or infrastructure. These are some worthwhile paper studies, but it isn't clear how this is helpful to larger DOE goals or to improving the overall technical readiness of the fuel cell system.
- This project serves objective E.
- Core components for an automotive fuel cell system are developed.

Question 2: Approach to performing the research and development

This project was rated 1.9 on its approach.

- This project primarily is obtaining humidification components which are off-the-shelf items, and then evaluating them.
- Any fuel cell developer has already done this.
- No technical criticality is identified. The two humidifier technologies are off-the-shelf, and the approach is to perform design verification testing. No research is evident.
- The program has focused on materials that are well known in the community, and which in general are not expected to meet system overall targets.
- This project does not seem to have an approach that, even if it succeeds in its stated goals, will enhance the prospect for fuel cell commercialization.
- The testing of planer materials has some value if combined with novel material development.

• This is a very large budget for a component that only changes the overall system cost by \$30 (for the radiator, slide 15). It is not clear how this effort is unique from what individual automotive original equipment manufacturers (OEMs) and stack manufacturers are developing on their own to meet customer requirements.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 1.9 based on accomplishments.

- There was a lot of discussion about radiator design; however, the automotive OEMs know how to make costeffective radiators. They make approximately 10-15 million per year.
- Humidification devices were purchased from other vendors.
- Little progress was observed.
- It is not clear from the results shown if microchannel radiators have some promise. Rather than focusing on a "value function," please show more test results and comparisons to baseline technology (commercial automotive radiator). Tests as-shown seem inconclusive.
- The project team appears to be making some progress, but nothing that fundamentally changes the cost model or system performance.
- Considering the budget for this project, more results and/or more substantiated results might be expected. The project team purchased and validated humidification devices and developed a cooler.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated 1.7 for technology transfer and collaboration.

- The DOE and the fuel cell technical team do not count as partners; all projects are reviewed by these organizations.
- There does not seem to be any real collaboration with ANL, other than providing some data to ANL for modeling purposes.
- Any effective collaboration is not seen.
- This program would benefit from exploring more novel materials to test for water transport, not just the wellknown Perma-Pure tubular membrane humidifier and Emprise Enthalpy Wheel.
- Collaboration indicates minimal exchange with other research and manufacturing institutions. It appears that there is one-way delivery of information to Argonne's modeling effort, and participation in technical team reviews, but very little interaction with stack integrators.
- The FreedomCAR Technical Team is the only partner in the project. It is assumed that DOE, even though quoted as a partner, cannot be considered a development partner.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.1 for proposed future work.

- Future work consists of modifying a test stand and testing components. There are no development activities to this project.
- Planer membrane testing may hold some value.
- The program should fully document test results. Do microchannel radiators provide a marked advantage?
- The project team should document: expected cost vs. conventional automotive radiators, delta-P vs. airflow, Q transfer vs. airflow (slide 11 just shows at one condition), and the expected impact of fouling.
- To be useful, membrane water transfer data should show operating conditions, molar flow rates, temperature, pressure, etc., not just relative humidity (RH).

Strengths and weaknesses

Strengths

- This is a solid hardware engineering study!
- The systems approach is good.

Weaknesses

- This project primarily evaluates existing technology and is not value added to developers.
- There are no development activities to this project.
- This is previously covered ground, with little potential to advance the state of the art.
- The project has minimal impact on DOE's overall goals.
- The value/cost ratio might be improved. For a smaller budget, the project had scored higher. In case the development is indeed that expensive, the presentation did not convey that message.

- SI units would be nice!
- This project's funding should be discontinued.
- In the remaining time, the project team should focus on testing planer membrane water transfer materials (if available) and document the radiator results in comparison to baseline conventional automotive radiators as noted previously.

Project # FC-40: Nitrided Metallic Bipolar Plates

M.P. Brady, T.J. Toops, and P.F. Tortorelli; Oak Ridge National Laboratory

Brief Summary of Project

The overall objective of this project is to demonstrate the potential for metallic bipolar plates to meet the automotive durability goals at a cost of <\$5/kW. Oak Ridge found promising initial performance of stamped and nitrided Fe-20Cr-4V in cyclic single-cell fuel cell tests. Durability studies of stamped and nitrided foils benchmarked to untreated stainless steels and graphite are underway.

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of **3.7** for its relevance to DOE objectives.



- This project addresses barriers A, B, and C under task 5, the development of bipolar plates.
- Bipolar plates play a critical role in meeting DOE stack and system targets.
- Metal plates for proton exchange membrane fuel cells (PEMFCs) have been thought necessary to achieve compact, automotive hardware.
- Several auto original equipment manufacturers (OEMs) are fielding metal plates, and that activity strongly supports DOE goals. Even so, the majority of PEM stacks today are using carbon-based plates, and these materials have also proven satisfactory for both performance and durability.
- The objectives of the project address the cost/durability of bipolar plates in a very good manner, and the breakout of the process steps and cost association of performance and cost with each step are commendable.

Question 2: Approach to performing the research and development

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

- It is appreciated that the team is going after a low-cost ferritic alloy such as Fe-20Cr-4V or Fe-23Cr-4V. Ni and Mo are highly price-volatile. The drive to shorter pre-oxidation/nitridization thermal cycles for low cost can also be appreciated.
- The project's approach is to develop a nitrided stainless steel that is cheap durable and can be stamped.
- The project team took an excellent approach to low-cost, rugged, and durable bipolar plates.
- The team should check the influence of worked or scratched surfaces on corrosion resistance.
- It would have been better if the presentation began with a review of the extensive previous work on the nitriding of stainless steel, and the use of such corrosion protection concepts in PEM stack engineering. Instead, the presentation tended to assert that new approaches were being pioneered, which is not the case. It would have been better if the presentation showed test results using earlier nitriding recipes, and then explained why the new concepts are an improvement.
- The "embossed" flow field shown is a long way from the precise geometries required to assure uniform flow and good stack performance. It was not apparent that the alloy selected, and the nitrided foils, are sufficiently ductal to allow such successful stamping.
- The approach to examining the alteration of metal alloys with respect to the resistivity and corrosion, relative to targets, is well-illustrated. The use of analytical techniques to determine the relationship between process, alloy surface change, and properties is very good. The correlation of surface properties and analyses of alloys which meet performance goals (but maybe not cost) and the alloys currently being investigated needs to be done to



gain better insight into what is causing the irregular surfaces; how much difference exists; and the limited nitridation of the Cr at the surface. Examination of the surfaces of the grooves to determine how much variation exists between the unstressed and stressed surfaces after the nitridation process needs to be determined. Examining smooth foil surfaces does not give a complete picture.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.7 based on accomplishments.

- It was a good idea to increase Cr content from 20 to 23 weight percentage in an attempt to form more continuous CrxN and CrxOy layers for better protection. There is concern that the interfacial contact resistance (ICR) is still too high.
- A nitrided steel has been identified, but the milestone for demonstration of a 15 cm² single cell has been postponed.
- The team has made good progress evaluating new lower-cost stampable metal alloys, despite reduced FY09 funding.
- The materials science was okay, although the results were not novel; however, there is a long way to go from the current status to large size, uniform, and corrosion resistant plates.
- The investigation of process steps and analyses has provided insight into problems associated with irregularities at the alloy surfaces; however, the need to increase the membrane thickness to conduct successful single cell tests suggests that the surfaces of the plates are entirely too rough for the membrane integrity (additional cost of thicker membranes, with higher resistance, has not been included in the projected cost estimates). The go/no-go decision of Milestone 2 cannot be complete until a solution to the inadequate nitridation of the surface is understood, because the single cell tests do not suggest that either a cost-effective process or an acceptable alloy has been determined. Perhaps a treatment of an alloy that meets the performance goals should be examined to determine the degree of surface differences.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.1** for technology transfer and collaboration.

- I was glad to hear that there will be collaboration with an OEM soon; this is critical. Also, perhaps a bit closer collaboration with LANL on the actual fuel cell testing would be to overcome some of the technical difficulties there.
- A large team of diverse skill sets has been assembled. An OEM should be involved; however, whether or not Milestone 2 is passed, in theory the OEM could help attain the milestone.
- ORNL needs some stack design competence in this program. Precise flow channels are required to achieve uniform flow distribution among the many plates in a PEM stack. The degree of uniformity is a design issue which must be addressed early. This team needs a partner who is competent in computational fluid dynamics and knowledgeable in fluidic design engineering of PEM stacks.
- It appears that the list of participants and their roles are appropriate, but the costs associated with each and the degree of participation is not clear. Perhaps more analysis work needs to be done; for example, examine potential welding/joining problems of plates, variations in surface across a stamped plate, and alterations in the treating process.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.9 for proposed future work.

- I would recommend adding the following to the go/no-go evaluation:
 - Analyze aged membrane electrode assemblies (MEAs) and product water from fuel cell durability tests to look for evidence of leached metals;
 - Analyze the metal foils for near-surface compositions following stamping, and compare with beforestamped to ensure no issues develop in that process that could interfere with the efficacy of the oxidation and nitridation treatments;

- Measure the surface energy of the nitrided surface;
- Work with OEMs to ensure the formability of these alloys enables stamping at length scales and tolerances relevant to automotive flow field designs.
- I'm not sure why they are now proposing to test commercial steels. Wasn't the point of the project to develop new steels? Also, the team should be evaluating larger surface area steels of at least 25 cm².
- It is logical to evaluate more readily available commercial alloys and the potentially lower-cost plasma arc lamp.
- Durability in the fuel cell environment (low ICR and high corrosion resistance) and stampability must remain the primary criteria (over alloy cost).
- Formability and joining with an industry partner for a demonstration on short stack is a must.
- Many have shown that Au-coated stainless steel delivers an okay performance. Although gold may be thought costly, Au-coated test articles might be useful for looking at planarity, resistance, and other stack design issues. Once a gold-plated plate design is shown to be useful, then the nitriding gun can be focused.
- The exposure of stainless steel plates to open circuit (high) voltage seems sacrificial. Steel corroded at high potentials; the low energy form of iron is Fe₃O₄, after all. There is no need to test to destruction, and adequate durability can be demonstrated by sensible testing. Operating a stack well past the "red line" seems risky, and proves nothing other than a sense for adventure.
- Extension of the second milestone go/no-go decision is correct, but a detailed plan for both examining options and definition of a resolution is necessary before the third milestone should be considered. After all, that is what a go/no-go decision is for. If the funds from the third phase can be readjusted to accommodate a resolution to the second phase problems, this would help reach an important goal as to what is needed to achieve an improved plate material.

Strengths and weaknesses

Strengths

- The team has demonstrated good science and collaboration with an alloy manufacturer.
- New steels with nitridation look promising.
- Nitrided metal plates, once successfully developed, have many advantages over other bipolar plates (thin, high mechanical strength, high electrical and thermal conductivity, light weight, and low-cost).
- Clearly, ORNL knows steel as well as anyone, and Allegheny Ludlum is a quality concern as well. ORNL has coating tools and experience.
- A well-defined approach to examining an alternate, potentially cost-effective method for producing metallic bipolar plates has been defined. The combination of alloy analyses, cost projections, and single-cell tests has been done in a coordinated fashion.

Weaknesses

- The team has issues with fuel cell testing, and there is a lack of involvement of an OEM.
- Single-cell testing should be on an industry standard of 25 or 50 cm², not 15 cm².
- There have been many, many attempts at nitriding stainless steel for PEM plates. It would have been more convincing if ORNL reviewed earlier work, and spoke to how new ideas (ORNL ideas) showed hope for greater success.
- Although the costs may be increased, a metallic alloy that has the necessary attributes, i.e., surface properties and nitridation success, should be included in analyses to illustrate specific differences between alloys under investigation and proven material. Industrial materials might be included as potential options.

- More hard work on the new steels is needed.
- A table in the Annual Merit Review presentation showing the bipolar plate technical targets and project status would be helpful.

- It makes more sense for ORNL to work with an existing quality stack developer, and fabricate and coat stainless steel parts to replace the bipolar plates within those designs, which are already known to yield adequate stack performance. Indeed, it might be possible to assemble a hybrid stack made from mainly existing proven plates interlaced with some number of "experimental" nitrided plates.
- Delay Milestone 2 go/no-go decision until a reasonable plan of action to resolve surface inadequacies has been defined. Reallocate funds from the third milestone to illustrate potential improvements with tests on a single cell being adequate. Determine additional properties such as welding and uniformity of nitridation control to illustrate the merit of the proposed approach. Costs projections seem to be well-understood.

Project # FC-41: Next Generation Bipolar Plates for Automotive PEM Fuel Cells *Orest Adrianowycz; GrafTech International Ltd.*

Brief Summary of Project

The overall objective of this project is to develop the next generation of automotive bipolar plates based on an engineered composite of expanded graphite and resin capable of operation at 120°C. The goals for Year 1 are to 1) develop a graphite/polymer composite to meet the 120°C fuel cell operating temperature; and 2) demonstrate the manufacturing capability of new materials to a reduced bipolar plate thickness of 1.6 mm. The Year 2 goals are to 1) manufacture high-temperature flow field plates for full scale testing; 2) validate performance of new plates under automotive conditions using a short (10cell) stack; and 3) show viability of the published cost target through the use of low-



cost materials amenable to high-volume manufacturing.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.0** for its relevance to DOE objectives.

- Bipolar plate materials are one of the largest fractions of fuel cell cost. Bipolar plate materials benchmarking is important.
- Bipolar plates are an integral part of every fuel cell stack and contribute significantly to performance, durability, and cost.
- The availability of bipolar plates for 120°C operation is an important prerequisite to successful testing and implementation of 120°C membranes and membrane electrode assemblies (MEAs).
- Bipolar plates play a key role in stack cost, mass, and size (packaging), and is thus an important and proper area for DOE support.
- The bipolar plate is a considerable contribution to the overall weight and volume of the fuel cell, and DOE targets are clearly established to address cost and performance; this program addresses those concerns, but it's not immediately clear how this is directly leading to cost reductions, or improved manufacturability.
- While the plan to fabricate graphite plates, which can operate at 120°C and achieve a thickness of 1.6 mm is described, and is commendable, these do not correlate with the generally stated barriers that are being addressed as described on the first slide. Isn't the plan to develop a novel plate that can provide higher temperature operation without sacrificing any performance or cost attributes?

Question 2: Approach to performing the research and development

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

- The cost advantage for both materials and manufacturing processes, and a generic formability limit should be evaluated before full-scale stack fabrication and testing.
- One disadvantage of this bipolar plate material is volumetric power density.
- A 1.6 mm of plate thickness as a target is not a proper metric, because it is dependent on flow field design variations. Gas impermeability of embossed materials is more appropriate and generic.
- The project exhibits a logical task structure.
- The team has a clear grasp of requirement targets and has addressed them systematically.

- The team could benefit from prioritization of these targets. Key targets include, but are not limited to: ultimate (high production volume) cost and stack volume [bipolar plate (BPP) thickness] potential, durability and conductivity. These need to be addressed specifically in the program and final report.
- The team needs to address detailed manufacturability metrics- key attributes in the bipolar plate. The bipolar plate has a few key parameters related to performance: resistance and feature tolerances, which, in turn, affect plate-to-plate pressure drop variability. Permeability is more of a safety/efficiency performance metric that will have a small effect on overall cell electrical performance.
- The way that the bipolar plate affects the overall stack is in manufacturing cost, some electrical resistance, and in flow distribution. Once the minimum level of electrical resistance has been met, further improvements tend to be dwarfed by MEA resistance. Therefore, cost and manufacturing tolerances for flow distribution will be key. These issues need to be addressed more specifically.
- The fabrication approach, identification of the proper binding materials, testing physical properties and assembling/testing single cell test units all address the correct means for evaluating high-temperature graphite plates. The plan for testing the fuel cell at high temperatures without insuring a high-temperature MEA seems to be shortsighted. Since the tests could not be completed at high temperatures, it would seem that operating at a temperature as high as the MEA would accommodate for a longer period of time for analyses, at least under conditions that could be compared to other plates, would be reasonable. The cost analyses were not found/shown. Planning to do a ten cell stack at 120°C seems like an unreasonable expectation, since no MEA capable of 1,000 hrs was identified.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.8 based on accomplishments.

- Bipolar plate materials with Bensoxazines resin, corrosion resistivity, formability (emboss) and cost should be evaluated.
- For formability, the formability limit should be identified.
- Is gas impermeability (0.8 mm) verified with embossed materials? If not, it should be tested with processed materials, as 0.8 mm is too thick for automotive applications; it should be less than 0.3 mm.
- MEA test data is very poor, why?
- Cost estimates have not been reported yet.
- Progress has been made in materials (graphite and resins).
- I still have questions on cost and volume (achievable power density).
- What is the anticipated cost in large-scale (500K systems/yr) production?
- BPP thickness is given as 1.6mm, which still may be challenging for packaging. Is this best anticipated for this material set? If not, how much can this be reduced?
- The PI states that composite plates are superior to metal in conductivity, but metal plates are much thinner, offsetting this. What counts is the overall area-specific resistance (ohm-cm²). How do their plates compare to metal here?
- The team needs to state contact and overall resistance.
- They have characterized several carbons and resins, but haven't quantified how these changes will affect cost. As mentioned before, the bipolar plate will not have much of an effect on performance, except for ohmic losses (if the plate is very poor), and will have more of an effect on cost.
- While the mechanical tests that were described to be done were reported as successful, and mechanical test data were shown, no test data on the single cell at a temperature commensurate with the MEA and at high electrical potential were demonstrated, no gas permeability data were exhibited, no high-temperature coolant durability data were provided, and no cost comparisons were generated. This project is not complete until the supporting information is provided. No 10 cell stack should be tested until the goals for the tests are defined to be useful, i.e., at a temperature commensurate with the MEA stability.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.0** for technology transfer and collaboration.

• A generic requirement of bipolar plate design would be helpful to evaluate the formability limit.

- Cell testing at 120°C appears to have been problematic.
- The team that has been assembled seems to be appropriate with the expertise available; however, the choice of high-temperature MEA before tests started, and the choice of single cell test conditions seems out of character for the fuel cell members of the team.

Question 5: Approach to and relevance of proposed future research

This project was rated **2.3** for proposed future work.

- For this project, materials benchmarking is good enough. Full scale testing seems to be superfluous.
- Stack testing for 1,000 hours may provide some indication of durability characteristics, but longer-term testing will be required to fully validate the plates.
- What is the heat capacity (mass C_p , kJ/K/cm²) of the bipolar plate?
- What is the anticipated cost in high-volume production?
- What is the PI's estimate of minimum possible thickness while meeting the other DOE targets?
- Please supply area-specific electrical and thermal conductivities (ohm-cm², W/cm²/K).
- The project seems to be moving in the direction of corroborating a good judgment, but doesn't address the overall needs of the technical community, or help in predicting cost reductions. We need to see cost reductions at a moderate volume, and changes in either raw material cost or processability that can be quantified.
- The project is 90% complete, and the remaining planned work will wrap-up the project.
- While the project is nearly, if not entirely, completed, there certainly needs to be more information provided to support any claims that the choice of materials for graphite plates can meet the gas impermeability, coolant durability, scale-up possibility, and cost targets for the described <0.8 mm thick graphite plates. The results provided look promising, but supporting information is necessary. The final stack tests should demonstrate at least a long duration test at higher potentials and a temperature commensurate with a stable MEA (at least 80°C), and post-test analyses should be compared to current graphite plates at those conditions. Supporting cost analyses must be provided.

Strengths and weaknesses

Strengths

- The team has identified alternative materials for the bipolar plate. The use of natural graphite is a cost and performance (electrical conductivity) advantage vs. graphite composite materials.
- The team's expertise in graphitic materials and plate development is good.
- The project team is moving quickly towards a finished product.
- They have a good plan to develop/fabricate high-temperature graphite plates.

Weaknesses

- This material has a generic disadvantage for stack power density. The cost advantage vs. metallic bipolar plates is questionable.
- Estimating plate cost appears to have been postponed till the end of the project. There will probably not be enough time to make changes before the end of the project if cost estimates do not meet DOE targets.
- There are fundamental limitations of graphite composite based materials vs. metal plates (thickness, cost?).
- Performance results are very scattered (slide 21), and don't instill confidence that the process or cell assembly is under control.
- The previous year's comments suggested "Poor channel formability might create problems for certain stack designs and/or create high contact resistance or poor fit if tolerances of the plates aren't well controlled."
- The project team gave qualitative assurances that these issues would be treated, but we see no quantitative data on tolerances, or throughput.
- The team made a poor selection of MEA for fuel cell tests and/or fuel cell test conditions.
- There was a lack of supporting data to verify claims for cost, gas impermeability, high-temperature coolant durability, and reproducibility of plate fabrication within tolerances.

- It would be good to focus on materials testing with more detailed materials requirements. The FreedomCAR Fuel Cell Technical Team may be able to provide detailed attributes and metrics. Full scale testing is not recommended.
- Testing cells at 120°C should be carried out at higher current density, at least 400 mA/cm².
- We need a cost analysis. What is the ultimate potential cost per plate?
- The team needs to determine the ultimate minimum thickness due to the importance of volumetric power density (automotive packaging).
- The above two, plus durability, will determine if graphitic/composite bipolar plates can compete with metal plates for automotive fuel cell commercial viability.
- The project team needs to add manufacturability/reproducibility studies so that yield, process time, etc. can be quantified by outside reviewers.
- The project team needs to complete the final report with data that supports or clarifies the claims listed above.

Project # FC-42: Low Cost, Durable Seals for PEM Fuel Cells

Jason Parsons; UTC Power

Brief Summary of Project

The objective of this project is to develop low-cost, non-silicone, durable seal materials and sealing techniques amenable to high-volume manufacture of proton exchange membrane (PEM) stacks. The project goals are to 1) improve mechanical and chemical stability of seals to achieve 40,000 hours of useful operating life; and 2) obtain a material cost equivalent to or less than the cost of high-performance silicones in common use. Material properties meet most ultimate program goals – FCS2 is expected to meet all program goals.



Question 1: Relevance to overall DOE objectives

This project earned a score of 2.7 for its relevance to DOE objectives.

- Low-cost, durable seals are needed for PEMFCs.
- While the need for durable, cost-effective seals is clear for fuel cell applications, the general low level of disclosure of the actual elastomers and specificity of the seal development to UTC's fuel cell design make this project's outcome of somewhat questionable value to the overall fuel cell program.
- The project is focused on two key barriers in fuel cell's durability and cost.
- The project is relevant to the DOE fuel cell program objectives. Cost effective, durable seals are necessary for successful commercialization of the technology.
- Although fuel cell efficiency and durability are worthy topics, gaskets for PEM stacks necessarily are of less importance. This is especially so for a company that has claimed an important position in the global PEM stack industry, and necessarily has extensive PEM seal technology under its belt.
- The need for durable, stable seals for fuel cells is necessary, and objectives regarding temperatures and time of tests generally align with the DOE goals. The description of materials/polymers that are under consideration for testing and evaluation should be identified in order to establish a well-defined database for future use in evaluating other options.

Question 2: Approach to performing the research and development

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

- The project team took a clear approach of material selection and evaluation.
- *Ex situ* accelerated stress tests should be correlated to fuel cell data for proven rapid material evaluation.
- The general approach for rapidly screening candidates and various levels of testing that have been established are very good and make for an effective development program. There is concern that leachability tests have not been performed early in material evaluation. Also, the focus on the Integrated Molded Seal MEA element of this program may be too ambitious and specific to UTC.
- The general materials development, compatibility, and testing approach seem sound for the seal development. Their approach has limited testing in fuel cells to confirm the compatibility and applicability of these materials, in particular, under fuel cell cycling conditions.
- The approach is sound and very thorough. *Ex situ* tests are being employed to screen candidate materials. The testing includes both physical and chemical characterization under conditions expected in operating fuel cells. An extensive database of material characteristics has been compiled for the few candidates under consideration.

UTC is planning to develop a unitized MEA seal package that requires the seal material to be compatible with the MEA materials. Other MEA materials may not be compatible. The project was selected on the premise that the seal material would be available to other developers. If the material is not compatible with other MEA material sets, this benefit would be lost.

- The real goal is to edge-seal an MEA. Although it was not mentioned, this would mean bonding to the membrane, which is not necessarily dimensionally stable, tends to flow under compressive stress, and is difficult to bond. There was no consideration of the most difficult engineering task, forming the seal.
- Even with supported membranes, one still needs to seal on an acidic, chemically, and dimensionally unstable polymer.
- Henkel contains technical excellence that should have been tapped early. It is most likely that they already know the problem, or know folks who do. Every successful stack manufacturer has figured out the sealing problem.
- Cost may still be an issue; if so, costs should have been addressed.
- The tests that are outlined for evaluating the polymer candidates are in-line with the fuel cell conditions. However, no description is provided on the process that the initial polymeric materials undergo that would be useful in determining what parameters could be important in the future advancement of sealing materials. The integration of efforts is a logical approach. Only static tests are described, but cyclic tests should be included in the screening process, since all automotive fuel cells will see significant cyclic operation.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.7 based on accomplishments.

- The project team conducted a good material evaluation, and materials were developed that meet most goals. However, as the material formulations are not provided, this limits the value of this project to other developers.
- For this project to have general value, much more detail should be disclosed regarding the formulations that have been evaluated. It will likely be helpful to know how the various elastomer chemistries that were evaluated by Henkel performed against the various project requirements. It would also be nice to see hydrogen permeability measured for these materials.
- They have made good progress in developing hydrocarbon elastomers with the target properties to provide for liquid injection moldable (LIM)-type seals. They appear to have conducted limited testing in real fuel cells.
- Progress is evident, but it is difficult to assess the effectiveness of the project. Only three materials advanced to the testing stage. It is not known how many others have been screened. Even a notional idea of the composition of the materials is not known. It is not clear if the materials are hydrocarbons, fluoropolymers, or blends. Testing at Virginia Tech appears thorough and capable of stressing the materials in an appropriate manner.
- This simple task began in April 2007, and still is not completed. There is no way that the described existing work should have cost the funds spent to date.
- The Fenton reagent is a very aggressive agent, based on oxidation with the ferric ion. Many perfectly good sealants will be attacked by the aggressive ferric ion, because most everything in the Merck Index is.
- A Fenton reagent test seems foolish—simply evaluate under anticipated worst conditions.
- The team's progress is commendable. A cyclic test comparison is missing. Marginal limits in hardness, 100% modulus, and tear strength suggest that supporting information should be provided as to why these limits were found to be acceptable, e.g., couldn't do better in timeframe, material wasn't found to be better, process was limiting. Do the accelerated tests adequately illustrate failure under nominal fuel cell conditions where surface area exposure is limited and penetration of reactant (peroxide, for example) is an important issue?

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.0** for technology transfer and collaboration.

- There is a good set of partners with clear interactions, especially with Virginia Tech.
- Good balance exists among the elastomer developer, the OEM, and materials characterization experts.
- This is a good team to address the development and testing of new seal materials. The project team seems to have a well-coordinated and close collaboration.

- Three organizations, in addition to UTC, are involved. Input from other developers regarding their seal requirements is not evident.
- Virginia Tech seems to be assigned simple materials characterization testing, a rather commercial task that could be done by any number of companies, and certainly by UTC.
- The choice of experimenters and participants seems to be appropriate for the defined tasks. Coordination of the information developed and team interaction has not been shown, and communication of results and analyses is to be determined in tests to be conducted. It would be appropriate to show the coordination of tests chosen to other materials besides silicon polymers that could be potential candidates for baseline comparisons, since some of the properties of the chosen materials are marginal (see slide 9).

Question 5: Approach to and relevance of proposed future research

This project was rated 2.3 for proposed future work.

- It seems that the overall outcome of the project would be strengthened by more focus on full-scale durability testing in fuel cells with more traditional over-molded design.
- The development of the elastomer materials addresses the chemical and physical characteristics of the seal targets. However, the testing of the seal materials is planned in single cells and not stacks, which may not provide all the information on the durability of the seals. Further, it is not clear if the program plans to test the seals under real world cycling conditions in the fuel cell testing.
- There is much to do in very little time.
- The latest and presumably best material appears that it will not be released in time for the final short stack build.
- The details of activities of bonding the fuel cell membrane to the gasket in ways that permit the membrane to shrink and swell without stress cracking was not addressed and needs to be.
- It would be beneficial to show the correlation of accelerated tests, and some combinations of tests, with actual operations. Cyclic tests should be included in testing protocol. A wide range of fuel cell operating parameters still needs to be evaluated. Material compositions should be identified to some extent, even if proprietary information cannot be shared. The correlation between the analyses of final test articles, tests conducted, and materials examined should be targeted in order to provide a solid conclusive analysis of materials potential for use in fuel cells. Are gas permeability tests considered?

Strengths and weaknesses

Strengths

- The out-of-cell testing/accelerated testing is good, and correlation to actual fuel cell performance should be emphasized.
- It appears to be a solid team with strong complimenting expertise to tackle these technical issues, and they have worked effectively together.
- The team has made good materials developments and testing of the elastomeric materials with the target properties of an LIM-based hydrocarbon seal material.
- The team has a good understanding of the requirements for seals, at least in the case of UTC's needs. It is a good, capable team.
- Industrial and academia partners were chosen with good skills for this project.
- A wide range of evaluation tests were covered.
- Tests are to include actual fuel cell assembly for correlation to static tests.

Weaknesses

- Material formulations eventually need to be publicized; otherwise, this project has limited value to any other fuel cell developers.
- There was not enough disclosure of the materials being examined. The project is perhaps too specific to UTC's applications.
- There was limited testing of fuel cells and stacks to ensure the material met the fuel cell durability.
- It is hard to justify the resources for the limited information generated.
- The classification of gasket compositions (FCS1, etc.) served no purpose. Commercial gaskets are beginning proposed. There was no description of why the existing UTC PEM stack gaskets are unsatisfactory and need

replacement, nor was there any description of what others are using for gaskets. It is easy enough to buy a commercial stack and shine a Fourier transform infrared (FTIR) on the gasket.

- Interim single-cell fuel cell tests of material should be included to verify the validity of accelerated tests.
- No cyclic condition tests were included in accelerated tests.

- It seems essential that more details of the elastomers evaluated be shared. It also is important that hydrogen permeability be measured for at least the final candidates and reported.
- The team should explore a means to get the FCS3 material into the test stack.
- The project should conclude in September 2009.
- Cyclic tests of materials should be included to examine effects compared to static tests.
- Some single-cell fuel cell tests might be conducted in the interim to ensure that acceleration tests are as applicable as possible.
- Discussion of why marginal limits were found to be acceptable for continuation of tests should be provided.

Project # FC-43: Diesel Fueled SOFC System for Class 7/Class 8 On-Highway Truck Auxiliary Power Dan Norrick; Cummins Power Generation

Brief Summary of Project

The objectives of this project are to 1) demonstrate on-vehicle and evaluate a solid oxide fuel cell (SOFC) auxiliary power unit (APU) with integrated on-board reformation of diesel fuel; 2) develop a transparent method of water management for diesel fuel reformation; 3) develop controls to seamlessly start, operate, and shut-down the SOFC APU; 4) evaluate hardening the SOFC APU to enable it to operate reliably in the on-highway environment; and 5) develop the overall system for performance, size, cost, and reliability targets.



<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of **3.4** for its relevance to DOE objectives.

- This project is very relevant to the Fuel Cells subprogram's emphasis on SOFC fuel cell power systems for mobile auxiliary power applications. The primary fuel is diesel as appropriate for highway truck applications.
- This project uses a challenging approach to truly test the feasibility of SOFCs as power conversion devices. The objectives of building and mounting a complete fuel cell system on a truck, reforming diesel fuel, solving water management, and simulating vibration conditions are all part of a true test of feasibility. This is an excellent demonstration of the SOFC part of the Hydrogen Program.
- This project focuses on the development of diesel-fueled SOFC-based APUs for truck applications and fully supports DOE RD&D objectives.
- This project appropriately supports DOE objectives for APU applications.
- This truck APU is an excellent introductory application for SOFC technology.
- The truck APU application would provide an environmental mitigation technology not strictly a principle goal, but one that might gain in importance. Fuel savings was well demonstrated, but it is not clear that multiplied by the number of trucks it makes a significant contribution to reducing imports.
- This project supports the DOE targets and plans for truck APUs in existing and re-directed program described by Sunita Satyapal on May 18, 2009.
- The project should demonstrate a viable SOFC solution to anti-idling.

Question 2: Approach to performing the research and development

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

- The project has adopted a logical approach to total system design and hardware performance optimization that incorporates a total system perspective.
- This project identifies and focuses on the most significant barriers of SOFC development. Many SOFC demonstrations are conducted by stack developers who are experts in the development of cells and stacks, but not necessarily the complete system. In this case, Cummins has significant experience in building portable power systems. Here, they have developed a fuel cell power system that responds to the current weaknesses of their SOFC component, while taking advantage of its strengths. Cummins recognizes that a SOFC system must include battery storage to become practical. Their second design emphasizes this further with larger batteries. This is an honest approach to the demonstration of SOFC systems.

- The "system" approach to performing the work is logical and reasonable. Progress has been made on addressing thermal management of the system and diesel reforming. No information was given regarding the identified barriers of transient operation, power density, and vibration and shock tolerance.
- Technical barriers have been well defined and are appropriate for truck APU applications. These include thermal management, degradation, zero-water diesel reforming, mechanical robustness (shock and vibration), and cost.
- This project comprehensively addresses the application requirements.
- The project team has set an initial 5-year (10 khr) lifetime target, but a final10-year target.
- Cummins has been working in this area for a number of years, so they are well aware of the challenges and opportunities associated with SOFCs for mobile applications. Overall, the work plan is appropriate. It would have been good to see some information regarding how they plan to deal with S impurities in the fuel.
- The project appears to contain all the critical technology pieces to achieve a successful system along with the key technical requirements identified. It is not clear from the presentation whether the specific technology chosen can meet the goals.
- The project team uses an excellent overall technical approach based on Cummins extensive experience in APUs.
- Sulfur tolerance was not discussed.
- 1.5 years is the payback time.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.9 based on accomplishments.

- Good progress has been made since this project was restarted. Scale-up of output power per module is significant.
- Much progress has been made in overall SOFC systems development. However, the stack technology chosen for this project has slowed the overall progress of the project. It is not clear which is the main cause, however; in general the SOFC has not made enough advances on fuel and sulfur tolerant anodes. Also, although the tubular SOFC stack design is known for its ease of gas sealing and is tolerant to thermal cycling, cell interconnection becomes more complicated and expensive.
- DOE needs to support development of new ideas on electrode, cell, and stack designs to solve these issues. The past several years have focused on the demonstration of fixed design technologies.
- The key technical accomplishments to date include completion of overall system design and hot box design. The degree to which progress has been made toward the other defined objectives (such as transparent method of water management for diesel reformation and controls) is unclear from the presentation.
- Analysis and design is 95% complete, subsystem test and development are 90% complete.
- The project team is slightly behind on milestones, but not to a degree unexpected in an R&D program. In addition to technical difficulties, funding appears to have lagged.
- The project team is not meeting DOE program targets (100 W/kg, 100 W/L aggressive) on power density, balance-of-plant (BOP) parasitic losses, efficiency, and fuel; however, the team has indicated progress.
- Subscale stack testing reflects good thermal-cycling performance. Long-term testing is being done with one cycle per day.
- The project team demonstrated waterless CPOX with ULSD, which is significant to the application.
- Overall the project shows good progress. It's not clear whether the tubular technology from Protonex will be able to offer satisfactory performance in terms of mechanical robustness and low degradation rates.
- Good tight packaging. Technology development has resulted in components that fit well into the tight package. However, this is based on conceptual-level pictures. The actual component performance was not adequately discussed or shown. The individual cell performance is unclear and therefore an estimate of the cost is impossible. No cell dimensions were given. O/C of 1.3 is good but the length of test was not specified. It is well known that a fuel processor can adsorb carbon deposition for several hundred hours before failure.
- The project team is making good progress.
- The project team has developed a transparent method of water management for diesel fuel (ULSD) reformation and of controls to seamlessly start, operate, and shut down SOFC APUs. These are significant accomplishments.

- <1% degradation in 10 thermal cycles was attained by the SOFC.
- The simplest possible fuel processor is being used. It is extremely compact with an inexpensive design. It has demonstrated stable operation of a SOFC stack on ULSD with no added or recovered water. This is excellent. The project team has demonstrated tight thermal integration.
- The SOFC APU is only a little bigger and heavier than diesel APU.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.4** for technology transfer and collaboration.

- Meaningful collaborations are in place and active with partners who complement the skills and expertise of Cummins. It should be noted however that one of the original partners is currently inactive.
- Cummins has organized a capable team to achieve the project's objectives. It is clear that Protonex has a significant role in the SOFC component development. It is unfortunate that the OEM could not stay active in project.
- Collaborations have been excellent.
- Excellent integration with Protonex LLC and International Truck and Engine Corp. (no longer present). Welldefined roles and responsibilities. International helped define technical requirements earlier in the project - vital given the ultimate commercialization goal.
- The collaboration between Cummins and Protonex appears to be well coordinated.
- Good collaboration between a packager (Cummins) and technology provider (Protonex) and an OEM, although the OEM is currently inactive.
- Collaborators in this project include Cummins Power Generation, Protonex LLC, and International Truck and Engine Corp.
- Cummins knows APUs and the market and its customers. They are the best.
- This is good team containing excellent industrial partners. However, the truck company is now inactive.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- The proposed future work builds on this year's progress and will continue to move the project toward its established goals and objectives. Prototype fabrication and testing is critical to successful completion of this project.
- Cummins is moving forward with its partners in the new design by addressing the shortcomings of the SOFC with a flexible BOP. The overall weight and volume of the system will be improved by reducing the size of the power conditioning and the number of fuel cell modules. This creates the need to rely more on battery storage, which seems to be a fair trade-off.
- Proposed future work is reasonable; however, a detailed plan with defined objectives and schedules may be needed.
- Not much detail was presented on risk mitigation and design alternatives.
- Overall, the proposed future work is appropriate; it would have been helpful to hear more about how the issues with the SOFC cells/stacks are going to be tackled.
- Looking at a battery/fuel cell system is good. This is an old and relatively obvious system, but it is good that it is the future direction. Not much else was discussed.
- The project team's future plans include:
 - System BOP design;
 - Protonex delivery of SOFC sub-assemblies;
 - System checkout ready for vehicle install; and
 - o Vehicle tests.
- The project barriers are well defined and excellent approaches have been used to resolve them.

Strengths and weaknesses

Strengths

- This is a good system design that is flexible to the abilities of the SOFC module.
- The hybridization between the SOFC and battery storage is inevitable, and other SOFC developers should admit this need. DOE should consider supporting development of advanced SOFC advanced battery systems to take advantage of their synergistic benefits. By themselves, neither can answer all power systems needs.
- The project has a strong system development approach.
- The project focuses on key technical areas: SOFC; diesel reforming; and thermal management.
- The application has strong market potential. Cummins is a leader in the market.
- Development of the transparent method of water management for diesel fuel (ULSD) reformation and of controls to seamlessly start, operate, and shut-down SOFC APU are significant accomplishments.

Weaknesses

- There is risk in connecting all of the cells in series.
- In general, it is expensive to interconnect tubular cells together and have good power output.
- Emphasis has not been placed on addressing certain issues such as SOFC degradation and start-up/shut-down of the fuel cell.
- Cost targets should be explicit, with regular analysis and reporting to DOE. Cost targets should be tied closely to the intended application alternatives (e.g., diesel APUs). Cost is an integral and critical element of a technology development program using DOE support for public benefit.
- To be fair, the SOFC stack is the critical element. Insufficient detail was provided to permit a substantial assessment. A non-disclosure agreement would be required.
- Efficiency may not meet target 35%. System is only 18% at 1500 W. SOFC needs improved power density. Fuel utilization is only 63%.

- The Cummins team is on the right track for real-world SOFC demonstration. However, to reach the deployment stage, DOE needs to allow for more fundamental work for the SOFC stack developer to develop high-sulfur and other impurity tolerant anodes, as well as economical cell interconnection. In addition, research needs to be done to develop high-volume manufacturing processes and procedures to reduce cost.
- The project team should use metrics to measure progress for each technical task.
- The project team should evaluate degradation (if any) and start-up/shut-down of the hot box, especially the SOFC.
- The project team should consider anode recycle to enable ATR reforming. This is more complex and expensive, but has greater operating margins and efficiency. Has this been considered? R&D Dynamics has high-temperature recycle blower prototypes. Perhaps the current SOFC stack hot box/module design precludes this option? Insufficient detail was provided in the presentation.

Project # FC-44: Solid Oxide Fuel Cell Development for Auxiliary Power in Heavy Duty Vehicle Applications *Gary D. Blake; Delphi*

Brief Summary of Project

The objectives of this project are to 1) develop auxiliary power unit (APU) system requirements and concepts with major truck original equipment manufacturers; 2) design, develop and test the needed subsystems for the approved concept; and 3) build and demonstrate a diesel-fueled truck APU system for the Department of Energy. Delphi is currently focused on 1) completing the solid oxide fuel cell APU hardware build and procurement; 2) initiating the subsystem hardware testing and design iterations; and 3) performing subsystem testing and controls development, initial solid oxide fuel cell APU system brass board integration and design iteration. Delphi is on target for



meeting timing and budget and is committed to introducing solid oxide fuel cell diesel technology in full-scale production for heavy duty truck applications.

Question 1: Relevance to overall DOE objectives

This project earned a score of **3.6** for its relevance to DOE objectives.

- The successful design, building and testing of a mobile APU will demonstrate the solid oxide fuel cell (SOFC) as a viable energy conversion device. The 5 kW scale, use of diesel fuel, and vehicle application all provide a rigorous tests of the technology. This project completely supports the SOFC part of the Hydrogen Program.
- This project aims at developing SOFC-based APUs for heavy duty trucks and fully supports DOE RD&D objectives.
- This project appropriately supports DOE Hydrogen Program goals for the APU application. The project team is keeping spin-off applications/alternative uses of the technology in mind.
- The APU has good applicability from an environmental perspective, which may reach a federal level of attention in the future.
- Reviewing the presentation of Sunita Satyapal on May 18, 2009, this project supports the new fuel cell systems program direction APUs.
- Truck idling emits 11-million tons of CO₂; 200,000 tons of NO_X; and 5,000 tons of particulate matter and consumes >1 billion gallons of diesel fuel.
- The Delphi unit will exceed the 35% efficiency goal for 2010.

Question 2: Approach to performing the research and development

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

- Delphi has an aggressive approach toward meeting and exceeding the DOE targets.
- Delphi is addressing two critical issues of sulfur and carbon, which are significant barriers to SOFC deployment.
- The planar SOFC is more sensitive to mechanical shock, vibration, and thermal cycling in a mobile application; however, it has a better power density than other designs. What kind of shock damage will the stack survive?
- This project focuses on the approaches to address the technical barriers relating to diesel reforming and system integration. Work on SOFC degradation and start-up/shut-down is needed.

- DPS 3000 enhancements address key technical barriers: fuel compatibility, power, and efficiency. Autothermal reforming (ATR) is key to meeting the DOE efficiency target and is enabled by anode recycle.
- System integration appears to reflect a near-commercial product.
- Scale-up of cells to >100 cm² in active area is very technically feasible and will result in system simplification and lower cost very appropriate.
- Some of the key issues specific to an APU are being addressed. A focus on carbon and sulfur issues is a good focus. Excellent focus on original equipment manufacturer (OEM) demos with complete system packaging.
- The work is well designed, feasible, and integrated well with other efforts, especially with SECA. All barriers defined and removed.
- Truck cab heating will be totally separate initially. Further full integration is being considered.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.6** based on accomplishments.

- Delphi has been working on similar systems for some time. It seems that the diesel reforming barrier has been significant and that the endothermic (H₂O, CO₂ recycling) reformer unit will help solve this issue.
- The short-term on-truck data looks promising.
- Is there longer lifetime and thermal cycling data?
- Significant progress has been made in hardware demonstration, especially in OEM demos (PACCAR/Peterbilt and Daimler/Freightliner).
- The data is well characterized for application operational requirements and the 2.5 4 kW power requirement.
- Delphi's internal targets closely match DOE targets with the exception of start-up time. However, the project will not achieve stated DOE goals of 100W/kg and 100W/L. Those targets are perhaps overly aggressive.
- The project team is on track with respect to key milestones.
- Vibration testing has shown promising results with respect to SOFC stack robustness.
- The project has demonstrated prototypes in on-truck operation (short term). Excellent performance.
- Maximum stack test duration to date is 5,000 hours, 1%/1000 h degradation. The ultimate target is 20,000 h.
- The project presentation did not address cost status a key metric in the technology's viability.
- Power densities look excellent. Cell area was specified. Using a recycle to perform a reformation rather than partial oxidation is difficult but was apparently achieved. There was significant positive impact relative to efficiency. A full system test with two OEMs was successfully accomplished. Given the importance, more discussion was needed on diesel reforming, accomplishments, and difficulties.
- Two major highly visible successful OEM demos were completed in 2008. Completed 3 or 4 milestones 50 % on fourth critical milestone.
- Degradation is less than 1% in stack 5,000 h testing in stack accomplished.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **3.6** for technology transfer and collaboration.

- The collaborator's specific contributions to the project are not clear other than the role of consultant.
- The project team works with PACCAR and Volvo Truck. Further OEM input is expected. Multiple demonstrations have taken place with truck OEMs (Peterbilt and Daimler).
- Excellent development of system requirements through multiple OEM input and demonstration.
- PACCAR, Volvo Truck, and Electricore Inc. are key automotive partners included in the project. Project is professionally coordinated.
- Team involvement of OEM allows load profiles -potential power requirements of 2.5 kW and 4.0 kW to be meaningfully determined.

Question 5: Approach to and relevance of proposed future research

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

- The future work is focused on finishing subsystem testing and completing design and development of the whole system.
- Specific barriers have not been outlined and addressed in future plans.
- Proposed future work is consistent with technical progress. The focus of "subsystem testing and development iterations" is unclear.
- Future plans include full APU system testing in 2009 and commercial package in 2010. These plans are appropriately aggressive; it appears that Delphi has the progress-to-date, focus and capabilities to achieve these targets.
- Activities are focused and are critical to commercialization.
- The project is over 70% complete. Future work for FY09 and FY10 is well defined and executable:
 - Finish subsystem testing and development iterations;
 - Conduct 24-month critical decision milestone review;
 - Complete system module testing and development;
 - Begin full SOFC APU system testing;
 - Build commercial packaged SOFC APU; and
 - Demo test, 24-hour truck user profile using battery interface and vehicle simulation (20,000 hour planned, 5,000 hour completed).

Strengths and weaknesses

Strengths

- Delphi has long and broad experience in developing SOFC systems.
- Delphi has used its experience with natural gas (NG) systems to successfully redesign for a diesel system.
- Delphi has a strong system development approach.
- Delphi has a strong focus on subsystem and system demonstration.
- State-of-the-art SOFC technology has very high performance.
- Excellent professional team.
- High APU efficiency supporting program goals.

Weaknesses

- Fundamental issues of SOFC anode durability are a weakness and have not been discussed in the presentation. Are these issues being addressed?
- Cost targets and estimates have not been addressed.
- It appears that several critical issues such as performance degradation and control including start-up/shut-down may not be adequately addressed in this project.

- Delphi is on track for real-world SOFC demonstration. However, for developers to reach the deployment stage DOE needs to allow for more fundamental work on SOFC anodes to develop high-sulfur and other impurity tolerance and direct reforming of CH₄ and C₂s in the stack. Research needs to be done to develop high-volume manufacturing processes and procedures to reduce cost.
- Some work needs to be initiated in this project to assess degradation, control, and start-up/shut-down issues.

Project # FC-45: Solid Acid Fuel Cell Stack for APU Applications

Hau H. Duong; Superprotonic, Inc.

Brief Summary of Project

The main focus of the project is to develop a solid acid fuel cell (SAFC) stack that operates on diesel reformate with performance characteristics approaching or exceeding most of the U.S. Department of Energy's 2010 technical targets for an auxiliary power unit. This project has demonstrated SAFC functionality and stability on methanol, propane, and diesel fuels. Future work will include the scale up of SAFC membrane electrode assembly size and quantity, building a 300-W SAFC stack, and designing a 3-kW SAFC system.

Question 1: Relevance to overall DOE



objectives

This project earned a score of **2.6** for its relevance to DOE objectives.

- This project introduces a novel electrolyte similar to the phosphoric acid fuel cell (PAFC). The PAFC had problems of low power density, handling of electrolyte before start-up, and platinum electrodes. These made the technology expensive.
- The ability to function in high CO makes the project worthwhile. However, the heavy loading of platinum electrodes makes it difficult to reach DOE cost objectives.
- Why this concept is better than phosphoric acid fuel cells isn't clear. What are its benefits compared to PAFC?
- The project focuses on the development of solid acid fuel cell stacks and fully supports DOE RD&D objectives.
- The project's stated goals appear to support the DOE Hydrogen Program's goals for APUs; however, there is no connection between the project details and a particular application of national interest. Furthermore, the state of development is quite immature to the point that meeting the 2010 goals is impossible.
- The APU represents an excellent potential application for SOFC technology.
- APU applications have good environmental characteristics and may reach a federal level of attention in the future.
- This project is not projected to meet the APU 2010 target of 35% efficiency.
- Conductivity is not equivalent to PEMFC.
- It is unclear what cathode and anode were used with CsH₂PO₄ proton-conducting electrolyte.
- Even higher temperature, 200°C operation does not offset the lower temperature PEFC system inefficiencies.

Question 2: Approach to performing the research and development

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

- The approach of this project was to fabricate and demonstrate cell and stack operation. Over the duration of the project, cells have been scaled from 3 to 15 to 125 cm². Twenty cell stacks have been built and tested in a variety of carbonaceous fuels. Long-term and thermal cycling tests are proposed.
- The main issue of heavy platinum loading on electrodes still needs to be addressed.
- There is no indication of how this concept addresses the cost barrier. This concept doesn't appear to offer any cost advantages compared to PAFC.
- No clear approaches have been shown to address the issues of durability and performance improvement under practical conditions (e.g., high fuel utilization).

- The presentation did not address, in a substantial manner, the technical barriers (cost, durability, and stability), paths for resolution, and status.
- The project is fairly limited in scope, focusing just on the fuel cell stack. Given some of the significant limitations of the solid acid electrolyte, it's not clear that it's a good candidate material for the APU application. The material is brittle and weak at room temperature. Even if it is more plastic at operating temperatures, the authors did not present any evidence that cells/stacks based on this material will survive thermal cycling. Cost issues were not satisfactorily addressed.
- Technical targets appear adequate for an APU application. Much of the discussion of technical characteristics of solid acids appeared to be idealistic rather than based on extensive data. Conductivity was given on a 1/T plot, which is not a good way to present data in a time-restricted review. It is difficult to draw out technical characteristics. The presenter did not seem to understand key technical aspects of the technology. The presenter discussed tolerance in relation to the electrolyte but did not discuss electrode tolerance, which is generally the more critical issue. The presenter gave the impression that this is an electrolyte development project not even close to an APU project. Some modeling results were presented. Electrodes and other stack components are not addressed. The use of diesel at 250°C could be fairly problematic given there is higher hydrocarbon slip. It is very difficult to control carbon deposition at this temperature.
- The overall approach is good. The project team has made the right level of advancement and scale-up.
- Stability in water vapor and carbon dioxide in anode and cathode gases may need to be addressed. CO and H₂S tolerance was tested.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **2.4** based on accomplishments.

- The project team has made impressive progress in the past year in cell power density. It has increased to 0.65 W/cm² at 250°C. The 20-cell stack has been tested on synthetic reformate, propane, methanol, and diesel reformate fuels.
- The project has demonstrated tolerance to reformates. However, the degradation rate of ~0.06%/hr on reformate is a significant weakness and will have to be greatly improved to meet the durability target of 2,000 hours.
- V/I polarization curves show a current density max of about 350 mA/cm² on hydrogen, and about 300 mA/cm² on reformate. This isn't consistent with claims of power densities of ~320-600 mW/cm² on slide 6.
- Progress has been made in demonstration of operation on a variety of reformate gases and stack design/fabrication.
- In light of the Hydrogen Program's 2010 targets, this is a very immature technology.
- The production target of 500 mW/cm². Claims to have achieved >600mW/cm² this year; however, details are not provided. All data presented is substantially lower.
- Power density appears low (most data ~100 mW/cm²), with substantial degradation 0.06%/hr on reformate. Newer performance is ~1-2%/100 hr. is very high.
- There appears to be durability issue. Can this technology be viable for commercial application?
- Numerous milestones have been missed.
- Cost entitlement and potential are not addressed.
- Much of the work is focused on development of fabrication procedures for the SAFC MEA. Thus far, the areal power densities obtained are quite low (requiring many cells to obtain the desired stack power level), and while a very limited amount of results over time were reported, it's clear that the degradation rates are very high.
- Other than a few plots related to the electrolyte, very little actual technical discussion is present. A project can't successfully evolve when primarily focused on electrolyte characterization to a 300 W stack. There are major material issues that have to be addressed before this could even be considered. There is no mention of any work other than characterizing the electrolyte. The level of technical discussion does not support the later milestone discussion. It is highly questionable how the milestones were met.
- Progress has been made. Stack performance on all reformates except diesel are acceptable.
- The project team tested one or more (unclear) 20-cell, 20-cm² cell stacks for 16 days on reformates. Reformate was not done *in situ*. It can only be assumed that the fuels were externally reformed and predicted compositions were used. Non-commercial platinum anodes and cathodes were used.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated 2.4 for technology transfer and collaboration.

- California Institute of Technology (Caltech) was the inventor of the technology.
- Richard Mistler, Inc. (Mistler) helped in coating formulations.
- Superprotonic should consider teaming with a fuel cell system integrator.
- Collaboration with other academic institutions (outside Caltech) and fuel cell developers is encouraged.
- Collaboration included Caltech and Mistler.
- It was hard to tell how much coordination/cooperation exists between the partners.
- The project team collaborated with an academic institution. There was no indication of industry input or interest.
- Coordination with Caltech is appropriate.

Question 5: Approach to and relevance of proposed future research

This project was rated **2.1** for proposed future work.

- Investigators plan to scale up stacks to 300 W and 3 kW. Long-term and thermal cycling short stack tests should demonstrate cell durability.
- Platinum loading needs to be reduced.
- The plan for future work is logical, but it's doubtful that the project team has the time or resources to complete the work as planned.
- The proposed future work appears to be too general and needs to include metrics and work on durability improvement.
- The proposed work focuses on scale-up; however, the small-scale data presented suggests that the technical barriers for the 3-kW APU application are much more fundamental than would allow scale up. There is also the issue of stability/degradation and the ability to operate for 10-20,000 h. These are issues that will likely take considerable resources and time to address.
- It seems inappropriate to devote resources to the design of a 3-kW stack/system unless the authors can show significantly improved power density and stability for their current cells/stacks (and also provide some evidence that the materials and fabrication processes involved in preparing the stacks could be cost-effective; it was noted that noble metals are utilized as electrode materials).
- The future work appears unrealistic with a large focus on full stacks verses technology characterization. There are substantial degradation questions that were not adequately addressed. Until these questions are fully addressed, there is little reason to proceed further.
- Future stack plans for FY2009 are appropriate:
 - Build and characterize a 300-W SAFC stack operating on hydrogen fuel;
 - Employ micro-porous layer deposition using tape casting;
 - Employ anode and cathode catalyst layer deposition using sprayer;
 - Employ CDP (25-50 micron) electrolyte densification using compaction; and
 - Fabricate 125-cm² SAFC MEA using above processes.
- Additional work will require more funding.

Strengths and weaknesses

Strengths

- CO poisoning is reduced by increased temperature.
- Aluminum bipolar plates can be used.
- Solid electrolyte has plastic mechanical behavior at operating temperature.
- The use of a solid proton-conducting electrolyte instead of liquid phosphoric acid is a benefit.
- The cell and stack fabrication is a strength.
- Operation has been demonstrated on a variety of reformate gases.

Weaknesses

- The electrolyte is water sensitive.
- There is high platinum loading in electrodes.
- There was no mention of the projected cost impact of the electrolyte.
- A number of technical barriers relating to durability, performance in reformates, and stack design/operation have not been addressed.
- The state of development is quite immature to the point that meeting the 2010 Hydrogen Program goals is impossible.
- Low efficiency is a weakness.

- This project needs to include a cost evaluation.
- Significant effort should be focused on reducing the use of platinum.
- The project team should work to identify causes for observed performance degradation and develop approaches to mitigate this issue is strongly recommended.
- Assessment was difficult because the details were not presented. However, a detailed cost entitlement analysis supplemented with a cost estimate based upon actual experimental data would be useful in assessing the state of development. Explicit degradation targets based upon a techno-economic analysis are needed.
- Longer term R&D project requiring more time and funding.

Project # FC-46: Low-Cost Co-Production of Hydrogen and Electricity

Fred Mitlitsky; Bloom Energy

Brief Summary of Project

The objectives of this project are to 1) demonstrate cost-effective, efficient, reliable and durable solid oxide fuel cells (SOFCs) for stationary applications; and 2) determine the economics of hydrogen and electricity co-production for comparison to stand alone hydrogen production facilities. The Alaska site build was completed. The planar SOFC system is installed and operational with key metrics achieved. The full-scale hydrogen pump has been integrated with the planar SOFC. Partial pressure swing adsorption design and prototype testing are being performed in parallel. The cost of hydrogen was analyzed using the DOE H2A model.



Question 1: Relevance to overall DOE objectives

This project earned a score of 2.9 for its relevance to DOE objectives.

- This project, the demonstration of stationary SOFC power generation under realistic operating conditions, is very relevant to the Fuel Cells subprogram goals and objectives.
- Using carbon-based fuels, steam, and a fuel cell to make hydrogen is an interesting idea. However, there can be some question on its economy. A comparison between this and other hydrogen/electricity generating technologies would be useful as an initial determination of the viability of the approach. This project has the potential to support hydrogen production goals.
- The focus of this project is on SOFC system tests. Is the overlap with SOFC programs supported by DOE's Fossil Energy (FE)?
- This project explores SOFCs for co-production of H₂ and electricity. It is not clear how this effort addresses the stated DOE Hydrogen Program's fuel cell goals. Is it categorized as an auxiliary power unit (APU) project?
- Overall, the relevance to DOE objectives (improvement of hydrogen generation and fuel cell technology) is good.
- Co-production of hydrogen and power seems of less significance than producing either hydrogen or power. It is not clear that supplementing gasoline with natural gas (NG) accomplishes much from a strategic fuel standpoint.
- This project should:
 - Demonstrate cost-effective, efficient, reliable, and durable planar solid oxide fuel cell (PSOFC) systems for stationary applications;
 - Demonstrate the feasibility of a delivered cost of hydrogen below \$2.50 per gge DOE target; and
 - Give information concerning the economics of hydrogen and electricity co-production to compare with stand-alone hydrogen production facilities.

Question 2: Approach to performing the research and development

This project was rated 2.7 on its approach.

• The approach taken for this project is straightforward and directed toward field-testing of a fairly standard SOFC power system. The approach supports the project goals and objectives.

- The technical barriers relating to fuel processing, stack operation, and hydrogen purification are not clearly spelled out.
- It seems that the capital equipment cost of two independent systems can be expensive. Why is this technology preferable to other hydrogen/electricity generation technologies?
- The project appears to focus on demonstration. A number of technical barriers have not been addressed (e.g., fuel cell performance and durability). The benefits of hydrogen production by electrochemical pumps or swing absorption for this system are unclear.
- Demonstration and scale are appropriate.
- The project integrates independent third party validation and economic analysis.
- The presentation was very "high level," making it difficult to assess the specific approaches being used to overcome challenges.
- The project team discussed application of H2A model but didn't discuss methodology or detailed assumptions. The team did discuss some global results and attributed them to the H2A model, which does not provide confidence in results. A system diagram was presented. It requires two fuel cells and a reformer to produce hydrogen and power when in principle the reformer is producing hydrogen. It would seem better to either use the reformer followed by purification or electrically driven electrolysis. The system doesn't indicate purification system or external heat source for the hydrogen concentration.
- This project uses an elegant, simple design for difficult environmental conditions.
- Excellent approach for operation:
 - 25 kW power;
 - Operation on natural gas;
 - Operating at 480 V;
 - Grid parallel operation;
 - \circ Remote monitoring; and
 - \circ One-year demonstration.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.7 based on accomplishments.

- Significant progress was achieved during the past year focusing on the field-testing, including extensive operational data collection.
- There is no description of what point the developmental work started or discussion of specific technical barriers, so it is difficult to evaluate what progress has been made.
- Each SOFC stack was rated at 25 kW and 480 V (slide 9). What fraction is the power conditioning section of the system? Is it included in the cost estimate?
- Although the stack technology is presented as a black box, the system setup and testing was quick (9-13 hours from delivery). However, there are no details of what lessons were learned during the setup of the system.
- Progress has been made toward demonstration of fuel cell operation and co-production of electricity and hydrogen. No work was indicated in the presentation to demonstrate or improve durability.
- Intermediate goals and objectives are reasonable and appropriate, reflecting appropriate progress.
- The project team claims the 50 kW PSOFC system has been operational since December and met key metrics. Testing of a full-scale H₂ pump integrated with 25kW SOFC stack should start soon.
- The project team's accomplishments include:
 - 45% LHV NG efficiency (electric only), peak of 51.1%;
 - Approximately 70% availability target, actual in the >90%;
 - Grid-parallel operation;
 - Greater than 2,000 hours of operation to date;
 - 95% fuel utilization in anode recycle mode;
 - \circ 19kg/day H₂ production;
 - PPSA will enable 80% fuel recovery, 95% CO₂ removal; and
 - \$4.53 H₂/gge, at 33% electrical efficiency (proposal was \$4.82 target), if get 0.12\$/kWh credit, get \$0.97 H₂/gge.
- The fact that the authors were able to get two separate 25 kW systems up and running is encouraging. Unfortunately it's hard to tell how successful/viable those demonstration units really are. We were provided

with power output over time, but not given information regarding changes in operating voltage and/or current during that time that were made to sustain the desired power level, so it's not possible to tell what sort of degradation may have been occurring. Also, the average AC efficiencies reported in the tables appear to be inconsistent with the efficiencies shown in the graphs in the supplemental slides.

- The project team discussed pressure swing adsorption (PSA) as a significant technical accomplishment without explanation. PSAs are standard equipment. Very little presentation of, (if any) technical or economic results. 45% electrical efficiency on natural gas is not particularly good for a 25 kW system. Electrical efficiency should be in the 50% + range. There was a substantial reduction in power a few months into the test, but no explanation given. This could be major problem. Impossible to judge future viability of technology from data given.
- SOFC run data looks good running for over 4,000 hours and operating at 38.2 kW.
- Project achieved 97% availability.
- Project team demonstrated remote monitoring.
- Good progress has been made to-date.
- 45% lower heating value (LHV) net electric efficiency in electric-only mode is possible. DOE should examine method used to calculate electrical efficiency.
- There was little discussion about H₂ and PPSA.
- It is unclear whether the degradation level was measured.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.3** for technology transfer and collaboration.

- There are good collaborations in this project. The partners played an important supporting role and provided independent validation.
- Bloom Energy seems to have assembled an effective team to build and demonstrate its technology; however, it is not clear what each team member's contributions are.
- The project may benefit from interactions with organizations working under FE programs.
- This project includes diverse partners and good coordination:
 - The demo was done at Udelhoven Oilfield System Services;
 - \circ H₂ Pump LLC has been providing hardware;
 - Giner Electrochemical Systems, LLC has been providing equipment; and
 - University of Alaska has provided independent validation.
 - The collaboration between partners appears to be well coordinated.
- The project team was able to install the system at an airport, which required coordination with several local organizations.
- Excellent team for manufacture, equipment, and testing:
 - o Bloom Energy: Planar SOFC system, hydrogen testing, and project management;
 - Udelhoven Oilfield System Services, Anchorage, AK: General Construction;
 - H2 Pump LLC, Latham, NY: Equipment;
 - Giner Electrochemical Systems, LLC, Newton, MA: Equipment; and
 - University of Alaska, Fairbanks, AK: Independent Validation.

Question 5: Approach to and relevance of proposed future research

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

- Plans for future work focus primarily on completion of the field demonstration and appear to be adequate to achieve this objective. In addition, the economic analysis will be updated and completed, and the hydrogen production component will be demonstrated.
- Since specific barriers are not mentioned, it is difficult to determine if planning for future work is focused on overcoming barriers.
- The plan for future work does not provide details or proposed approaches/directions to address certain barriers such as degradation and performance or system operation improvements.

- The key element in going forward is a detailed assessment of the capital cost of the system, including the SOFC stack and balance-of-plant (BOP). This analysis should take into account the degradation and overall life of the SOFC stack as the technology stands today.
- The proposed future work appears to be in line with the overall project goals.
- There was no real discussion of future work.
- This is a nice project that has identified and managed risks.
- Plans to overcome barriers and have a successful demonstration are excellent:
 - Complete PSOFC system demonstration (1 year);
 - Complete hydrogen production demonstration;
 - Complete PPSA build, test, and investigation; and
 - Complete cost and economics analysis.

Strengths and weaknesses

Strengths

- The system seems to be well designed and developed by the performance data, but not enough of the right data is given to be sure.
- The system power output appears stable, based on the data given.
- The project team's hardware and prototype demonstration is a strength.
- This is an excellent team and a simple design.

Weaknesses

- The SOFC and H₂ separation units do not appear to be heat integrated. It is not clear how efficient and cost effective the system is.
- Approaches to resolve certain barriers are lacking.
- DOE should examine the method used to calculate electrical efficiency.

- The project team should be more forthcoming on the technology and cost barriers.
- In addition to the demos, part of the effort should be focused on addressing certain key barriers in the development of such a system. Metrics and detailed plans need to be developed.
- DOE should examine the method used to calculate electrical efficiency.

Project # FC-47: Development of a Novel Efficient Solid-Oxide Hybrid for Co-Generation of Hydrogen and Electricity Using Nearby Resources for Local Application

Greg Tai, Mike Homel, Bruce Butler, and Anil Virkar; Materials and Systems Research, Inc.

Brief Summary of Project

The overall objective of this project is to develop a low-cost and highly efficient 5 kW solid oxide fuel cell (SOFC)-solid oxide fuel-fed electrolysis cell (SOFEC) hybrid system co-generating both electricity and hydrogen to achieve the cost target of <\$3.00/gge when modeled with a 1,500 gge/day hydrogen production rate. The project focuses on materials research and development, stack design and fabrication, and system design and verification. The 2008-2009 objectives for this project were 1) 5 kW SOFC-SOFEC hybrid system development and 2) 5 kW SOFC-SOFEC hybrid system evaluation.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.2** for its relevance to DOE objectives.

- This is a clever idea for using carbon-based fuels and steam to make hydrogen. However, there may be some questions on its economy. A comparison between this and other hydrogen/electricity generating technologies would be useful to determine the viability of the approach. This project has potential for meeting the Hydrogen Program's goals.
- This project aims at developing a system capable of co-producing electricity and hydrogen and fully supports DOE RD&D objectives.
- It's not clear how this project supports the stated DOE Hydrogen Program's fuel cell goals. Is it considered an auxiliary power unit (APU)?
- The subjects of the work (improvement of SOFC and hydrogen production technology) are relevant to DOE objectives.
- Hydrogen production via water electrolysis in an SOFC is an interesting approach.
- The project will lead to the development of a low-cost and efficient 5 kW SOFEC-SOFC hybrid system cogenerating both electricity and hydrogen and achieving the cost target of <\$3.00/gge when modeled with a 1500 gge/day hydrogen production rate.

Question 2: Approach to performing the research and development

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

- The hybridized 13-cell SOFC/20-cell SOFEC stack design seems like a good design for heat integration. Is the gas manifolding too complicated to continue in this direction?
- How well is the 5 kW design thermally integrated?
- Does use of LSCM as a reversible cathode work well enough? This material has not been fully proved as an active, conductive electrode.
- The project approaches are logical and focus on resolving certain barriers in the SOFC followed by stack and other component designs, then integration and testing.
- This project looks at H₂ generation by electrolysis and a low-cost, efficient 5 kW SOFEC-SOFC system for cogeneration <\$3/H₂ gge.

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- Approach is fairly comprehensive, culminating in proof-of-concept fabrication and operation.
- The approach, involving hybrid SOFEC/SOFC stacks, is interesting and worthy of investigation.
- There is a good balance between component and system development. It would seem more efficient to produce power or hydrogen, versus combining the two functions. In most cases, the power-producing SOFC will not produce sufficient heat to substantially contribute heat to the electrolysis function (i.e., combining the functions doesn't necessarily produce a synergistic system). Having the power and electrolysis co-located could be very problematic in maintaining a balanced thermal profile within the respective stacks.
- The project team has a well-documented and conceived approach and a well-integrated SOEC/SOFC concept to optimize thermal integration.
- An excellent engineering analysis of performance and cost can be expected.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.3** based on accomplishments.

- The team has designed and fabricated the cells for a 5 kW SOFEC-SOFC hybrid system and has tested cells, 2cell stacks, a 13-cell SOFC/20-cell SOFEC stack, and a 40-cell 1 kW SOFC stack. The team will need to assemble the 5 kW unit and test it before July. More analysis and development of the cathode will probably be needed. What about the anode material? The Ni-cermet has poisoning and coking issues when using carbonaceous fuels.
- The key achievements include development of cathode materials stable under redox conditions, 1 kW stacks built and tested, 5 kW components fabricated, and long-term stability tests.
- The project appears to be progressing well towards 5kW construction and operation, given the apparent underfunding.
- Results to date are encouraging, although degradation rates appear to be quite high. It will be interesting to see if the approach can be extended to the larger 5 kW scale. More information regarding the expected cost of materials and fabrication would have been useful.
- The project team has presented a reasonably good level of technical detail. It is clear that the LSCM cathode is only moderately active –the price that is paid for claimed stability in reducing and oxidizing conditions. The electrodes can be problematic in this application. It may require extensive work to demonstrate that they are truly stable for extended life beyond a thousand hours or so. Extensive microanalysis work needs to be done. In particular, substantial transmission electron microscopy (TEM) work needs to be done.
- SOA SOFC performance with the LSCM electrolyte has been excellent.
- The project team has conducted long-term stability tests of hydrogen production to reduce cost. The team has finalized the design of hybrid modules with improved thermal and flow management.
- The project team has designed, fabricated, and tested main balance-of-plant components.
- The project team has fabricated cell/stack components for the 5 kW system.
- The project team has assembled and evaluated the 1 kW SOFC stack with the new design.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.2** for technology transfer and collaboration.

- The partners in the project are known for their experience in the field and can contribute to the project. It is not clear however why anode fracture and residual stresses are an issue as to require focused attention.
- Excellent coordination has taken place among resources with appropriate skill sets and capabilities:
 - University of Alaska at Fairbanks (UAF) anode failure and residual stresses;
 - Missouri University of Science and Technology (MST) cathode and seal materials development; and
 - University of Utah (UU) interconnect development.
- Project collaborations are difficult to evaluate; collaborations weren't discussed much in the presentation.
- There has been good collaboration for technology development. MST and UU add substantial component expertise.
- Excellent team:
 - UAF: anode fracture mechanisms and modeling of residual stresses (S. Bandopadhyay);
 - o MST: cathode & seal materials development (H. Anderson; R. Brow); and
 - UU: interconnect development (A. Virkar).

<u>Question 5: Approach to and relevance of proposed future research</u>

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

- The 5-kW unit still needs to be demonstrated and a cost analysis needs to be completed for this project. In addition, experiments and analyses demonstrating biomass or coal operation would help the idea obtain broader support.
- The plan is straightforward for the proposed future work. Development of test protocols, including startup/shut-down and controls and metrics, may be needed to support the 5 kW system demonstration.
- The future work is appropriate and includes system assembly and operation and cost analysis.
- The planned scale-up of the technology seems appropriate, but cost-effective resolution of performance degradation issues should take higher priority.
- Future work is focused on producing a full system with a cost estimate. It is questionable whether detailed component R&D is done, particularly in regard to stability of electrodes in a wide range of conditions and thermal integration and profiles in the stacks.
- Future work is well-planned and will result in testing and analysis:
 - SOFEC-SOFC hybrid module assembly, integration and burn-in;
 - o 5-kW hybrid system assembly;
 - System testing and evaluation;
 - o Implementation and optimization of system controls; and
 - Hydrogen production cost analysis using H2A model.

Strengths and weaknesses

Strengths

- Materials and Systems Research Inc. (MSRI) has extensive experience in fabricating solid oxide cells and stacks, and has excellent research-level stack production facilities.
- MSRI also has a good understanding of SOFC materials development and testing.
- Key technical barriers have been identified and addressed.
- The project team has used logical approaches.
- Excellent SOFC co-generation system performance and design.

Weaknesses

- Not much has been said about fuel processing and impurity removal. Impurities affect the operation of the fuel cell anode.
- More explanation of the design of the balance of plant is needed.
- System integration can be improved.
- The project team needs to eliminate some piping in the hybrid design.

- The viability of this approach should be addressed by a comparison of this and existing hydrogen generation technologies.
- The project team should consider expansion of the tests on the type of fuels used, especially biomass and coal.
- The project team should develop metrics/success criteria and test protocols.
- The level of detail and rigor in the cost analysis is critical to assess the true status of the technology, and should include such factors as system degradation and stack life.

Project # FC-48: Silicon Based Solid Oxide Fuel Cell for Portable Consumer Electronics

Alan Ludwiszewski; Lilliputian Systems, Inc.

Brief Summary of Project

The objective of this project is to improve the cost, durability, and performance of the MEMS-based solid oxide fuel cell (SOFC) technology for consumer electronics applications. Project objectives are to 1) increase the MEMS SOFC array power density toward a level sufficient to support a commercial product launch; and 2) improve vacuum sealing materials and processes to extend lifetime at higher operating temperatures.

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>



This project earned a score of **3.0** for its relevance to DOE objectives.

- There is a great need for increased energy density in power sources for portable consumer electronics. Fuel cell technology will allow longer operation times than a battery system, so there is an opportunity here to support the main objectives of the Hydrogen Program. Nevertheless, Lilliputian technology competes with direct methanol fuel cells (DMFC), which have made great progress and operate at room temperature. The PIs need to demonstrate and openly document high energy density, low cost and long life over and above DMFCs.
- The project focuses on the development of silicon-based SOFCs for portable consumer products and fully supports DOE RD&D objectives.
- This project supports the DOE Hydrogen Program's 2010 consumer electronics goals; however, it is difficult to see how this work supports national energy goals and interests security, availability, cost, and environmental.
- The subject of the project (portable SOFC power systems) is relevant to DOE objectives.
- It is not at all clear what role portable electronics plays in the hydrogen or R&D objectives.
- DOE Hydrogen Program stated objectives include development and demonstration of cost-effective, reliable, durable fuel cells for portable power applications.
- Initial product targets are below DOE 2010 targets for sub-50 W systems.

Question 2: Approach to performing the research and development

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

- There must be a practical limit to the cell size where current path length and heat integration give way to the small cell area. Generally, the larger the cell area, the cheaper the materials cost is until high current cannot be transferred out of the stack. It is not clear how well MEMS technology can achieve cost targets with so many small, interconnected units.
- Will the large MEMS fuel cell array depicted on slide 5 have a significant heat disposal requirement? What has been done to model and design for this?
- The fuel-type and cathodic activity determine the SOFC operation temperature. How do fuel impurities affect operation? How is the air and fuel flow controlled for each module? There is a significant materials issue preventing the silicon substrate from oxidizing. What is the role of the silicon substrate, is it really needed?
- It was unclear how cell performance and sealing would be improved; no details were given on approaches to address these issues.
- 1-25 W range for consumer electronics is not the optimal application for a high-temperature fuel cell. An assessment of the cost entitlements should show that.

- The work plan to achieve the specific objectives (higher power density and improved vacuum sealing) was well organized and consistent with other aspects of the technology.
- Seal work is detailed but not pursuing any new concepts. Clearly looking crystallized glasses; which has proven to largely be a dead end. The project is at a very low level of maturity. Much work needs to be done.
- Lilliputian Systems' MEMS-based SOFC technology could be a scalable, cost-effective, environmentally robust approach to addressing the consumer electronics application.
- The project team has an excellent understanding of markets.
- There are 192 cells in the array.
- The team used an electrical heater to start up the device. A 4-minute start-up time may be too long.
- No thermal cycling and degradation data discussed. 300 thermal cycles were attained of the necessary 3000 thermal cycles.
- SOFC component materials were not discussed.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **2.4** based on accomplishments.

- It seems that this technology has not progressed very far since 2002. Specific cell and stack performance information would allay this suspicion. What is the power density or power output? What do polarization curves look like? What is the fuel utilization and what happens to the heat from excess fuel?
- The issue of vacuum sealing is a difficult problem with an operating 800°C device in close proximity to the vacuum seal.
- Progress has been made on improving cell power but significant challenges remain on sealing.
- 300 thermal cycles have been completed to date. Seal degradation 250-hour life, need at least 2 khr. Apparently a modified glass composition was developed; however, it could not be tested in the timeframe of the project.
- It appears that increased power density was obtained, although it's not clear if the achieved power density is sufficient for the proposed application. Also, power degradation rates should have been addressed in the presentation. Sealing obviously remains problematic, and it doesn't appear that the authors have identified a viable sealing material at the present time. Finally, it's hard to tell if the proposed technology can be cost-effective.
- Considering it would normally take several hundred million dollars to develop a relatively new fuel cell design, this project is at the expected level of low maturity. Nothing was said about thermal issues, which would be a major question in this size. Self-sustainability is very tenuous and potentially reduces efficiency substantially. However, good detailed work on a few aspects such as producing the silica backbone and seals although ultimately the glass approach has been more or less shown to be a dead end.
- Significant progress in very difficult undertaking was made in:
 - Increasing array power density (2.5x) toward that of button cell benchmarks (this is major accomplishment now at low end of commercial product);
 - Exceeding the target of 50% power improvement with improvement of 85% attained;
 - Implementation of automated dispense equipment with custom control enhancements, increased throughput, and improved repeatability;
 - o Improvement of vacuum seal temperature/lifetime performance;
 - o Improved understanding of die/sealing glass interactions; and
 - Manufacturing fuel cell chips with the improved processes and a 2.5x improvement in power.
- The total current tested lifetime was not discussed. Items like cooling and fuel utilization were not discussed. Butane fuel processing was not discussed.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **2.9** for technology transfer and collaboration.

- Alfred and MS&T are helping with the vacuum sealing and electrode issues. No original equipment manufacturers (OEM) have been identified who will provide production capacity and cost reductions.
- Alfred University and Missouri S&T bring excellent credentials with respect to glass science and sealing.

- The collaboration with partners appears to be well coordinated and productive.
- Alfred University and Missouri S&T provide excellent SOFC component support. There has been no industry interaction.
- The project team has partnered only with universities: Alfred University, Missouri S&T, and no industrial partner.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

This project was rated 2.4 for proposed future work.

- It is not clear how much the performance needs to be improved to achieve a viable product. The PI is retaining important performance data in order to be able to determine where the main issues lie.
- Scale-up to volume production may be too early? It is unclear whether all the issues relating to manufacturing have been resolved.
- This project is complete. Future work should focus first and foremost on cost if everything worked well, is this application of SOFC technology economically viable? Does it offer significant advantages over other existing and under-development technologies for the application?
- Not much detail was provided regarding future work, but it makes sense for the focus to remain on power density (and stability) and sealing quality.
- Several hundred million dollars in development is likely pending. The project team is nowhere near having a product as implied.
- Project complete.
- The target first product is a USB charger capable of delivering 2.5 W with the following single cartridge performance: power density: >25 W/l; energy density: >500 Wh/l; and run time per cartridge: >30 h.

Strengths and weaknesses

Strengths

- For applications with extremely small power requirements (mW levels), this technology may become viable.
- SOFC power densities should be larger than other technologies if the operation temperature of 700°-800°C can be successfully contained in such a small space.
- The tiny cell size could makes it easier to increase voltage output without losing power by minimizing current path lengths.
- Cell design and manufacture.
- Understanding of customer requirements.

Weaknesses

- It is not clear from this presentation that the technology can be scaled large enough to provide 3 4 W of net power.
- It is not clear how the heat is disposed for large 22 x 8 cell arrays.
- It is not clear which fuel has been demonstrated with the technology. Has butane been demonstrated?
- Approaches to address technical barriers are unclear.
- Seal stability and isolation from silicon structure (interaction between silicon structure and the SOFC) is a key issue.
- Low power density.
- No industrial partner.

- The PI needs to be more forthcoming with performance data.
- The PI needs to state clearly how this particular technology can meet performance requirements because of their design, materials and methods.
- Emphasis on establishing all of the key cell operating characteristics is recommended.
- Future work should focus first and foremost on cost if everything worked well, is this application of SOFC technology economically viable? Does it offer significant advantages over other existing and under-development technologies for the application?
- Focus on durability and stability, if the economics justifies the application.

Project # FC-49: Biogas Fueled Solid Oxide Fuel Cell Stack

Praveen Cheekatamarla; NanoDynamics Energy, Inc.

Brief Summary of Project

The main objective of this project is to develop and demonstrate a 400 W stack module using advanced solid oxide fuel cell (SOFC) technology. Specific objectives include 1) electric power generation from different biogas compositions; 2) costeffective SOFC manufacturing; 3) increased energy efficiency; and 4) the capability to integrate in to commercial systems such as micro combined heat and power and portable power. A new SOFC fabrication route was successfully implemented. Future work will focus on thermal shock resistance and long-term endurance of the SOFC operated on biogas.



<u>**Ouestion 1: Relevance to overall DOE**</u> <u>objectives</u>

This project earned a score of 3.1 for its relevance to DOE objectives.

- This project using a renewable fuel source coupled to SOFC electrical generation is relevant to the goals and objectives of the Fuel Cell Program.
- The use of biogas as fuels for SOFCs is a critical move to demonstrate that SOFCs can be an efficient green technology. This project helps expand the scope of the Hydrogen Program.
- The project fully supports DOE RD&D objectives.
- It is not clear which of DOE Hydrogen Program's fuel cell goals this project is pursuing the "low weight" and thermal cycling suggest a truck APU. But if that is the case, why biogas?
- This project is relevant to DOE objectives (i.e., fuel cell technology for portable power, renewable energy).
- Presumably, the DOE Biomass Program has a use for such a fuel cell. Certainly, the U.S. Department of Agriculture (USDA) does in rural areas. Relevance to the Hydrogen Program appears tenuous.
- The project supports DOE EERE through:
 - Research and development of advanced fuel cell technology for portable power applications;
 - Renewable energy-biogas is a domestic, renewable resource, which offsets the use of non-renewable resources with corresponding emission reduction and energy security benefits; and
 - Viable, cost effective process for efficient power generation.

Question 2: Approach to performing the research and development

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

- The technical approach adopted in this project focuses on the development of advanced SOFC technologies coupled with a biogas fuel input. The approach includes design, fabrication, and testing of a complete power system.
- Most of the project effort is spent on developing a tubular SOFC "stack" or module. Tubular SOFC designs are interesting because of the ruggedness of individual tubes and the elimination of seals. The major problem with tubular cells is economically interconnecting tubes together to increase voltage output.
- The major point of the project is the use of biogas to fuel SOFCs. Is there a need to develop an integrated fuel processor to generate biogas from bio-solids? Are there potential partners who could help address this important issue?

- Approaches to performing the work are reasonable. Ways to improve fabrication yields need to be developed.
- The project identifies cell yield as a barrier; however, based on the presentation, it is in no way evident what is novel about this SOFC technology that causes there to be cell fabrication yield issues.
- The approach is to fabricate "lightweight" cells, but no comparison or reference is provided.
- Critically, the approach does not address cost in any meaningful way.
- Emphasis on optimization of fabrication yields seems premature. It's more important to focus on fundamental challenges (performance degradation over time, increasing fuel utilization, thermal cycle stability, demonstration of 400 W stacks).
- Statements of approach were highly generalized and unsubstantiated; therefore the reviewer was not able to render a judgment.
- The project team identified most barriers. The team is somewhat inexperienced.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.6 based on accomplishments.

- Good progress since the beginning of the project at the start of FY09. Advances in performance, using H₂ fuel, are impressive. Operation on synthetic biogas will require additional optimization.
- The technical accomplishments that have been made so far are designing, fabricating and testing single cells in hydrogen and simulated biogas. Single cell performance is good, >0.525 W/cm², assuming the cell voltage is not less than 0.7 V. Stack design and building is in the early stages. The real test will come in interconnecting cells into bundles or modules. A 400 W stack will need a minimum of 42 cells. The PIs should give more information on standard operation metrics (e.g. fuel utilization, polarization curves, or current at 0.7V) to demonstrate that cell performance meets DOE objectives.
- Technical achievements include electrode material and microstructure optimization, biogas testing, and stack operation. Current cell fabrication yields (at about 50%) are too low and need to be improved.
- Presentation of power and volumetric density does not address the balance-of-plant (BOP), which is substantial in terms of volume and weight. No comparison to other SOFC technologies is presented. How does this compare to the state of the art? The power density of >500 mW/cm² on a button cell is not impressive in this regard, even on biogas. Power and efficiency numbers appear to be presented on a stack basis. They have little bearing on ultimate system performance, given that state-of-the-art SOFCs can operate at very high fuel utilizations and voltages with economically viable power densities. In this case, a meaningful discussion of cost is not presented.
- Thermal cycling performance appears good although this may be only a button cell, in which case it means little in regards to full system cycling capability.
- Progress has been made, but performance over time and the ability to scale up to stack level have not yet been adequately addressed.
- Technical accomplishments were highly generalized and contained no specific information to review. Some volumetric power density data was presented but it was based on single cell volumetric power density, which is fairly irrelevant given that most of the volume will be BOP. Areal power density would be relevant but not reported. Information presented is very vague.
- New SOFC fabrication route was successfully implemented. This process offers numerous benefits over conventional methods: cost effective, automated, lightweight fuel cells with high energy efficiency.
- SOFCs with high volumetric (3.4 kW/liter), gravimetric (2.56 kW/kg), and specific power densities (0.63 W/cm²) were fabricated and tested on hydrogen and biogas. Is this SOFC only single cell? Tubular design is not correctly accounted for.
- SOFC cells demonstrated up to 50% and 40% electrical efficiencies on hydrogen and biogas respectively.
- There is only 32% stack/system efficiency on H₂.
- However there were achievements:
 - \circ Achieved >525 mW_e/cm2 producing >9.5 We on biogas;
 - Thermal shock tests: 120 cycles conducted; and
 - Endurance tests: 250 hours achieved.
- Unfortunately, there is a 12-16% drop in power with biogas as the fuel. There is only 26% efficiency.
- Stack/cell design, sizes, and fuel utilizations were not adequately identified.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **2.1** for technology transfer and collaboration.

- Collaboration consists primarily of a university partner who provided analytical support for the project. Other collaborations, including a user community representative, would be useful.
- The State University of New York (SUNY) at Buffalo partner is not a subcontractor. This project could use system development partners, especially in the reforming of biomass to biogas.
- The only collaboration is with SUNY on an as-needed basis.
- The project uses some university support on an as-needed basis; otherwise, all work is done in house.
- Informal collaboration only with SUNY.
- Virtually no collaboration. However, it is commendable that the project procures microanalysis as needed.
- Collaborations were limited to SUNY.

<u>Ouestion 5: Approach to and relevance of proposed future research</u>

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

- Plans for future work appear to be adequate, although the design and testing of the 400 W stack will be challenging and new technical issues will likely be encountered.
- The stated next steps of this project are to demonstrate long-term cell performance and to collect tubular cells into modules with high power output. More importantly, a critical step should be to show that real biogas, including all its impurities, can be internally reformed in this SOFC system while maintaining high power output. The project team should have better addressed how it plans to accomplish these important tasks.
- The purpose of certain proposed tasks is unclear:
 - Fabricate 150 cells with integrated catalytic layer (what is the justification for the number?); and
 - CFD analysis of 1.2 kW (is the focus is on 400-500 W stacks?).
- Future work should focus primarily on durability (long steady-state run times), degradation (which was not discussed), and particularly cost.
- The proposed future work is okay overall, except for emphasis on fabrication yield.
- Determining what barriers exist is very difficult, but list of future actions is reasonable.
- Plans are probably aggressive:
 - \circ 400 W stack build 11/2009;
 - CFD analysis of 1.2 kW stack –10/2009; and
 - Stack feasibility demonstration on biogas.

Strengths and weaknesses

Strengths

- Good cell performance in simulated biogas.
- Good thermal cycling performance of single cells.
- Cell manufacture.
- Electrode materials and microstructures.
- Stacks operating on biogas.
- Achieved $>525 \text{ mW}_{e}/\text{cm}^{2}$ producing >9.5 We on biogas. Utilization and cell/stack size is unclear.

Weaknesses

- No provision is made for demonstrating cell/module performance in real biogas.
- No clear design is mentioned on how cells will be bundled to increase voltage output without loosing power or thermal cyclability.
- Cell reproducibility and long-term stability not well defined.
- Overall, too little information was presented about the cell construction and materials to permit a full assessment of performance and cost potential.
- There is little endurance testing.

• The lack of experience presents a possible challenge in building large stacks.

- The project team should incorporate plans to study operation on actual biogas; e.g., biogas from water treatment operations.
- The project should be expanded to include biogas production method and demonstration of technology using real biogas
- The project team should focus on long-term operation (at least 1000 hrs) with demonstration of no coking on biogas.
- Future work should focus primarily on durability (long steady-state run times), degradation (not discussed), and particularly cost.

Project # FCP-02: Component Benchmarking Subtask Reported: USFCC Durability Protocols and Technically-Assisted Industrial and University Partners

Tommy Rockward, Rod Borup, and Fernando Garzon; Los Alamos National Laboratory

Brief Summary of Project

This task supports LANL technical assistance to fuel cell component and system developers as directed by DOE. This task is expected to include testing of materials and participation in the further development and validation of single cell test protocols. This task also covers technical assistance to the U.S. Council for Automotive Research (USCAR) and the USCAR/DOE Freedom Cooperative Automotive Research (FreedomCAR) Fuel Cell Technology Team. This assistance includes making technical experts available to the Technical Team as questions arise, focused single cell testing to support the development of targets and test protocols, and regular participation in working and review meetings.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.8** for its relevance to DOE objectives.

- This program supports the DOE R&D objectives.
- It is important to provide consistent measurement methodology/protocol to pursue fuel cell R&D.
- Support for developers from national labs is important to maintaining a highly scientific approach while keeping characterization and analysis costs within a reasonable budget for those developers that cannot afford in-house equipment and dedicated personnel.
- The project fills a void in the Hydrogen Program by making advanced testing and analytical procedures available. The LANL fuel cell team has the expertise to assist other research organizations.

Question 2: Approach to performing the research and development

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

- This project provides a wide variety of support to companies and universities engaged in fuel cell R&D.
- Basically, the team is consulting as required.
- Activity to standardize testing methodology/protocols would be good.
- The approach appears to be to wait for DOE or others to contact LANL for assistance. LANL could be more proactive and identify those researchers having limited capabilities and offer assistance to them.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 3.3 based on accomplishments.

- This project has supported many important efforts.
- The presentation/poster identified a number of interactions that produced positive and beneficial results to other research organizations. The examples for analytical capability are impressive.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **4.0** for technology transfer and collaboration.

- Excellent collaboration.
- Excellent collaboration with a wide range of partners and institutions.
- There are a large number of collaborators.

Question 5: Approach to and relevance of proposed future research

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

- The project team has a good plan. The approach builds on current successful interactions.
- New.
- Future work is not identified on the charts. Holding a class in 2009 appeared to be the only planned future work.

Strengths and weaknesses

Strengths

- Excellent collaboration.
- This project provides a wide variety of support to companies and universities engaged in fuel cell R&D.
- Testing and consulting capability.
- Experienced researchers and facilities at LANL.

Weaknesses

• The project team needs to focus on future work.

- The project team should develop standardization of measurement methodology and protocols for fuel cell materials evaluation.
- The project team should continue this activity and become proactive in support of other Hydrogen Program projects.

Project # FCP-03: Research & Development for Off Road Fuel Cell Applications *Richard J. Lawrence; IdaTech*

Brief Summary of Project

The objectives of this project are to 1) have Toro measure loads and report vehicle modifications and specifications, 2) report on shock and vibration profiles and lifetime, 3) complete shock and vibration of the fuel cell system, 4) have Donaldson measure contaminants and develop an air filter, and 5) install a proton exchange membrane liquid-fueled system in a golf course maintenance vehicle. The critical assumptions for this project are that the fuel cell system can 1) physically fit into the vehicle, 2) can provide the required energy during field-testing, and 3) function under applications' shock and vibration loads. Potential solutions include 1) modifying the vehicle, 2) improve controls and response;



and, 3) incorporate shock and vibration test results.

Question 1: Relevance to overall DOE objectives

This project earned a score of 2.8 for its relevance to DOE objectives.

- This project provides off-road demonstration of a methanol fuel cell maintenance vehicle.
- Fuel cell demonstrations in a real-world, near-term application are important to outreach and support commercialization of fuel cells.
- This project is relevant to the DOE Fuel Cell Program in that it provides a demonstration of the technology in a near-term application and can potentially provide feedback to the materials development efforts within the DOE program. Reforming technology will likely become more important in the DOE program for stationary applications. The project addresses issues related to air filtration and shock and vibration.
- The project pursues the goal of an early market introduction and penetration. Additional scientific goals are not pursued.
- Objectives A through E are not addressed.
- The project could align with most aspects of the Hydrogen Program and DOE RD&D objectives, but as presented the information would not be very supportive of the Hydrogen Program.

Question 2: Approach to performing the research and development

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

- The vehicle uses an IdaTech 3-kW methanol fuel cell. (R&D on this existing technology started in 2004 and was stopped in 2005.) A maintenance crew uses the vehicle at a golf course. The fuel cell was already developed using DOD funding, so the project made good use of the existing system. Shock and vibration testing provided useful data.
- Golf course maintenance vehicles are a good near-term market that can help increase fuel cell sales volume and therefore reduce cost.
- The approach is practical, which keeps the project simple. The project uses existing technology developed at IdaTech and modified it for this application. The poster did not present air filtration and shock and vibration targets and how well IdaTech met them. The engineering of the system in the vehicle appeared to be well done.

- The project focused on making a system work, and the system was successfully proven to work. Though the system works as a hybrid advanced design, methods for determining the battery size versus the fuel cell size were not applied. An average systems efficiency of 40% was stated, but not substantiated.
- From a market introduction point of view, the project focuses on a very reasonable application.
- The approach was not clearly stated in the presentation. The approach appears to be to install a fuel cell in a golf maintenance vehicle and drive it around the golf course.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.6 based on accomplishments.

- The project team has obtained about 60 hours operation on a golf course (start/stop, not continuous). A lot of spurious shut down problems have hindered data collection. Many wiring problems have been encountered. Voltage and power curves were obtained, but they are not very understandable.
- The testing was performed under realistic conditions.
- The vehicle is up and running on a golf course. Operating data is being acquired that will assist in further development of the power plant. The intended customer Toro is actively involved in system definition. Data showing the effectiveness of the air filtration would be helpful.
- The system was proven to work.
- Fuel cell performance was measured, but were strain gauges attached to the fuel cell system? What was the level of vibration? What methods were used to dampen the vibration or shock to the system? The technical accomplishment presented was "see it works." This is not at the level of understanding of why it works. There is no data on fuel or fuel efficiency.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **3.2** for technology transfer and collaboration.

- There have been interactions with Toro, although Toro is not really an active participant. University of California at Davis (UC Davis) is a partner. Rivers Edge golf course provides a demonstration site.
- Collaboration appears to be good. Toro is actively involved in developing the definition and specification for the final commercial product. However, what is not clear yet is whether or not the fuel cell maintenance vehicle will be commercially viable. UC Davis performed shock and vibration testing on a vibration table.
- A good industry consortium exists, yet the approach left the impression of being too practical not sufficiently making use of existing cutting edge design insights and tools. An academic partner might help improve this weakness. The potential role of that institute is suggested to be control and hybridization of fuel cells.
- There has been outstanding collaboration within the scope of the effort.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.8 for proposed future work.

- The project team will do public outreach at the upcoming Bend High Desert Conference. On-board data acquisition will be installed this year and should provide better quality data, 24 hours per day. The project team wants to get into parks and recreation.
- The additional testing planned appears to be appropriate. A second-generation system will be built that will incorporate learnings from the vehicle currently undergoing testing. Design for manufacturability will be developed with input from Toro.
- The project is basically finished. Insofar, this point is not really relevant any more.
- What barriers are being addressed by the future work? How will this effort be calibrated? What will full time data acquisition acquire; vibration data, shock data, or fuel cell performance data?

Strengths and weaknesses

Strengths

- The project team uses fuel cells developed under government funding. Some good public outreach. Lots of electrical problems have been encountered and this provides insight into the use of "already developed" equipment.
- The is a near-term application.
- So far, the project has developed and tested hardware in a maintenance vehicle on a golf course. A second-generation vehicle is planned.
- There has been strong industrial cooperation, which even survived a period in which DOE did not fund the technology.
- IdaTech makes a good fuel cell system.

Weaknesses

- More systems are needed to provide a "real" test.
- Only one ultimate user is represented on the team.
- The team has used a very industrial trial and error approach.
- What fuel was used? The project team claims liquid fuel was used, but the type of fuel used was not provided in the presentation. The presentation did not answer the questions required by DOE (e.g., future work did not identify barriers). What were the performance indicators for the technical accomplishments?

- The project team should get information on cost objectives commercialization plan none was presented. For continued funding, this should be made clear.
- The project team should evaluate whether controls and battery selection (type, size) can be improved to reduce cycling of the fuel cell stack, and therefore improve its life.
- The project would benefit from more input by the ultimate user community (e.g., golf course superintendents and other building owners). If the community is enthusiastic about the vehicle, it would strengthen the business case for Toro.
- The project team should add an institute to the project with practical experience in control and hybridization of fuel cells.
- This program needs a major overhaul. Vibration and shock data should be quantified. Fuel efficiency needs to be identified. Design characteristics for dampening shock and vibration should be discussed.

Project # FCP-04: Renewable and Logistics Fuels for Fuel Cells at the Colorado School of Mines

Neal P. Sullivan, Robert Braun, Anthony M. Dean, Robert J. Kee, Ryan O'Hayre, and Tyrone Vincent; Colorado School of Mines

Brief Summary of Project

Objectives for this project are to 1) develop solid oxide fuel cell materials for robust operation on diesel fuel; 2) identify optimal hydrocarbon fuel reforming strategies; 3) create thermally stable fuel-reforming catalysts and supports; 4) employ system modeling to optimize auxiliary power unit configurations; and 5) utilize modelpredictive control to integrate system hardware.

<u>Question 1: Relevance to overall DOE</u> <u>objectives</u>



This project earned a score of **3.0** for its relevance to DOE objectives.

- Multitask (5 different groups) solid oxide fuel cell (SOFC) auxiliary power unit (APU) systems for heavy vehicles. Improve performance, increase efficiency, and develop understanding of thermal management and transient operation.
- SOFC APUs are relevant to the Hydrogen Program.
- The project includes different tasks that address various aspects of operating solid oxide fuel cells on diesel or biofuels, including sulfur tolerance, tar-free fuel reforming, and durable reforming catalysts and supports.
- General objective of broadening fuels suitable for SOFCs is certainly relevant. DOE's refocus on stationary, portable, and auxiliary power in the near term increases the project relevance.

Question 2: Approach to performing the research and development

This project was rated 2.3 on its approach.

- Develop new SOFC materials for greater durability and robust operation using low sulfur diesel fuels. Evaluate various fuel reforming and integration strategies. Do cell and stack modeling to understand operational characteristics, thermal management. Translate physics-based models into system models. Optimize APU configuration and performance. Projects attempts to do wide-ranging fuel cell APU effort, and ultimately bring results from different groups together to improve fuel cell APUs using diesel fuel.
- The approach seems similar to work performed under SECA- investigation of non-nickel catalysts, modeling efforts, etc.
- The heat exchangers efficiency was surprisingly low improving this should be a top priority.
- There are two significant barriers to SOFC: durability and cost. The project is examining only durability. It would be more applicable if the project team could quantify how its improvements in durability and efficiency will help in cost reduction.
- The project team does not seem very focused. The team is modeling two similar but different reforming approaches: CPOx and ATR. The team is also trying to develop catalysts for two very different fuels: diesel and biomass tar.
- The project is exploring nickel-free anodes for sulfur tolerance, and barrier layers for durability.
- It is not clear how proton-conducting ceramics fit the overall project objectives; such proton conductors operating at 400°-700°C would require a very significant effort to develop effective non-precious-metal cathode electrocatalysts. Also, a proton-conducting fuel cell would require a very different system configuration from

that of an oxide-ion-conducting fuel cell, since the product water would be produced on the cathode side rather than the anode side.

- Reaction modeling is being used to understand, and then inhibit, the formation of tar (high molecular weight compounds) during fuel reforming. Thermally stable supports and catalysts are being studied for activity and durability.
- System modeling is being used to identify opportunities for system performance improvements.
- Activities within subtasks seem appropriate. However, the project scope is likely too broad to significantly advance the state of the art. The project proposes to explore everything from fuel reforming and fuel cell catalysts (and reforming catalyst supports), to operating strategies, to system modeling, to model-predictive control of operating systems.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **2.5** based on accomplishments.

- The project team developed improved microstructures for LSCM anode materials and evaluated protonconducting electrolytes. The team modeled an ethylene kinetic model for APU conditions and showed underpredicted CH₄ and over-predicted C₄H₆. Better understanding was gained on how to prevent deposit formation through better upstream mixing. The team synthesized catalyst supports and new catalysts that resist deactivation at high temperature. The team developed a better understanding of inefficiencies in system using CPOX reforming, recuperator, SOFC APU. The team also developed a better understanding of thermal management – this is the best part of this project. The team performed single tubes response modeling using simplified predictive control model.
- The data indicates that the catalysts selected do not perform very well.
- Progress from last year is difficult to determine, since no baseline data was presented.
- This project is only about halfway through its scheduled term. Therefore, complete results are not in yet. Some of the desired materials structures have been obtained but the materials have not been tested for activity.
- Modeling of the reforming processes is yielding useful insights into the factors and parameters that promote (and inhibit) tar formation, such as unsaturated and aromatic precursors and imperfect mixing of reactants.
- Fuel cell and system modeling is also providing helpful insights into potential for system-level improvements and process control.
- The short time since the project started and the overly broad scope (lack of focus) have led to limited results to date.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated 2.8 for technology transfer and collaboration.

- The project team interacted with three companies: Protonex (provides data), Reaction Systems (provides catalysts), and CoorsTek (low-cost materials processing). However, there wasn't any strong collaborative research and it is not clear how results are transferred to industry that would do commercialization.
- There appears to be good collaboration between the team participants.
- In addition to several co-investigators at the Colorado School of Mines, the collaborators include an industrial SOFC developer, a catalyst developer, and a major ceramics supplier (for the proton-conducting materials development task?).
- Collaboration and coordination appears good at the project level, but it may be limited within subtasks.

<u>Question 5: Approach to and relevance of proposed future research</u>

This project was rated 2.5 for proposed future work.

• The project team plans to continue to evaluate next generation SOFC materials, including new anodes and new proton conducting electrolytes; expand reforming strategies and experimental evaluation of feed gases, such as toluene; and conduct catalyst aging and stability tests. The team will perform CFD modeling of systems and

propose optimal configurations of liquid fueled SOFCs. The team also plans to further evaluate and refine system control models.

- It is good that they intend to begin examining long-term durability.
- It is unclear whether the project team has a catalyst that is active enough to warrant long-term testing. The team may benefit from screening a few more candidates while performing long-term testing on the best so far.
- The project team needs to improve the heat exchangers.
- The project team has not demonstrated a suitable catalyst for diesel reforming, yet it intends to begin investigating catalysts for a more difficult fuel (biomass tars). The team needs to focus its effort. If this is for a diesel truck APU, then the effort should focus on diesel.
- The materials prepared in Task 1 will be characterized in terms of some of the performance characteristics. There is little or no indication, however, of how the performance will be improved.
- In Task 2, a combination of reaction modeling and experimental validation will be pursued to define the conditions necessary for fuel reforming without tar (or coke) formation.
- The catalysts' reforming activity will be characterized in Task 3, along with aging tests, but there was no discussion of how catalyst improvements will be achieved.
- In Tasks 4 and 5, efforts will continue to improve the performance and control models. The effectiveness of some of the approaches, such as high-fidelity CFD modeling or modeling for "robust operation," is not clear within the context of the overall tasks.
- The subtask plans are good, but at the overall project level, this effort is likely too unfocused to succeed.

Strengths and weaknesses

Strengths

- Good academic groups are present in most areas of this project.
- The project is well funded. The modeling work is interesting.
- The project is following a multi-prong approach to address different aspects of diesel and biofuels-based fuel cell systems.
- The project team is working with industrial companies that are developing ceramics, catalysts, fuel cells, and systems. Thus, any materials or process improvements achieved in the laboratory can be tested out in an operating platform.
- This is a multi-disciplinary team with appropriate test equipment and use.

Weaknesses

- This project is very spread out and difficult to coordinate. The project team has used a scatter approach that attempts to cover most aspects of fuel cell APU. Newer proton conducting materials will take a long time to bring to commercialization. There is no commercialization plan. There is no real plan for long-term testing.
- The project team lacks focus. The team should select one fuel and select one reformer technology [autothermal reforming (ATR) or catalytic partial oxidation (CPOX)].
- Amount of progress on catalyst work is unclear. The Protonex APU (600 W) seems low power for a truck APU where 1 5 kW would be more reasonable.
- One weakness appears to be the activity investigating proton-conducting electrolyte materials. As mentioned above, if such materials are developed successfully, very substantial additional developments will be needed in electrocatalysts, fuel processing, and system configurations. It would not be a simple transition from an oxide-ion conducting fuel cell system to a proton-conducting fuel cell system.
- Project scope is too broad to advance the state of the art.

- The project team should focus the project into one or two areas probably thermal modeling. More collaboration with industry and transfer of results are needed.
- If this is for a diesel truck APU, then the team should focus on diesel and not get distracted by examining a fuel that will never be used in the vehicle application. Biomass tars are cleaned up in the biomass gasification process so they do not foul the rest of the system. The team should focus its modeling efforts on a single reforming technology (ATR or CPOX). The team needs to consider larger systems for a truck APU since 600 W is too small.

- Finally, the Solid State Energy Conversion Alliance (SECA) program did a lot of work in this area. The project team should engage some of the prominent SECA participants to benefit from past work.
- The overall program appears to be structured well, with the exception, perhaps, of the subtask on developing proton-conducting ceramics for use as the electrolyte in ceramic-based fuel cells.
- The project team should focus down to one or two closely related tasks.

Project # FCP-06: Fuel Cell Research at the University of South Carolina

John W. Van Zee; University of South Carolina

Brief Summary of Project

The objectives of this project are to 1) develop novel materials (e.g. Nb-doped) for improved corrosion resistance and improved fuel cell components; 2) develop a fundamental understanding of performance loss introduced by fuel contaminants and durability loss fuel induced by contaminants; 3) develop a fundamental understanding of the degradation mechanisms of existing gaskets and the performance of improved materials; 4) develop a fundamental understanding of acid loss and acid transport mechanisms; and 5) predict performance and lifetime as a function of load cycle.



<u>**Ouestion 1: Relevance to overall DOE**</u> <u>objectives</u>

This project earned a score of 3.0 for its relevance to DOE objectives.

- This project is concerned with the durability of stack components, cost of catalysts, electrodes and seals, and hydrogen quality.
- Corrosion resistive catalyst support is important area to improve membrane electrode assembly (MEA)/catalyst durability.
- Non-platinum group metal (PGM) catalyst can be a transformational technology for fuel cell commercialization.
- The project is addressing issues of stable non-carbon supports for electrocatalysts, fuel impurity effects on fuel cells, seal durability, and water vapor adsorption/desorption in PBI-based high-temperature polymer electrolyte fuel cells.
- All of these tasks are important for fuel cell development, except that the task on hydrogen impurity effects applies primarily to transportation fuel cell systems, and the task on water sorption in polybenzimidazole (PBI)-based MEAs applies only to non-automotive systems.

Question 2: Approach to performing the research and development

This project was rated 2.8 on its approach.

- It appears that DOE selected four subprojects on durability of seals, development of non-carbon catalyst supports, H₂ impurity effects, and characterization of PBI-H₃PO₄ membranes.
- The project team needs to identify rationale for titania-based materials selection for catalyst support.
- The project team needs to more specifically identify what kind of modeling development could be pursued for H₂ quality.
- Generic characterization of seal materials is good.
- This project is investigating doped-titania as electrocatalyst supports as potential replacements for carbon.
- The impurity effects task has focused primarily on carbon monoxide (and more recently on CO plus ammonia), which has already been investigated at length by others.
- In the task on gasket/seal durability, the project is essentially characterizing the aging performance of commercial sealant materials, with the objective of predicting seal life, rather than developing improved sealant materials.
- For the PBI water and acid loss measurements, the project team has devised an experimental apparatus specifically suited for these measurements.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated **3.0** based on accomplishments.

- The project is wrapping up. The tasks on gaskets and seals and non-carbon supports have been completed. The project has collected and published data on the effects of CO and NH₃ in fuel hydrogen. The project has also developed techniques for the measurement of isotherms and rate constants for interaction of water vapor with PBI-H₃PO₄.
- More analysis is necessary for alternative catalyst support data and investigation of performance enhancement.
- In the task on carbon-free electrocatalyst supports, a 25atom% Nb content in titania is more than a typical "doping" amount. Also, 4% hydrogen peroxide production is greater than what fuel cell developers would like to see. The project team has observed good performance and, apparently, good stability (preliminary).
- In the task on fuel impurity effects on fuel cells, they have observed slow rates of poisoning by CO, leading to hysteresis in short-time measurements.
- In the task on water sorption in PBI membranes, the team has determined water vapor sorption and desorption rates as functions of temperature. The team has not, however, reported any electrochemical performance or membrane conductivity data.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **2.8** for technology transfer and collaboration.

- Each of the four subtasks had significant collaborations and partners.
- The project has extensive collaborations with industry and national laboratories.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.8 for proposed future work.

- The project is in the final stages. The plan is to continue to disseminate results through presentations and journal publications.
- For seal, focus on generic materials characterization for fuel cell seal, not seal concept, such as gasket.
- This project is scheduled to end by October 2009. As such, no additional work is planned in the task on dopedtitania-based electrocatalyst supports and in the task on seal durability.
- The project team will complete the work on CO and combined CO and ammonia poisoning.
- The project team will conduct additional work on water balance measurements in PBI membranes during transient operation of single cells.

Strengths and weaknesses

Strengths

- Qualified PI and team members.
- Extensive experience.
- Good lab facilities.
- Good collaborations.
- Controlled experimental capability for catalyst activities.
- The project has an extensive array of analytical and characterization tools available for the different tasks.

Weaknesses

- The project is a collection of four unrelated subtasks.
- The four major tasks are quite disparate and there appears to be little interaction between them (or among the different groups of researchers addressing the four tasks).
- There was no indication of how the results of this work are being, or will be, used by other researchers or fuel cell developers, except for the task on fuel impurity effects on fuel cell performance.

Specific recommendations and additions or deletions to the work scope

• The various tasks in this project are 90% to 100% complete. Therefore, it is difficult to make any meaningful recommendations for the remaining work yet to be completed.

Project # FCP-07: Development of Kilowatt-Scale Fuel Cell Technology

Steven S.C. Chuang; The University of Akron

Brief Summary of Project

The overall objective of this project is to develop a kilowatt-scale coal fuel cell technology. The results of this research and development efforts will provide the technological basis for developing megawatt-scale coal fuel cell technology. The objective for 2008 was to investigate the factors governing the anode catalyst activity for the electrochemical oxidation of carbon in coal. The objectives for 2009 were to 1) evaluate the long-term anode and cathode catalyst activity as well as interconnect durability; and 2) refine the coal injection and fly ash removal systems.



Question 1: Relevance to overall DOE objectives

This project earned a score of 1.5 for its relevance to DOE objectives.

- This doesn't appear to fit in with the rest of the Hydrogen Program. It appears to fit in with the Office of Fossil Energy (FE) since it uses coal.
- Properties focused on direct electrochemical oxidation of coal, with no hydrogen production or utilization involved. It represents an interesting and potential alternative to coal burning for electricity generation, but it is at its infancy. It does not fit into stationary or early market applications or transportation.
- It is hard to see the relevance of this project to the DOE EERE Fuel Cell Program. If such a coal-fueled system could be successfully developed, it would be more germane to the DOE FE effort. It would appear to apply to larger sized generators. Emissions of CO₂ will be less than from a coal-fired power plant due to increased efficiency, but will still be substantial.
- This project employs solid carbon as a fuel in a solid oxide fuel cell (SOFC) fuel cell, and as such, has little direct relevance to the bulk of the hydrogen fuel/proton exchange membrane (PEM) fuel cell centric DOE program. However, this should not be viewed as a negative, but rather as a positive aspect. This is a novel approach that has potential to be a useful technology and is worth further investigation. If the Hydrogen Program is to be re-focused to consider other fuels and approaches with the potential for greater impact in the near term, then this effort may very well be much more in line with next year's program objectives.

Question 2: Approach to performing the research and development

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

- This project uses a high-temperature fuel cell, with direct coal with a saturate H₂O anode feed, Anode catalyst is a large 3-5 micron particle size.
- This is an interesting approach, essentially examining the direct conversion of coal in a high-temperature fuel cell, however the likelihood of success is rather small, and there appears to be no actual data showing that the direct conversion of coal is achievable.
- The concept demonstration is in its infancy. Proof-of-concept still needs to be demonstrated. Many issues remain to be revealed let alone addressed, such as oxygen crossover and anode poisoning.

- The project is overly ambitious for the funding provided. The investigator is focusing efforts on finding stable anode materials in the coal dust environment. However, ash-free coal is planned for a large part of the testing. It is not clear how the fly ash removal system will be able to cope with the fly ash that is released in the reaction.
- At the current stage of development, the project approach is good. The PI is addressing appropriate issues (catalysis, materials development, cell fabrication).

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.5 based on accomplishments.

- 500 hours of durability have been demonstrated with hydrogen.
- The project team is currently using Ni anode and will add Cu + Ce in the future.
- A coal injection system has been designed.
- Durability data to date is questionable as it is not with coal and is not with the catalyst intended for eventual use.
- A large fraction of the funding has been spent already this does not seem to match the timeline or results produced. The major accomplishments appear to be to have identified two significant problem areas of anode poisoning and oxygen crossover, as expected for this crude fuel oxidation.
- For the limited funding and time since the beginning of the project, progress is reasonable. Anode stability over 500 hours has been demonstrated using hydrogen as fuel. A long-term test is planned with low-ash carbon as fuel. Fabrication of the coal injection system and fly ash removal systems is underway.
- The PI is making good progress towards understanding the implications of the unique aspects of this effort. For example, dealing with a solid fuel this eliminates the benefit of a porous, three dimensional electrode structure as is common with gaseous reactants. As a result, a thin, non-porous anode structure is preferred, and is being pursued. This in turn should allow for a lower operating temperature, which will enhance production of CO₂ relative to CO, and simplify the interconnect and sealing issues. The progress along these lines is promising. However, there is still considerable thought needed to address the fuel utilization/fly ash mitigation problems associated with this concept. The solution to these problems will likely not be as simple as having a vertically oriented anode as the PI believes.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated 1.5 for technology transfer and collaboration.

- The project team is working with the Ohio Coal Development Office and First Energy; however, it is not clear how the interactions with the other partners are contributing to the development of the system.
- There are no apparent partners with SOFC experience.
- Three collaborators are mentioned but their roles are not clear. The Ohio Coal Development Office is probably a funding agency and is not likely to bring technical expertise to the project.
- More contact with other groups that are perhaps more experienced in SOFC technology would be helpful. The overall progress of the effort would probably be faster. However, given the unique nature of the problem (oxidizing a solid fuel in an SOFC), it may not make a huge difference. Again, the bigger issue will be in regards to fuel handling for eventual stack designs, particularly how is the stack to be configured to allow for high fuel utilization, how is fuel to be fed into a stack, etc. There are significant engineering issues here that need to be addressed to move this concept out of the laboratory. At this point, its not clear to me if Chemstress (who as a partner on this project, should have some responsibility for these issues) is really a partner and will be intimately involved in the design of a stack, or if they are merely in place to build a stack eventually.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.5 for proposed future work.

- Anode catalyst formulation will be modified to minimize carbon formation on the nickel catalysts.
- Coal injection system has been designed; and will be implemented.
- It is not clear how the anode catalyst will be developed. The project team should pick one major problem and address it with the limited resources remaining, e.g., how to remove fly ash residue from the anode surface or

interface with the carbon fuel. This project seems doomed to failure if solid impurities like trace metals in the coal build up on the catalyst surface, preventing coal oxidation.

- The project team should try using ultra-pure carbon and if the time decay of the polarization curves on slide 9 are still seen, then there is an even more significant fundamental issue than contamination poisoning.
- The future work is laid out and addresses the most significant issues challenging the concept. The path to developing interconnects that cost 50% less than present interconnects is not well laid out. The proposed future work is ambitious for the amount of funding provided.
- The PI's plans for addressing near-term issues relating to catalysis, interconnects, and cell fabrication are logical. Longer-term issues relating to stack design and operating have not been adequately addressed as mentioned above. This is definitely an area where considering many alternate possibilities would be helpful and should be encouraged.

Strengths and weaknesses

Strengths

- Good funding partners.
- This is a novel approach using a readily available fuel: coal.

Weaknesses

- Direct conversion of coal has not yet been demonstrated.
- Progress to date is with hydrogen, which is already proven. There doesn't seem to be evidence that the direct coal conversion will work.
- There is a lack of partners with high-temperature fuel cell expertise. The PI should ask the SOFC community for help.
- Power density is low and there is not much discussion on how it will be increased.
- How to handle/deliver a solid fuel within a fuel cell stack?
- How to handle/exhaust solid waste (fly ash) produced within the stack?
- Solid fuel coupled with 2-D anode architecture suggests that the limiting current density achievable may be relatively low. Will very large area electrodes be required? If so, what is the ultimate power/energy density achievable with this approach?
- At what scale is this approach really feasible? Is it appropriate for kW size systems such as APUs? Or, is it really only suitable for stationary applications? My hunch would be that more conventional SOFC systems using natural gas or liquid fuels would be more widely applicable, but good engineering could prove otherwise.

Specific recommendations and additions or deletions to the work scope

• The project team should review the work on materials development in the DOE-FE SECA to determine relevance to this project.

Project # FCP-08: Center for Fundamental and Applied Research in Nanostructured and Lightweight Materials

Julie King; Michigan Technological University

Brief Summary of Project

This project involves fundamental and applied research in the development and testing of lightweight and nanostructured materials to be used in fuel cell applications and for energy storage. The research covers three areas, including: 1) heat and water management; 2) development of new electrode materials; and 3) enabling technologies for membrane synthesis.

Question 1: Relevance to overall DOE objectives

This project earned a score of **2.6** for its relevance to DOE objectives.

• The project has 3 different approaches



(and in some cases with multiple tasks under each approach). The approaches based on thermal and water transport, catalysis, and advanced membranes all have some relevance to fuel cell needs. Each of the approaches presented is limited in that they have not shown that they address the critical needs for any of these areas (e.g., conductivity (thermal or electrical) or cost data for injection molded bipolar plates; mass activity and durability for catalysis; and high-temperature/low relative humidity performance and durability for advanced membranes).

- Each of the project areas is relevant to DOE objectives.
- The program serves objectives A through E.
- It is a materials' program in which different materials are being developed. Not every material addresses all of the DOE targets yet, the whole program does.
- Each of the individual projects has relevance to DOE program goals; however, much of the actual effort seems to be materials of interest to the PIs, and not of direct relevance the DOE Hydrogen Program. The carbon foam/nickel hydroxide work is an example. Advances in carbon foam materials could certainly contribute to DOE Hydrogen Program goals, but there does not appear to be any reason for diluting this effort with a foray into battery materials. Similarly, the electrospinning work seems to be completely focused on spinning a polymer (PLLA) that is not of interest to the fuel cell community.
- Only part of this project appears to be relevant to DOE objectives. The part on "new electrodes" would be more appropriate for a battery or ultracapacitor project than a fuel cell effort.

Question 2: Approach to performing the research and development

This project was rated 1.8 on its approach.

- The study is disjointed and has little cohesion and no synergy amongst the various PIs. Even within a specific topic area there are widely different tasks such as injection molded flow fields and fundamental aspects of water transport under water and thermal transport. The study of electro-catalysts based on heterogeneous catalysis of ethylene hydrogenation on non-conducting supports is not an appropriate platform for investigation. Using polymers with poor durability like poly lactic acid will not allow useful materials to be developed.
- The approach for areas 1 and 3 is reasonably good. The area 2 approach is not as good. More attention should be focused on the electrochemical reactions that are of interest H₂ oxidation and O₂ reduction instead of examining ethylene hydrogenation. Also, depositing metals on non-conductive oxide supports is not relevant for fuel cell electrocatalysis.

- Research targets are on bipolar plates, thermal cyclability, and gas diffusion layer (GDL) improvement. Means to achieve this are: development of testing protocols, tailoring of a carbon foam as a support for fuel cell electrodes, development of nanostructured polymer membranes, and enabling of rapid production techniques.
- Three main areas are targeted: Heat and water management, new electrode materials, and technologies for membrane synthesis.
- The approach is well structured and fully addresses the barriers in an appropriate way. The membrane development seems to be very basic and should be observed yet, not questioned at this stage.
- The focus is not completely on fuel cells but also on batteries. Although synergies are appreciated, the program might be more focused.
- Bipolar plate work is adequate, although more attention could be paid to mechanical properties now that thermal and electrical property goals are in sight. Very little relevant progress on other topics, so it's difficult to assess the approach.
- This project is a collection of three independent research efforts that are unrelated to one another. Approaches within these individual "sub-projects" are not particularly new and do not assure progress towards the technical barriers. Only the development of improved bipolar plates and selected aspects of hybrid-membrane deposition offer certain promise.
- Significant fraction of electrode materials seems to be tailored for batteries and ultracapacitors instead of fuel cell electrodes. This approach does not promise good progress in a fuel cell project.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 1.8 based on accomplishments.

- To date, data generated by the project shows no progress towards addressing any objectives or barriers.
- Technical accomplishments are few, and some of the presented accomplishments do not demonstrate significant progress toward goals. Doping of fluorescent materials into electrospun polymer fibers has not been shown to constitute progress toward DOE objectives.
- Bipolar plates: the in-plane conductivity meets set targets, yet through-plane the conductivity is in the order of 1/3 to 1/5 of the target. Testing of thermal conductivity shall be finalized by December 2009. Respective optimization is not mentioned. As for the GDL, an environmental testing chamber was fabricated as the first contact angle measurements were carried out. A GDL compression device was fabricated and taken into operation; no major results were attained yet.
- Carbon material development for electrodes targets batteries, supercaps, and fuel cells. A production method was developed. It is irritating that properties mentioned in this review are not relevant to fuel cells. The specific advantages of the carbon over existing solutions were not clearly presented. The addition of nickel to the anode is not self-explanatory, nor has it been explained. This section might need some clarification.
- Catalysts with core-shell structures here called pseudomorphic overlayers are a development with high potential and first success has been made. Yet, again it seems necessary to keep it on a fuel cell track in a fuel cell program.
- The production technology for membranes focuses on fibers. It should be clarified how these fibers will eventually contribute to the formation of a gas tight membrane.
- Assessing the program, it needs to be taken into account that the program just recently started and very little funding was spent in FY08. For the time and money given, the results are very good and thus rated outstanding. For FY09, support should be given to keep the program on a fuel cell track.
- Only the composite bipolar plate effort can make any claim of real progress towards addressing a DOE goal/barrier. The rest are making small steps with materials that are not relevant.
- The absence of fuel cell performance data, electrochemical test results (e.g., for electrode materials), as well as of membrane conductivity and performance data, make evaluation of the technical progress in this project very difficult.
- Charge and discharge data, appropriate for batteries, are irrelevant for fuel cell electrode development. Also irrelevant are results pertaining to ethylene hydrogenation.
- The purpose of building catalytic layers on electronically non-conductive supports is difficult to justify. The reasons for measuring hydrogen adsorption or, for that matter, development of anode catalysts in general, and using precious metals in particular are also unclear.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **2.0** for technology transfer and collaboration.

- It is a single institution collaboration. Even within the project, no cohesion or synergies exist.
- Specific contributors to the program are developing the different materials. There might not be a strong interaction between the different partners yet, but this is not necessarily to be expected at this point in time. Dana Corp. provides technical input. RIT and GM were mentioned as well. There is a common DOE project with GM.
- In conclusion, the program is well tied to industrial experience and requirements.
- This is a collection of individual efforts that are not linked in any meaningful way. The individual PIs do not seem to have a good familiarity with prior work in several of the areas they are investigating.
- There appears to be no collaboration between various, distinctively different parts of this effort. The effort clearly lacks coordination.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.2 for proposed future work.

- Proposed future work is a continuation of work to date, which has shown no progress in overcoming barriers and has little relevance toward advancing the program.
- Future work is reasonable.
- Only the bipolar plate work is relevant and some thought has been given to future plans (collaboration with Dana Corp.).
- Similar to the development effort to date, proposed research is not particularly novel. Also, continued fragmentation of this project does not promise significant progress.

Strengths and weaknesses

Strengths

- For the most part, the project is relevant to DOE objectives.
- New ideas for materials' fabrication.

Weaknesses

- Approaches and results to date provide no value in advancing the field or addressing key issues in fuel cell technology. There is no synergy in the project.
- Some of the work being done seems unlikely to yield anything useful in terms of DOE objectives. Catalysts supported on non-conductive oxides are irrelevant to fuel cells.
- A strength and weakness is the classical approach of materials' scientists to develop materials for a range of applications. The project needs to keep its focus on fuel cells.
- There is no over-arching theme to bring individual elements together.
- This project has a very poor focus on DOE needs/goals.
- It is very hard to get a good read on a program when one PI is presenting the work of 6 PIs, but has essentially no familiarity with the details of the work of the other PIs.
- At best, these efforts duplicate work being performed elsewhere or previously.

- This project should be cut. Nothing of merit exists to build upon from the work presented.
- If project cannot be cancelled, insist that the effort be focused on materials relevant to fuel cells not batteries, not bio, and not catalysts supported on alumina.
- There seems to be little justification for maintaining funding for this project.

Project # FCP-09: Engineered Nanostructured MEA Technology for Low Temperature Fuel Cells *Yimin Zhu; Nanosys, Inc.*

Brief Summary of Project

The objective of this project is to develop a novel catalyst support technology based on unique engineered nanostructures for low temperature fuel cells that 1) achieves high catalyst activity and performance; 2) improves catalyst durability over current technologies; and 3) reduces catalyst cost. Nanosys, Inc. has developed novel inorganic, nano-technology support structures using functionalized silicon nanowires that 1) are highly durable, have high conductivity and large surface area; 2) have controlled porosity which improves catalyst utilization; 3) have improved Pt alloy catalyst particle dispersion on the support; and 4) maintain well controlled catalyst loading characteristics.



Question 1: Relevance to overall DOE objectives

This project earned a score of **2.8** for its relevance to DOE objectives.

- The project proposes to address the three most critical barriers for MEA components, but only focuses on one component of the membrane electrode assembly (MEA) the catalyst and support, and for direct methanol fuel cell (DMFC) small portable applications.
- This project is relevant to the DOE goal of reducing carbon corrosion and increasing catalyst stability. Replacing carbon black with another material or composite may achieve a more thermally and oxidatively stable catalyst support.
- This is nice work that addresses new support materials for fuel cell electrodes that avoids the use of carbon. This is very relevant for MEA durability. Some of the tests shown are not so relevant and need to be modified for open circuit voltage (OCV) and humidity cycling with hydrogen feed. Work is focused on DMFC, which is not relevant to DOE goals. Behavior with low relative humidity (RH) hydrogen would be more relevant.
- This project addresses the critical barrier of catalyst and catalyst/support durability. Testing has been for DMFC applications, while DOE Hydrogen Program has been focused on H₂ fuel and systems that have potential crossover between H₂ and DMFC.
- This project is relevant to the development of DMFCs for portable power applications. However, its focus is only marginally relevant to improving the performance of these devices.

Question 2: Approach to performing the research and development

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

- The project is based on the vapor liquid solid approach for generating nanostructured catalyst support fibers. This is an inherently slow and costly approach. The high-temperature SiC coating process may also add additional cost limitations.
- The project lacked detail on why it was expected that the particular Si/C/nitride support would be expected to be more stable than state-of-the-art materials.
- "New" support materials still contain carbon. Discussion on why oxidation/corrosion is expected to be less would be helpful.

- No standard C corrosion data was available. No electrochemical area (ECA) measurements were available. No end-of-life (EOL) microscopy was available.
- The project team has done good work making electrode supports, dispersing catalysts, etc. Tests are focused on DMFCs, which is not so relevant. Behavior with low RH hydrogen feed would be better. Also, the project assumes dimensions of the nanowires are optimized. Some transport modeling is needed to develop a model that could place the choice of dimensions on a sound footing.
- The approach leads to more controlled porosity and appears to provide better catalyst dispersion. Potential cycling tests cycle over a limited potential range at relatively low potential where corrosion is not expected to be an issue. Potential hold test is at a low voltage, again where limited corrosion is expected. DOE suggested protocols involve cycling between 0.6 and 1.0 V for catalyst durability studies, and a potential hold test at 1.2 V for catalyst support durability.
- This project seeks to improve the stability and utilization of the anode catalyst in direct methanol fuel cells by replacing the amorphous carbon support or unsupported PtRu catalyst with PtRu deposited on carbon-coated SiC nanowires. The main issue with the stability of DMFC anodes is not support-related but is the stability of the catalyst composition itself. This project does not address this issue.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.6 based on accomplishments.

- The project is complete and some milestones were achieved. However, the project team did not overcome the DOE barriers targeted.
- Si/C nitride nanowires production technology was previously known.
- Formation of nanoparticles technology was previously known.
- The project successfully married the technologies and produced Si/C nitride wires with catalyst.
- The testing plan was very lacking and could have used some more fundamental testing.
- The accomplishments are impressive for the work performed, but some important durability studies are absent including OCV and humidity cycling with hydrogen feed. There is too much DMFC work here for the program goals.
- Results suggest some advantages in catalyst dispersion, increased surface area, porosity, and durability. Durability testing has been performed under relatively mild conditions; so testing under DOE protocols to see if the durability advantage remains is of interest.
- The project has accomplished quite a bit in a short amount of time. However, it is not clear how this project addresses the overall barriers because it focuses on replacing the anode support for DMFCs where the support stability is not the biggest issue with the anode. Utilization may be improved with this approach, but the overpotential for this reaction is extremely high leading to low cell power densities. This is the main issue with the cathode catalyst and not its utilization. This approach would have been more interesting and have made a bigger impact if it were pursued for the cathode catalyst.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **1.6** for technology transfer and collaboration.

- This project appears to have been done very quickly with earmarked funding from the company's congressional members. There has been little interaction with other partners for the technology advancement.
- No collaborations were noted.
- Collaborations with others have been non-existent. The project team needs to collaborate with some electrochemical modelers to determine what the best dimensions of the support structure should be and then vary the dimensions accordingly.
- Collaborators were not listed. No collaborations were apparent.
- No apparent collaborations, with the exception of acquiring materials from component suppliers.

<u>Question 5: Approach to and relevance of proposed future research</u>

This project was rated **2.3** for proposed future work.

- Not applicable because the work is completed.
- The project team discussed testing in H₂ fuel cells. This should have been part of original project plan.
- The project is at an end. Plans for the future were not given.
- The project is complete. No future work has been planned.
- Water management with this catalyst layer would be of interest. Does the large pore size help or hurt water management?

Strengths and weaknesses

Strengths

- This project is one of very few that directly targets the replacement of "classical" carbon supports with a different material. The use of Si/C nitride materials may well prove much more thermally and oxidatively stable.
- Nanowire structure capability is shown and the ability to disperse conventional catalysts is demonstrated.
- The project team used a novel approach for catalyst support development. The project team has shown the ability to synthesize very small and monodispersed PtRu nanoparticles.

Weaknesses

- The project plan for testing is suspect. DOE goals are primarily for hydrogen proton exchange membranes (PEM). H₂ fuel cell testing should have been carried out. Classical carbon corrosion and ECA experiments should have been done. In-depth microscopy at beginning-of-life (BOL) and EOL, thermal stability, and oxidation testing should have been done.
- The project team assumed nanostructures are the best dimensions. There is a lack of electrochemical modeling and expertise to determine whether the right dimensions are achieved. Demonstrating ability to change dimensions at will for various applications would be good.
- Durability was measured under mild conditions.
- The project team is not addressing the main issue for DMFC anodes.

- The project team should add a electrochemical modeler and expertise. The project team should collaborate.
- The project team should perform durability tests under DOE suggested protocols.

Project # FCP-10: Alternative Fuel Cell Membranes for Energy Independence

K.A. Mauritz and R.F Storey; University of Southern Mississippi

Brief Summary of Project

The objectives for this project are to 1) synthesize stable aromatic hydrocarbon polymers containing acid ion-exchange groups tethered to the backbone via fluorinated side chains; 2) synthesize benchmark aromatic hydrocarbon membranes for properties comparisons and refinement of synthetic methods; 3) establish Nafion® benchmark data to which novel hydrocarbon membrane data can be compared on an all-other-things being equal basis; and 4) establish hydrocarbon membrane structure-property, proton conductivity characterizations and fuel cell performance data as relating to the Hydrogen Program proton conductivity technical achievement milestones.



Question 1: Relevance to overall DOE objectives

This project earned a score of **3.2** for its relevance to DOE objectives.

- The project targets the synthesis of new membrane materials for operation at low relative humidity (RH) and high temperature for H₂ proton exchange membranes (PEM), which are main goals for the DOE PEMFC development plan. The project team has a good plan for membrane design, synthesis, and in-depth testing.
- This program supports the DOE R&D objectives.
- This project is relevant as it looks at preparing new materials and looking at behavior under degradative stress. These techniques are valuable and relevant. The syntheses are also interesting. However, there is no rationale given for how these membranes will achieve the conductivity goals under low RH, and it is not at all clear how these materials that depend on the presence of water will be any better than Nafion®.
- The project team should address barriers identified in DOE Multi-Year RD&D Plan (found at http://www1.eere.energy.gov/hydrogenandfuelcells/mypp/): durability, cost, performance, water transport in the stack, thermal and water management, air management, or start-up/shut-down time, and energy.
- Determining baseline properties of Nafion® for comparison to other membranes does not really address these barriers. Others have pursued preparation of sulfonated poly(arylene ether sulfone) (sPAES) systems. Phosphonic acid derivatives and are of interest, but it is not clear what equivalent weight materials are being prepared. The project team needs to work at low equivalent weight/high ion exchange capacity.
- Development of hydrocarbon membranes with improved functionality is relevant to DOE program goals.

Question 2: Approach to performing the research and development

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

• The approach includes the synthesis of modified diphenol-type monomers to be used as comonomers in sulfone-ether type copolymers. The target modified monomers include tethered acid groups that may provide more mobility and free-volume in the material and enhance proton conduction. The initial target of using phosphonic acid (or a,a'-difluorophosphonates) may not be the best approach given that the pKa of these groups is much greater than that of sulfonic acid groups. Researchers discussed the use of sulfonate or a,a'-difluorosulfonate acid groups, which may be a better alternative.

- Collaboration with a world-class membrane testing group will prove most useful once viable membranes are produced. The testing group is currently running baseline experiments with commercially available materials.
- The project team has a good approach using materials that should have good stability.
- The degradation work and application of the dielectric relaxation methods are very useful. The development of the methods is tested on Nafion® and the syntheses methods are sound. The work promises to produce valuable results of use to others in the program.
- The strategy for addressing the technical barriers is not clear. Determining baseline properties of Nafion® and determining how it degrades may be useful, but unless you have strategies to address its failures, the work does not lead to advances. The strategy for addressing mechanical failure of Nafion® and low conductivity at high temperature/low RH is not apparent.
- DOE membrane targets are not identified, nor seemingly used to guide development. This seems to be more of a university learning activity at this point, rather than a high-functioning R&D effort with experienced investigators. Plans are okay, but the reviewer needs to see progress to make a judgment.

<u>Ouestion 3: Technical accomplishments and progress toward project and DOE goals</u></u>

This project was rated 2.4 based on accomplishments.

- The project is currently in early stages and consists mainly of monomer and polymer synthesis. Synthesis of new materials can be quite time consuming, so it is understandable that viable membranes are not yet in hand.
- The a,a'-difluorophosphonate diols may be interesting, however, it is recommended that the group move toward sulfonate and a,a'-difluorosulfonates.
- Successfully carrying out sulfone-ether condensation copolymerizations can be very tricky, particularly in ensuring that monomer ratios are correct and that polymerizations are completely dehydrated. It is suggested that the project team consult with J. McGrath at Virginia Tech.
- It is too early in the program to evaluate accomplishments.
- Progress is good although not clear regarding the polymer synthesis. Testing of methods is satisfactory and adequate.
- Membrane development work has not made membranes to test yet. Characterization work has observed features in dielectric loss spectroscopy with ageing, but it is not clear exactly what the features are due to.
- Progress to date is limited to some backbone synthesis and to characterizing a Nafion® membrane electrode assembly (MEA) as a benchmark (with nothing new apparent). Backbone has not been functionalized. Necessary equipment has been procured but is not yet operable.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **2.0** for technology transfer and collaboration.

- Good teaming of synthesis and characterization expertise. Partners are within the same university. Outside collaboration is suggested to strengthen project.
- An industrial partner would help in the evaluation of these new materials.
- Not much collaboration is evident. There should be more interaction with others in the program.
- Some collaborations are in place.
- The only apparent collaboration is unfunded activity (scope not defined) with the Illinois Institute of Technology. There is no mention of DOE membrane working group collaboration or coordination.

Question 5: Approach to and relevance of proposed future research

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

- The future work plan is well defined. Synthesis of new monomers will lead to work on copolymerization and membrane production. Membrane testing by dielectric spectroscopy and various microscopy techniques are planned.
- A good plan was presented.
- Future plans do not directly address DOE milestones.

- Proposed future work will move membrane development along. The project team needs to concentrate on low equivalent weight materials.
- The project team's plans seem adequate, but progress needs to be demonstrated.

Strengths and weaknesses

Strengths

- The teaming of synthesis and characterization expertise is strong. The materials approach accounts for the dual goal of low RH conductivity and temperature stability. Multiple synthetic targets have been suggested to reduce risk.
- Synthesis of new and interesting materials although incremental from those at Virginia Tech.
- Dielectric relaxation and mechanical measurements of degradation behavior are valuable.
- The characterization technique is novel.
- Polymer science and property characterization.

Weaknesses

- Moving from new monomer synthesis through membrane production can be very time consuming. Strict project planning must continue to ensure materials are produced with sufficient time to carry out the characterization.
- The project team has used of phosphonate functionality as proton conductors (less-acidic, higher pKa than sulfonate).
- This project does not address DOE milestones. No attempt has been made to understand proton transport mechanisms.
- The strategy for addressing barriers (durability and performance) is not clear.
- The lack of progress would indicate that the project did not have the capability to perform the work at the time of project award.

- The project team should focus on sulfonic acids.
- The project team should suggest ways to increase sulfonic acid content (lower EW).
- The project team should suggest ways to mechanically stabilize membranes (cross-linking, support material).
- An industrial partner would help in the evaluation of these new materials.
- The project team should address low RH conductivity.
- This project should be evaluated next year to see if meaningful progress is made.

Project # FCP-11: Extended Durability Testing of an External Fuel Processor for SOFC

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

Brief Summary of Project

The overall objectives for this project are to 1) conduct long-term tests in a relevant environment for the external fuel processor for a 1 MWe solid oxide fuel cell generator; 2) determine long-term performance of critical components including catalysts, sorbents, heat exchangers, and control valves; 3) evaluate the impact of ambient temperatures (hot and cold environments) on performance and component reliability; 4) determine system response and performance of process controls for transient operation; and 5) identify any long-term failure mechanisms.



Question 1: Relevance to overall DOE objectives

This project earned a score of 2.4 for its relevance to DOE objectives.

- Durability studies are always of value; however, this study is looks at components two of which are only to be used during start-up operations. This system is not designed for frequent start-ups.
- This project examines durability and performance of a solid oxide fuel cell (SOFC) fuel reforming system for combined heat and power (CHP).
- This project provides demonstration of SOFCs (1 MW) using natural gas as fuel for stationary power. It provides demonstration of fuel processing subsystem durability and performance information. It also plans to provide demonstration of all season operation.
- 1 MW is the scale that Fossil Energy is looking at, so the principle (long-term testing) is applicable to the Fuel Cells Program. The scale is a bit large.
- The objective of this project is to test an external natural gas fuel processor for a 1 MWe solid oxide fuel cell power generator. This appears to be of low relevance to the overall hydrogen and fuel cell program for the following reasons:
 - o No megawatt-scale SOFC systems have been developed and demonstrated yet.
 - It is not clear what the application of interest would be.
 - It is not clear why water-free fuel processing is needed for stationary distributed power plants of this scale.
 - o It is not clear how this scale SOFC system fits in with the other activities in the Hydrogen Program.

Question 2: Approach to performing the research and development

This project was rated **2.2** on its approach.

- The study uses the full-scale components.
- It is not clear how degradation that is not visually detectable will be identified, or what (if any) post-test characterizations are planned.
- Durability and performance including start-up/shut-down are measured.
- The system intends to use pipeline natural gas.
- This system uses a very complex method to start-up the CHP unit, including O₂/N₂ separation, a syn-gas subsystem, and a start-gas subsystem. It also includes electric heaters for preheating and standby.
- The project team demonstrated multiple start-ups (10) and 1200 hours for the syn-gas subsystem.

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- The project team demonstrated 24 start-ups and 200 hrs for the start-gas subsystem.
- The project uses an already developed Rolls Royce SOFC, 1 MW unit built up from four 250 kW blocks. A catalytic partial oxidation (CPOX) is used to start up and system uses internal reforming to run using pipeline natural gas.
- This is a full-scale demonstration.
- If this unit was intended for a distributed residential area, then transient, load following behavior would also be of interest.
- The described approach was to test two different fuel processors for reforming natural gas, an internal one to generate the fuel gas for the SOFC (without an actual SOFC in the system to be tested), and an external one used essentially for start-up of the internal one. It was not explained why the external (start gas) fuel processor needed to be located outdoors.
- There was no discussion of a test plan, of the data to be obtained, or how those data would be used. The only metric given was the number of hours of operation and the number of start-ups and shut-downs.

<u>Ouestion 3: Technical accomplishments and progress toward project and DOE goals</u></u>

This project was rated 2.4 based on accomplishments.

- The project has just started.
- Durability testing has not yet been started and appears to be behind schedule.
- The project just started so there are few results.
- The project began only a few months ago, and hardware has been fabricated and installed.
- This project started very recently (in January 2009). The early months have been spent in building the indoor test rig and enclosure and in preparing specifications for the external test facility.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **1.8** for technology transfer and collaboration.

- Collaboration with an educational institution will help train students. The project team should use a metric to quantify student involvement.
- There has been minimal collaboration up to this point.
- Eventual collaboration is intended with the Ohio Department of Development and Stark State College of Technology.
- There have been no obvious collaborations.
- The partners seem to be coordinated and doing well. It is too early in the project to determine how close the collaboration is.
- No technical collaboration was identified with any organization other than Rolls-Royce Fuel Cell Systems.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.8 for proposed future work.

- The main activity is testing the system for time on-stream and cycling.
- Plans for post-test studies are not well defined.
- The proposed future work appears as a serial methodology: testing the syn gas system, testing the start-up gas system, and then testing durability.
- At end of the project, the team will have 8000 hours of durability testing of the system, including desulfurizer. The project team will have winter/summer operation data. The team wants to do parallel operation with multiple fuel cell blocks. This will require a distributed air and fuel system and this could provide useful systems data.
- Subject to the weaknesses in the approach mentioned above, the proposed future work is to complete the project as planned.

Strengths and weaknesses

Strengths

- The project addresses durability of components.
- The project team has incorporated an academic institution, which will help train faculty and students.
- The project team uses an already developed system to test and get durability data. This project provides a good demonstration of SOFC running on pipeline natural gas.
- This is a large-scale demonstration project that will provide important data for industry on this technology. By operating outside, weather impacts will be evaluated. By operating year-round, temperature extremes and changes in natural gas composition will be evaluated.
- The personnel involved have an extensive background in fuel processing and they understand the issues and challenges involved.

<u>Weaknesses</u>

- Post-test studies are not clearly communicated.
- The team is relying on electric heaters for standby and preheating.
- This project employs a very complex system, with a standard reforming unit, a syn-gas subsystem, a start-gas subsystem and O₂/N₂ separation membrane.
- Durability testing has not yet been started.
- The project team has no commercialization plan.
- The project team is focusing on natural gas. It would be interesting to examine biogas in addition.
- The lack of technical objectives in the test plan is a significant weakness.
- The project needs to identify what maximum rate and extent of performance degradation would still qualify as a successful durability test.

- The project team should define a metric to gauge faculty/student participation.
- The project team should include an analysis on what it would need to do in order to work on biogas.
- The project is very similar to Fossil Energy's SECA and FutureGen projects and would benefit from increased interaction with those programs perhaps by adding a partner from the SECA program.
- Since the project is so similar to SECA, it may want to be transferred to Fossil Energy.
- The project team should develop a technical test plan with quantitative performance targets over time.
- The project team should identify the classes of applications for which these megawatt-scale SOFC systems are targeted.

Project # FCP-12: Hydrogen Fuel Cell Development in Columbia (SC)

Kenneth Reifsnider; University of South Carolina

Brief Summary of Project

The general objective of this project is to contribute to the goals and objectives of the Fuel Cell element of the Hydrogen, Fuel Cells and Infrastructure Technologies Program of the Department of Energy by enhancing and supplementing the fuel cell research and development efforts at the University of South Carolina. The project research activities focus on the following technical objectives: 1) the development of metal-free oxygen reduction catalysts to reduce cost, facilitate manufacturing, and enhance durability of fuel cells; and 2) the development of redox stable mixed ionic and electronic conductors for bi-electrode supported cell symmetrical solid oxide fuel cell designs, to reduce cost by simplifying



manufacturing, enhance durability, and greatly reduce sensitivity to thermal cycling.

Question 1: Relevance to overall DOE objectives

This project earned a score of 2.8 for its relevance to DOE objectives.

- The project focuses on fuel cells.
- There is a very diverse portfolio of projects presented in this one poster. All of the projects are relevant to DOE's objectives in different applications of fuel cells. The impurity project would become more relevant if it were to focus on materials not already studied by other laboratories.
- Each of the project areas is relevant to DOE objectives.
- This project is an unfocused, broad and seemingly unconnected effort to "support" other funded work at the University of South Carolina. The shotgun approach is unlikely to yield relevant results to the DOE program.

Question 2: Approach to performing the research and development

This project was rated 2.8 on its approach.

- The scope is very broad and should be focused. The goals should be more specific. Milestones and project outcomes need to be more specific.
- The solid oxide fuel cell (SOFC) work should collaborate with the Solid State Energy Conversion Alliance (SECA) and leverage what has been done in this area.
- In general, the approaches pursued in these diverse projects do contribute to overcoming the barriers to commercialization of polymer electrolyte fuel cells (PEFC) and SOFCs to varying degrees. The SOFC projects appear to be at an early stage, but are following relevant approaches. The impurity project should focus on other impurities not already covered in DOE's portfolio of impurity projects.
- The approach is good overall, although insufficient information was presented for each of the project areas to fully demonstrate a good approach for attacking the various technical barriers.
- The approach to individual identified tasks is okay; but the unfocused, broad and seemingly unconnected effort is unlikely to result in significant advances toward DOE goals and technology targets.

Ouestion 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 2.4 based on accomplishments.

- The project has just begun, so it is hard to judge.
- The non-platinum electrocatalyst project is generally effective, but makes claims regarding optimization of pyrolysis temperature that are not substantiated by the data. Is catalyst activity optimized at pyrolysis temperature of 1000°C? Or would even higher temperatures yield higher activities?
- Ammonia and sulfur dioxide contaminants have been studied extensively by LANL. With so many potential contaminants and given the urgency of determining the effect of these contaminants, it would be best for the University of South Carolina (USC) to examine contaminants that have not been or are not being studied by other organizations.
- Various results have been presented but it is difficult to gauge to what extent they represent significant progress during the project period. For instance, presenting the demonstration of a role of N content in C-based catalysts in determining activity doesn't seem like significant technical progress. Others have previously shown this, and anyway, the relationship is very complex.
- I was unable to assess what portion of the progress presented was accomplished under the funding from the program. The poster and presenter comments would indicate much of this work was authorized and funded elsewhere.

<u>Ouestion 4: Technology transfer/collaborations with industry, universities and other laboratories</u>

This project was rated **2.0** for technology transfer and collaboration.

- The partners seem limited to those located in South Carolina.
- A list of collaborators was given, but it was not clear what these collaborators contributed to in the presented work.
- I was unable to assess what portion of the collaborations presented was resulted from funding from the program. The poster and presenter comments indicate much of this work was authorized and funded elsewhere.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.4 for proposed future work.

- The seals work does not seem very ambitious.
- The project team needs to identify the anticipated outcomes and benefits of its work. The modeling looks useful, but benefits from other areas of the work are not as clear.
- The presentation was lacking in proposed future work. Only a few broad statements were presented.
- Few plans for future work presented. Future work is in general relevant and reasonable, but decision points and alternate development pathways were not mentioned.
- To the extent the future plans presented are to be accomplished under existing funding for this program the proposed work is good. The scope is too broad, however, for meaningful advances under this funding in this time period. It is unclear what portion of the plans presented is to be accomplished with funding from the program.

Strengths and weaknesses

Strengths

- The project team is leveraging from many past projects.
- The project areas are generally quite relevant to DOE objectives and the approach seems good.
- Scientific breadth and approach to individual tasks.

Weaknesses

- The scope is too broad.
- The project is poorly designed and unlikely to significantly advance the technology toward DOE goals and targets by itself.

- Focusing of scope would help this project make meaningful progress.
- The project team should include more partners or at least interactions with institutions located outside of South Carolina.
- The SOFC work should engage the SECA program to leverage the SECA knowledge.
- The project team should consider drastic scope reduction and improve focus and deliverables. Otherwise, this project should be terminated.

Project # FCP-13: Martin County Hydrogen Fuel Cell Development

Jeffrey Bonner-Stewart; Microcell Corporation

Brief Summary of Project

The objective of this project is to transfer a microfiber fuel cell technology's manufacturing process from a research and development level to a manufacturing environment and evaluate various parameters including speed and product quality. Unicell throughput was increased due to enhanced systems and programming without affecting Unicell performance.

<u>Ouestion 1: Relevance to overall DOE</u> <u>objectives</u>

This project earned a score of **1.8** for its relevance to DOE objectives.

- The manufacturing technology under development seems applicable only to relatively low total power "stacks."
- Cost was not mentioned.
- Objectives are not tied to any fuel cell performance metrics. The person presenting the poster couldn't provide details.
- The project involves scaling up an R&D level technology to a mass-produced product. In principle, this is relevant to DOE objectives.
- The subject of this project may have some relevance to DOE objectives; however, the lack of technical content in the presented material makes assessment of the effort practically impossible.

Question 2: Approach to performing the research and development

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

- Scaling up of the manufacturing process is important, but there is not enough information on how scaleup/optimization was achieved.
- Insufficient information is given about the intended applications and system packaging issues for those applications.
- The presenter did not have sufficient knowledge of the technical aspects of the project and of the cell construction.
- My only take away from the poster presentation and discussion with the presenter was that this project uses an alternative approach for fabricating fuel cells and stacks. No information was available on any performance requirements or important performance metrics.
- The approach is good in terms of addressing technical barriers to scale up.
- While extrusion of a complete MEA in a single nanofiber appears attractive, this presentation gives no information on whether the approach has been successful to date and does not allow one to judge prospects for overcoming various barriers that this project is targeting.

Question 3: Technical accomplishments and progress toward project and DOE goals

This project was rated 1.5 based on accomplishments.

- The goals of the project were met although the general applicability is not evident.
- Throughput was maintained with no adverse effect on quality.


- I'm not sure how to assess or evaluate the impact of the accomplishment, because it is not compared against any metrics.
- The lack of technical performance data (other than power per "unicell" of unspecified size) renders technical evaluation of this project's progress impossible.

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **1.5** for technology transfer and collaboration.

- There appear to be no technical partners. Input from catalyst and membrane developer/manufacturer would have been beneficial.
- The technical collaboration that took place with North Carolina State University (NCSU) is unclear.
- The roles of two partner organizations in this project, Martin County Economic Development and NCSU, are not explained. All work appears to be have been carried out by Microcell.

Question 5: Approach to and relevance of proposed future research

This project was rated 1.8 for proposed future work.

- This program ended in March 2009. There is no DOE-funded future work planned.
- Proposed future work needs to include measurable performance metrics.
- The future plans appear reasonable on the surface, but the limited information on the poster and the limited technical knowledge of the presenter leave some questions unanswered.
- Relevance of proposed future work cannot be evaluated in the absence of technical content in this year's presentation.

Strengths and weaknesses

Strengths

- The project is making reasonable progress towards the goal of scaling up an R&D level technology to a manufacturing environment.
- Neither project strengths nor weaknesses can be determined in the absence of any substantial technical information.

Weaknesses

- It was nearly impossible to assess the status of the technology readiness level or the objective of this project from the context of a fuel cell power generator.
- Further work on refining the technology on the R&D level should be done before any efforts at mass-production are made.

Specific recommendations and additions or deletions to the work scope

- This whole project needs to be thought through and its justification needs to be reevaluated.
- The funding of this project should not be continued.

Project # FCP-14: Fuel Cell Balance of Plant Reliability Testbed

Vern Sproat; Stark State College of Technology

Brief Summary of Project

The objectives of this project are to 1) develop test beds to address the challenge to the fuel cell industry of the durability and reliability of components that comprise the complete system (balance of plant); 2) develop the test plan to address the candidate balance of plant components and basic test bed design for long term operation; 3) use collaborations with component manufacturers to develop and enhance final product performance; 4) develop statistical models for extremely small sample sizes while incorporating manufacturer validation data for future evaluation of candidate components; and 5) use the test beds to enhance the education of the technical workforce trained in proton exchange membrane fuel cell system technology.



Question 1: Relevance to overall DOE objectives

This project earned a score of 2.3 for its relevance to DOE objectives.

- This project seeks to generate a reliability database for proton exchange membrane fuel cell (PEMFC) balanceof-plant (BOP) components. It is also relevant to producing a workforce trained in fuel cell technology.
- Published (PEM) fuel cell system data indicate that non-stack components account for about 67% of unplanned system shut-downs/failures. Better understanding of these failures is needed.
- It has been difficult, both in direct discussions and reviewing the presentation, to ascertain what the primary goals of the project are and how the project team intends to meet these goals. The fuel cell BOP can be improved, and educating a potential future workforce for related studies has value; unfortunately, based on the data and approach presented, it is unlikely this project will be effective at addressing either of these areas.

Question 2: Approach to performing the research and development

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

- The project has taken the approach of assembling test beds to generate the reliability database, working with a manufacturer to design meaningful tests, and developing statistical models for small sample sizes.
- The BOP components to be evaluated are not listed so it is difficult to fully assess the approach. If the project includes only fittings, tubing, transducers, and gauges, then its usefulness is diminished.
- It is hard to imagine that these test beds would be better or provide more information than the BOP component developer/supplier's in-house testing.
- The barriers and approach are completely unclear. Three test beds have been presented without insight into what they focus on and how the application of them and data obtained will lead to improved understanding or advances. The project represents incompletely formulated ideas without substance. The project seems to focus on test bed design and fabrication without any guidance presented to what this means and how it will be implemented.

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<u>Question 3: Technical accomplishments and progress toward project and DOE goals</u></u>

This project was rated 2.0 based on accomplishments.

- The project is just getting started. Turnover in project management has apparently impeded progress.
- Progress is reasonable for the recent start date of August 2008 and low funding to date.
- To date, no progress has been made (except perhaps that a hydrogen safety plan is now in place).

Question 4: Technology transfer/collaborations with industry, universities and other laboratories

This project was rated **1.3** for technology transfer and collaboration.

- The project team is working with Lockheed Martin to design three test beds, one of which will be located at the partner's site.
- This project desperately needs participation of BOP component developers and system users such as compressor/blower suppliers, humidifier manufacturers, and auto original equipment manufacturers (OEM) to help define operating conditions including cycles.
- Lockheed is a partner and few other institutions are also mentioned, however, their roles aren't clear. At this point no progress has been reported so it is not possible to comment on this aspect of the project.

Question 5: Approach to and relevance of proposed future research

This project was rated 2.0 for proposed future work.

- It appears that the plans for future work are still being refined.
- The future work follows logically.
- Plans have no coherence and it is uncertain what studies will be undertaken and what systems will be designed. From the materials presented, it is extremely unlikely the project will advance the program or address barriers.

Strengths and weaknesses

Strengths

- Collaboration with Lockheed Martin.
- Availability of the student body.

Weaknesses

- The PI is still gaining familiarity with the fuel cell technology, the state of the art, and technology needs.
- This project lacks direction and a clear plan for making any impact.

Specific recommendations and additions or deletions to the work scope

- DOE needs to redirect this program by closely aligning it with the fuel cell system analysis and cost projects that DOE is sponsoring.
- The project needs better identification of the anode-side BOP components and the needs for performance and reliability data.
- The project should be cut. There is nothing of merit to build on from the work presented.