2018 – Hydrogen Fuel R&D
Summary of Annual Merit Review of the Hydrogen Fuel R&D Sub-Program

Summary of Hydrogen Fuel R&D Sub-Program and Reviewer Comments:

The Hydrogen Fuel R&D sub-program comprises early-stage research and development (R&D) to reduce the cost and improve the reliability of technologies used to produce, deliver, and store hydrogen from diverse domestic energy resources. The sub-program is divided into two categories: (1) Hydrogen Storage R&D and (2) Hydrogen Production and Delivery R&D. The latter includes seedling projects under the Hydrogen Generation Consortium (HydroGEN), which is part of the U.S. Department of Energy (DOE) Energy Materials Network (EMN).

In fiscal year (FY) 2018, production projects focused primarily on early-stage R&D for advanced water-splitting materials and systems funded through HydroGEN. Production pathways under investigation included advanced high- and low-temperature electrochemical water splitting, and direct solar thermochemical (STCH) and photoelectrochemical (PEC) water splitting. In FY 2018, delivery projects focused on liquefaction technology; materials compatibility research for infrastructure applications, such as pipelines; research on compression technologies, such as linear motor reciprocating compressors, metal hydride compression, electrochemical compression, and coatings for compressor seals; and hydrogen dispensing technologies, such as wireless communication, meters, and hoses. Hydrogen storage projects in FY 2018 focused on materials-based hydrogen storage R&D through the Hydrogen Materials—Advanced Research Consortium (HyMARC). HyMARC is an EMN consortium comprising a core national laboratory team and individual seedling projects that benefit from access to the core team's capabilities. The sub-program continued early-stage R&D in advanced tanks through development of precursor fibers for low-cost carbon fiber. All projects under the Hydrogen Fuel R&D sub-program continued to be evaluated with respect to their potential to meet DOE's cost and performance targets for the near and long terms.

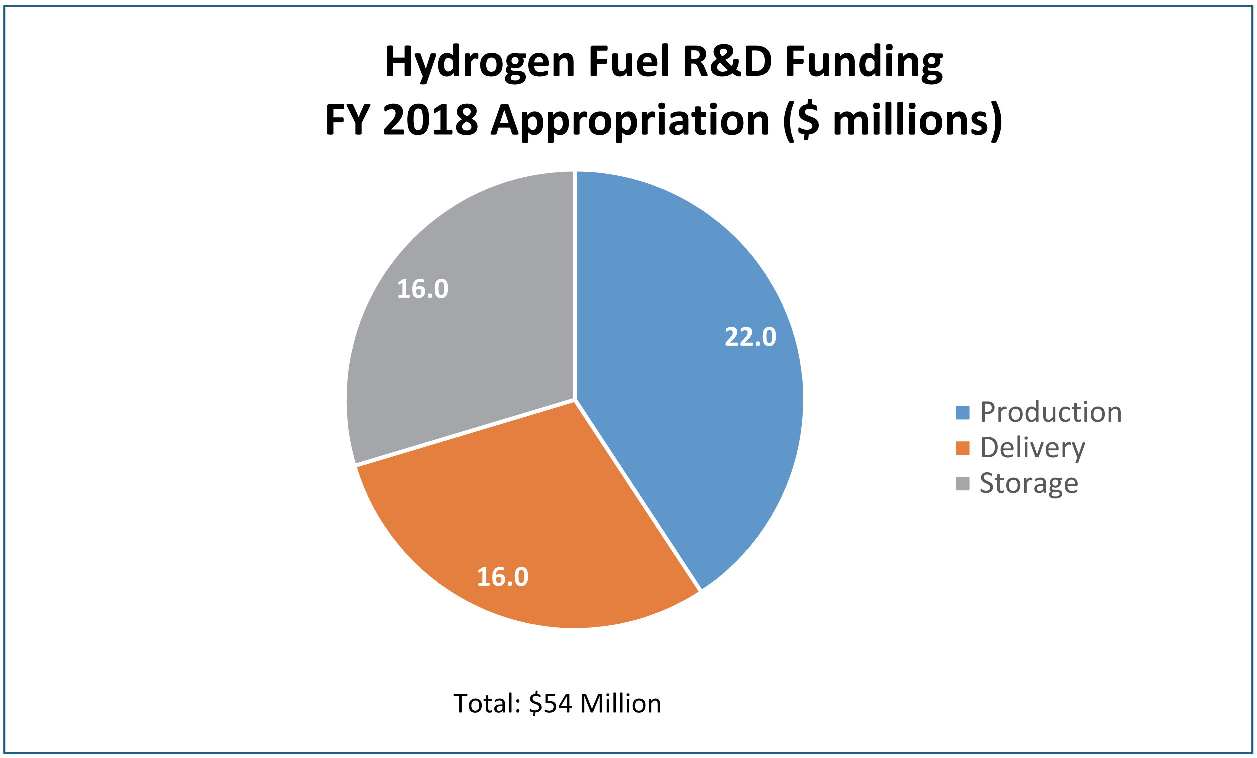
For the hydrogen production projects in the sub-program, reviewers were impressed with the effective collaboration of the HydroGEN Consortium and its cogent use of networking and productivity enhancement tools such as the shared data hub. The seedling water-splitting projects funded in conjunction with HydroGEN were praised for their strong interaction and successful leveraging of national laboratory capabilities through the HydroGEN framework. Reviewers were also impressed with the use of modeling and technoeconomic analysis efforts to guide experimental work toward meeting DOE cost targets. Reviewers commented that they would like to see increased interaction between the projects themselves as they move forward. Among hydrogen delivery projects, research on pipeline materials compatibility to enable large-scale infrastructure was particularly commended. Projects with research on electrochemical and/or metal hydride compression were praised for their potential to achieve higher reliability than conventional mechanical concepts, but the project teams were encouraged to consider the economic viability of these approaches. Projects on hydrogen dispensing were encouraged to collaborate with relevant industry stakeholders and ensure that technology prototype development is informed by early-stage materials research (e.g., materials compatibility evaluation and modeling of hose prototype performance). Fueling technologies for heavy-duty vehicles were recommended as an area for future research.

Within the hydrogen storage portfolio, reviewers noted an adequate balance of resources, priorities, and technical goals. Given the new focus on the H2@Scale efforts within the Hydrogen and Fuel Cells Program (the Program) as a whole, reviewers encouraged closer integration of early-stage R&D efforts, including hydrogen storage R&D, with H2@Scale infrastructure activities to maximize R&D impact and increase the likelihood of success for the overall Program. Reviewers commended the sub-program's management and openness to engagement, communication, and collaboration with stakeholders to ensure R&D work remains valuable to industry and relevant to the hydrogen and fuel cell market. HyMARC continued to be regarded as a key endeavor to leverage foundational scientific understanding and world-class resources and facilities across multiple institutions. The consortium was also recognized as a catalyst for groundbreaking advances in hydrogen storage materials with the potential to meet the sub-program's ultimate goals. As more HyMARC seedling projects are selected, reviewers encouraged continued and careful coordination across the HyMARC portfolio to prevent overlap in activities and maximize results.

Hydrogen Fuel R&D Funding:

The FY 2018 appropriation for the Hydrogen Fuel R&D sub-program totaled $54 million. Of these appropriations, $22 million was allocated for hydrogen production research, $16 million for hydrogen delivery research, and $16 million for hydrogen storage research, as shown in the figure below. Projects funded in the hydrogen production R&D portfolio are expected to accelerate materials development for advanced water-splitting technologies toward meeting DOE targets, and this emphasis is expected to continue into FY 2019. Nineteen hydrogen production projects were reviewed, with overall favorable scores ranging from 2.9 to 3.7, with 3.4 as the average score. Funding in hydrogen delivery focused on hydrogen pre-cooling technologies (e.g., cryocoolers), compression technologies, liquefaction technologies, and launch of the H-Mat consortium focused on materials compatibility research. Fourteen projects were reviewed, with a minimum score of 2.8, a maximum score of 3.6, and an average score of 3.3. In FY 2019, H-Mat is expected to remain a priority area of research. The hydrogen storage R&D portfolio was represented by twelve oral presentations and eighteen posters (including three Small Business Innovation Research [SBIR] projects) in FY 2018. Out of the twelve projects reviewed, nine focused on materials development, two on analysis, and one on engineering. In general, the reviewers' scores for the projects were good, with scores of 3.6, 3.1, and 3.4 for the highest, lowest, and average scores, respectively.

Each of the following project reports contains a project summary, the project's overall score and average scores for each question, and the project-level reviewer comments.

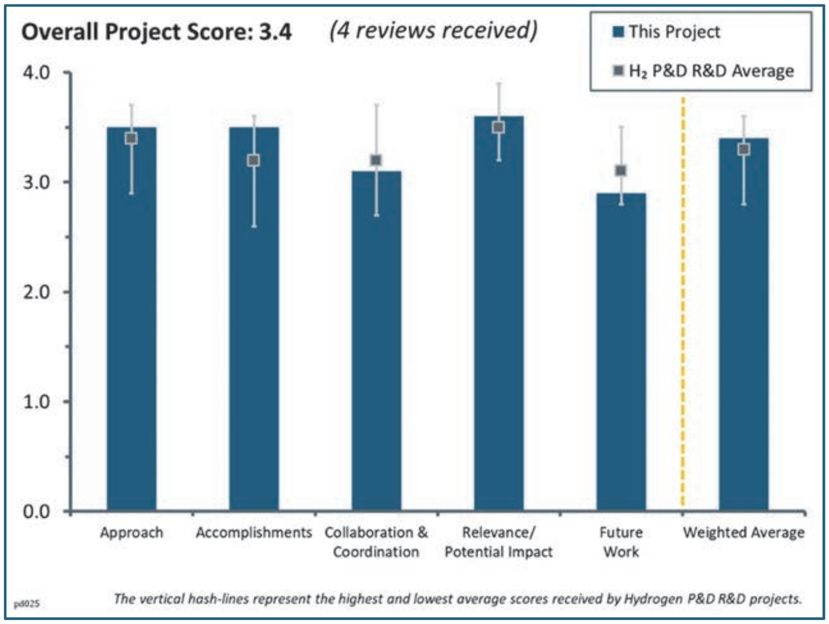


Project #PD-025: Fatigue Performance of High-Strength Pipeline Steels and Their Welds in Hydrogen Gas Service

Joe Ronevich; Sandia National Laboratories

Brief Summary of Project:

The primary objective of this project is to evaluate the potential for modern, high-strength steels to facilitate reductions in the cost of hydrogen pipelines. Specific goals are to (1) characterize fatigue performance of high-strength girth welds in the presence of hydrogen gas and compare performance to that of low-strength pipe welds, and (2) establish models that predict pipeline behavior as a function of microstructure in hydrogen to inform future development.



Question 1: Approach to performing the work

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

- The experimental evaluation of the resistance of high-strength pipeline steel welds to fatigue crack growth is necessary for cost reduction. Similarly, an understanding of how this resistance relates to materials microstructure will lead to the development of steels with improved resistance. Consideration of the residual stress contribution shows a careful approach to the project objectives. The phenomenological model used to ascertain fatigue crack growth behavior needs further evaluation because it is not predictive.
- The goals of the work were clearly identified, and the project has a rigorous approach to achieve these goals. The lone consideration here is the concern that the fatigue crack growth rate (FCGR) testing is focused on the weld material; a further justification of the "weakest" link plane along the weld would provide more confidence that this approach is capturing the most relevant properties for failure.
- The project's approach is clear, logical, and detailed. The identification of actionable results and how to adapt those results into commercial practices would be helpful.
- The presenter provided a clear procedure for starting and completing the project.

Question 2: Accomplishments and progress

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

- The results could clearly lead to achieving DOE hydrogen delivery cost goals. For example, the finding that "using X100 (instead of X52) can result in 42% cost reduction for 24' pipe operated at 110 bar (1600 psi)" seems significant.
- Relevant data has been collected, important experiments linking microstructure to properties have been performed, and logical modeling approaches have been put forth. In all, the project has made strong progress toward the stated goals.
- All experimental results of the project were well presented. In particular, the result shown on slide 8 in which the behavior of the X100 steel is shown to be the same as that for low-strength steels is impressive. There is a problem with the result shown on slide 13; the numerically calculated stress ahead of the crack tip is not compatible with small-scale yielding behavior, and as such, it cannot scale with the stress

intensity factor associated with the compact tension specimen. Hence, the numerical results presented are not transferrable because of the absence of similarity. The comment on the slide regarding infinite and finite domain is irrelevant. It is not clear why the synchrotron studies shown on slide 14 are necessary. Evaluating elastic strains in a fracture specimen is just an elastic calculation.
- The materials are suited for hydrogen storage, not for transport.

Question 3: Collaboration and coordination

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

- Strong research community collaboration was shown. However, industry and company-specific engagement was not presented.
- The collaboration with the University of California, Davis, on residual stress assessment is important.
- The collaborations within this project are adequate.
- This project has strong collaboration with other team members, particularly with the National Institute of Standards and Technology and University of Arizona. It is not clear that the results from the collaboration with Oak Ridge National Laboratory have yielded any insights, considering the lack of relevance of the elastic field results at scales that are relatively large, compared to the relevant plasticity-driven damage mechanisms in the micron-to-submicron size scale relevant to the hydrogen embrittlement damage process. Also, while there is scientific interest in understanding/modeling the role of microstructure in the hydrogen embrittlement damage process for these steels, it is important to ensure that the modeling is focused on a relevant question. The data (particularly the results on slides 8 and 9) show that the growth rates are generally agnostic to the microstructure (this is consistent with literature); as such, it is necessary to better justify the relevance of microstructure-scale modeling for this project.

Question 4: Relevance/potential impact

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

- The use of existing infrastructure (such as existing steel pipelines) for hydrogen transport/storage will be critical in the integration of this technology. Furthermore, the efficiencies gained by the use of higher-strength steels would be critical; such work is relevant to the qualifications of such ideas.
- This research is central to achieving DOE hydrogen delivery cost goals.
- The experimental component of the project is extremely important and impactful. It is difficult to assess the impact of the modeling work, as it relies on a phenomenological fatigue crack growth model.

Question 5: Proposed future work

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

- The proposed future work to develop test protocols to measure fracture toughness of welds and heat-affected zones and to broaden this project to influence other steels used in hydrogen infrastructure is valid and important. The project team could consider developing a commercialization roadmap.
- The proposed future work has good potential for the storage of gaseous hydrogen.
- Further correlation between grain size/FCGR would be interesting. In particular, identifying the relevant grain structure feature, as well as how/why this does/does not scale across different steel microstructures would be interesting. While the orientation-dependent trends observed on slide 10 are convincing, it is unclear why similar trends are not observed on slide 9. The additional Gleeble specimens will be useful in identifying this. The calibration of the diffusivity measurements is tricky. As always, it is hard to understand the transfer function between bulk diffusivity metrics and diffusion through a highly dislocated structure at the crack tip. The project team's publishing goals are good.
- The proposed work on slide 16 is all descriptive, without a single explanation. The presentation did not clarify how single-grain response could be related to the FCGR (da/dN) versus the change in stress

intensity factor (dK) curves for welds. In addition, the presentation did not clarify how the hydrogen effect would be treated in the modeling of the single-crystal response.

Project strengths:

- This is a relevant topic with a good approach and good progress, led by a team with the appropriate expertise to address issues. This is a unique and rigorous experimental effort. It incorporates residual stress into the driving force calculations. The work also consists of unique correlations of microstructure and FCGR behavior; there is a strong link between the scientific work and a useful engineering output via informing the governing specification.
- Strengths include:
  - Remarkable experimental findings
  - Actionable results for hydrogen pipe materials that can be expanded to other materials
  - A plan for peer-reviewed publication of results
- This project's strength lies in the experimental evaluation of fatigue crack growth of high-strength welds.

Project weaknesses:

- There are no material weaknesses.
- There is tenuous relevance of the diffraction work on the technical topic and a lack of consistency between the effects of grain boundaries for the different orientations, as well as a lack of impact of the boundaries for the Gleeble material. Better justification for the microstructure-scale modeling is necessary when the data shows that the FCGR behavior is agnostic to the microstructure. There is work to be done here, but the concern should really be about why there are similar growth rates despite changes in strength (thus the nature of the local crack tip stress gradient) and diffusivities between the different materials that would lead to different crack tip stresses and H-profiles. It would be good to know whether the model can predict this, as well as predict instances where this may not be the case. Technical concerns include the following:
  - The project needs to establish whether it is the weld metal or the heat-affected zone (or another part of the weld) that will actually fail if the team were to run a dog-bone sample in this orientation. This leads to the question of the rigor of putting the notch in the weld metal, rather than at another part along the weld. This will be even more important in the next phase of the work if fracture toughness is evaluated.
  - As one incorporates the residual stress into the dK calculation, the maximum thermal performance (R–value) will inherently change. However, the magnitude of R-value change is not stated. It is unclear if the R-value could be pushed low enough to the point where it becomes relevant (owing to closure of other R-dependent mechanisms).
  - It is unclear why the grain-dependent behavior is seen for the orientation but not for the Gleeble. The exact meaning of high-angle grain boundary (HAGB) is not clarified. It is also unclear whether these are prior austenite grains.
  - Recent work on strain gradient plasticity suggests that the local stresses that were calculated in figure 13 may be too low and too diffused. These calculations are qualitatively consistent with the abundance of crack wake transmission electron microscopy (TEM) analysis that shows a highly localized dislocation structure focused within roughly one micron of the crack wake. It is important to consider/refute this as the current models are interpreted.
  - The assertion that the growth rates outrun the diffusivity was not clearly articulated.
  - It was not clear how the project team monitored the crack length during the work on the Gleeble sample. If this was done via compliance, then it is unclear if there is any concern about inhomogeneity within the bulk compliance as the crack progresses because of the gradient in the microstructure. The sensitivity of the results to such an effect is unknown.
  - There are many issues with the superposition model reported in figure 11. This is a criticism not of the project but rather of the relevance of having such a model in a standard.
  - The devil is in the details on slide 16. The incorporation of the environmental–mechanical aspects of the loading into the X-face is not explained.
- The modeling and simulation within this project is very general and not targeted to the specific characteristics of the weld microstructures.

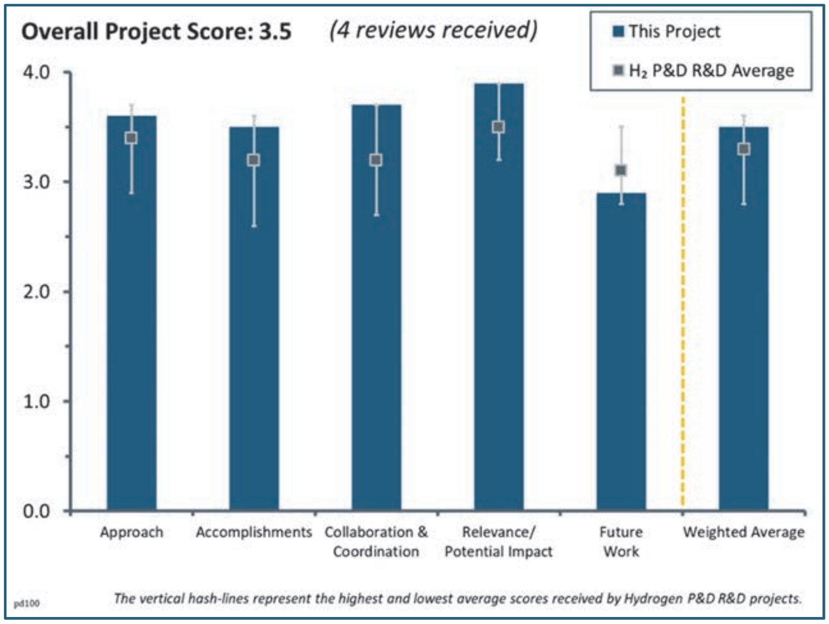
- This project's approach is not practical for the transportation of high-pressure hydrogen because of the excess weight of thick steel wall pressure vessels.

Recommendations for additions/deletions to project scope:

- Understanding weld microstructures experimentally is a very important objective and needs to be pursued. The synchrotron studies are irrelevant to the project and have not revealed anything related to fatigue crack growth.
- One important addition to the project scope could be the development of a field demonstration to validate cost savings and performance. This could help drive the adoption of improved practices based on the research results.
- The modeling effort should be focused to better align with the observed data trends. The diffraction work should be either eliminated or repurposed.
- It is recommended that the team look for other materials, such as carbon composite.

Project #PD-100: 700 bar Hydrogen Dispenser Hose Reliability Improvement

Kevin Harrison; National Renewable Energy Laboratory



Brief Summary of Project:

The objective of this project is to characterize and improve 700 bar refueling hose reliability under fueling conditions expected at heavily utilized hydrogen fueling stations. The National Renewable Energy Laboratory designed a test system that subjects refueling hose assemblies to pressure, temperature, mechanical, and time stresses. The high-cycling test reveals the compounding impacts of high-volume 700 bar fuel cell electric vehicle (FCEV) refueling, which has yet to be experienced in today's low-volume market.

Question 1: Approach to performing the work

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

- The project team identified an issue of clear importance to hydrogen infrastructure, especially in early days of rollout, in that dispenser hose failure will have an impact on not only station economics but reliability and therefore customer experience. The team has adopted a good approach to investigating the problem by using both accelerated testing and post-mortem analysis.
- The project is very relevant and directly applicable to industry needs. Hoses are a common failure point and a high-cost maintenance item, owing to frequency. More importantly, public safety is directly affected. The capability this project provides is appreciated; Shell intends to leverage this project.
- The testing facility and protocols are outstanding.
- Using a six-axis robot that emulates human motion when interacting with the hose, the duration of fills, and the pressure profile did real interactions justice. However, it would be desirable to address different ambient temperatures, as it is to be expected that temperature shocks are more severe in different environments. Therefore, the impact of temperature shocks could have been discerned more precisely.

Question 2: Accomplishments and progress

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

- Good failure mode analysis was provided, helping to inform new product development to solve reliability issues and reduce cost.
- The project has covered all objectives: testing and detailed identification of failure.
- The progress that has been made on the specific hose tested is significant. It remains a little unclear what was responsible for the surface blistering (i.e., the plastic failure) and what polymers might solve the problem. For future progress, it would be nice to include a hose other than the one from SpirStar to be able to draw inferences from different hose designs (e.g., different wire braidings or steel overwraps).

- The project should significantly increase the number of post-mortem analyses in order to assess type and frequency of failure modes. If these hoses have indeed been failing at high rates, there should be plenty to examine.

Question 3: Collaboration and coordination

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

- Other research institutes working on the problem from another angle are part of the cooperation. The issue of integrating another hose supplier has been addressed. Especially remarkable is the cooperation with the International Organization for Standardization's Technical Committee 197, Working Group 22 (ISO/TC 197/WG 22).
- There is excellent collaboration with industry and suppliers to test the most relevant products and share operational experience.
- All collaborations shown on the relevant slide are outstanding, with each one serving a specific purpose.
- For the size of the project, there is an impressive list of collaborators making high-quality contributions. Expanding the list of hose providers is suggested.

Question 4: Relevance/potential impact

This project was rated **3.9** for its relevance to/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- Frequent hose failures will give hydrogen refueling stations a bad reputation. If this project is successful in developing a strategy to greatly increase hose reliability, the project will have a meaningful impact on the uptake of FCEVs by avoiding the development of negative interactions, and it will have done so at a modest cost. The potential impact-to-cost ratio is high.
- The project is already making measurable impact in practice. Shell has a focus on changing hose suppliers and possibly helping to develop a new product with suppliers. This project and the test facility are a large part of assisting suppliers to develop less expensive and more reliable solutions.
- There can be no doubt about the relevance, as hose failures are an issue for every hydrogen fueling station operator. Apart from nozzle failures, hose failures are the most common reason for hydrogen fueling station downtime, which undermines consumer trust and buy-in. A remedy to this problem, therefore, is badly needed.
- Having identified the way failure appears and advances, the project can identify mitigation strategies.

Question 5: Proposed future work

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

- The proposed activities in the future cover the most vital challenges, namely using a hose from a different manufacturer and using dynamic mechanical analysis (DMA) rheology. However, it would be desirable to also address different plastic composites in more detail, if that is possible. This may be an issue, as manufacturers may not be inclined to share such information, but it may be possible to ask them to provide different prototypes. In the future, different ambient temperatures in which the robot works should also be addressed in order to learn more about thermal shocks in very cold (as in winter in the Northeast) and hot (as in summer in California) conditions.
- The listed potential upgrades are worth pursuing, provided that the reasons for failure have been identified and analyzed.
- The project should be more ambitious in its plan for testing the polymeric materials used in hoses. Increasing focus on DMA at cold conditions is commendable, particularly if it will involve temperature cycling. However, the team should consider how it could combine temperature cycling with hydrogen pressure—if not in the DMA, because of technical or safety challenges, then in a separate test rig where a DMA sample could be exposed to temperature and hydrogen pressure cycling, then subsequently

tested in the DMA or other mechanical property testing for failure. The project team should consider also adding a notch or other controlled flaw to simulate the stress concentration from the steel wire overlap.
- Future steps were unclear.

Project strengths:

- This project is a good use of DOE funding to address a critical issue for hydrogen delivery. The research plan incorporates accelerated testing and post-mortem analyses. The focus was on understanding the root cause of failures. There was an appropriate focus on properties of construction materials and the influence of cycle conditions, especially thermal shock.
- This project is highly relevant. The testing design is excellent and innovative, emulating real-world conditions very well. The choice of partners/cooperators was outstanding.
- This project was well executed with good collaboration and is relevant for cost-reduction goals.
- The project has a systematic and well-thought-out experimental approach to testing.

Project weaknesses:

- There were hardly any weaknesses.
- There were not enough post-mortem analyses to date to really understand the range and frequency of failure modes. The materials property testing protocols need to be more creative to better expose those materials to temperature and hydrogen pressure cycles that can encourage growth of cracks and holes due to stress/strain.
- There was a lack of understanding of failure initiation and evolution.
- This project should be communicated widely to industry; not many know about it.

Recommendations for additions/deletions to project scope:

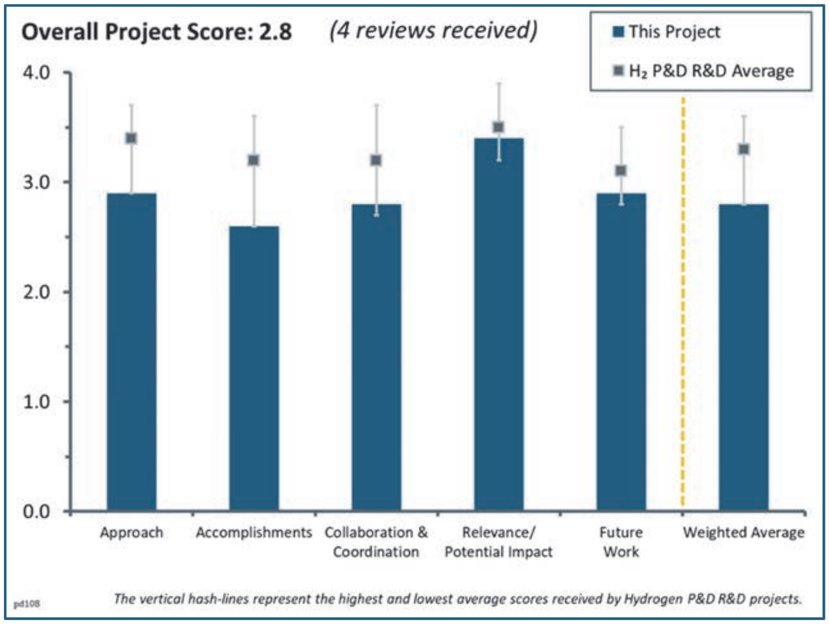
- The project should collaborate more closely with experts on hydrogen interactions with reinforced polymers. Observed failures need to be understood so that mitigation strategies can be suggested. A solid mechanics researcher with expertise in viscoplastic deformation and failure analysis should be added to the team.
- Japanese hoses should be included in the testing and post-mortem analysis plans, particularly if they use different materials.
- A focus on crimping and the effect this has on reliability would be an improvement.
- Increased variation in crucial parameters, which would provide the ability to draw inferences, would be good.

Project #PD-108: Hydrogen Compression Application of the Linear Motor Reciprocating Compressor

Eugene Broerman; Southwest Research Institute

Brief Summary of Project:

The objectives of this project are to (1) improve isentropic efficiency of high-pressure hydrogen compression above 95% by minimizing aerodynamic losses, (2) reduce capital costs to half those of conventional reciprocating compressors by minimizing part count, and (3) reduce required maintenance by simplifying the compressor design to eliminate common wear items.



Question 1: Approach to performing the work

This project was rated **2.9** for identifying and addressing barriers, project design, feasibility, and integration with other efforts.

- The project is focused on compressor efficiency and cost, which are major challenges in hydrogen infrastructure.
- High-efficiency lower-cost hydrogen compressors are needed to reach hydrogen refueling cost goals.
- The approach needs to be more clearly defined. Listing milestones from three years ago does not help reviewers determine the approach to this year’s work. The milestone list provides reviewers with an explanation of high-level tasks that have been completed, but the list is not clear about what was done this year versus past years. After looking up last year’s U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program Annual Merit Review (AMR) presentation, it looks like all that needed to be completed this year was the test stand construction, commission, and bench-scale performance testing. It is unclear why these three tasks are not the focus of this presentation. It also is not clear why building the first low-pressure stage of the hydrogen compressor is the appropriate approach to building this three-stage compressor. The integration of the multiple stages along with the pressure requirements of the medium- and high-pressure stages is not trivial. The strategy to start with the low-pressure stage individually may be correct, but the project lead has not indicated why this is the case.
- The project takes a technical approach to device development.

Question 2: Accomplishments and progress

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

- The accomplishments are fine, given the goals and approach of the project.
- Improvements in speed, design, and sealing were implemented, but significant improvements in efficiency have not yet been made.
- A clear picture needs to be presented of this year’s accomplishments, important discoveries, and innovations. Work from years ago and simple maintenance tasks should not be highlighted. There is a need to focus on progress around isentropic efficiency, motor control issues, and the completion of next steps.

The presentation was a disappointing highlight of what might otherwise be good work. Raw notes for each slide are provided for the project team to improve the presentation for next year.

- Slide 6 from AMR 2016 is not relevant to current progress.
- Slide 7 from AMR 2016 is not relevant to current progress.
- On slide 8, it is unclear why these components failed; perhaps they were sized improperly. It is uncertain what is the root cause of this failure was and whether there were lessons learned.
- Slide 9 is not an accomplishment—the content is neither interesting nor impressive.
- On slide 10, motor control would be essential to operation. The approach to remove the linear variable differential transformer (LVDT) and replace it with electromagnetic sensors seems reasonable, but unfortunately, it did not work. Solving this problem is essential to future success.
- On slide 11, it seems failures would be expected if the researchers knew they did not have full control of the motor. Given the inaccuracies in motor control, the graph provided looks okay. The number of measurements taken is unclear, and it is uncertain whether this is just n = 1. Multiple iterations could help smooth the data.
- Slide 12 states, "Commissioning issues sufficiently resolved," but how and when are not explained. It is uncertain whether motor control is still an issue. The approach of designing the first test to be able to make changes and pack later is good.
- The results on slide 13 look good and have finally hit one of the main components of the project, which is a high isentropic efficiency. More details are needed on how the 80%–90% isentropic efficiency is calculated and how many tests were performed to get to this efficiency. The compression ratio will need to increase for the other stages.
- Backup slides 30 and 31 indicate that LVDT and motor control are still an issue and that they have not been solved. It is not stated whether the data provided are only from short-duration tests that ensure damage to the compressor does not happen.

- Accomplishments over the previous year seem modest. The flow rate has not met the 10 kg/hour ±10% target, but the efficiency appears as though it will meet the target (>73%). Much like the responses to last year's reviewers' comments, concerns remain over the efficiency. It appears as though there is significant work remaining regarding (re)design, purchases, controls, and testing. It is not clear whether the remaining budget is sufficient for the remaining efforts.

Question 3: Collaboration and coordination

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

- The new partner, Libertine, is expected to help solve significant remaining feedback and control challenges.
- The collaborations shown on slide 15 seem to be okay.
- The project team has done an okay job of utilizing their collaborations. The team needs to focus on leveraging Libertine to achieve high efficiency moving forward. A project partner that can help solve the motor control issue is needed.
- The collaborations with part and component suppliers and partners are very good, but the project lacks collaboration with potential users of the linear compressor.

Question 4: Relevance/potential impact

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

- Lower-cost, low-maintenance hydrogen compression is essential to offering hydrogen to fuel cell vehicles at a competitive price.
- If this project is successful, the compressor could help reduce the burden of compressor reliability that exists today.
- The project team discussed how they would get their three-stage design down to 2.54 kWh/kg, which is approaching the Multi-Year Research, Development, and Demonstration Plan target of 1.6 kWh/kg. The team needs to leverage Libertine to achieve this. The original design and concept is there, but the progress

is lacking. A high-efficiency hydrogen compressor that could get up to 875–900 bar would have a big impact on the industry.

- The project is relevant, but it is not certain that it will lead to viable technology for real-world hydrogen compression.

Question 5: Proposed future work

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

- It appears the project team understands the challenges ahead and is set to take them on. The detailed challenges and barriers section was appreciated.
- The proposed future work is in line with the approach.
- It would be nice to quantify this particular motor's energy consumption (as indicated on the Proposed Future Work slide) in comparison with the 1.6 kWh/kg and include the starting and ending pressure.
- The provision of more detailed steps toward achieving the project targets is desired.

Project strengths:

- The unique and novel design of this compressor is intriguing. It is focused on compressor costs, which are a burden on the deployment of hydrogen systems.
- The project compressor has a simpler design and higher efficiency compared to conventional reciprocating compressors.
- The project goals are in line with industry needs, and accomplishments around the isentropic efficiency look promising.
- The design proposed in this project can lead to real-world technology.

Project weaknesses:

- The safety and safe operation of the test rig is a concern. Slide 9 shows a leaky pressure safety or relief valve found with a Snoop liquid leak detector on a (relatively) low-pressure nitrogen fitting. It is great that the leaks were found, but the label pressure safety valve (PSV) 100 (product specification to 500 psi) does not match the ink-pen-written setpoint of 1360 psi on the side of the valve body. The purple wrap on the PSV seems to confirm the ink-pen setpoint is within the range of the Swagelok PSV spring set, so the label needs to be changed to read PSV 130. This seems petty, but for an owner/operator of hydrogen systems at pressures approaching 13,000 psig, attention to detail is critical, and documentation (even labels) helps reduce the chance of accidents.
- Material failures in the presence of hydrogen (e.g., valves and seals) are likely to occur, but the project does not address those failures.
- Durability testing is not addressed.
- The project team should focus on what is important to the project and reporting to industry. If slides from previous AMRs are used, they should be marked accordingly.

Recommendations for additions/deletions to project scope:

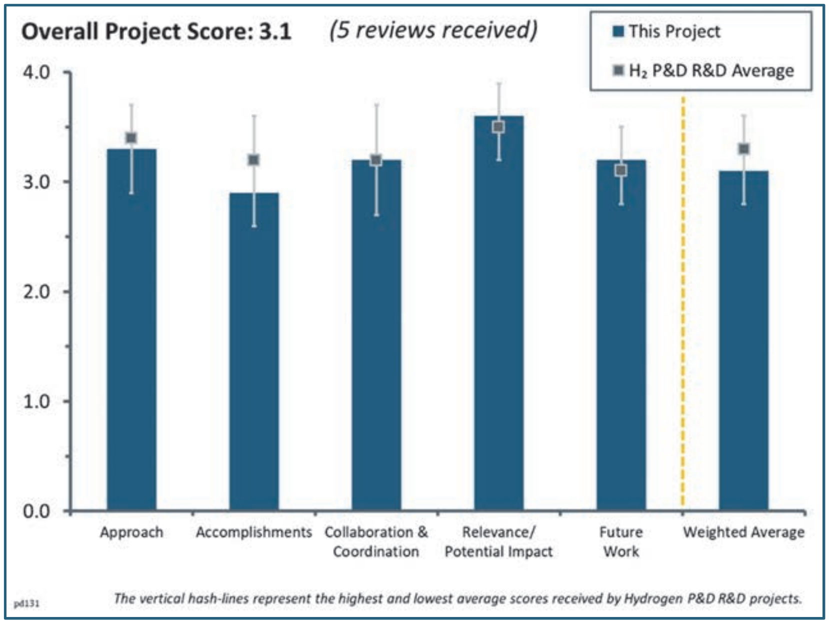
- The project team should focus on getting Stage 1 dialed in and what the plan will be to move to Stages 2 and 3 of the compressor. Issues around high pressure and how that will change compressor design should also be addressed.
- Durability testing must be added to validate component improvements.
- It is recommended the project team have an understanding of potential component failures and mitigation strategies.

Project #PD-131: Magnetocaloric Hydrogen Liquefaction

Jamie Holladay; Pacific Northwest National Laboratory

Brief Summary of Project:

The Pacific Northwest National Laboratory (PNNL) magnetocaloric hydrogen liquefaction system is expected to be considerably more energy-efficient than the Claude cycle. At 30 tons per day, the latter shows 40% efficiency, while the former is projected to be 70%–80% efficient. In this project, investigators will demonstrate the PNNL system liquefying ~25 kg/day. At industrial scales, the concept is expected to have a figure of merit (FOM) >0.5 (as compared to the Claude cycle system's FOM of <0.3). The project will also identify a pathway to a larger-scale system with an installed capital cost of less than $70 million.



Question 1: Approach to performing the work

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

- The project will develop a magnetocaloric process for liquefying hydrogen. These materials will couple with magnetic fields and operate as a solid-state refrigerant. Current technologies operate at less than 40% efficiency, have a capital cost of $70 million, and require 10–15 kWh/kg H_2. The project will achieve 60%–85% efficiency and operate at 5–6 kWh/ kg H_2. To achieve these metrics, the project will demonstrate magnetocaloric hydrogen liquefaction from 285 K for the first time at 10–25 kg/day. The project approach involves engineering and designing new installations with bypass flow, new ferromagnetic materials, and a 6T magnetic field. The two-stage design will lower hydrogen from 285 K to 120 K in the first stage, and finally to 20 K in the second stage.
- The team is looking at improving the baseline design by exploring bypass flow, different materials, and a magnetic field. The focus on starting from room temperature is fine, but a good deal of effort is being spent on a 70 K cooling system from room temperature, and it is of concern that much more trouble is ahead at cryogenic temperatures. This concern was expressed last year, but the principal investigator seems to be very confident that this will not be an issue. More science-/engineering-based arguments would have been welcome here.
- The approach is grounded on solid physical principles, and the team has identified problems with the current design (joule heating when the material moves into the magnetic field). The source of G-10 cracking was identified with the unexpected change of the coefficient of thermal expansion of the housing. However, the solution to the cracking problem was specific to the design under consideration and not a general approach to any future design developments.
- The approach has been reasonable, given the changes in plans because of development issues. Reducing the overall scope may be necessary for the team to achieve success by the end of next year.
- The demonstration of the concept is good. More work on the feasibility of the concept in practice would be helpful. It is not clear what the system to support the core concept looks like.

Question 2: Accomplishments and progress

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

- The project has the potential to drastically improve state-of-the-art technology for hydrogen liquefaction. An anticipated two- to three-fold decrease in energy input would reduce production costs and is better than the 2017 DOE target of 12 kWh/kg H_2.
- The project experienced some hiccups (G-10 cracking, valve design), which is totally understandable and well documented. The researchers were proactive in addressing those issues, and FOM analysis was a great addition to this work. A few concerns remain (on slide 20: the total ideal work and why its value differs from the first row are both unclear), including a comparison with today's benchmark, although this is partially addressed in the backup slides (slide 37). A detailed breakdown for both options, side-by-side, would be great.
- The team did a commendable job in making progress and overcoming technical hurdles. G-10 cracking due to large coefficient of thermal expansion (CTE) effects has been mitigated. The Ames Laboratory did a great job in potentially reducing the production costs of the magnetic materials. The addition and development of diversion valves was also noteworthy. The demonstration of bypass flow helps the efficiency of heat exchange, but more progress needs to be made in the magnetic refrigeration performance. It is unlikely that the full set of objectives will be achieved by the end of fiscal year (FY) 2019.
- The presenter claimed that the FOM for the device was 0.73 and that bypassing reduces it to ~0.5. No explanation was given for such a high FOM. It is not clear how such an extraordinarily large FOM can be achieved with the project's design, which involves two heat exchangers. In addition, the presenter's argument on low cost, despite the use of rare earth metals, needs further elucidation.
- It is unclear whether this approach will be cheaper/smaller/more responsive/etc. than current liquefaction approaches. It was not clear what the targets for this technology were.

Question 3: Collaboration and coordination

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

- The project team ran into trouble in 2017 when the system experienced mechanical failure. A crack developed in the regenerator housing during the cool-down process. The FY 2018 work was then refocused to address this problem. It was determined that a force imbalance associated with the movement of magnetic material in and out of the magnetic field contributed to the cracking. This problem has since been fixed by adding a flexible and expandable polytetrafluoroethylene (ePTFE) packing material between the layers and the housing. Additional work has addressed force imbalances. The magnetic field has been increased to 6T, and the cryogenic cooler has kept the magnet at ~5 K. First- and second-quarter milestones have been completed, including design and initial cool-down operation, achieving a 50 K temperature drop. New diversion valves were developed and constructed to control the flow between individual layers. No commercially available components were obtainable, so this required in-house fabrication. This new setup allowed for cooling down to 203 K. The design for the two-stage system has been completed.
- It looks like the work is going smoothly between the three main partners (PNNL, Emerald Energy NW, LLC. [EENW], Ames Laboratory). The Caloric Materials Consortium, CaloriCool (Energy Materials Network), seems to be another great avenue for collaborations.
- Collaboration with Ames Laboratory on materials issues is important.
- The longstanding partnership with Ames Laboratory and EENW is good. At some point, however, it would be good to see some private money being invested as matching funds.
- There was no collaboration (or it was not apparent) with industry; this is important for understanding the technical and commercial requirements for the technology. The project does have good collaboration among academia and institutions, however.

Question 4: Relevance/potential impact

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

- The project has good collaboration with Ames Laboratory and Iowa State University for materials characterization and synthesis, as well as with EENW for design and modeling, experimental tests, and data/cost analysis. The project is also engaging the CaloriCool consortium to encourage development of new magnetocaloric materials.
- It is a potential game changer if high-FOM hydrogen liquefaction can be achieved via magnetocaloric refrigeration, since liquefaction costs are a major issue in commercial hydrogen usage. There is also a potential reliability benefit due to lack of moving or rotating equipment requiring lubrication.
- Small-scale liquefaction systems are very relevant for unlocking low-cost access to locally distributed, renewable hydrogen. An explanation of how this technology will achieve that (in terms of economics, permitting, footprint, plausibility, etc.) is necessary.
- Alternative techniques to liquefy hydrogen are necessary, and the active magnetic regenerator liquefier cycle certainly represents one of the best options thus far.
- The project is very important to the Hydrogen and Fuel Cells Program mission, but the claim of an FOM equal to 0.73 is difficult to believe. The investigators must present a solid and detailed quantitative argument to support such a claim.

Question 5: Proposed future work

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

- The proposed future work involves the validation of the multistage concept to reduce temperatures from 285 K to 120 K, and then from 120 K to 20 K. FY 2018 plans also involve engaging industrial partners, redesigning and testing the valve system, and designing and building an eight-layer system. FY 2019 plans involve testing the eight-layer stage to achieve 120 K and then having a final demonstration of 120 K to 20 K liquefaction. Finally, FY 2020 plans involve hydrogen gas liquefaction, completion of technoeconomic analysis (TEA), and the licensing of patented technology.
- It is very clear what the next steps are to overcome some of the challenges identified in this phase. This future work is certainly relevant for advancing the technology readiness level.
- The determination of the FOM must be an important part of the future plans of the project, especially as it relates to the new materials synthesis and the heat exchanger design.
- The proposed future works seem a little too optimistic. Given the troubles experienced thus far, it is likely that the Gen-3 design effort, including the heat exchanger and ortho/para catalyst, will be much more significant than anticipated. The project is expected to last only one quarter into FY 2020, but the project team expects to carry out a good deal of work within that timeframe. Refocusing the scope of work may be discussed with the Fuel Cell Technologies Office (FCTO).
- It appears the scope for the next year has been changed to focus only on the first stage. That is fine, as long as progress is being made. However, the second stage will be more challenging than the first. Perhaps the team might give some thought to doing an initial cooldown using liquid nitrogen (LN2) to get the magnets below the Curie temperature.

Project strengths:

- This project’s main strengths lie in the promise of this technology to help minimize liquid hydrogen (LH2) costs and the simplicity and elegance of a magnetocaloric refrigeration system, as compared to typical cryogenic liquefaction cycles. Much good progress was made, both in advancing the original objectives, as well as in overcoming technical hurdles.
- The project has a knowledgeable team working on a complex and innovative technology that could be a game changer for hydrogen deployment. Issues are methodically addressed and include hands-on

demonstration and engineering solutions. This project should definitely keep being funded beyond the present contract.
- This is a very ambitious project that has the potential to transform hydrogen liquefaction. The project team has had some initial setbacks, but solutions have been found. The approach is unique.
- The project has a very compact liquefaction system and could be a very simple operation. The project also has a nice demonstration of applied engineering and science.
- The project's strengths lie in its technical design and industry and laboratory collaborations.

Project weaknesses:

- The setbacks associated with the initial cooling indicate additional unforeseen challenges in the future. This will likely require redesigns and possible delays. The project also relies on relatively expensive and rare materials, although the team does provide some evidence that less expensive magnetic materials could also work. It is unclear how scalable this technology is, but a full TEA may provide insight.
- The project team really needs to describe a vision for how this core technology would be integrated into a full system and what the commercial targets are for the technology. The team needs to explain the product/ commercial challenges with the system. For example, the technology uses large amounts of rare earth materials; the team needs to address whether this is a problem.
- The presentation could be improved. Complicated phenomena should be expressed in simple ways for the public and reviewers to fully appreciate the work being performed. For example, the process flow diagrams on slide 18 are unnecessarily complex, with a small font. The diagrams look like the same figure pasted twice, with the only difference being the J-T valve.
- This is a very challenging technology, and DOE has been funding it on some levels since the early 2000s. After all this time, there are still challenges in achieving cryogenic temperatures, much less LH2 temperatures.
- It is hard to accept the reported FOM magnitude of 0.73. The investigators must make a detailed presentation of how they came up with such an extraordinarily high FOM.

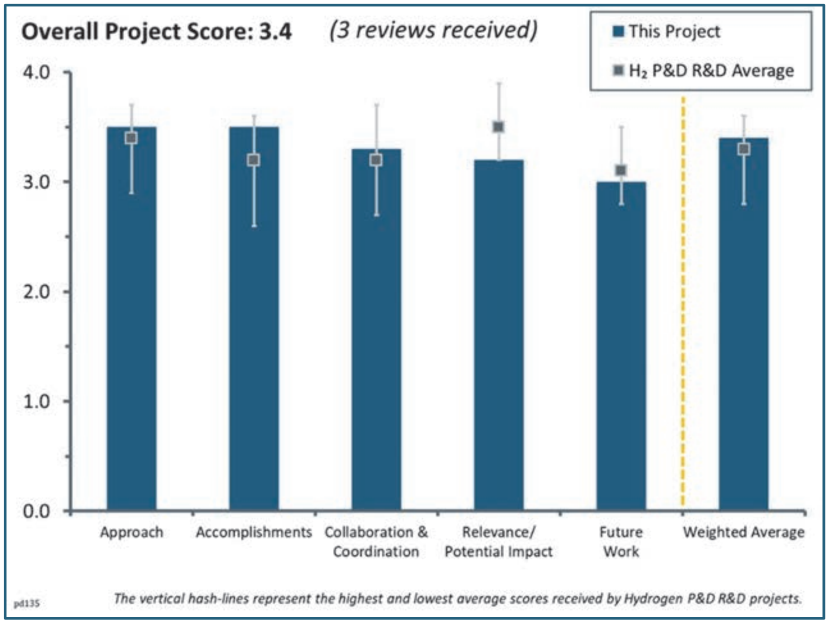
Recommendations for additions/deletions to project scope:

- No additions/deletions to the project scope are recommended. Perhaps the work should have been focused on cryogenic temperatures (e.g., LN2) first for accrued learning, but it is too late now to re-scope the entire project. The project team demonstrated cooling/liquefaction to 230 K a couple of years ago, albeit using a much less efficient design.
- There should be more efforts made in the lower-temperature stage. This will be more challenging thermally, but the first stage can be approximated by a LN2 supply.
- TEA should be considered in FY 2019 rather than FY 2020.
- It is recommended that the project team do more work on the system design.
- It is recommended that the team put more focus on the FOM.

Project #PD-135: Liquid Hydrogen Infrastructure Analysis

Guillaume Petitpas; Lawrence Livermore National Laboratory

Brief Summary of Project:



Liquid hydrogen (LH2) has many benefits for the hydrogen infrastructure, especially at large scale. However, the high cost of liquefaction, integration of LH2 with refueling stations, and transfer and boil-off losses pose challenges to broader use of LH2. This project aims to better understand and quantify the transfer and boil-off losses along the LH2 delivery pathway. To accomplish this, the project team simulated the LH2 pathway (from liquefaction plant to end use) using a thermodynamic model to estimate, then mitigate, the transfer and boil-off losses that occur at each step along the delivery pathway. Real-life driving and parking data were collected from a large population to use as an input for the model. The project also identified the major hydrogen boil-off sources and investigated potential recovery technologies and processes—from technical and cost perspectives—to eliminate and/or reduce these losses.

Question 1: Approach to performing the work

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

- The research approach used was appropriate and led to a first-order analysis of the boil-off losses in handling LH2 fuel. The basic thermodynamic models allowed an improved understanding of the problems associated with using hydrogen fuel in liquid form, and resulted in recommendations that could significantly avoid boil-off losses. The project was well designed, and the scope was relevant to the goals of the Hydrogen and Fuel Cells Program and well aligned with H2@Scale activities.
- The development of models to simulate LH2 processes and losses is a valuable tool to help identify major loss mechanisms and evaluate the impact of changing systems.
- The modeling approach is a reasonable approach. However, it was not clear whether the model was validated. The project team had some data that could have been used to validate the model. It is not clear if the team used hydrogen recovery analysis to try to determine whether hydrogen purification was required. Since it is likely that the recovered hydrogen would require purification, the purification costs should have been added. The project team contacted two gas companies, which was good. However, they did not contact users such as NASA or refueling stations. Those users, especially NASA, might have provided much insight.

Question 2: Accomplishments and progress

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

- The project accomplished the desired goals. A versatile model was constructed that allowed for the analysis of boil-off losses from liquefaction to car dispensing, as well as onboard. Furthermore, the project provided

guidelines within regulation that may result in significant hydrogen savings from not venting LH2 trailers. The model was used to simulate the performance with real driving patterns, and the conclusions indicate that, when handled appropriately, boil-off losses can be minimized to only a few percent of LH2 stored.
- It appears that all original technical objectives have been accomplished, and the models have been made available for others to use/validate.
- The analysis suggests that hydrogen boil-off losses were acceptable in most cases. The hydrogen recovery may not make economic sense, and the team did not examine whether there was a safety reason to recover the hydrogen. The identification of top-fill operation as a preferred mode is important. The boil-off recovery system could reduce the cost to end users by 50%.

Question 3: Collaboration and coordination

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

- This is a small project, so major collaboration is difficult, but the principal investigator (PI) reached out to other government and industry experts to gather their feedback.
- The collaboration was fruitful, as evidenced from the positive outcome of the project.
- The project team engaged two gas companies. It is surprising that pump companies and companies using LH2 were not contacted. The team should have contacted companies who are currently using LH2, such as NASA and the many locations using LH2 for their hydrogen-powered forklift fleets.

Question 4: Relevance/potential impact

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

- The findings from this project will help inform the decision on pathways for hydrogen distribution. LH2 has high potential for economic savings from lower transportation costs, thanks to the higher energy density of the fuel compared to compressed hydrogen gas. Also, the model results could help design better LH2 handling systems for onboard LH2 tanks.
- LH2 analysis like this is necessary to enable effective use of LH2 for delivery.
- The model is a good planning tool, but until it has been validated by data for real-world hydrogen storage and delivery losses, its benefit is limited. The impact has been magnified by making the model available to others.

Question 5: Proposed future work

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

- It appears this is a final deliverable project, and there is no follow-up work proposed.
- This project was completed, so no future work is necessary.
- The project is ending, so there is minimal future work. It is recommended that the researchers contact users of LH2 beyond the gas companies.

Project strengths:

- A simple thermodynamic model was developed that allowed the team to assess the extent of LH2 boil-off losses; recommendations from the model could help save significant amounts of LH2 in the future. The results from this project have high potential impacts for the implementation of LH2 as a viable fuel.
- The project team is investigating a very important topic for DOE. The team has experience working with LH2, and the project has contacted two major gas companies in its work.
- Overall, this project was very successful in developing a tool to begin to predict hydrogen losses in the distribution chain.

Project weaknesses:

- There are many institutions that use LH2, beyond the gas companies, that the team could have contacted. For example, the project team could have talked with NASA about how to mitigate this challenge. With the amount of hydrogen NASA uses and NASA’s strong safety culture, this would have been a very interesting discussion. The team could have also contacted companies that use hydrogen-powered forklifts and are having LH2 delivered. It does not appear that the project team validated the project model.
- While the model is useful and the PI did a good job in simulating different driver usage profiles, more real-world data is necessary to continue the validation of this model. Also, little data on boil-off recovery benefits was obtained.
- The proposal to work on computational fluid dynamics models for modeling top-fill losses might not be that important, given that the extent of the losses from boil-off is limited.

Recommendations for additions/deletions to project scope:

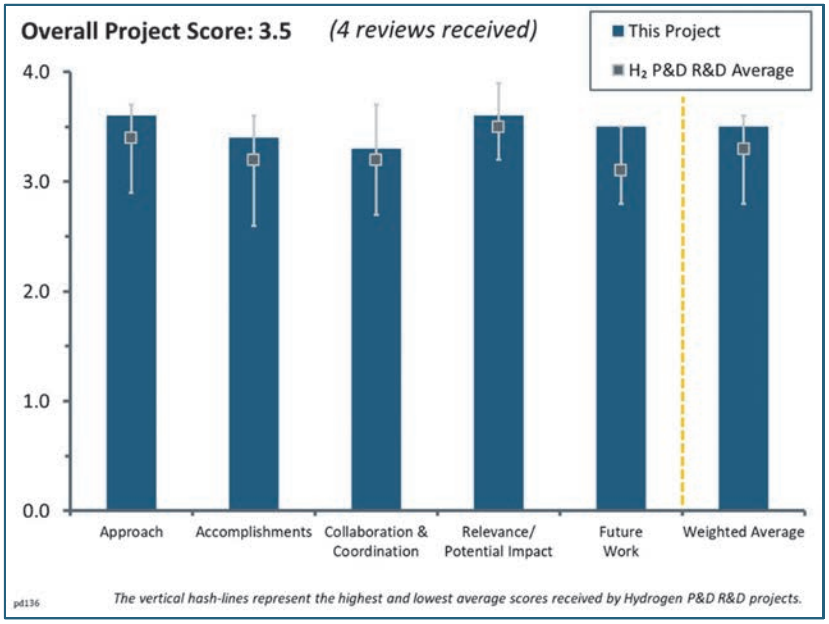
- Possibly there should be follow-up work to instrument delivery trucks and refueling stations to help validate the model over a longer period of time, along with continued refinements in the model to accommodate actual fluid behavior.
- There are no additional suggestions to the project scope.

Project #PD-136: Electrochemical Compression

Monjid Hamdan; Giner ELX, Inc.

Brief Summary of Project:

This project will develop and demonstrate an electrochemical hydrogen compressor (EHC) that is lower in cost, higher in efficiency, and more durable. Specifically, the project will (1) fabricate hydrocarbon membranes with enhanced properties for use in EHCs, (2) improve EHC water and thermal management, (3) optimize stack hardware and demonstrate cell performance, and (4) build a prototype system. Development of reliable and low-cost, high-pressure hydrogen systems is needed to enable market penetration of fuel cell electric vehicles.



Question 1: Approach to performing the work

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

- The project identifies and addresses key issues for electrochemical compressors of water management, thermal management, and mechanical strength. The approach of utilizing hydrocarbon membranes should provide benefits of lower osmotic drag and lower cost. The water management membranes should improve water management and lead to better durability. The project milestones are logically laid out to lead to demonstration of a compressor that meets the project targets.
- The team had a good understanding of the whole system; the team did not focus just on membranes or one specific technical challenge. Focusing on delivered cost, reliability, and performance is essential. The team's ability to paint a full picture, then focus on low-technology-readiness-level (low-TRL) research, is appreciated.
- The team had a logical approach to addressing major barriers of electrochemical hydrogen compression including advanced membranes, water management, and cell and stack scale-up.
- The project has a sound approach.

Question 2: Accomplishments and progress

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

- The new membrane showed a 30% reduction in electrical energy usage (kilowatt-hours per kilogram of hydrogen) at $1 A/cm^2$, 350 bar. The project has reduced electro-osmotic drag (EOD) by 30% compared to the perfluorosulfonic acid (PFSA) baseline. The project has reduced back diffusion of hydrogen by 50% compared to baseline PFSA. The project has tested cell design and cell components at up to 5000 psi. Energy consumption per kilogram of hydrogen is still far from the target of 1.60 kWh/kg.
- This project is making excellent progress toward Year 1 milestones. The team already achieved the Year 1 go/no-go milestone.
- Successive progress has been made throughout the year, leading to promising results.

- There is good progress according to the plan; however, this project may be behind others developing a similar system. The team has to accelerate progress to be competitive and relevant.

Question 3: Collaboration and coordination

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

- The team has very good collaboration with academia, the national laboratories, and private partners.
- The collaborations between the partners appear to be working effectively.
- The collaborators are clearly listed, along with their roles.
- The team needs better collaboration with end users and operators. Mechanical compression is getting cheaper, quieter, and easier to use, year over year. The team should be sure to set the right targets and be clear about what the right use cases are for this technology; it will help focus development.

Question 4: Relevance/potential impact

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

- Compressors are a major cost for hydrogen refueling stations and are also one of the parts most responsible for station downtime. The electrochemical compressor can potentially address reliability and cost concerns. The proposed work has the potential to reduce cost and improve efficiency of EHCs.
- Electrochemical compression has excellent potential to be the lowest-energy and lowest-cost option for compressing hydrogen to required refueling pressures.
- Alternative compression technologies are of high interest to the industry. A solid-state system has advantages in terms of noise and footprint; there are concerns over how this technology scales. It would be interesting to understand the potential for these cells to scale up to 120+ kg/h at 70 MPa output pressure, which would be relevant for where the industry is going with high-demand regions (e.g., Japan, California, China).
- This project can have enormous impact if scale-up challenges are overcome.

Question 5: Proposed future work

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

- Plans address critical issues of back diffusion, durability testing, and cell area scale-up.
- Future work is focused on completing membrane studies, scaling up to 300 cm^2 active area, and increasing operating pressure. Further membrane development should reduce back diffusion and increase efficiency.
- It will be interesting to see how the technology scales. Focusing on mechanical integrity of larger cells may be of interest for increasing output of individual stacks.
- Future work clearly addresses the challenges necessary for the project’s success.

Project strengths:

- Membrane materials and packaging are clearly the limiting factors to scaling up a viable stack. The team is focusing on the right things.
- The project approach is well defined. Project progress has been promising, and future work is clear.
- The project’s hydrocarbon membrane should reduce cost, EOD, and hydrogen back diffusion.
- The project has a strong approach and is making excellent progress.

Project weaknesses:

- The team needs to accelerate fielding viable high-capacity units. Development in parallel on aspects other than membranes could help. For example, structural integrity of larger areas and high-output units is needed. In addition, this project could benefit from collaboration with others in the community; a large part of the issue here is material science. The team could benefit from focusing externally on centers of excellence to accelerate development.
- Aromatic hydrocarbons can be hydrogenated under some conditions, and the high hydrogen pressure and presence of platinum-group-metal catalysts are conditions that make this reaction more likely. It is not clear whether the project has evaluated the membranes to see if this is occurring and what the impact on durability would be. This could be a second degradation mode (in addition to mechanical failure).
- Overlooking the effort required to scale up from 50 cm^2 to 300 cm^2 is a project weakness.

Recommendations for additions/deletions to project scope:

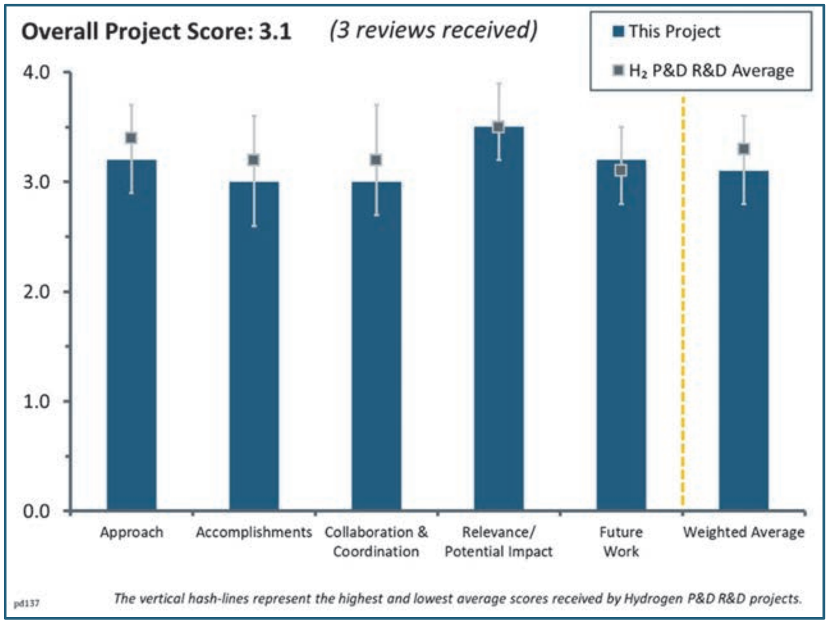
- The team should add pressure cycling testing to the stack test plan, especially for 300 cm^2 stacks.
- The team should look for any evidence of hydrogenation of the aromatic polymer backbone.

Project #PD-137: Hybrid Electrochemical–Metal Hydride Compression

Scott Greenway; Greenway Energy, Inc.

Brief Summary of Project:

There is a need to increase the reliability, reduce the cost, and improve the energy efficiency of gaseous hydrogen compressors. This project seeks to address this challenge by developing a hybrid electrochemical–metal hydride (EC-MH) compressor. The project will analyze and screen potential hybrid compressor systems and materials, conduct experimental tests, develop a hybrid compressor system model, and build a prototype unit.



Question 1: Approach to performing the work

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

- The overall approach of this work is strong. The methodology addresses most of the conceivable steps for improving the overall technology readiness level (TRL) and directly addresses cost goals and U.S. Department of Energy (DOE) targets. While not part of the scope of work, it would be worthwhile to indicate the likelihood of reducing some of the major costs such as membrane electrode assemblies (MEAs) and metal hydrides, as these are the largest cost drivers and have historically been difficult to lower. As it stands, it appears as if the biggest obstacle to making this technology economically feasible is left unexamined. Currently, the documentation presented listed MEA costs at five times higher than the cost required to meet DOE targets. Likewise, metal hydrides are three to four times higher than the costs required to meet DOE targets. These issues should be addressed, at least in the abstract, as to how the system could be made more cost-effective.
- The project approach combines an existing EHC and MHC.
- The combination of an electrochemical hydrogen compressor (EHC) and a metal hydride compressor (MHC) has potential to increase efficiency using heat from the EHC for the MHC. The work should take into account thermal and mechanical properties of the membranes. Nafion 117 has a glass transition temperature of around 120°C–130°C, which is considerably below the temperatures being considered for the pump operation (up to 170°C). Mechanical properties of the membrane above the glass transition temperature are expected to suffer, and the membrane's durability at these temperatures with a pressure differential across the membrane is suspect. It is not clear that the project gives enough attention to controlling water content in the system and integrating the water control with the rest of the system. The EHC needs to remain well hydrated to operate at the temperatures described, and the EHC will require a pressurized system with high water partial pressures to maintain good conductivity in the Nafion membranes. The metal hydride system requires very low water partial pressures (likely sub-parts-per-million [sub-ppm] to meet durability requirements). The system diagram shows a pressure swing adsorption unit in the design, but it is not clear that the requirements and performance of this pressure swing absorption (PSA) system are integrated with the rest of the system, and the experimental plans for testing PSA systems under the relevant conditions for their hybrid compressor are not described. Water management will be crucial for this design and needs more attention.

Question 2: Accomplishments and progress

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

- So far, the progress of this work is good. There has been strong development in the experimental stage of the project. Models have performed well and are fairly descriptive of the work that has been accomplished. Some small issues were seemingly not addressed in the presentation (high material costs, thermal durability of some materials, etc.).
- Initial results have led to a down-selection of membrane types and indicated polybenzimidazole (PBI)–phosphoric acid membranes are not a good match, owing to physical properties of the membranes and issues with protecting the system against phosphoric-acid-induced corrosion. Finned heat exchange design appears appropriate for metal hydrides. Experiments indicated Nafion stability at high temperature varied parameters during the test (temperature and pressure) and did not return to initial conditions, so it is not apparent that no performance degradation was shown over the test. Over the first 25 hours, the team indicated a constant voltage, but the temperature was increased from 130°C to 145°C over this time period, which would result in changes in the membrane conductivity from the increased temperature. A temperature increase alone would lead to an increase in conductivity and should result in a lower required voltage for the same current. A complicating factor is any changes in hydration level, which was not sufficiently described. It is not clear what relative humidities or water partial pressures are being used in the experiments or modeling. Nafion conductivity is dependent on the water content.
- Operating Nafion far above the glass transition temperature does not bode well for long-term durability of EHCs.

Question 3: Collaboration and coordination

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

- These are good collaborations for MHCs.
- Collaborations within the project appear to be operating. Collaborations with those doing metal hydride and MHC work appear to be in place and operating well. The project would benefit from collaborations with the membrane community and those working on polymer electrolyte membrane (PEM) fuel cells, electrolyzers, and EHCs. There does not appear to be any collaboration with PEM membrane suppliers or research groups. Supported and doped PEM membranes would offer some advantages.
- The extent of collaboration seems reasonable, though vague terms are used to describe the frequency of collaboration. Further, the scope of work for some collaborators seems extremely limited, to the point of questioning the value of having multiple groups working on specific aspects. Other groups must be taking the lion’s share of the work, although what work they are completing seems undefined. It is often beneficial to incorporate the assignment of labor onto the technical slides in which the work is reported.

Question 4: Relevance/potential impact

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

- The potential impact is high if this project is successful and can increase efficiency and reduce cost of hydrogen compression. Hydrogen compression is one of the major costs associated with hydrogen delivery.
- Low-cost, larger-scale hydrogen compression is needed.
- This work is useful, as it relates to several aspects of the Hydrogen and Fuel Cells Program Multi-Year Research, Development, and Demonstration Plan. The project will potentially advance several areas, although, given the lack of a large-scale model and the potential cost inhibitors, there is the chance that it will not have a large impact in the long run. Existing and planned future work may make this clearer, and the project team should be congratulated on the foresight to plan for these long-term and large-scale issues.

Question 5: Proposed future work

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

- The project's future work is clearly outlined and established, and it seems relevant and feasible to improving the project and accomplishing established goals.
- Future work on the MHC system appears appropriate. While Nafion 117 may be appropriate to prove the concept, durability of Nafion at temperatures >150°C under pressure differentials is suspect because of its low glass transition temperature, especially if membrane thickness is reduced. Work on the EHC system operating at T>150°C should look at supported membranes and alternative membranes with glass transition temperatures higher than Nafion's (one possibility is Nafion doped with inorganic oxides). Water management needs to be given a higher priority in work moving forward, as it is critical to the operation of this system.
- The scale of the prototype demonstration system in Tasks 2.1 and 2.2 should be clearly stated. Durability testing and cycling testing should be included in the plan.

Project strengths:

- The project shows a complete effort, running modeling all the way through experimental tests and scale-up. The results seem promising, and milestones are being completed with seemingly good efficiency. The design of the experiment seems complete, and individual tests all seem to be showing good results, with the end goal directly and completely related to improving the work requested by DOE.
- The concept for a hybrid EC-MH compressor offers some potential advantages and opportunities to improve efficiency.
- This is an interesting hybrid compression approach.

Project weaknesses:

- The economic analysis has some small shortcomings—namely, financial assumptions are buried in the results. For example, the type of economic analysis is unclear. Further, there is limited discussion of whether the required specific material costs are reasonable for future efforts. The extent of collaboration could be better indicated. The authors could give an idea of how a functional, large-scale system will benefit the end-use application. For example, they could explain whether this system would lower compression costs, reduce filling time, etc. and indicate by how much various parameters would affect the end user.
- This hybrid approach requires temperature matching, which requires PEM membranes to be operated outside of their normal operating range, with uncertain long-term durability.
- The project could use assistance in the area of membranes and membrane performance. The project has put too little effort on water management, which is a critical barrier for this concept. The team's effort on water management has to be increased.

Recommendations for additions/deletions to project scope:

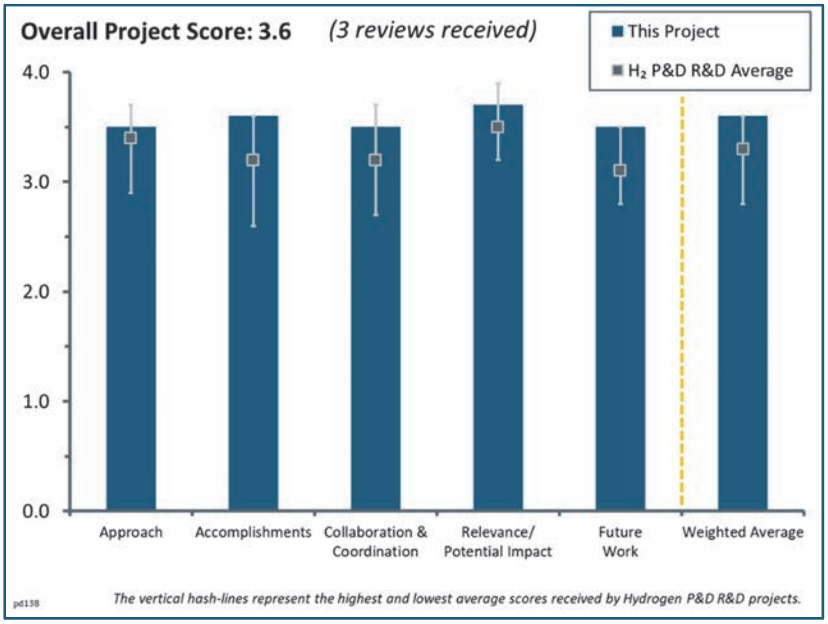
- The team should add durability testing and pressure cycling to the prototype testing protocol. The team should include the breakdown of kilowatt-hours per kilogram for the EHC and MHC systems (based on actual measurements) to allow easier evaluation of progress toward the DOE target.
- It is recommended that the project find a membrane supplier, research group, or PEM electrolyzer/fuel cell system group with which to collaborate.

Project #PD-138: Metal Hydride Compression

Terry Johnson; Sandia National Laboratories

Brief Summary of Project:

The objective of this project is to demonstrate a two-stage metal hydride compressor with a feed pressure of 50–100 bar delivering high-purity hydrogen gas at an outlet pressure of 875 bar or more. The project will identify at least two candidate alloys for both the low-pressure and high-pressure stages, complete a detailed design for the compressor, and build a prototype compressor. The developed technology seeks to address the need for less expensive and more reliable compressors for hydrogen fueling stations.



Question 1: Approach to performing the work

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

- The two-stage metal hydride compressor under development is promising. The design effort was carefully planned, and progress of the overall system integration plan is on track. The system-level focus will allow the team to advance from technology readiness level (TRL) 2 to TRL 5.
- Critical challenges (e.g., thermodynamical properties of metal hydride alloys, overall energy efficiency, system design optimization) have been identified and are addressed in a perfect way.
- This project has a consistent and linear approach: (1) identify materials, (2) check thermal conductivities, (3) verify with a system-level model, (4) design the prototype, and (5) build the prototype.

Question 2: Accomplishments and progress

This project was rated **3.7** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Accomplishments and progress so far for this project are excellent to outstanding. Scientific and technical challenges have been identified, and excellent progress has been demonstrated. The project has accomplished high-pressure characterization of these alloys, along with development of the world-class instruments needed for this task, and progress is very good. The project is on a very good track. The identification of possible heat pumps to be used to reach the required energy efficiency is an important milestone. Analysis and optimization of the design of the system are excellent.
- The project team met all milestones since the last DOE Hydrogen and Fuel Cells Program Annual Merit Review. Much work seems to have been done in simulation and design.
- There is progress toward most of the objectives of system design and integration targets. The materials selection work is somewhat under-emphasized. It was not clear whether the 20-minute target for the half-cycle proposed is already achieved. In the future, greater clarification of the performance gaps of different aspects of the project, a clearer separation of predictions from modeling, and a discussion of the results achieved so far will be needed. The team's milestone 7.1, currently 50% complete, will be an interesting challenge for the team.

Question 3: Collaboration and coordination

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

- The team members are very knowledgeable and are close to the "best-in-class" in metal hydrides. Collaboration between Sandia National Laboratories, Oak Ridge National Laboratory (ORNL), and Hawaii Hydrogen Carriers, LLC, seems smooth and fruitful and is a great combination of skills.
- The project shows outstanding internal and external collaboration (especially with project PD-137).
- Leveraging other funded projects, such as PD-137 and PD-171, is mentioned. The team has many more opportunities for collaboration, both inside the organization and with other institutions such as ORNL and Ames Laboratory, to help achieve the targets.

Question 4: Relevance/potential impact

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

- This project is highly innovative and most probably will bring about a breakthrough in high-pressure hydrogen compression technologies. Because of capital investment and maintenance expenses, compressors are the most expensive components in hydrogen fueling infrastructures. If the project is successful, highly reliable compressor technologies with lower maintenance costs and longer lifetimes could be developed. The potential impact of this project is enormous.
- This a very important technology. The progress made will go a long way toward changing the market for compression and energy consumptions using metal hydrides.
- Alternatives to mechanical compressors are needed. Metal hydride compressors appear to be an okay candidate, and this work is very important as a means to settle this question once and for all.

Question 5: Proposed future work

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

- Project plans clearly build on past progress and are sharply focused on critical barriers to project goals. Project plans are a logical extension of what has been achieved so far and what has to be done before project end.
- Proposed future work is consistent and looks reasonable for achieving successful end-of-project goals.
- Although all necessary elements are there in the planning, it is not clear whether some critical bottlenecks can be removed without some re-planning of the tasks and bringing in experts in alloy selection. Right now, the risk of not achieving the target seems high.

Project strengths:

- The project has a strong engineering connection and has made important progress. The challenges are fundamental in nature in materials performance for the high-pressure alloy and energy targets. Irrespective of the potential challenges, the project will improve the state of the art and can be rebooted in the future if the data and capabilities are maintained in a retrievable format after the end of the proposed performance period.
- This project has a great team with a scope consistently oriented toward demonstration of a continuously operating prototype up to 875 bar. There is a clear path toward the end goal, and there have been great accomplishments thus far.
- This is an outstanding project performed by an outstanding team of highly excellent researchers with outstanding perspectives.

Project weaknesses:

- Project weaknesses are not major. The challenging nature of the concept is the high-pressure stage. The high-pressure stage has greater dependency on materials availability, the maturity level of the materials, and how much of the development can be shaped by the current team. The team needs to make more of an effort to engage experts inside and outside the laboratory for help.
- Metal hydrides rely on waste heat, which is not even available at a station. However, this aspect is not really a project weakness. More publications could be a plus.

Recommendations for additions/deletions to project scope:

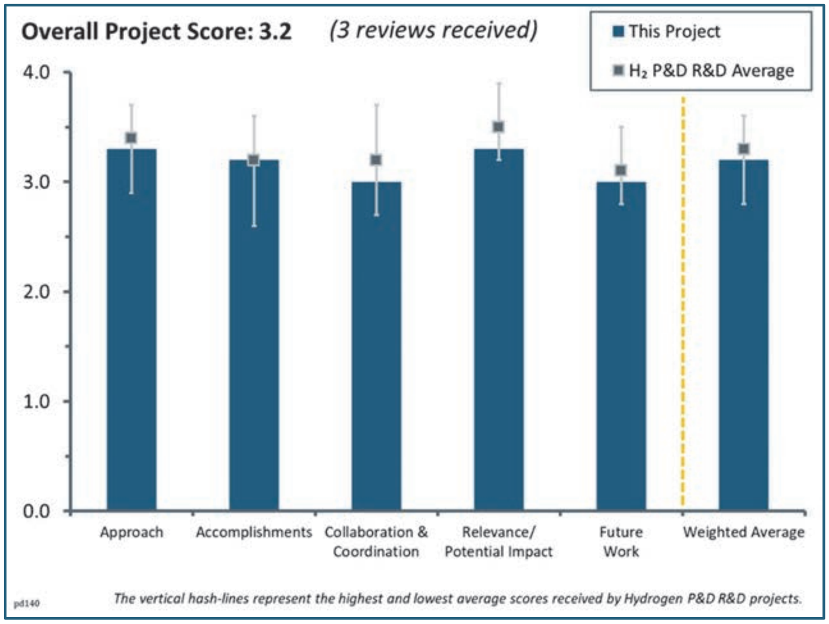
- There are no recommendations for additions/deletions to the project scope.
- Recommendations include more collaborations on the high-pressure alloy phases and a stronger design effort beyond the helical design of the heat exchanger. The nuclear reactor community has lot of experience in this problem. Other parts of the National Nuclear Security Administration/DOE laboratory system can be engaged in support of the heat exchanger design. A good modeling effort might suggest better designs. The fabrication/manufacturing challenge will be less important if performance is not optimal. Overall, the team is aware of the issues and can make course corrections depending on resource availability and the go/no-go points coming up.
- To demonstrate the highest possible energy efficiency, the coupling with the heat pump would be essential. The necessary funding for this should be added to the project.

Project #PD-140: Dispenser Reliability

Michael Peters; National Renewable Energy Laboratory

Brief Summary of Project:

Hydrogen fuel dispensers are a top cause of maintenance events and labor time at hydrogen fueling stations. This project seeks to identify the proper balance between dispenser costs—both capital and operations and maintenance (O&M) costs—and performance. The project consists of three major tasks: (1) a technoeconomic analysis of capital and O&M improvements to the chiller/heat exchanger, (2) reliability testing of dispenser components, and (3) development of an open-source and free hydrogen fueling model.



Question 1: Approach to performing the work

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

- The project tests hydrogen polymer dispenser reliability under real-world conditions by checking for hydrogen leaks and pressure tracking. The project is a field study that can be typecast as a performance approach to safety and reliability. As such, this project has merits.
- The device-under-test (DUT) components are comprehensive. The accelerated testing system, leak detection, and materials testing are very well conceived.
- The approach appears reasonable for the limited costs associated with this task. It would be beneficial if the presentation had more details on the types of failures, such as internal versus external leaks, valve failures, and contamination. Using multiple dispensers allows for maximum tests in a minimum amount of time. Selecting multiple suppliers for DUTs also increases the quality of data.

Question 2: Accomplishments and progress

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

- The accomplishments reflect an impressive effort to set up and establish a detailed experimental design and a state-of-the-art test apparatus. The actual results could prove to be invaluable.
- Good collaboration with manufacturers has resulted in the testing of multiple components. Slide 15 showed that the project infrastructure had been well designed, and slide 18 showed that the project's approach can successfully replicate real-world dispensing conditions. No information was provided on the hydrogen wide area monitor pressure sensor. On slide 13, it was stated that material testing was to be done at Sandia National Laboratories (SNL), but no additional details were given. From the presentation, one could not conclude that the two laboratories interacted on the project. Similarly, the statement that the reliability of components was statistically assessed by researchers at the Colorado School of Mines cannot be assessed; no details were given as to how the statisticians assessed mechanical failure.
- Progress is being made, but it is a little slow. The test set-up has been completed and commissioned, and the dispenser components are now ready for testing. A simple schematic of the dispenser, DUTs, and

recirculation loop together would be beneficial to understand the level of effort put into design and assembly.
- The presenter appeared very convinced and provided data to prove his point. However, the reviewer's knowledge of this technology is limited.

Question 3: Collaboration and coordination

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

- Industry involvement may be challenging because of the competitive nature of this field. Involvement by refueling station developers might be helpful.
- There is good collaboration with SNL in the area of polymer materials characterization, and baseline samples have been taken.
- The project has good collaboration with Weh Technologies, Inc., and Walther–Praezision. However, non-disclosure agreements need to be set in place properly, or else the data obtained from the project will not be available to the public as the "Management Plan" claims.

Question 4: Relevance/potential impact

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

- This project represents a high-quality effort that is crucial to kick-starting the fuel cell electric vehicle market.
- Collaboration with industry and real-world testing conditions can provide data for assessing the reliability of dispensers and dispenser materials.
- The presenter provided a good, convincing picture of this technology.
- The project team should deliver good additional information on device reliability that could be used to help develop better component alternatives. This project relies too much on the results of this specific round of testing. It would be better to include continued investigations on real-world stations and issues that the dispensers are having; this would give the team a more statistically significant sample to investigate. Also, the team should include failure analysis results on real-world dispenser problems.

Question 5: Proposed future work

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

- The project has a great test plan and system design—now it is time to execute it. A national test center for hydrogen refueling technology could be established, based on the body of work and systems. The test apparatus could be productized for use by manufacturers.
- Now that all the preliminary work is complete, the next phase of the project (actual testing) is the most important and should be given time to continue. It looks as though the project is slated to be complete by the end of September, but consideration should be given to the team's continuing testing until multiple failures have occurred and there is enough data to draw conclusions.
- The presenter provided a good, convincing picture of this technology.
- It is not clear what materials analysis will be provided to materials manufacturers (slide 22). It is also not clear that a well-thought-out process is in place for such an analysis.

Project strengths:

- This project identifies the most likely problems with reliability on hydrogen fueling stations (dispensers) and develops a test rig to be able to try to determine failure modes. Overall, the budget is small compared to the potentially positive results that could help minimize station down time.
- The project's scope, test design, and test system are excellent.

- The project's strengths lie in its real-world failure assessment and close collaboration with industry.

Project weaknesses:

- This project has no material weaknesses.
- More details could be given on the types of failures, and some effort should be made to continue monitoring real-world stations for issues as they arise. More details could also be given on the design of the overall test set-up, including schematics.
- A collaboration plan with SNL is lacking. Furthermore, there is no plan for how the project will document failure or what the properties of materials that need to be monitored are.
- The project's weakness is that the devices are not being tested commercially (i.e., outside the laboratory).

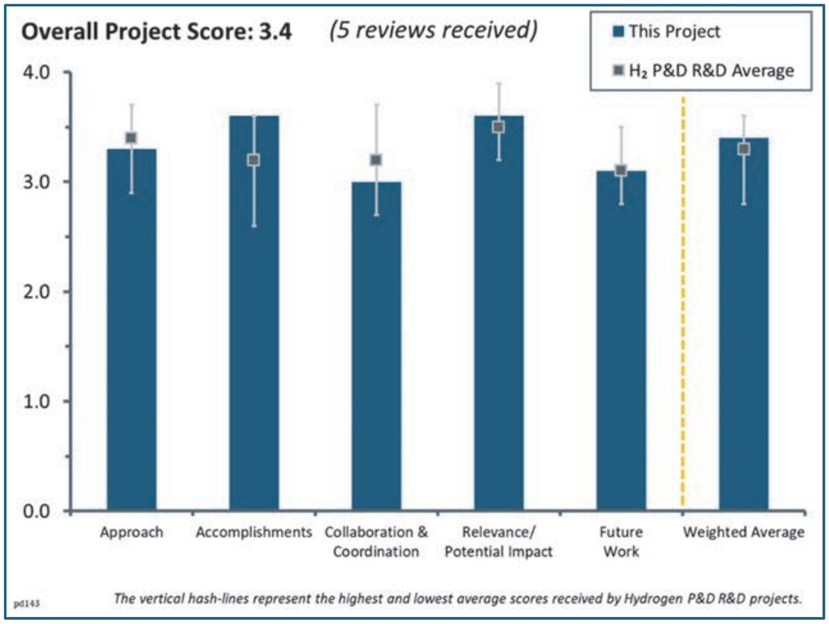
Recommendations for additions/deletions to project scope:

- There are no recommendations—the presenter provided a good convincing picture of this technology.
- Testing of these devices should continue until the planned tests are complete. These data should then be compared to real-world dispenser failure data that should continue to be compiled in a database.
- The project does not seem to have a well-defined plan on future direction. In particular, the project lacks objectives and an approach of how to document failure based on a set of indices/parameters that can be used as measures of reliability and safety. The future work reported on slide 23 is vague and lacks specificity and targets.

Project #PD-143: High-Temperature Alkaline Water Electrolysis

Hui Xu; Giner, Inc.

Brief Summary of Project:



This project aims to develop high-temperature (HT) molten alkaline electrolyzers with improved electrical efficiency and reduced cost. The electrolyzer will operate in the temperature range of 300°C–550°C. Specific project tasks include (1) development of porous ceramic oxide matrices, (2) incorporation of molten hydroxide electrolytes into the porous matrices, (3) selection of anode and cathode catalysts, (4) assembly and testing of single cells, (5) construction and testing of a 1.8 kW electrolyzer stack, and (6) system and economic analysis.

Question 1: Approach to performing the work

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

- The project team is investigating the use of molten hydroxides in porous metal oxides for use in HT alkaline electrolysis. The approach could potentially lower the energy requirements for hydrogen production, improve reaction kinetics, and reduce steady-state voltages. The approach for making the materials is fairly simple and straightforward, which is an inherent advantage for commercialization efforts. The performance of these systems, based on the project data and milestones, is rather good, and the systems have reasonable stability. The authors have addressed some corrosion issues with their cells that were limiting performance (slide 15). The cells "nearly" achieve the metric of <1.50 V at current density of 1.0 A/cm^2 or <1.40 V at 0.6 A/cm^2 (slide 16). This is a fairly impressive performance and very reasonable progress for this effort.
- The overall approach of infiltrating hydroxide into porous alumina and zirconia is good. This project has promising potential for large-scale hydrogen production with a low-cost electrolyte/matrix.
- The project has an interesting approach to benefit from higher operating temperature (faster kinetics, lower catalyst cost).
- The project has a solid foundation and a thorough approach.
- The project appears to use a relatively simple approach. Several metal oxides from Zircar Zirconia, Inc. (Zircar) were tested for compatibility as porous matrix phases for HT alkaline OH- electrolytes. It is unclear whether the technical targets are much of a challenge, aside from achieving 90% lower heating value (LHV) efficiency, which may be claimed based on an energy balance with some generous assumptions. For such a relatively simple scope, more detail would be expected on the individual components. Perhaps significant portions of information are being withheld to protect intellectual property. For instance, the modus operandi is not explicitly identified, which makes it challenging to consider the degradation mechanisms within them.

Question 2: Accomplishments and progress

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

- The authors identified technical targets of (1) composite electrolyte OH- conductivity of >0.1 S/cm, from 300°C–550°C, (2) per-cell area-specific resistance of ≤0.2 ohm-cm^2 from 300°C–550°C with a 200-micron membrane, and (3) stack electrical efficiency of >90% LHV H_2 with a current density of 1.2 A/cm^2. Technical targets 1 and 2 were carried down to the four milestones and go/no-go decision listed on slide 5. Technical target 3 on stack efficiency did not seem to be included in the current milestone set. The authors have achieved all of the milestones listed for this performance period. They have "nearly" achieved the performance target for the go/no-go decision of single cell performance of V <1.50 V at 1.0 A/cm^2, or 1.4 V at 0.5 A/cm^2 (data on slide 16). Hitting the four milestones is excellent progress. Additionally, the team is incredibly close to the technical targets for the go/no-go decision, which is also good-to-excellent progress. In general, the progress toward the defined technical targets and project milestones for the period is excellent. It is assumed that the technical target of stack electrical efficiency >90% LHV H_2 with a current density of 1.2 A/cm^2 will be addressed in later years for the project, but there were no details about this in the slide deck or verbal presentation.
- This project has outstanding progress in the conductivity increase of the matrix and the reduction of cell voltage and resistance, especially considering the challenges of operating at higher temperatures.
- The project has made very good progress in making the electrolyte matrix and testing of cells. Progress in cell performance over a fairly short period is very impressive.
- There is good progress to date, good responses to review comments, and good mass/energy balance. The team's progress toward project and DOE goals is substantial.
- The presentation mentioned 100% progress for Tasks 1–4, but there was no meaningful discussion of the catalyst development in the presentation, only a mention that the catalysts were optimized. There was no discernable discussion of the percent increase in activity/corresponding decrease in polarizations associated specifically with the hydrogen evolution reaction and oxidation evolution reaction catalyst. Overall, minimal concrete detail was given on progress.

Question 3: Collaboration and coordination

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

- The authors list the University of Connecticut (UConn) as a collaborator for fundamental studies of matrix coarsening and corrosion. This data is illustrated on slide 17. From the supplementary slides, it seems that the team has added UConn to address previous review comments regarding the stability of these electrocatalysts at higher temperatures. The project team has taken a good step toward addressing previous reviewer comments. The results with UConn seem aimed at addressing issues with the current collectors. UConn's role on this project seems small, but the authors are making progress and hitting their milestones. Additional collaboration would make sense only to address knowledge/skills/capability gaps on the team.
- The project has excellent collaboration with the UConn and Giner ELX.
- It appears as if collaboration has been limited, or at least, the presentation communicates that collaboration has been limited.
- Giner ELX is collaborating with P. Singh's group at UConn. It appears that slides 17–18 are based on UConn work. It would be advisable to leverage the UConn team's skills more fully; it is unclear whether the matrix solubility study was planned for MO-3 only. It is also unclear what is unexpected regarding the corrosion determined by the metal stability tests. Energy balances with Giner ELX are not necessarily collaborative efforts since there is a pre-existing link.
- UConn seems to be the only collaborator. Energy Materials Network nodes should be engaged for independent evaluation.

Question 4: Relevance/potential impact

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

- The project is making good progress toward its technical goals, which will help enable the large-scale deployment of hydrogen for energy applications. The project is relevant and impactful to the H2@Scale concept. The performance of the materials is reasonable and is hitting the technical targets associated with the go/no-go decision, which also makes it relevant and impactful to H2@Scale.
- Although the efficiency may be lower, this approach has the potential to make large-scale electrolyzers meet the DOE's targets. The cost of hydrogen is a concern with the high operating voltage and the corresponding energy requirement.
- This work has the potential to reduce the capital and operating cost of water electrolyzers.
- The project consists of very relevant work. The true quantitative assessment of corrosion is uncertain. If corrosion is truly a minor issue, this work seems very promising.
- This alternative electrolyte route to steam electrolysis could have a noticeable impact if the cost models are reliable. However, it is unclear that assumptions such as "90% heat recovery in the heat exchangers" are feasible.

Question 5: Proposed future work

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

- The work seems to focus on performance increase. Because targets seem to be mostly met, some effort may shift to durability/corrosion. It seems that a thinner electrolyte may not be absolutely necessary; rather, a stable system at current performance levels may be sufficient to move forward. Performance could then be improved at a future time, once long-term durability issues are fully sorted out.
- The project team's future plans focus mostly on additional optimization and fine-tuning of the current approach. This is a reasonable path forward since the system seems to be meeting performance targets. In general, the slide on future plans (slide 23) does not include much detail on what will be done. There are no technical targets, alternate paths, barriers, or timelines presented on the slide to make it easier to assess the team's path forward.
- The future plans consist of addressing major challenges of stability associated with this class of materials.
- The future work is not very specific. It is unclear how matrix stability will be addressed. It seems that only three matrix oxides, supplied by Zircar, are in consideration, so it is unclear whether there are any custom formulations being considered with compositional or morphological changes. It is unclear whether the ternary electrolytes are expected to improve both conductivity and matrix/metal stability or if they will likely exacerbate the corrosion challenges. The plans for addressing the expected sealing challenges are not communicated.
- Corrosion mitigation of stainless steel is a concern, and the mitigation plan was not clearly defined.

Project strengths:

- This is a unique approach to hydrogen production that offers opportunities for lowering the energy requirements of hydrogen production. The methods for making the materials for this system are simple and straightforward, which will be beneficial for commercialization efforts. The authors are hitting all of their technical targets and milestones.
- The project leverages existing paradigms (OH- electrolyte solutions, HT operation of oxide-type cells, materials available from a commercial powder supplier) to demonstrate an alternative alkaline electrolyzer technology.
- The project has a novel approach that promises lower capital and operating costs, compared to polymer electrolyte membrane water electrolysis (PEMWE). This promises stability data at a 30-hour level at a good current density (600 mA/cm^2).

- The project has shown very good performance, and the team has a good approach for a potentially practical system.
- This is a good team with good progress.

Project weaknesses:

- The project does not involve much external collaboration. The current collaborator, UConn, has a relatively small role in the work. It is not clear whether the team needs additional help from external parties. The future plans and challenges were not very detailed, which made assessing the next year of work difficult. The energy and cost balance were a bit confusing on the slides and during the oral presentation. The comparison between HT alkaline electrolysis and PEMWE was difficult to follow. The assumptions and comparisons were not obvious. The project could be improved with a bit more consideration in detailing the cost/energy balance aspects of the work.
- The project does not share much detail on the methodologies and phenomena utilized, or what specifically is being targeted by the approach to improve the active components. This gives the impression that the electrolyte and matrix formulations are being explored by a relatively unambitious mix-and-match approach. This approach may yield some demonstrable progress in the technology, but it is unlikely to provide any breakthroughs.
- The local temperature of cells needs to be measured to avoid overestimating the cell performance from local temperature increases in the exothermic mode of operation.
- Longer-term testing is required to assess material stability.
- It seems as though collaborators were underutilized.

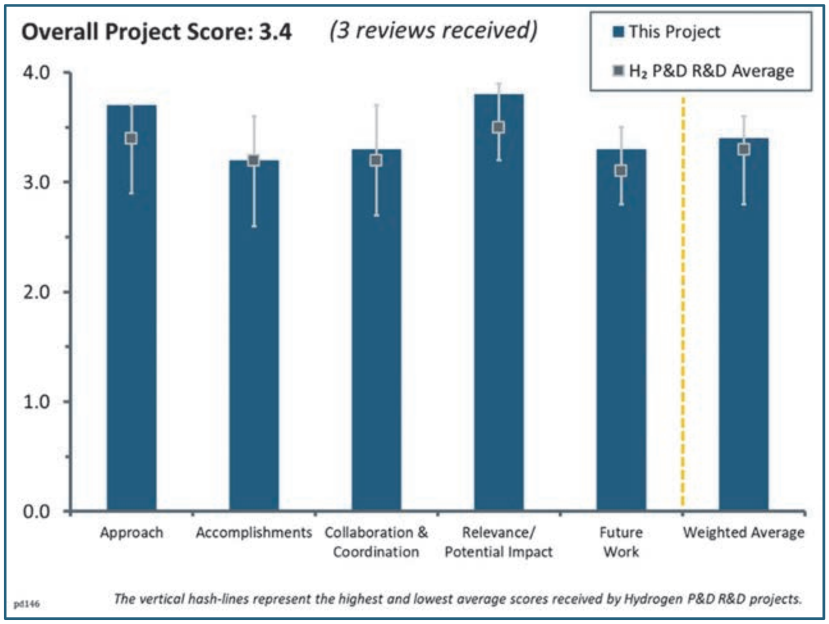
Recommendations for additions/deletions to project scope:

- The project could provide a more detailed cost/energy balance to better identify how this approach compares to more traditional technologies, such as PEMWE.
- It would be advantageous to include more analysis of the root cause of metal and matrix corrosion so it can be addressed.
- It is critical to address corrosion of stainless steel bipolar plates and inactive metal components.
- It may be wise to prioritize durability over performance; at present, performance seems adequate, and durability seems good.
- The project team should initiate long-term testing of the matrix and cell to gain a better understanding of material stability.

Project #PD-146: Advancing Hydrogen Dispenser Technology by Using Innovative Intelligent Networks

Darryl Pollica; Ivys Inc.

Brief Summary of Project:



The primary objective of this project is to develop a robust and cost-effective system for dispensing and measuring hydrogen; the system is meant to further enable widespread commercialization of fuel cell electric vehicle (FCEV) technology. Key project activities include (1) development of robust sensor hardware and algorithms that improve accuracy based on empirical testing and enhanced meter temperature measurement; (2) development, testing, and demonstration of the use of dedicated short-range communications (DSRC) for use in vehicle refueling; and (3) simplification and cost reduction of flow control and hydrogen pre-cooling systems.

Question 1: Approach to performing the work

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

- The project team is focused on critical dispensing issues: safety, accuracy, and cost. The project has a great set of collaborators to accomplish the objectives.
- The approach is comprehensive and well-focused.
- The approach is sound, although the description lacks detail. From the presentation alone, it is somewhat unclear how all of the equipment interacts and fits within the scope. For example, on the slide titled “Approach (4),” the Coriolis meter is not shown in the diagram, yet it appears in the slide titled “Approach (3)”; it is clearly a key part of the system.

Question 2: Accomplishments and progress

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

- The accomplishments achieved so far are good. The Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan (MYRDDP) target for accuracy has already been achieved. It appears further progress is required to lower the total cost of the system to meet future MYRDDP targets. Several mentions of no-cost extensions are present, and the project schedule should be maintained.
- The project team’s accomplishments are impressive.
- The principal investigator (PI) provided a good, convincing picture of this technology.
- This project is a year behind schedule and will likely need even more time beyond that. There is great progress on improving dispenser accuracy, although more information should be provided on (1) potential challenges (e.g., reliability and long-term accuracy under real station versus laboratory conditions), (2) how the team will assess the robustness of the design to those challenges (preferably beyond long-term field

testing), and (3) contingency plans. The presentation was completely silent on progress toward the cost-reduction objective. Although interaction with an automotive original equipment manufacturer (OEM) was identified in the summary, this critical aspect was not addressed in the plan.

Question 3: Collaboration and coordination

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

- There was a sufficient number of agencies and private companies listed in the collaboration and coordination section of this presentation.
- This project has an appropriate and qualified set of partners. The team has demonstrated collaboration with a Coriolis meter company, but strong interactions with other partners have not yet been demonstrated. This is primarily because project activity seems to be concentrated toward the end of the project, and the project has been delayed. It is suggested that the team engage the National Renewable Energy Laboratory and Air Liquide to learn about known challenges to station reliability, thereby minimizing surprises during site demonstration.
- The project has strong collaborators with excellent backgrounds in this field. An important aspect that is not made clear includes how frequently the collaborators met, shared data, or ran experiments for other team members. It does appear that future collaborations will be frequent in Phase 2.
- FCEV refueling station developers and vehicle/part OEMs should be included as advisors to gain market insight and improve the team's understanding of metering technology.

Question 4: Relevance/potential impact

This project was rated **3.8** for its relevance to/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The project is directly relevant to the MYRDDP goals. Project improvement has the potential to advance infrastructure goals.
- The results of this effort will be critical to the future success of the FCEV refueling market.
- Achieving accurate and reliable hydrogen dispensing at a reasonable cost is critical to the success of hydrogen refueling stations. Unfortunately, the cost axis was not addressed in this presentation.

Question 5: Proposed future work

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

- The development of a plan for improved low-cost manufacturing could be included in future work.
- The future work seems appropriate for this project. However, no work is suggested for planning on the addition of proper receivers to OEM vehicles. It appears that the PI expects OEM manufacturers to put the appropriate technology onboard vehicles in the future, though it is advisable to research and support this assumption.
- The plan for validating the dispensing equipment is good, but it appears the team underestimates the timing of the field testing phase or will not be able to do it justice because of its being cut short. It is strongly suggested that, if faced with this choice, the team seek to extend the project for adequate testing time rather than leave the demonstration partially finished. Several aspects are missing from the plan, including automotive OEM engagement and plans for how to close any shortfall in meeting the cost target.

Project strengths:

- The project seems well-laid-out. Excellent collaborators have been arranged for the project, and the proposed changes directly address the MYRDDP. The proposed technology changes take into account both cost and security.

- This project addresses a critical need. The project has an innovative approach, a good plan to demonstrate the technology, and a good collaboration team.
- The relevance, approach, accomplishments, and presentation of this project are all excellent.

Project weaknesses:

- The PI indicated that the addition of DSRC technology would lower costs, but no cost data was provided. Even a simple technoeconomic analysis would be beneficial. There is a given assumption that the vehicle manufacturers will include the proper technology in new vehicles. Should that not be the case, this technology will be rendered somewhat useless. The presentation indicated that no-cost extensions have been granted, perhaps more than once. This could indicate potential schedule problems.
- There is no OEM engagement. The technology will be useless if not adopted by the OEMs. It seems the team has not yet addressed the cost objective (based on information provided). There is an apparent presumption that Coriolis technology is sufficiently robust to work in real station environments for a reasonable lifetime (many years).
- The engagement of vehicle OEMs and refueling station developers could be helpful.

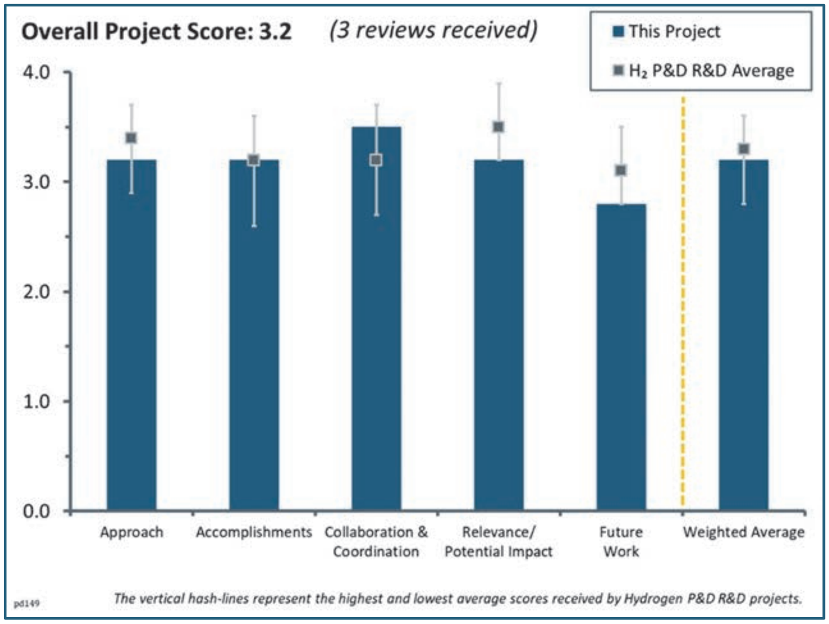
Recommendations for additions/deletions to project scope:

- No changes to the project scope are recommended.
- The presenter provided a good convincing picture of this technology.
- DOE should look for ways to take advantage of the improved dispenser accuracy before the proposed communications standard is adopted.

Project #PD-149: Hydrogen Dispensing Hose

Jennifer Lalli; NanoSonic, Inc.

Brief Summary of Project:



This project aims to develop a hydrogen hose for fuel cell electric vehicles (FCEVs) that is (1) engineered to be flexible and enable hydrogen delivery at less than \$2/gge, (2) durable in conditions of roughly -50°C and 875 bar for H70 (70 MPa) service, and (3) reliable and safe for conducting approximately 70 fills per day for more than two years. NanoSonic, Inc., is partnering with two national laboratories, a standards development organization, a local government, and industry to implement and test a cost-effective, metal-free, high-pressure hydrogen hose design that meets the above criteria, resists hydrogen embrittlement and contaminant leaching, and endures mechanical fatigue.

Question 1: Approach to performing the work

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

- The project, scope, focus, approach, and accomplishments are all excellent.
- The approach used for the work, performed from the third quarter of fiscal year (FY) 2017 through the second quarter of FY 2018, was well-thought-out. Many issues were considered thoughtfully, such as relevant performance properties of the produced hoses, partnerships that leverage expertise to fill knowledge gaps, and expectations on performance needed to get to the next stage. It is possible that some detailed modeling and analysis of the pressurized hose-fitting combination would have predicted the issues related to the fitting (i.e., slipping and lack of connection that would necessitate crimping). In hindsight, it appears that the fitting aspect presents a barrier that may prevent project success. Increased rigor on the analysis side might have enabled addressing this sooner.
- Goals were established to meet H70 pressure requirements (for burst and durability) and achieve low cost. Testing was not aligned to evaluate whether the hose will actually meet the needed pressure. Thus far, the failure has been at the fittings, but this failure has precluded determination of whether the composite hoses are feasible. It is concerning that there is an advancement of production capability prior to there being any proof that the hose can meet H70 burst requirements. It is unclear from the presentation whether the composite hose is compatible with the fittings. There was not a clear path forward as to how this severe challenge with the fittings is going to be overcome. The motivation for moving away from the current hose configuration was not clearly stated. Also, if hydrogen embrittlement of the metals in the current hoses is the issue, it is not clear that the current solution to use metal fittings will fully address the issue.

Question 2: Accomplishments and progress

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

- The project achieved the following critical criteria:
    - 3500 bar hydrostatic burst strength (>50,763 psi) held for one minute
    - 875 bar pressure cycle at (50,000x at -50°F and 50,000x at 85°F)
    - No contaminant leaching, competitive cost, mechanical durability, and environmental lifetime
- Progress has clearly been made. Pressure testing has been used to identify a failure mode not anticipated (i.e., slippage of fittings) and to quantify the pressure loads at which this failure occurs. Cardinal Rubber & Seal, Inc. (Cardinal) developed a very good engineering solution to bypass this failure mode. The principal investigator (PI) and her team have also identified supplemental activities (e.g., digital image correlation [DIC] and dynamic mechanical analysis [DMA] time–temperature super-positioning [TTS]) that provide insight on component performance through leveraged use of laboratory capabilities. Clearly, more testing is needed to determine whether this engineered solution is durable and enables the hose to reach the desirable maximum pressure limit.
- It is clear that work has been done, but the best results reach only 60% of the required goals. Thus far, the progress would not justify continued investment in the next steps of production and testing. Prior to further investment in this technology, there needs to be data showing the feasibility of the hose's being able to handle the operating loads.

Question 3: Collaboration and coordination

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

- The PI has engaged and added partnerships such that all those involved provide meaningful and substantial contributions to the development of this technology. These contributions are designed to leverage appropriate capabilities of each partner, thereby making effective use of the partnership itself.
- The project could involve FCEV station developers.
- The company has several collaborations and is reaching out to involve local suppliers and eventual customers, but it is not clear in all cases that these collaborations address the most pressing issue of meeting baseline burst pressure requirements. It is not clear how the Cardinal testing is relevant, as there is an inherent limitation on the company's testing capabilities; the testing is well below the pressures needed for the application.

Question 4: Relevance/potential impact

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

- Obviously, the results of this effort are critical to the commercial success of the FCEV refueling market.
- This effort has significant relevance to improving durability and lowering the component costs associated with fueling infrastructure. The project stands as an exemplar on how material properties and material interfaces can limit performance and proposes a scope of work to investigate these limitations (even though it may not have intended to do so).
- This project seems more like company-specific product development outside of the scope of the current research and development program. There are existing products that meet this requirement. It is not clear (1) that this technology provides a better solution, (2) that the lack of this product will inhibit the application of this hydrogen technology, and/or (3) that this approach will even be successful.

Question 5: Proposed future work

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

- The proposed future work is very important.
- The proposed future work of DIC for multi-strain imaging may provide insights on failure modes and mechanisms that occur at the fitting–hose interface. The DMA TTS work proposed will also provide some information regarding long-term testing and the durability of the component under abnormal handling conditions. That said, it is not clear to what degree that further burst pressure testing will be used and when it will occur. Perhaps that testing depends on the outcome of the DIC analysis. More clarity on the path forward—and what that timeline is now changed to—is warranted.
- The primary issue of not even coming close to meeting the H70 burst pressure requirement was not clearly recognized. There was no clear path forward for how this deficiency will be addressed.

Project strengths:

- The well-defined goal and the approach are project strengths. The project has outstanding partnerships that leverage each contributor's expertise and capabilities to achieve progress and further insight on hose performance. Given reliability issues that face the infrastructure industry, this effort should have a large impact once a final hose design and manufacturing method are established.
- Project strengths include good collaboration in the development process, good facilities, clear goals, and abundant data generation.
- The project plan, execution, and diligence in addressing challenges are impressive.

Project weaknesses:

- The project could have uncovered some of the critical issues related to fitting–hose interface behavior through inclusion of modeling and analysis of the manufactured system. It is not clear that this modeling and analysis was done. Also, the presentation did not provide backup materials, reviewer-only slides, or a data management plan. Finally, given the obstacles uncovered this year, the path forward and schedule are not sufficiently detailed to address how to get back to the original plan of high-pressure testing (or to address whether the project needs to do so).
- The project team did not clearly communicate the issues with the current technology and the unique engineering gap that this product will fill. The work has yet to come close to demonstrating whether the hose is even feasible; there is no clear path forward for how the fittings issues will be addressed. Many different parallel paths are being evaluated, and it was not clear which is the critical path and whether there are decision point criteria for continuation with each. Many technical issues remain unresolved:
    - Whether the metal fittings, which might still be used, are candidates for hydrogen embrittlement—or whether this has even been evaluated
    - How much costs will increase because of all of the modifications of the fitting process
    - What the plan to stop the slippage is
    - Whether the slippage is simply the result of the inherent differences in the composite hose and the fitting materials, and if so, whether this would preclude the achievement of the required pressures
    - Whether there is an alternate path for evaluating the hose's efficacy, thereby justifying all of the effort on the fitting modification
    - Whether there is a known quality control process for coupling these fittings to a composite hose, if/when an acceptable fitting is found
    - Whether there is any consideration of external degradation of the composite due to atmospheric environmental exposure (e.g., salt, ultra-violet radiation, etc.)
    - Whether relevant hydrogen-based degradation mechanisms of the composite are known, or whether tests have been performed

Recommendations for additions/deletions to project scope:

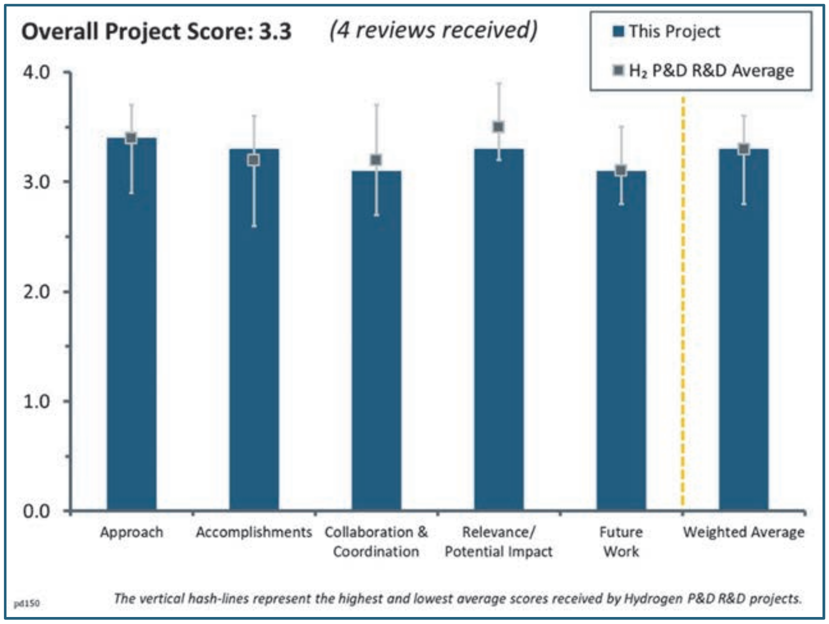
- Modeling and analysis of the manufactured system to uncover some of the critical issues related to the fitting–hose interface behavior could still be added to gain further understanding of anticipated DIC results. Also, modeling and analysis might predict further issues should the fitting–hose interface issue be resolved.
- The project should put a primary focus on demonstrating product feasibility and limit the scaling up of production capability until the product can demonstrate feasibility.

Project #PD-150: Coatings for Compressor Seals

Shannan O'Shaughnessy; GVD Corporation

Brief Summary of Project:

Seal failure is a major contributor to hydrogen compressor maintenance, adding significant downtime and cost to compressor operation. The goal of this project is to improve seal life in hydrogen compressor systems by three to five times. The work focuses on two different types of coatings. For static seals, the project will develop barrier coatings that mitigate hydrogen ingress into the seals, which prevents premature failure. For dynamic seals, low-friction coatings that reduce wear and extend seal life will be developed. A room-temperature polymer vapor deposition process will be utilized to produce thin polymer coatings for both types of seals.



Question 1: Approach to performing the work

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

- The project has a sound approach to address a critical barrier that affects hydrogen compression, storage, and dispensing reliability. The methodology is both sound and well-thought-out, and it uses a mixture of insightful scientific investigation with clever engineered technology to understand and qualify the effectiveness of the barrier coating solution.
- This project contains promising material for reducing maintenance and cost.
- The issues with the current state of the art and the goals of the current work were established. Reasonable approaches were presented for both lubrication and barrier coatings to inhibit hydrogen permeation. The evaluation approach was reasonable; however, it could be improved by evaluation of the permeation *after* pressurization loading. There is some concern about the durability of a 2-micron-thick polymer/inorganic barrier coating in maintaining its function after pressurization and/or wear loading.
- The project team addresses the challenges with hydrogen material interaction as a barrier and assumes improved sealing treatments as the solution. However, the material has not yet been tested in hydrogen, and there are many assumptions related to hydrogen's and helium's being similar, based on the size of the gas. In fact, solubility has a significant role, and the two are completely different.

Question 2: Accomplishments and progress

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

- The project is developing promising materials for lowering maintenance needs.
- The project team has made good progress on performing coatings with the material. The material still needs to be hydrogen-tested. The barrier material needs to undergo pressure cycling, or at least a rapid decompression test after a 24-hour hydrogen soak test, to determine if there is any blistering or delamination of the coating. The coatings on the compressor seals are a good idea, but there has not been

enough work on the durability. The work at Powertech looks promising, but there is no characterization discussed based on the number of cycles endured or whether there was a transfer film on the mating surface. More durability testing on the coating is suggested. It would be good to determine the wear rate, and what impact the mating surface characteristics have on that wear rate.

- The project team has made good progress in making the materials efficiently. The project itself consists of good initial data, although the assumption of helium's being a conservative proxy for hydrogen is questionable. Hydrogen will likely have a higher diffusivity, which would be a relevant parameter. There was no significant reduction in permeability, even without possible mechanical-loading-induced defects. The team has produced excellent lubrication results.
- The project team has made substantial progress in characterizing the morphology, properties, and behavior of barrier coatings to prevent wear and hydrogen degradation. Further analysis in still warranted, such as the examination of the effect of contaminants/particulates on coating stability and the economic cost of the coating relative to system setup and other capital costs.

Question 3: Collaboration and coordination

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

- The project's collaborators have well-defined scopes of contribution that complement one another. These contributions leverage skills, expertise, and capabilities at each institution to gain further clarity on the robustness of this particular technology solution.
- This project has a solid group of collaborators.
- A fair number of references were provided.
- There was discussion of working with other partners such as PowerTech, but the contribution of partners was not obvious during the presentation. It was thought that GVD Corporation was doing the helium permeation testing, but it was Green, Tweed & Co., based on the collaboration table. The collaboration slide was the best at explaining this. Oak Ridge National Laboratory (ORNL) has good hydrogen permeability capability and experience that could be shared with GVD Corporation; it was not clear that this body of knowledge at ORNL was being leveraged.

Question 4: Relevance/potential impact

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

- The project has good relevance to technology barriers and presents a method to significantly enhance the reliability of fueling technology. The project also does a good job of anticipating potential issues with the proposed solution and shaping activities to pre-emptively address them.
- The project is relevant and could have a great impact and provide solutions to some current problems.
- The project aims to reduce the cost of maintenance.
- The project team address an important topic that is a cost-efficiency limiting problem. The impact will be more rigorous once the behavior after loading is known. The benefit of the initial coating was very modest. It seems as though this could be improved by additional layers, etc. This type of work could have a broader impact in other areas.

Question 5: Proposed future work

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

- The project is definitely ready to progress to hydrogen, as the presenter discussed, and the compressor testing is the right next step.
- The proposed future work appropriately addresses remaining knowledge gaps and anticipated modes of failure.
- Good short-term and long-term plans were provided.

- The project team could have challenges in meeting the 10-fold reduction in hydrogen permeability, based on initial results; this should really be tested after the system has been mechanically loaded. This work should include some study/characterization of the mechanical degradation of the coating.

Project strengths:

- The project is very clear on goals, approach, accomplishments/results, and roles of partners. Technology development demonstrates a good initial effort toward commercialization. The project has a reasonable data management plan, and technical details are well presented. The team has foresight on potential issues with the proposed solution and has planned actions to mitigate these issues.
- The project consists of an interesting and innovative technology that shows promise in coating, even in tight crevices. The lubrication results are excellent. The development of both an efficient process and a proper testing apparatus are strengths of this project. This technology has the potential for wide applicability and has a targeted insertion point.
- The project has some real value in addressing gas diffusion into materials and wear issues in compressors.
- This project is pretty good.

Project weaknesses:

- Permeation has not yet been evaluated under realistic conditions. Specifically, the effect of mechanically induced damage on such a small-scale polymer/inorganic coating system will be critical—yet the compatibility of the polytetrafluoroethylene (PTFE) and the polymer/inorganic with the hydrogen is not addressed. The project team has not stated whether there will be expected degradation and, if so, over what timeframe. There are also several technical concerns:
  - The team has not discussed whether there is a compatibility issue with these coatings possibly contaminating the hydrogen if they wear off.
  - It would be useful to compare this coating to competing technologies for permeation barriers.
  - It is unclear how the team plans to verify the uniformity of the coating. Also, little was mentioned on the application-induced defects of the coating—this seems like it could be critically important. It is unclear whether there are standard quality assurance practices for this and how this could impact the cost.
  - Perhaps the team could correlate the 20% increase in cost with a percentage increase in lifetime, thus making a business case that this product would be worthwhile.
  - The project team should also clarify whether there is a temperature issue with the durability and the performance of the coatings.
- The team should continue to address durability issues, especially with the barrier coatings on the seals. Rapid decompression after long-term soaks is a good test of whether the barrier is defect-free and the durability of the coating still stands. The wear rate and mating surface influence should be addressed. There are tribology standards, such as pin-on-rotating-disc or reciprocating pin-on-disc, that could be utilized for wear testing. Different sliding surface roughness could influence the wear rate; it is not uncommon for transfer films to build on the mating surface that provides low friction and wear. This should account for mass loss. If not, the mass would be lost as a particulate, which would not be beneficial to the particulate levels in the hydrogen.
- The only real weakness is the lack of detail on the cost of barrier coatings relative to capital costs. Some knowledge is obviously there, but the details are omitted. Also, clarity is needed on total permeability reduction for hydrogen exposure. This was not always clear in the presentation.
- The results have limited applications.

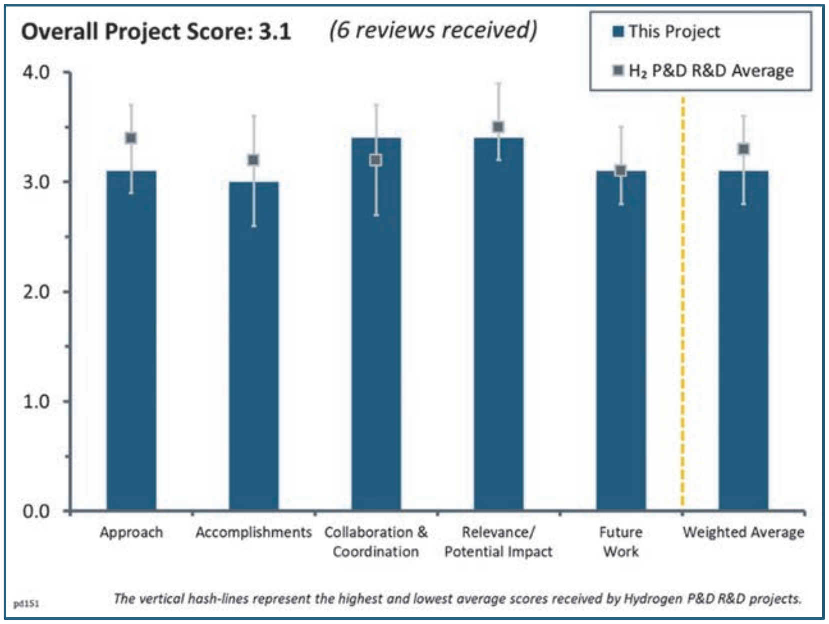
Recommendations for additions/deletions to project scope:

- No scope changes are needed.
- It is strongly recommended that the coatings be evaluated after mechanical loading.
- It is recommended that the team add surface roughness characterization on the mating sliding surface.

Project #PD-151: New Approaches to Improved Polymer Electrolyte Membrane Electrolyzer Ion-Exchange Membranes

Earl Wagener; Tetramer Technologies, LLC

Brief Summary of Project:



The project seeks to develop improved polymer electrolyte membrane (PEM) electrolyzers that will minimize physical and chemical degradation and enable the cost-effective production of hydrogen, enhancing grid stabilization and facilitating remote renewable energy storage. Tetramer Technologies, LLC (Tetramer) will optimize ionomer molecular architecture and membrane configuration with a goal of developing a membrane material superior to Nafion in terms of performance, durability, and cost. A final, down-selected polymer material will be scaled up. The project team is partnering with Proton OnSite, which is providing insight on membrane requirements and testing membrane materials.

Question 1: Approach to performing the work

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

- This approach seemed reasonable for a Small Business Innovation Research (SBIR) project for developing an optimized ionomer. The project team worked on proving the feasibility of the ionomer process in Phase I before moving to making dozens of ionomers for testing in Phase IIA, at which point the project exceeded expectations and thus received Phase IIB funds to scale materials for prototyping an electrolyzer with Proton Onsite.
- The project team approach seems reasonable. Proton Onsite is a solid partner to direct this work. More information on the design iteration process and how the team approaches the next iteration for this project is desirable.
- The project has a very good approach that combines an appropriate backbone and conductivity along with durability.
- This novel approach has promise to significantly reduce cost.
- The authors are designing conductive polymer architectures for the ionomers/membranes in an electrolyzer. The polymer design, synthesis, and execution are very interesting. The authors do not provide any technical targets (for the polymers or the integrated cell) or milestones on the project. This makes it difficult to evaluate the approach fully because it is not obvious what targets the authors are aiming for, nor how their approach may or may not get them there.
- The project team does show some improved performance relative to Nafion (slide 6), improved swell control (slides 8 and 9), and improved down-selected membranes (slide 13); however, these are mostly relative targets that evaluate the system relative to Nafion or that highlight a net improvement relative to one of the project's own polymers. It would be useful to see absolute performance targets for the polymers and/or integrated cells. While the project team aims to improve the durability and efficiency of low-temperature water electrolysis cells/stacks, the presentation did not include specific goals in those areas. Showing voltage–current (V-I) curves without providing an example of how the improvements will affect

the efficiency and cost of Proton Onsite's 250 kW stack, for example, is concerning. Goals listed on slide 4 include "Work closely with Proton [Onsite]," which is a really good thing, but then "build a prototype [and] assess performance…in customer trials" will not happen with Proton Onsite. It is possible that Proton Onsite will want to see tens of thousands of hours of benchtop operation/performance before any stack ends up in a commercial (i.e., customer) system. The project team should work more closely with Proton Onsite and ask Proton Onsite to provide guidance on how to develop a plan to systematically achieve the project goals.

Question 2: Accomplishments and progress

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

- A new polymer that appears to exceed the performance of current membranes is a very promising development.
- It is always impressive to see an SBIR project progress the way this one has, from Phase I to Phase IIA, and now to Phase IIB.
- This aspect of the project is difficult to assess because the authors have not provided technical targets, milestones, or go/no-go decisions. The data within the presentation shows performance relative to Nafion or performance relative to other polymers in this study. The authors need to define some absolute technical targets for their polymers and cells. It would also be helpful to see some targets for costs and performance lifetime. The authors do make reasonable progress on the development of these materials. Slide 6 does show improvement relative to Nafion. The improvements in swelling (slides 8 and 9) and durability (slide 7) and the results on conditioning (slide 10) are noted as examples of progress on this project.
- The polarization curves help show the relative improvement over other candidate materials, but absolute targets would help explain how this work would improve over the state of the art. With Proton Onsite as a partner, this data should be easy to show next year. Slide 6 shows the polarization curve for Tetramer Series A (3 mil), which seems to be highlighted on slide 7 showing the 1000-hour durability run. On slide 6, the voltage of this cell at ~1.85 A/cm^2 seems to be around 1.8 V, while slide 7 shows the cell performance closer to 2.05 V. The reason for the apparent disparity is unclear.
- The project results have shown promising durability at 1000 hours. Hydrogen crossover needs to be further reduced. A cost analysis is mentioned, but no results are reported in the presentation. Little or no progress was made compared to the Phase IIB baseline membrane.
- This year, the project focused on incremental improvements when compared to the baseline for last year. The project team should focus on providing metrics for comparison for each graph provided. For example, the project team should show what an acceptable hydrogen crossover level is and how it compares against the measured level. The continuity between samples/graphs need to be clarified. It is unclear what the conditioning process is or how it can be used as a performance metric if it changes per test. Although this year's work was ambitious, it seems that only one sample performed better than the baseline, and it needed to be conditioned through five cycles to achieve this result.

Question 3: Collaboration and coordination

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

- The authors explained that they represent a polymer research and development company focusing on low-technology-readiness-level (low-TRL) efforts of approximately TRL 1–3. The team established a collaboration with Proton Onsite to help with incorporating these materials into an electrolyzer and conducting performance testing. Proton Onsite is also helping to address hydrogen–oxygen crossover in their cells. This is a smart collaboration and helps round out the technical capabilities of the team, providing a balance of strong polymer/chemistry skills and strong skills for engineering/deploying electrolyzers.

- Proton Onsite is a strong partner to help keep this project focused and informed. The project leads should consider asking the project manager at Proton Onsite for advice on how to approach this work more systematically.
- The project has a good collaboration partner who is a leading manufacturer with a commercial presence in this area.
- The collaboration with Proton Onsite is good.
- Proton Onsite is a solid partner for this project. The only small critique is that it is unclear how this project fits into Proton Onsite's vision moving forward.
- This project is not really aimed to be involved with other institutions. Proton Onsite is supposed to be involved later in the effort and has not been a major part of the project yet.

Question 4: Relevance/potential impact

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

- This project is right in line with the efforts toward what the Fuel Cell Technologies Office (FCTO) aims to do: lower the cost of hydrogen production. It could make units easier and cheaper to produce and get more electrolyzers out into the market, which would be a win-win for the renewable energy sector.
- The performance, durability, and cost of membranes are very important to advancing PEM electrolyzers.
- A promising alternative membrane will lead to quicker transition to commercialization.
- The collaboration with Proton Onsite will ensure this work is relevant.
- The efficiency and durability of low-temperature PEM electrolysis is aligned well with DOE and H2@Scale goals. However, even with modest efficiency improvements, products of this research may not be introduced into Proton Onsite's systems unless the project also meet cost targets. The presentation and work seem to be lacking in this area.
- The project supports the general goals of the H2@Scale program, specifically, to create technologies that enable the deployment of hydrogen energy. The project, as presented, is missing any links to the H2@Scale program goals, technical targets, etc. The project does not list any milestones or other metrics that could be used to assess how well it supports the program targets/goals/metrics. This is one aspect of the project that needs to be improved for next year's Annual Merit Review.

Question 5: Proposed future work

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

- The future work looks good.
- The proposed future work appears adequate.
- The project has a good plan for scaling up and working toward mass manufacturing. Hydrogen crossover is not addressed in future work.
- The project team should stay focused on the down-selected materials that they have found. This focus will help the project partner assess progress and meet the project's goals.
- Future work is described, briefly, on slide 17. It seems the plan is to continue with developing the material sets described in the presentation. The authors describe more casting trials to assess performance/ consistency. During the oral presentation, the authors mentioned how well this approach was working for assessing scalability. The authors also indicate they will perform a cost analysis. These general plans all seem fine at face value but lack useful detail. The future work plans would be improved if there were technical targets, metrics, or other performance goals included. For example, "optimizing cell design" is an okay future plan, but it would be improved if we knew what the performance targets were that the authors wanted to hit (even if the optimized cell does not get one there). In general, the lack of technical targets/ metrics in this work makes it difficult to assign a better score to many facets of this project. It would be an area to improve for next year's presentation.
- The project plans to do a down-select of the approximately 19 membranes the team is currently working on in Phase IIB. The materials will then be scaled for prototyping in a PEM electrolyzer with Proton OnSite.

More details into how that down-select will be done and in what timeframe would have been helpful in understanding how this project moves ahead in the near future.

Project strengths:

- This is a nice approach for developing polymer materials for PEM electrolyzers. The materials presented seem to perform better than Nafion in some aspects. The materials seem to be durable, performing for ~900–1000 hours of testing. The partnership with Proton OnSite seems to round out the technical skills of this team, making for a very robust and effective partnership.
- Improving efficiency while maintaining durability and reducing costs is critical for PEM electrolysis. The cells/stack make up a large percentage of the overall system cost, so the area of focus in this project is excellent. Tetramer has a strong partner with Proton OnSite, so Tetramer should tap into Proton OnSite's expertise in project management as well.
- This effort has a very specific and targeted task: progress beyond the capabilities of Nafion materials. The focused goal is an advantage in that it keeps the project on task, and it aligns with FCTO goals of getting more electrolyzers deployed.
- The project has a strong approach to polymers and strong experience with polymers.
- The approach, accomplishments, and partners are all strengths of the project.
- The project has a very solid approach to developing a new membrane.

Project weaknesses:

- The project does not list any technical targets, goals, or metrics for either its own work or from the H2@Scale program. It is difficult to assess this work without knowing targets. This is an area that should be addressed for next year's presentation. Currently, there is no cost analysis on the polymer materials or manufacturing costs. Even a simple analysis can serve as a "gut check" as to whether the systems can be commercialized if the performance can be engineered to meet technical targets. This is another area that should be addressed for next year's presentation. The project did not list any milestones or go/no-go decision points. It is difficult to assess progress without having the milestones included. This is a third area that should be addressed for next year's presentation.
- The project seems to lack initial cost estimates to ensure that the down-selected materials will help reduce the cost of the stack while increasing efficiency and maintaining performance. Showing a lot of V-I polarization curves without providing context of how the improved cells affect the goals (with quantified metrics in units that can be understood by people outside the project) is frustrating.
- No cost analysis was presented. The effort appears to lack measurement and quantification of membrane conductivity and of the mechanical properties of the improved membranes.
- It is not clear whether swelling is under control. Crossover is a concern.
- It is not entirely clear how the next part of the project will occur.

Recommendations for additions/deletions to project scope:

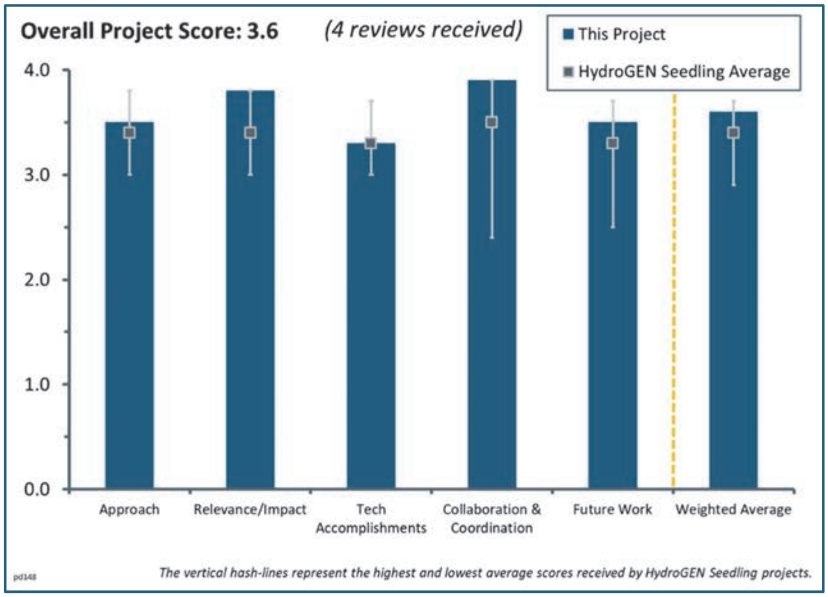
- No change in approach is needed.
- The project team should perform and present a rigorous cost analysis for the membranes. The team should also increase efforts to reduce gas crossover, especially given the reduced membrane thickness. Finally, the team should perform and present quantitative characterization of the membranes.
- A detailed cost analysis of the materials and their manufacturing costs with comparison to current state-of-the-art materials being deployed in electrolyzers should be added to the project scope.
- The project team should clarify when and how the down-select will occur and determine what exactly Proton OnSite will do in this project.

Project PD-148: HydroGEN Overview: A Consortium on Advanced Water-Splitting Materials

Huyen Dinh; National Renewable Energy Laboratory

Brief Summary of Project:

The HydroGEN Consortium's objective is to facilitate collaborations between federal laboratories, academia, and industry to evaluate and accelerate the research and development (R&D) of innovative, advanced materials that are critical and necessary to advanced water-splitting technologies for clean, sustainable, and low-cost hydrogen production. Water-splitting technology pathways supported by HydroGEN include (1) photoelectrochemical, (2) solar thermochemical, (3) low-temperature electrolytic, and (4) high-temperature electrolytic.



Question 1: Approach to performing the work

This project was rated **3.5** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The HydroGEN steering team appears to be working collaboratively and effectively in setting up the Energy Materials Network (EMN), defining capabilities within the network of member laboratories, and defining the barriers to the technologies being pursued. It is still premature to evaluate the overall effectiveness of HydroGEN at this point in time in terms of the role HydroGEN has played in helping to advance the technologies being developed, through projects that utilize the capabilities within HydroGEN.
- Bringing all hydrogen pathways with water-splitting materials under one umbrella is a good approach that can perhaps lead to more focused efforts to efficiently explore and meaningfully assess technoeconomic performance of the materials. The project should consider setting a guidance for near-term success measures (different from U.S. Department of Energy targets) that is common to all pathways.
- The overall HydroGEN effort is undertaken by several dozen groups. The overall approach seems sound, with a mix of projects devoted to hydrogen generation technologies at varying levels of technical maturity: high-temperature electrolysis (HTE)/low-temperature electrolysis (LTE), photoelectrochemical (PEC), and solar thermochemical (STCH) routes.
- The work is well-designed and feasible. It contains useful networking/engagement tools.

Question 2: Relevance/potential impact

This project was rated **3.8** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- Given that HydroGEN is in the early stages of development, it appears to have a significant impact, considering that nearly half of its 80 nodes are being utilized and that multiple nodes are generally being utilized by a single project.
- Looking beyond the somewhat self-referential nature of this question in this case, HydroGEN fits well within the objectives of DOE and the Fuel Cell Technologies Office (FCTO). Hydrogen, as an energy

vector, impacts many industrial sectors: transportation, energy generation, improving the efficiency of industrial processes, etc.

- The apparent return on investment (i.e., time and money) is very good, and the initial reaction/enthusiasm from the community is impressive.
- The HydroGEN Advanced Water Splitting Materials (AWSMs) effort is in full alignment with the EMN and U.S. DRIVE Partnership objectives. The effort provides a forum and funding for breakthrough and incremental technology development opportunities.

Question 3: Accomplishments and progress

This project was rated **3.3** for its accomplishments and progress toward overall project and DOE goals.

- The fact that about half of the 80+ nodes (capabilities) are being utilized suggests that the interaction with the HydroGEN-supported R&D projects/community is a benefit toward helping DOE realize its goals. It is unclear how to put into perspective the number of users, page views, downloads, etc., as well as the publications and presentations, and whether these are helping DOE achieve its goals/targets. As HydroGEN "matures," a better metric would be clear evidence of how the nodes had an impact in making measurable/ quantifiable benefits toward advancing R&D to meet DOE goals.
- The presented work under consideration, which covered Consortium project updates for the four technology focuses (i.e., HTE/LTE, PEC, STCH), represented the overall HydroGEN program, as well as several projects within the program. This necessarily limited the communication of many distinct accomplishments in any one presentation in the afternoon session. There was the sense that there was abundant progress in the awarded projects, but perhaps a more coordinated effort between the four presentations to avoid redundant information (though there was not a huge amount) would have bought some more time to focus on a few more project highlights rather than relying on the poster sessions to convey that information to the audience. Some of the secondary, visible metrics, such as the use of the data hub (~250 data files in a year), should get more emphasis either on boosting participation or in communicating the complexity of the data contained within the hub. There is ambiguity as to what a single file contains: whether it is a single resistance measurement or a summary from an entire collection of measurements from a unique tool on the beamline. In short, if the scale of the databank were conveyed in person-hours per data file that makes up the ~250 total, that could strike an audience as more impressive/ appropriate than leaving the number of files to remain as an abstract concept, which risks sounding underwhelming.
- The launch of the searchable website on capability nodes and the data hub is an important initial accomplishment. That said, it may be a good idea to revise the effectiveness measure used for these data-sharing tools. The number of files shared does not mean much without some kind of quality check. Also, the ongoing parallel work by Proton and its project team in support of the benchmarking of AWSMs is important and long overdue.
- More time/data is necessary to fairly assess progress in this regard. The initial progress is very positive.

Question 4: Collaboration effectiveness

This project was rated **3.9** for its collaboration and coordination with HydroGEN and other research entities.

- The concept of the EMN has been relatively recently created, and HydroGEN is among the first programs to leverage it. The requirement to leverage the EMN, among other things, makes this inherently a strongly cooperative, collaborative project. Hopefully, the successes of this effort can serve as a model for other similar efforts in the future.
- HydroGEN is doing an excellent job leveraging resources and encouraging collaboration.
- There appears to be a strong sense of positive collaboration among the four national laboratories involved in HydroGEN.
- The organizational leadership structure design across the six national laboratories makes collaboration inherent and necessary across the entire AWSM R&D portfolio. However, it is not obvious whether there is a collaboration with other HydroGEN activities such as H2@Scale.

Question 5: Proposed future work

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

- The proposed work looks great. If possible, the project should accelerate the benchmarking effort so that the results are implemented by the AWSM teams sooner, although it is understood that this point is outside the scope of this review.
- Probably one of the most pressing issues under Proposed Future Work is the development of an effective data management program, not so much in managing the data but in presenting to the R&D community in a format that is of value to the community. Developing benchmarking standard protocols and metrics is another pressing issue to make sure that the protocols for evaluating the various technologies provide an apples-to-apples comparison. Regarding the alignment of core national laboratories with the go/no-go decisions of seedling projects: the role of HydroGEN is to "help" these technologies succeed. One comment, which may or may not be under the "control" of HydroGEN, is the consistency in go/no-go metrics, in terms of a consistency in the quantitative metrics for how far the metrics go toward pushing technology development forward.
- The HydroGEN team should keep up the momentum. It is important to pay close attention to metrics and be sure to adjust to retain interest.
- Most of the projects funded by HydroGEN are still ongoing. The focus was more on current work and results.

Project strengths:

- The collaborative aspect of requiring projects to work with the EMN, creating a search engine for capabilities, and creating and utilizing a data hub are all strengths of this project. The strengths are all tools that may help the impact of the awarded projects to extend beyond their individual groups and to last beyond the scope of the funded projects. Also, it is staggering to see how involved certain node principal investigators are with so many of these projects.
- There is good collaboration among the national laboratories involved in HydroGEN, without the appearance of any turf battles over expertise that may be located within more than one partner laboratory. The identification of a large number of nodes (capabilities/expertise) that are being highly utilized at this early stage suggests that there is good value in the nodes identified so far.
- The project's strength lies in solid leadership and organizational structure, as well as the inherent collaboration across the six national laboratories.
- The strengths of this project lie in the collaboration, resource utilization, and the data storage/sharing (in theory).

Project weaknesses:

- The categorizing of nodes as Categories 1–3, with Category 1 being the most developed and Category 3 being the least developed, is a good strategy. What is in question is how the nodes are funded. For example, if a node is not initially called out in a proposal, it is not funded. That is fine for Category 1 nodes that are not funded because they are the most mature technologically, but for Category 3 nodes, it could be an issue if the node were to be of value to a later funding opportunity announcement awardee but was not fully developed to the point of being of value. Data management in terms of how much data has been made available so far seems very limited, given the level of funding and the number of nodes participating in the projects; the form in which the data is made available seems to be an issue. It is uncertain how to even determine how effective the data management has been to date and how effective will it be going forward.
- The category readiness level classification was confusing. There is an upward increment of technology readiness levels (TRLs) to show increasing maturity, whereas the commercial readiness levels (CRLs) decrease. The project presenters would simply speak of "readiness levels," and the meaning was unclear without more context. On the other hand, the CRL counting down to 1 makes the boundaries of the project scope clear. The classification tops out at CRL 1, so there is clarity in that fixed endpoint.
- Although individual projects have milestones and go/no-go points, the AWSM Consortium lacks clear success metrics at a higher level to guide its pathways and projects.

- The data use metrics were unclear. The metrics of user "engagement" were also uncertain; it is unclear how many of the users are participating and at what level.

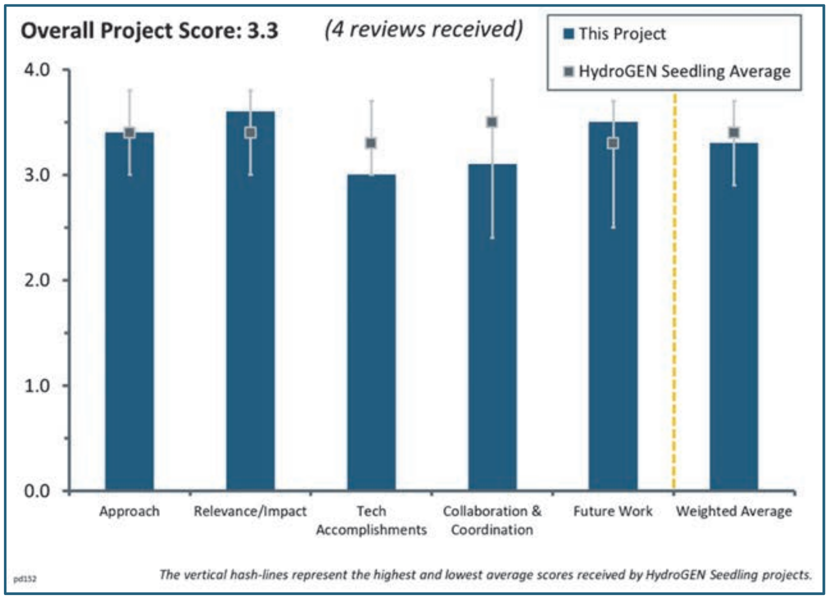
Recommendations for additions/deletions to project scope:

- The project team should consider making a distinction between long-term technologies (e.g., STCH, PEC) and near-term ones (e.g., LTE)—perhaps in some form of TRL numbers, graphics, colors, or financial tags. That way, the reader has a reasonable understanding of the relative commercial readiness and R&D effort level of the various pathways. The project should consider setting near-term success measures or expectations that are common to all pathways.
- Clear metrics for user engagement and activity are recommended.

Project #PD-152: Proton-Conducting Solid Oxide Electrolysis Cells for Large-Scale Hydrogen Production at Intermediate Temperatures

Prabhakar Singh; University of Connecticut

Brief Summary of Project:



The primary objective of this project is to identify novel materials and processing techniques to develop cost-effective and efficient proton-conducting solid oxide electrolysis cells (H-SOECs) for large-scale hydrogen production at intermediate temperatures (600°C–800°C). New proton-conducting electrolytes, tailored hydrogen and oxygen electrodes, and optimized cell designs for lowering the electrode polarization and resistive losses will be developed. Following synthesis and characterization of new electrolyte and electrode materials, they will be used for the fabrication of SOEC single cells and tested for performance and durability. Degradation mechanisms will be developed and materials chemistry and component structures will be optimized to mitigate any degradation.

Question 1: Approach to performing the work

This project was rated **3.4** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- This is a good project. It should seek to map physical outcomes as a function of, or a correlation to, chemical "income." The project could probably improve upon the vertical interaction among team members. For example, the undergraduate student has never had a direct technical conversation with the chief scientist at PNNL. While it is not expected that such interactions should be an emphasis of the project, it does seem like a missed opportunity. The metrics to demonstrate project impact are slightly more evolved than those seen in other projects. For example, instead of just saying the system needs to be "stable," the project states that the performance degradation rate must be less than 4 mV/1000 h. This specificity is commendable. Some additional thought may need to be expended on the benefit of having "uniform bulk phase composition." Matter moves as waves, and the periodicity of "defects" should not be overlooked as having no role in this. The technology need not obtain 100% densification. Indeed, having such perfectly dense material may be detrimental to the system. This is nice from a practicality perspective. The idea of using a getter to keep chromium content down in the system seems effective. The preliminary work of varying the chemical constituents in the atmosphere during densification of the electrolyte seems to allow some divergence in the electrical conductivity at higher temperatures. To date, this work has involved only dry and wet conditions of air and nitrogen. The addition of other interesting oxidants could lead to more changes in conductivity.
- This is a sound approach. The sintering aid seems to be working well. It would be good to see some assessment of a sintering aid "sink" or "final resting place." Given that most of the ZnO vaporizes and some trace Zn must remain, the question remains about where the final ZnO mass accumulates and whether there are any materials that must be avoided to avoid later contamination, degradation, or otherwise detrimental outcomes.

- Evaluating systematic screening of sintering aids is a good approach. Exposure to hydrogen needs to be done to make sure that the sintering aid oxide does not precipitate out along the grain boundaries—and if it does, that it is not detrimental to electrical and mechanical properties.
- The high-performing proton conductor $BaZr_{0.1}Ce_{0.7}Y_{0.1}Yb_{0.1}O_{3-\delta}$ (BZCY-Yb) and ZnO sintering aids have been in high-temperature proton conductor literature for several years. It is unclear where the innovation is, other than in the high-throughput compositional analysis that will be done at NREL and the atmosphere composition effects during sintering. The approach seems incremental but could yield some improved processing steps for high-temperature electrolysis.

Question 2: Relevance/potential impact

This project was rated **3.6** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- Apparently, dropping the sintering temperature by even 100ºC for the electrolyte greatly changes the sintering behavior. It also leads to cheaper heating elements, cheaper insulation, and other benefits. Being able to operate between 550º and 750ºC is important because this brings the technology into a space in which some material development efforts may be shared with concentrating solar power, which is another renewable energy technology that is also targeting 750ºC operation. In the long run, this could help develop the critical mass needed to drive costs down. The milestones seem okay, being SMART (specific, measurable, assignable, realistic, and time-related). The go/no-go decision point is especially strong. The value of the achievement could be better emphasized by perhaps also including some comparison of the change(s) versus the state of the art (i.e., a relative percent change).
- Yttrium-doped barium zirconate (BZY) is notoriously difficult to densify, so any improvements in processing could benefit other uses of the material. Lowering the sintering temperature and times will lower processing costs and throughput times.
- This project is extremely relevant with a high impact and makes a very good case for hydrogen production via high-temperature H-SOECs.
- Proton conductor-based electrolysis aligns with DOE objectives. The opportunity to produce dry hydrogen is very attractive.

Question 3: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and DOE goals.

- Meeting the conductivity target is a good accomplishment. The approach to modifying the dopants to achieve target conductivity is well-thought-out and well executed.
- The project goals are on track. Good progress has been made on milestones.
- The addition of ZnO produces a visually stunning result in sintering at 1350ºC. Tin has also been evaluated. The strategy/scientific rationale behind which elements are selected for the sol gel synthesis could have been better articulated. While it is obvious that NREL is providing support for the "...investigation of combinatorial libraries of Y-substituted BaZrO3," it is not clear whether the effort is purely combinatorial. In other words, it would be good to know whether all the combinations defined by mathematics are being tested or whether some scientific intuition (or better yet, some scientific hypothesis) is being tested. Three of the five milestones have been met so far for budget period 1. It seems likely that some of these milestones were perhaps not challenging enough. The project should consider incorporating some "stretch" milestones in the future such that, if these are met by the researchers, it will be considered an outstanding achievement. If, however, the stretch milestones are not met, the project would not be punished. This will provide some internal calibration for what the project team truly considers challenging in this vein of research.
- There is no apparent evidence of significant experimental progress. It seems that some fabrication basics are still being ironed out. The material sets at play are not novel, so more detail is to be expected on the processing studies, as well as more progress on them. It is unclear why an oxygen-ion-conducting SOEC (O-SOEC) is being tested as a benchmark. The materials and temperature of operation are different. It

would make more sense to compare H-SOECs using more conventional high-temperature processing (or simply literature data) as a benchmark.

Question 4: Collaboration effectiveness

This project was rated **3.1** for its collaboration and coordination with HydroGEN and other research entities.

- Collaboration seems to be effective based on milestone progress and reports from each group. Work appears thorough, but also independent.
- Good interaction is occurring with the nodes.
- NREL work will be valuable when the data from the laboratory can be used to inform the University of Connecticut (UConn) fabrication and data collection. INL helped UConn make dense electrolytes. It is to be hoped that these collaborations with capable EMN nodes will bear more fruit in the months to come.
- The collaboration with INL to receive half cells appears effective. The role of PNNL is not clear from the presentation. PNNL could contribute in the immediate term by determining the stability of the electrolyte compositions in a steam environment at the target operating temperature.

Question 5: Proposed future work

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

- The proposed future work is a logical extension of the existing work. Some of the proposed future work addresses the comments and concerns listed in prior review criteria. There is a good use of SMART metrics for future work. The impetus for the technical and scientific undertakings seems clear, insofar as it relates to the project objectives, but the scientific hypotheses being tested could be better (explicitly) defined.
- There is good momentum, and project collaboration seems to be functioning well. The future work goals are reasonable and seem achievable at present.
- The Phase II work plan includes some specific targets and seems rational.
- The proposed work is adequately enumerated. Stability in steam is planned. It is critical to evaluate the stability of both the electrolyte and the possible precipitates from the post-hydrogen-exposure sintering aid. While the hydrogen produced is expected to be dry, conducting a stability test in a hydrogen–steam environment is recommended. It is also important to characterize the proton transference number in the proposed operating temperature range. These materials are known to exhibit proton, oxygen, and electronic conduction, all of which vary with temperature and oxygen partial pressure. The investigation of such properties will help decide suitable operating conditions to achieve high efficiency.

Project strengths:

- This is a good project that is leveraging both new materials and new ways to combine those materials. It is pursuing these innovations with a mindfulness for reducing the demands placed upon the manufacturing infrastructure. Innovations developed at the national laboratories are being exploited effectively. The principal investigator is clearly an expert in the project team's field of research.
- Collaboration with other nodes is good. The plan adequately addresses potential pitfalls of this materials set.
- The high-throughput capabilities at NREL should help to provide a strong direction for this project, assuming that the compositional space is rationally defined.
- This is a good team, and there is good collaboration.

Project weaknesses:

- This project seems to suffer from a lack of focus and progress. Materials processing for lowered temperature fabrication of dense parts can be impactful, but it must be done meticulously and documented and communicated well to have meaningful impact.

- Explicitly stating the scientific hypotheses in sets of the null condition and the alternate would provide additional transparency into what is driving the experiments. The vertical integration of project participants could be improved.
- The project is very dependent on the sintering aid for electrolyte density. Attention to the sintering aid accumulation/detection is encouraged.
- The priority for materials stability testing needs to be high.

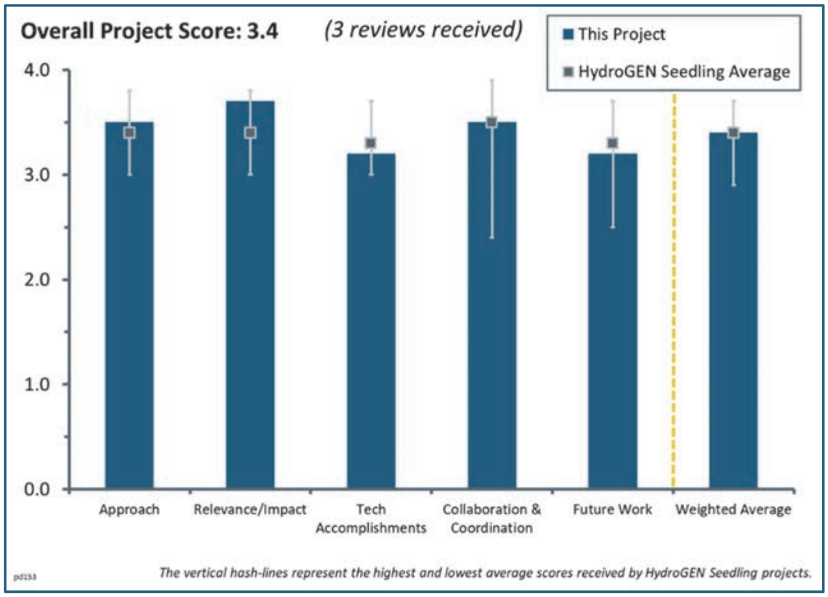
Recommendations for additions/deletions to project scope:

- Overall, the project scope is very good. Phase II needs to include a thermomechanical characterization of these electrolytes. They are known to be mechanically weak.
- The project may benefit from a more defined processing plan to help make faster progress in the experimental work.
- The project team should consider adding the ability to sinter under the influence of applied electric/ magnetic fields.
- The project should address a sintering aid sink, when possible.

Project #PD-153: Degradation Characterization and Modeling of a New Solid Oxide Electrolysis Cell Utilizing Accelerated Life Testing

Scott Barnett; Northwestern University

Brief Summary of Project:



Solid oxide electrolysis cells (SOECs) have the potential for high electricity-to-hydrogen conversion efficiency, but these cells lack long-term stability, particularly at high current density, and the degradation mechanisms in SOECs are poorly understood. The project aims to develop mechanistic degradation models that realistically predict long-term solid oxide electrolysis cell (SOEC) durability, using input data from accelerated electrochemical life testing combined with quantitative microstructural and microchemical evaluation. Also, a promising SOEC cell type with high performance will be further developed. The understanding achieved by combining experimental results and theory will be used to guide improvements in long-term SOEC durability.

Question 1: Approach to performing the work

This project was rated **3.5** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The approach is clearly laid out. The principal investigator's group is leveraging its degradation model experience with solid oxide fuel cells (SOFCs) and applying it to SOECs. The barriers are identified, and appropriate project partners at LBNL and INL were selected to improve the odds of success. The model selected predicts oxygen potential in order to anticipate conditions that lead to degradation, when compared to experimental testing.
- The extension of knowledge about SOFC degradation to SOECs is reasonable and logical. The model's methods/approach and boundary conditions are logical.
- The prediction of oxygen potential across the electrolyte is a useful tool for determining critical parameters in improving electrolyzer stability.

Question 2: Relevance/potential impact

This project was rated **3.7** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- Oxygen electrode delamination is a well-known challenge in electrolyzer development. This project addresses one aspect, namely, high oxygen partial pressure at the electrolyte–electrode interface. The estimation of interface oxygen pressure using measured electrode polarization provides predictive capability to select operating conditions without catastrophic cell failure. This model has applicability independent of the cell design and materials set.
- The potential impact of this project could be exceptional if it generates tools that could be used broadly in SOEC design. That would be an exemplary case of a Fuel-Cell-Technologies-Office-funded project

furthering not only its own niche goals but additionally benefiting the high-temperature electrolysis community at large.
- This project is very relevant, particularly considering the fact that higher current densities are not always detrimental. The coupling of current density and effective oxygen partial pressure at the electrode is important.

Question 3: Accomplishments and progress

This project was rated **3.2** for its accomplishments and progress toward overall project and DOE goals.

- Though the project is behind on many of its milestones (partially but not 100% complete), it appears that that could be due to delays in the cell fabrication that delayed planned testing, which is not uncommon. The approach is strong, so this project should be able to stay on track and deliver additional impactful information. The metal-supported SOECs at LBNL could be promising if further improvements are made. The INL-made cells should add an additional level of model validation as well.
- The modeling work seems to have gone well and is on the right track. However, the presentation does not show a pathway to accelerated testing—which is the title of the project. It would be helpful to define the accelerated testing, as well as the basis of such test methods, so that the test method ensures the degradation mechanism remains the same as in normal testing.
- Progress seems to have built up slowly, with some slipping, but overall, it is reasonable and promising.

Question 4: Collaboration effectiveness

This project was rated **3.5** for its collaboration and coordination with HydroGEN and other research entities.

- The cell developments in parallel at LBNL (metal-supported), INL, and Northwestern University add significant value to the degradation model efforts, assuming that there is a useful agreement of failure modes that can be linked back to the oxygen activity upon which the Northwestern University model is predicated. Hopefully, this yields a more robust model.
- The collaboration with LBNL to test metal-supported cells in electrolysis mode is appropriate, and the performance results are encouraging.
- The Northwestern University, INL, and LBNL collaboration appears to be functioning well. The use of data from Data Hub for model verification would be very encouraging.

Question 5: Proposed future work

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

- Incremental advances in the understanding of degradation mechanisms are very useful. No leaps in understanding are expected, but hopefully, reasonable predictions of SOEC lifetime expectations, or at least optimal operating points, will result from this. The model will almost certainly hit additional hurdles, but progress is reasonable, and future work seems manageable for the team.
- The future work gives some general details about the tasks that remain ahead. With much work remaining on the first iteration of the degradation model, this level of detail is understandable. However, there is much similarity between the proposed future work for budget period 2 and 3.
- The definition, justification, and execution of accelerated testing must be included. The effect of the ceria barrier layer may be useful in the model for oxygen potential prediction. The other tasks seem fine.

Project strengths:

- This project leverages the team’s experience with degradation model development and pairs that with SOEC development from INL and LBNL.
- The predictive model for oxygen potential is a useful tool. The new oxygen electrode performance and stability appear excellent.

- This is a great team with a diverse set of well-made cells. The researchers have a good data set with which to work.

Project weaknesses:

- The model may not account for enough variables to truly build degradation mechanism insight, but it should be able to predict reasonable expected lifetimes.
- This project's weakness is that it still requires a significant amount of degradation data to build and validate the degradation model.
- The accelerated testing protocol needs to be defined.

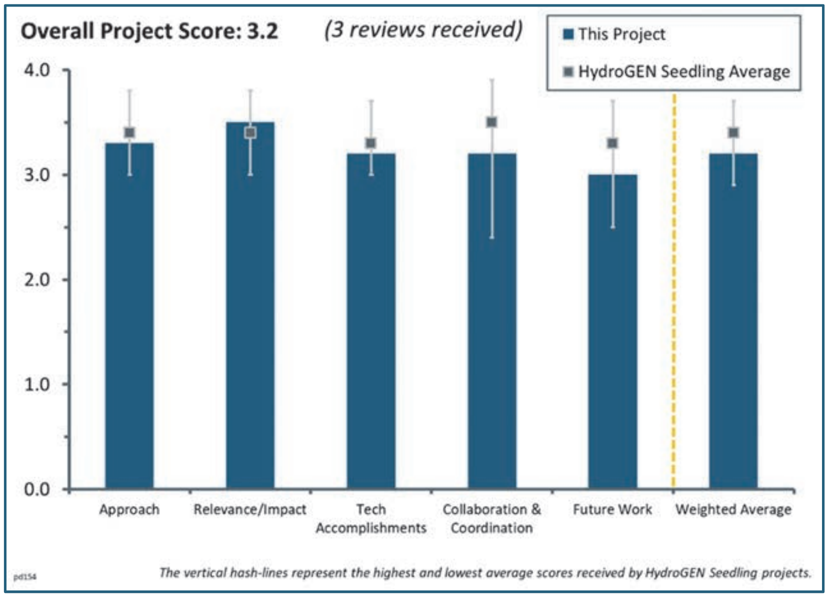
Recommendations for additions/deletions to project scope:

- The project team should keep up the good work.
- No additions/deletions to the project scope are requested.

Project #PD-154: Thin-Film, Metal-Supported High-Performance and Durable Proton–Solid Oxide Electrolyzer Cell

Tianli Zhu; United Technologies Research Center

Brief Summary of Project:



This project is developing a thin-film, durable metal-supported solid oxide electrolysis cell (SOEC) using a proton-conducting electrolyte at targeted operating temperatures of 550°C–650°C. This advanced SOEC will provide a highly efficient, cost-competitive high-temperature electrolysis process for hydrogen production. Initial efforts are on demonstrating the feasibility of the proposed concept by further advancing metal-supported single cells based on work completed previously for solid oxide fuel cell (SOFC) applications. Cell fabrication, especially electrolyte deposition via reactive spray deposition technology (RSDT) and suspension plasma spray (SPS) processes, is a focus.

Question 1: Approach to performing the work

This project was rated **3.3** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The approach for this project attempts to combine low-temperature deposition techniques to fabricate electrolytes of highly refractory yttria- and ceria-doped barium zirconate (BCZY) material on metal supports. This type of approach has been used successfully for metal-supported SOFCs in the past. The tasks for budget period 1 seem appropriate for evaluating whether the fabrication and testing progress is on track to deliver more refined data in budget period 2, or if unforeseen processing bottlenecks could jeopardize the success of the project.
- The overall approach is promising. Metal-supported design has the possibility of being a good option for hydrogen production at scale.
- The project follows a solid, logical approach and has a good team.

Question 2: Relevance/potential impact

This project was rated **3.5** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- The project team pursued a less expensive fabrication path to high-temperature proton-conducting electrolyzers for hydrogen generation. Addressing the cost issues while maintaining performance fits with the HydroGEN consortium's goals, and the project pulls in several Energy Materials Network (EMN) nodes.
- Proton-conducting SOECs are very relevant; it is hard to debate that fact. Whether cost/durability targets will be met may be debated, but this work seems to indicate that there is good progress in that direction.
- If successful, metal-supported cell design could be a very promising option for large-scale hydrogen production, in terms of cell scalability and low cost.

Question 3: Accomplishments and progress

This project was rated **3.2** for its accomplishments and progress toward overall project and DOE goals.

- Complex, multication materials are difficult to deposit on a support. The success of getting a dense layer on a metal support within a single phase is a good accomplishment.
- The project is on track to meet its stated goals and DOE goals.
- It is challenging to judge the progress of electrolyte fabrication without cell data. Focused ion beam (FIB)/scanning electron microscope (SEM) imaging can show miniscule dense regions but cannot capture the deleterious effect of pinholes or other defects in the membrane. Electrical performance is the key to verifying that electrolyte and electrode morphologies are in spec, so seeing that data is anticipated. Only one full cell test is shown, and it seemed to suffer a cell or seal failure and never reached a significant open circuit voltage, nor was it run in electrolysis mode. Those milestones are not due until the fourth quarter and are necessary for a go/no-go decision, but until that data comes in, it seems that fabrication progress is satisfactory.

Question 4: Collaboration effectiveness

This project was rated **3.2** for its collaboration and coordination with HydroGEN and other research entities.

- The team uses several EMN hubs. Lawrence Berkeley National Laboratory (LBNL) was utilized for its expertise in metal-supported cells, Idaho National Laboratory (INL) for fabrication, and the National Renewable Energy Laboratory for modeling work.
- Collaboration with INL and LBNL was appropriate to engage laboratories with the right set of skills.
- It is unclear how well the collaboration is functioning. The project is progressing well, but work seems very independent at the present time.

Question 5: Proposed future work

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

- The proposed future work has appropriate objectives.
- Negligible detail is given for tasks in budget periods 2 and 3. The project team should also consider how compositions or processes might be improved; some detail should be given, based on what is known now, i.e., at about 75% through budget period 1.
- The team has good performance targets, but it is uncertain whether targets can be met at the current pace of the project.

Project strengths:

- Metal-supported cells have the best potential for large-scale hydrogen production. Prior experience in an Advanced Research Projects Agency–Energy (ARPA-E) project of similar cell design gives the team background technical information on which to build.
- The project leverages technology solutions that have proven success for SOFCs, based on similar materials.
- This is a good team with a good start and good targets.

Project weaknesses:

- If the processing routes do not produce functional cells quickly, the project cannot effectively proceed past the first go/no-go decision point.
- Collaboration could be stronger or the strength better communicated. Sintering and barrier coating work needs to be strengthened.
- Electrolyte density appears to be an issue, but there was no specific approach defined to address this.

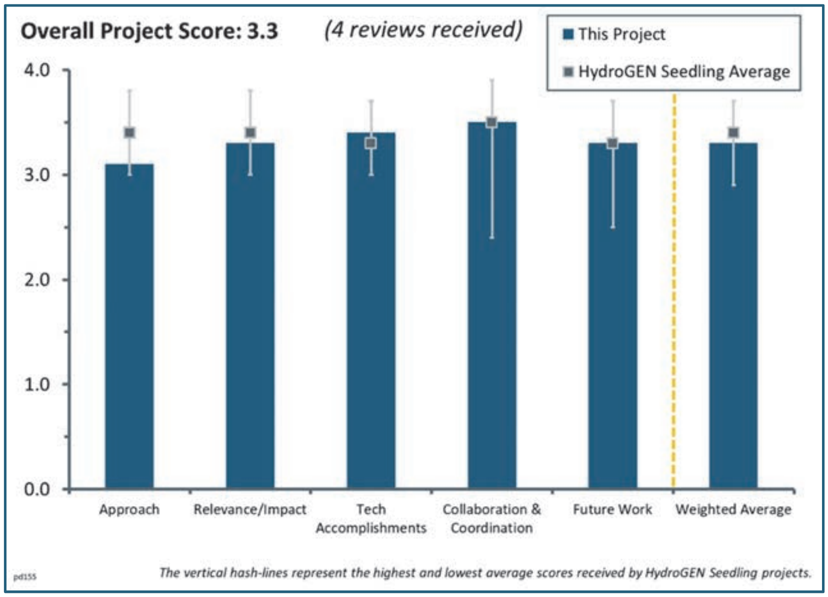
Recommendations for additions/deletions to project scope:

- No change to the scope is necessary.

Project #PD-155: High-Efficiency Polymer Electrolyte Membrane Water Electrolysis Enabled by Advanced Catalysts, Membranes, and Processes

Kathy Ayers; Proton OnSite

Brief Summary of Project:



This project will develop an advanced, highly efficient polymer electrolyte membrane water electrolysis (PEMWE) membrane electrode assembly (MEA) by addressing membrane, catalyst, catalyst layers, and their interfaces. Four areas affecting cost and efficiency that will be developed include (1) thinner membranes, (2) lower catalyst loadings, (3) optimized gas diffusion layer and porous transport layer materials and structures, and (4) increased operating temperature. Successful demonstration and integration of these four areas require a deeper understanding of the scientific and technical aspects of PEMWE MEAs. Proton Onsite will partner with Tufts University and Oak Ridge National Laboratory—with support from the National Renewable Energy Laboratory (NREL) and Lawrence Berkeley National Laboratory (LBNL)—to integrate advanced cell designs and materials and fundamentally characterize performance.

Question 1: Approach to performing the work

This project was rated **3.1** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- Researchers are addressing cost barriers associated with PEM technology through cell design and material optimization. Their final deliverable is stated to be an "advanced electrolysis stack producing H_2 at 43 kWh/kg and at costs of \$2/kg H_2." The project's approach involves optimizing the catalyst composition, developing thinner membranes, and optimizing the interfacial properties. These parameters are expected to improve water transport and prevent catalyst migration for improved system performance. The project is separated into three experimental tasks:
  - Task1: Membrane processing, with a goal of characterizing properties and measuring changes that occur during operation
  - Task 2: Advanced MEA fabrication, resulting in the development of formulations and deposition parameters, as well as the characterization of water distribution using X-ray computed tomography (CT)
  - Task 3: Catalyst development, involving synthesis of alloyed catalysts containing Ir and Ru and analysis of the catalysts with microscopy to evaluate performance

  These tasks all partner with HydroGEN consortium nodes to accomplish the stated goals. The approach is very reasonable, and the results are promising. For example, the project team's approach has successfully demonstrated good MEA performance at 1.85 V. The approach has also demonstrated that alloying Ir and Ru provides a more active and durable catalyst. The team's use of in situ CT allows for the imaging of catalyst layers and bubble formation on the gas diffusion layer. This will help with optimizing the electrode and reactor structure for improved performance. However, the approach still relies on expensive precious metal catalysts (Ir and Ru); a true reduction in cost barriers requires precious-metal-free catalysts.

- The project clearly identifies the challenges to durable and high-efficiency membranes for hydrogen generation through water electrolysis. The scope of work as initially outlined exceeds the allocated resources by identifying five major design parameters and multiple diagnostic tests. However, delving into the project reveals appropriately re-scoped work to yield meritorious data within existing resource constraints.
- Thinner membranes, advanced catalysts, and operation at higher temperature have good potential to improve efficiency.
- The approach is supposed to address critical barriers such as long-term durability and higher defect sensitivity. In the “approach: innovation” section, the researchers listed standard terminology that has been used over the years to address essentially the same issues. It is hard to see what would be the novelty of this approach that would distinguish it from past efforts, unless this is what the team terms a “holistic” view of the problems.

Question 2: Relevance/potential impact

This project was rated **3.3** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- The project aligns well with the DOE Hydrogen and Fuel Cells Program (the Program). The project team’s combination of membrane, cell, and catalyst optimization should meet the target production cost of $2/kg H_2 by the fourth quarter (Q4). Moreover, the development of in situ CT provides key insights into the structural changes that occur at the electrode surface and also shows how gas bubbles accumulate. This information can be used to further optimize cell design for improved performance and lower overall costs. The project team’s current work is extremely promising and is on target to meet DOE goals, but improvement could be made by eliminating or reducing precious metals in the catalyst structure.
- The project is relevant, given the potential for complex, integrated system benefits when the performance at component interfaces is more thoroughly understood. Within the available resources, this project has the potential to achieve the production target of $2/kg H_2 by reducing the inefficiencies at these component interfaces, as well as improving cell-level performance.
- Reaching the DOE hydrogen production goal of $2/kg H_2 is very relevant to hydrogen refueling infrastructure. The project also leverages the HydroGEN consortium.
- This project aligns well with the objective of the Program and the HydroGEN consortium.

Question 3: Accomplishments and progress

This project was rated **3.4** for its accomplishments and progress toward overall project and DOE goals.

- The project team has successfully met the Q1 and Q2 milestones and is making progress toward the remaining Q3 and Q4 milestones. Specifically, the team has exceeded Q2 performance metrics and was able to operate the cell with 2.2 A/cm^2 at less than 1.85 V. There is an excellent use of HydroGEN nodes to accomplish in situ CT and rotating disk electrode studies of catalyst degradation. This data will hopefully allow for further optimization of cell parameters for improved overall performance. The authors claim they are close to the go/no-go criteria (Q4 milestone) of 1.8 A/cm^2 at 1.7 V cell potential operating at 90°C. This would require an approximately 100 mV decrease in operating potential compared to the current performance data.
- Significant progress was made in the first year, considering the late start. Tomography insight into the electrode structure and water management is valuable for the future design of electrodes. The MEA performance of 1.85 V at 80°C was demonstrated within the first two quarters. The project seems to be on a good path to meet the Q4 projected milestone of 1.8 mA/cm^2 at 1.7 V.
- The hardware test results demonstrate that both the prime vendor and project partners have made notable progress. Catalyst performance and diagnostic test results are on track to effectively inform manufacturing technique development for scaling this performance from the catalyst level to the cell level with potentially thinner membranes.
- The team has met the Q2 milestone performance target of 800-hour durability at high current density.

Question 4: Collaboration effectiveness

This project was rated **3.5** for its collaboration and coordination with HydroGEN and other research entities.

- The project effectively leverages Energy Materials Network (EMN) nodes through the LBNL and NREL for catalyst activity screening and degradation measurements, as well as membrane hydration modeling and measurements. Moreover, cell characterization with in situ CT is a unique capability that provides insight into the catalyst layer structure and the formation/movement of bubbles across the electrode surface. This technique will provide extremely valuable information on how to potentially optimize the cell architecture to improve performance.
- This work both replicates catalyst performance demonstrated in previous research by other firms and increases the available relevant empirical data. Including both successful and unsuccessful catalyst formulations in the EMN leverages heritage efforts. Executing standardized testing increases the potential for replication of results by research partners, thereby augmenting the pace of development.
- The project effectively leverages the capabilities of two EMN nodes.
- There is excellent collaboration among the participants.

Question 5: Proposed future work

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

- Developing an MEA is not a trivial task. Overall, this activity proposes to investigate thinner membranes, novel OER catalysts, and refined electrode deposition and inspection methods to deliver this improved MEA to satisfy the DOE performance metrics. Near-term work focuses on membrane processing to incorporate catalyst advances into an interim MEA while investigating thinner membranes as an external partner's parallel effort.
- The proposed future work is in excellent agreement with the approach and ongoing work.
- There is a good plan in place to meet the go/no-go milestone.
- The project is on track to meet budget period 1 milestones and a target performance of 1.8 A/cm^2 at 1.7 V cell potential at 90°C. This will achieve the DOE goal of $2/kg H_2. The proposed future work/next steps involve additional membrane processing for mechanical evaluation at LBNL and in situ electrode imaging via CT. This tomography will be used to guide the modeling at LBNL. Parallel efforts will evaluate and define processing parameters to achieve thinner membranes, and high-activity catalysts can be integrated with NREL support. There is a desired reduction in operating temperature to meet target performance, although the researchers claim they are close to achieving 1.8 A/cm^2 at 1.7 V. This future work is reasonable, but it could be improved by considering how to reduce or eliminate precious metals from the catalyst composition.

Project strengths:

- This project is doing an excellent job of improving the performance of precious-metal catalyst systems and characterizing the evolution of catalyst layer and bubble distribution along the electrode surface. These efforts are well integrated with EMN nodes, and the collaboration seems to be working well. The work has currently produced very high activity numbers and is on track to meet DOE goals.
- The team executing this task is experienced and understands the multiple interfaces under investigation.
- This is a well-established effort executed by the leaders in this field.
- This project addresses critical issues related to PEM water electrolyzers.

Project weaknesses:

- This activity is attempting to cover a wide scope of work and has the risk of spreading resources too thin to complete the assignments. Decreasing membrane thickness typically decreases durability and challenges the system for high-pressure applications.
- The project team did not discuss the class of membrane materials used and how the risk of increased cross-over and reduced mechanical strength of the thinner membrane is mitigated.

- The only real weakness is the reliance on precious-metal catalysts. This could be addressed in future budget periods or be the subject of an entirely new project.
- A more diverse set of tools for characterization should be used.

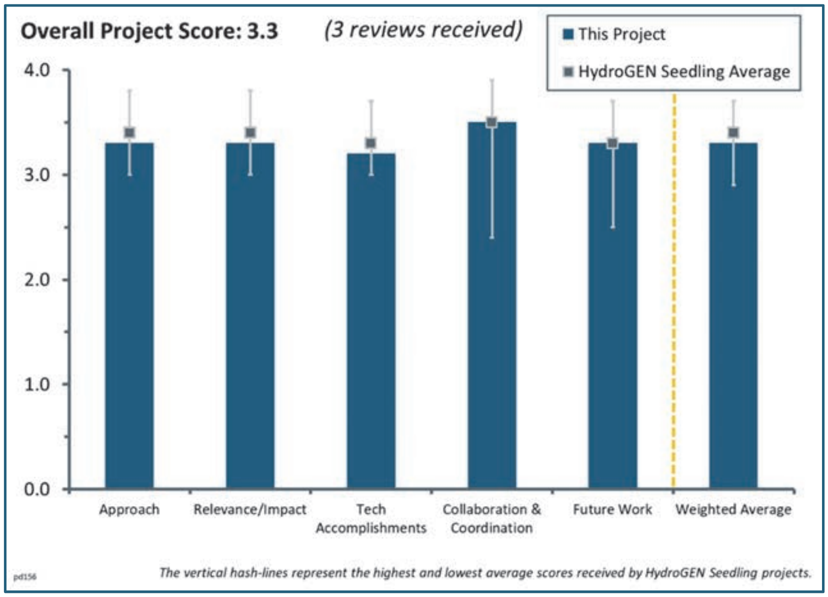
Recommendations for additions/deletions to project scope:

- The scope of the project is fine for a precious-metal-based catalyst system. However, once cell and membrane parameters have been optimized, future efforts should focus on creating catalysts with reduced precious-metal content.
- It would be valuable for the team to include this new membrane in a heritage high-pressure electrolyzer to investigate the potential for inclusion within high-pressure applications.
- It is recommended that the project team add metrics for evaluating the membrane: conductivity, mechanical strength, gas cross-over, etc.
- In dissolution studies, more sophisticated methods should be used, such as online inductively coupled plasma–mass spectrometry (ICP-MS).

Project #PD-156: Developing Novel Platinum-Group-Metal-Free Catalysts for Alkaline Hydrogen and Oxygen Evolution Reactions

Sanjeev Mukerjee; Northeastern University

Brief Summary of Project:



The aim of this project is to develop (1) stable, high-conductivity, and high-strength anion exchange membranes (AEMs) and ionomers, (2) stable and active platinum-group-metal-free (PGM-free) catalysts for hydrogen and oxygen evolution reactions (HERs/OERs), and (3) high-performance electrode architectures that together can begin to achieve the low-cost advantages of AEM electrolyzers. This effort is focused on materials development by tailoring synthesis and composites with supporting efforts in computation and characterization. The project work—and collaborations with the University of Delaware, Advent North America, the National Renewable Energy Laboratory (NREL), Lawrence Berkeley National Laboratory (LBNL), and Sandia National Laboratories (SNL)—strives to enable a clear pathway to achieving hydrogen costs of less than $2 per kilogram, with an efficiency of 43 kWh per kilogram, via AEM-based electrolysis.

Question 1: Approach to performing the work

This project was rated **3.3** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- This project is rather ambitious and plans to address improving AEMs, developing novel PGM-free catalysts that show stability and activity for both cathodic and anodic HER and OER reactions, and integrating these into high-performance electrolyzer electrode assemblies. The project impact and overall goal is to achieve a clear pathway to hydrogen production at <$2/kg with an energy efficiency of 43 kWh/kg hydrogen. Catalyst development will be conducted at Northeastern University. HER catalysts will include Ni-based oxides, functionalized mono-metallic, and nitrogen–carbon–metal catalyst stems. OER catalyst development will include double-layer metal oxides on Raney Nickel. In situ X-ray absorption spectroscopy (XAS) will provide key insights into catalyst electronic structure and performance. Membrane development and gas diffusion electrode development will be conducted by sub-awardees University of Delaware and Advent North America. The project will leverage NREL's expertise in membrane electrode assembly (MEA) and testing, LBNL's capabilities for small-angle scattering and transport modeling, and SNL for interfacial modeling. The project scope is large, but the work breakdown seems reasonable and effectively leverages the Energy Materials Network's (EMN's) capabilities for development of high-activity electrochemical systems.
- This is an excellent approach in tackling the most challenging issues in electrochemistry. The lead researchers rely on fundamental principles to resolve complex interfaces to design new materials for the HER and OER. In addition, highly sophisticated characterization tools are employed to get ex situ and in situ feedback. All three crucial aspects are being covered by this work: PGM-free catalysts, novel membranes, and electrode structures.
- The project has an exceptionally logical approach of dividing the problem into manageable sub-elements and distributing these sub-elements across a team of collaborators. It would be beneficial to have some

verbiage on how to eventually implement these innovations on the industrial scale to guide this work from the laboratory into practice.

Question 2: Relevance/potential impact

This project was rated **3.3** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- The project is extremely relevant to the HydroGEN consortium mission of sustainable and cost-competitive hydrogen generation at $2/kg. Moreover, the emphasis on precious-metal-free catalysts will further lower the costs associated with water-splitting systems. The project also effectively leverages HydroGEN capabilities through synchrotron-based X-ray characterization, MEA preparation, and modeling.
- This fundamental work solidly addresses the catalyst and membrane barriers to AEM electrolysis. Membrane durability and cumulative performance of the proposed formulation of the AEM MEA will determine the impact of this technology. It would be of benefit in future proposals to expand the list of potential salts in the feedstock water. This has the potential to reduce water-processing requirements, thus expanding the potential geographical regions into which this technology may be deployed.
- If successful, this project will have rather high impact.

Question 3: Accomplishments and progress

This project was rated **3.2** for its accomplishments and progress toward overall project and DOE goals.

- The project has successfully completed first quarter (Q1) and Q2 milestones of demonstrating high-performance, precious-metal-free catalysts for both HER and OER reactions. Synthetic scale-up has produced 5-gram batches and delivered them to consortium collaborators for X-ray characterization and rotating disk electrode (RDE) testing. Novel MEA materials have also been synthesized, and initial evaluations have begun. This project is well on its way to creating active, precious-metal-free water-splitting catalysts.
- Given that this project had a late start, significant progress has been made toward the DOE goals. For the first two quarters, the project met the milestones and delivered the three 5-gram batch samples of Ni-MO_x/C catalysts, as well as one batch of Ni-N_x/C HER catalysts, for RDE and single-cell tests. They showed overpotential of η=300 mV at 500 mA/cm^2 and 2 A/cm^3 (HER). In addition, a membrane based on PAP-TN was synthesized. Reaction and polymerization conditions are still to be optimized.
- The work clearly identifies improved non-PGM HER and preliminary OER catalyst performance under a range of applicable conditions with supporting empirical data from the collaborating partners. The project appears to be on schedule, with near-term work to elucidate the performance of the new ionomer and AEM formulations.

Question 4: Collaboration effectiveness

This project was rated **3.5** for its collaboration and coordination with HydroGEN and other research entities.

- The project has demonstrated good collaboration and use of EMN nodes. For example, the team has used modeling results from SNL to understand ionomer membrane properties. These results will be used to further optimize the ionomer composition to improve performance. Modeling and X-ray scattering results from LBNL have provided key information on AEMs as a function of hydration and applied current density through the electrochemical cell. NREL capabilities will be leveraged for MEA preparation and evaluation.
- There is excellent coordination between the collaborators.
- This project includes a comparatively large number of collaborators: three contract awardees and three national laboratories. Developmental materials, modeling results, and test results all flow among the group well enough to deliver punctually on the contract milestones.

Question 5: Proposed future work

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

- The proposed future catalyst development work is largely focused on optimizing catalyst composition, stability, and active site density. In situ X-ray absorption and Raman spectroscopy will provide information about catalyst composition, structure, and degradation under realistic working conditions. This information can then be used to further refine catalyst structure to optimize performance. Future durability testing and catalyst ink development will evaluate performance in full electrolyzer cells at elevated temperatures. This work will also accurately determine OER overpotentials to understand performance losses and optimization of fuel cell architecture. Additional modeling efforts will identify how the presence of specific electrolyte ions impact membrane and cell performance. Finally, full MEA optimization and testing will be conducted at NREL in fiscal year (FY) 2019.
- The project has positioned itself for completing the contractual milestones in a timely manner. The diagnostic, performance, and durability testing on the membrane and catalysts scheduled for the balance of this activity improve the data quality sufficiently for a thorough and accurate assessment of the developed technological elements.
- The proposed future work is very well aligned with ongoing efforts.

Project strengths:

- This is an excellent team with broad expertise and an approach that includes all relevant aspects in development of PGM-free electrolyzers.
- This project is successfully developing high-performing, precious-metal-free HER and OER catalysts for hydrogen production in MEA electrolyzers. This approach is key for realizing cost-effective and sustainable hydrogen generation. The project has many parts, but it has demonstrated good collaboration and has a high chance for success.
- The team is well-connected and understands where to collaborate to get either the knowledge or the analysis required to further the work.

Project weaknesses:

- This is a very strong project.
- No weaknesses were found.
- Requiring that source water be doped as an anolyte will likely add challenges to the system that is eventually deployed.

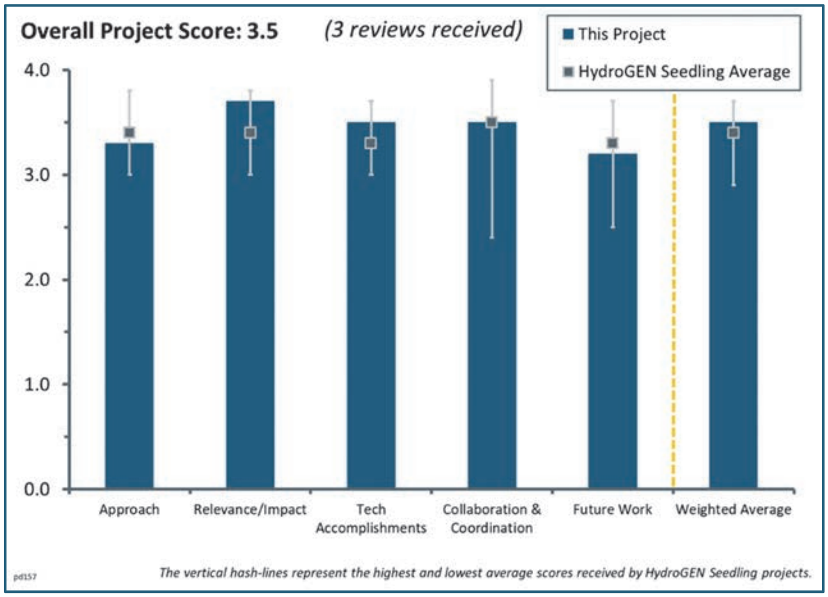
Recommendations for additions/deletions to project scope:

- It would likely be worthwhile to peruse the expanding EMN and HydroGEN data hub prior to executing FY 2019 activities. There is potentially some relevant data that already exists, which would enable this activity to advantageously redirect resources.
- The project scope is appropriate.
- The team should better delineate contributions among the participants.

Project #PD-157: Platinum-Group-Metal-Free Oxygen Evolution Reaction Catalysts for Polymer Electrolyte Membrane Electrolyzer

Di-Jia Liu; Argonne National Laboratory

Brief Summary of Project:



The objective of this project is to lower the capital cost of polymer electrolyte membrane (PEM) electrolyzers by developing low-cost, platinum-group-metal-free (PGM-free) oxygen evolution reaction (OER) electrocatalysts. The project is developing high-activity, high-conductivity, durable metal–organic framework (MOF)-based catalysts via both direct (e.g., solvothermal) and template (e.g., infiltration) synthesis approaches with one, two, or three transition metals. The most promising MOF-based catalysts will then be incorporated in a 3-D porous nano-network electrode (PNNE) architecture. The goal is to produce durable PGM-free OER catalysts with performance approaching that of current Ir-based PGM catalysts. Argonne National Laboratory (ANL) is partnered with Giner, Inc., and University at Buffalo (UB) and is leveraging national laboratories within the HydroGEN consortium.

Question 1: Approach to performing the work

This project was rated **3.3** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- This project will attempt to reduce hydrogen evolution costs by creating high-activity, PGM-free OER catalysts that can operate in acidic conditions. PGM-free OER catalysts composed of first-row transition metals (Ni, Co, Fe) have shown great promise in alkaline OER applications. However, these materials typically demonstrate severe instability in acidic conditions. This project uses metal–organic frameworks (MOFs) and zeolitic–imidazolate frameworks (ZIFs) containing mixtures of Fe, Co, Ni, and Mn to promote the OER in an acidic electrolyte. A variety of metal mixtures will be screened to determine the optimum composition. Current results show decent activity and remarkable stability for transition metals in acidic conditions. The project leverages Energy Materials Network nodes via computational modeling at Lawrence Livermore National Laboratory (LLNL) and Lawrence Berkeley National Laboratory (LBNL), electron microscopy at Sandia National Laboratories (SNL), and electrode optimization and catalyst characterization at National Renewable Energy Laboratory (NREL). Finally, the project partners with Giner, Inc., an industrial partner specializing in water electrolyzer technology. This is a well-rounded project with a good approach.
- This project has focused all of the team's efforts onto one problem (PGM-free OER catalysts) and has methodically evaluated a number of potential options. The only recommendation would be to move the migration from the rotating disk electrode (RDE) to the membrane earlier in the process.
- The approach for improving activity and durability is excellent. However, it is not clear how the project plans to reduce the cost of the OER catalyst to less than 1/20 of the cost of the Ir catalyst, which is the primary goal of the project.

Question 2: Relevance/potential impact

This project was rated **3.7** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- The ability to generate a cost-effective and high-performing MOF-based, PGM-free catalyst would both reduce the cost of electrolyzers and expand the deployment to support distributed hydrogen generation. It would also mitigate any supply issues associated with a PGM-based catalyst.
- The project is relevant to the HydroGEN consortium’s mission of developing low-cost, high-activity catalysts for water-splitting technologies. This project will directly address this mission by developing low-cost, high-activity transition metal catalysts for the anodic OER in acidic conditions.
- This project is essential to the overall DOE Hydrogen and Fuel Cells Program goals of reducing the cost of production. However, the lead researchers should consider quantifying and demonstrating the cost advantage of at least one of the materials under study, say, the Co-MOF- or Fe-ZIF-based catalyst, over conventional PGM catalysts.

Question 3: Accomplishments and progress

This project was rated **3.5** for its accomplishments and progress toward overall project and DOE goals.

- The project has demonstrated good progress to date by successfully completing two performance milestones and is 50% complete on a third. These milestones include the synthesis of nine new ZIF-based catalysts and two MOF-based OER catalysts. These catalysts show activity comparable to other literature examples of PGM-free OER catalysts and are approaching the performance of industry-standard Ir-black. Moreover, a Co-MOF OER catalyst shows much lower degradation compared to Ir. The project team is very close to meeting the go/no-go milestone of demonstrating a PGM-free OER catalyst with an overpotential <350 mV or 15 mV higher than Ir-black at 10 mA/cm^2 in an acidic electrolyte.
- The project team has demonstrated multiple PGM-free catalysts from different partners, with reasonable performance on a RDE with a very low pH electrolyte. The team is currently setting up to migrate from RDE testing to PEM testing.
- The results from the activity and durability test of the ANL catalyst in an acidic environment, compared to a conventional PGM-based catalyst, look encouraging.

Question 4: Collaboration effectiveness

This project was rated **3.5** for its collaboration and coordination with HydroGEN and other research entities.

- The project has so far shown good collaboration with HydroGEN consortium capabilities. The project includes modeling efforts from LLNL and LBNL, the advanced electron microscopy of catalyst materials from SNL, and catalyst testing from NREL. NREL will also support catalyst characterization and high-throughput electrode optimization to maximize catalyst performance. Ongoing modeling at LBNL and LLNL will provide realistic structural models to improve understanding of catalyst performance.
- This project includes federal laboratories, commercial vendors, and partners from academia. Participants work toward their strengths within a logical and methodical plan. Specific procedures are identified to submit results to the HydroGEN consortium.
- This project has good collaboration with NREL on establishing baseline activity and durability testing. The computational modeling collaboration with LLNL and LBNL, as well as the transmission electron microscopy (TEM) work with SNL, also look fine, although the impact or link of those results to observed activity or durability data is lacking.

Question 5: Proposed future work

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

- The proposed future work is centered on improving the performance and durability of Co- and Fe-based MOF catalysts via compositional control (doping), as well as improving the design of PNNEs through an electrospinning deposition technique. The optimized catalysts will be incorporated into the membrane electrode assembly (MEA) and tested by Giner, Inc., to achieve the go/no-go milestone criteria. This future work is appropriate.
- The work plan is logical and focused on the stated goal of reducing the cost of PEM electrolyzer catalysts. The eventual testing of the PGM-free catalysts in a PEM MEA will reveal any stability or performance issues.
- The proposed work on MEA fabrication and PEM electrolyzer testing with Giner, Inc., is a logical next step. The continued development of MOF-based OER catalysts by both ANL and UB also makes sense. However, it is curious that the future plan does not include any work on ZIFs. It is unclear whether this is an oversight or the project team has decided to abandon ZIF-based catalysts, despite claims that the "UB team is making excellent progresses [sic] in activity and durability improvements for FeMx-ZIF-8 and FeMx-ZIF-8/Oxide-based catalysts." Also, if the performance tests on activity and durability of the current OER materials are acceptable, it is unclear why the team would start a new synthesis method with graphene, atomic layer deposition, etc., rather than just continue to improve on the current method.

Project strengths:

- The project has shown impressive performance and stability for PGM-free OERs in acidic media. The catalysts are very stable and are approaching the performance of state-of-the-art Ir-based catalysts.
- This project concentrates resources to address a single problem: the rate-limiting OER. There is no evidence of resource dilution on tangential activities.
- At this point, the project strength is perhaps the synthesis and testing of the PGM-free OER catalysts.

Project weaknesses:

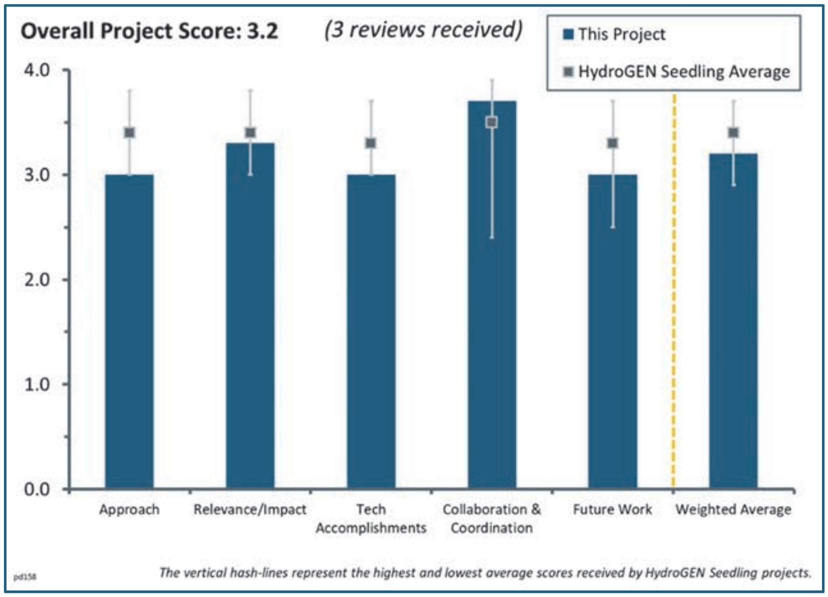
- The slow progression from RDE to MEA may slow the down-selection process for catalysts within the activity, thereby potentially consuming resources that could be devoted to other activities.
- The project lacks a direct link to catalyst performance, although the goal is to reduce cost by 5%.

Recommendations for additions/deletions to project scope:

- There are no modifications to recommend.
- The project scope is appropriate.
- The lead researchers should consider quantifying and demonstrating the cost advantage of at least one of the materials under study, say, the Co-MOF- or Fe-ZIF-based catalyst, over PGM catalysts.

Project #PD-158: High-Performance Ultralow-Cost Non-Precious-Metal Catalyst System for Anion-Exchange Membrane Electrolyzer

Hoon Chung; Los Alamos National Laboratory



Brief Summary of Project:

The primary objective of this project is to develop low-cost, active, and durable platinum-group-metal-free (PGM-free) oxygen evolution reaction (OER) and hydrogen evolution reaction (HER) catalysts with high performance in an anion-exchange membrane (AEM) water electrolyzer. The HER and OER catalysts being developed are based on Ni-La alloys and $LaSrCoO_3$ (LSC)-based perovskite materials, respectively. The catalysts and electrodes will be carbon-free, and a pure water feedstock (i.e., no added electrolyte) is targeted. In addition to utilizing HydroGEN consortium national laboratory capabilities, the project team will partner with Pajarito Powder, LLC, for catalyst scale-up activities.

Question 1: Approach to performing the work

This project was rated **3.0** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The project team is developing PGM-free OER catalysts from $LaSrCoO_3$ (LSC)-based materials for operation in alkaline water electrolysis systems. The approach involves using anion exchange membranes in conjunction with an organic cation (butyltrimethylammonium [BTMA+]) in the electrolyte to promote the OER in perovskite-based catalysts. These PGM-free catalysts are projected to eliminate the performance degradation associated with the organic components of the membrane, which can poison the active sites of traditional precious-metal catalysts. Understanding the interfacial phenomena that occur at the catalyst–membrane–electrolyte will help improve OER systems.
- In an attempt to simplify the overall system, the project team addresses the alkaline electrolysis cell as a whole to avoid the complications with recirculating an alkaline solution. This guided development of the membrane, the ionomer, and both HER and OER catalysts leads toward a low-cost integrated system. It would be helpful to identify the source of the contaminating benzene that would compromise the catalyst.
- The approach is based on the utilization of perovskite materials and Ni-La alloys in alkaline electrolyzers. This is not a novel or original idea; however, it is worth exploring in this applied project.

Question 2: Relevance/potential impact

This project was rated **3.3** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- The project is relevant to the HydroGEN consortium's mission to develop cost-effective, precious-metal-free catalysts for hydrogen production. Since OER is a major energy component of water splitting, the identification of robust, PGM-free catalysts with improved stability will directly achieve these goals. The elimination of PGM-based catalysts will also allow for the use of cheaper components and hydrocarbon-

based membranes. The expected roughly 50% reduction in stack costs will help obtain the target of <$2/kg H_2.
- This project has the potential to eliminate the recirculating electrolyte to simplify the overall system, minimize carbon in the membrane electrode assembly (MEA) to improve longevity, and utilize PGM-free catalysts to decrease cost and increase the number of deployable systems to improve the chances of achieving the DOE target metrics.
- The potential impact to DOE goals might be significant if this project delivers all projected milestones.

Question 3: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and DOE goals.

- The project has shown good progress; the team has completed fiscal year 2018 first- and second-quarter (Q1 and Q2) milestones involving the establishment of catalyst synthesis equipment, the synthesis of HER and OER catalysts, and the setup of the alkaline electrolyzer testing station. Q3 and Q4 milestones are on track, including the go/no-go performance criteria. The current data shows improved durability and has comparable activity to a state-of-the-art IrO_2 OER catalyst. Six perovskite OER catalysts have been compared with IrO_2 in both 0.1 M KOH electrolyte and 0.1 M BTMAOH electrolyte. The perovskite catalyst showed slightly reduced OER activity in 0.1 M KOH but had better performance. The perovskite catalysts showed greatly improved performance in 0.1 M BTMAOH compared with IrO_2.
- In the first year of funding, this project made significant progress in the establishment of synthesis equipment for both OER and HER catalysts, as well as in the AEM system setup. The first-year milestones and go/no-go decision are expected to be met and possibly exceeded; the project is predicted to result in significant AEM water electrolysis technology progress.
- By and large, the test data and progress are solid and illustrate that the activity is on track to continue with success. The notable exception, however, is the durability data. In the end application, an electrolyzer will likely be generating hydrogen for extended periods of time over a range of production rates. From that perspective, the cycle data well illustrates one aspect of durability: cycle life. The data generated by an hour-long steady-state test dubiously represents the other element of durability: stability. This is a fundamental research project, so accumulating hundreds or thousands of hours is neither feasible nor practical. It should be feasible, however, to demonstrate catalyst stability for more than an hour.

Question 4: Collaboration effectiveness

This project was rated **3.7** for its collaboration and coordination with HydroGEN and other research entities.

- The project effectively leverages four nodes. For example, the density functional theory and ab initio calculations node at Lawrence Berkeley National Laboratory provides computational input for the catalyst design and synthesis; the node also helps provide atomic-level details on catalyst reactivity. Initial results have predicted a much stronger binding of BMTA+ at IrO_2, compared with the LSC perovskite. The surface analysis cluster node at the National Renewable Energy Laboratory (NREL) provided X-ray characterization of catalysts. Separators for the hydrogen production node at Sandia National Laboratories supplied state-of-the-art alkaline membranes and ionomers. Hydrogen in situ test capabilities for the hydrogen production node at NREL provided the project with MEA fabrication and an in situ electrolyzer test. These tests have produced initial performance data that shows improved perovskite-S performance compared with IrO_2 at potentials greater than 1.7 V.
- With four government laboratories and a commercial vendor progressing toward an affordable system, it would be challenging to identify a better example of the integrated teamwork envisioned by the HydroGEN consortium.
- This project consists of excellent coordination between the participants.

Question 5: Proposed future work

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

- The proposed future efforts are reasonable; the planned work includes the study of the impact of other organic alkaline electrolytes, such as TMAOH, and the study of their impact on rotary disk electrode HER/OER with PMG-free catalysts. The planned experiments will probe phenomena occurring at the PMG-free catalyst and anion-exchange ionomer membrane, including in situ X-ray absorption spectroscopy (XAS). This fundamental information will lead to improved catalyst design for testing in AEM water electrolyzer tests. The project partner, Pajarito Power, will scale catalyst synthesis into the 25 g/batch range.
- With the progress made to date, the integrated alkaline electrolysis system testing at the suite of national laboratories will provide valuable results that should guide further design efforts and inform the balance of the HydroGEN consortium.
- The proposed future work is well aligned with ongoing efforts.

Project strengths:

- The project team has effectively synthesized a series of PGM-free, perovskite-based OER catalysts that show comparable performance to state-of-the-art IrO_2. The team has also shown the impact of organic alkaline electrolytes on OER performance. Incorporating these concepts into working systems will ultimately provide a route to high-activity and low-cost water-splitting technology.
- The team uses perspective from the end application to guide the project. This reduces the risk of developing an architecture that requires excessively complex and expensive deployed systems. This project also successfully leverages the capabilities of multiple partners.
- The project is well focused and well executed, with clearly defined milestones and objectives. It seems that all projected targets in the first year will be met.

Project weaknesses:

- There are notable schedule pressures on the project participants. The brevity of the presented catalyst stability data renders this one data set dubiously relevant.
- Not much work other than prolonged cycling is being done for careful durability studies.
- The overall performance does not appear as high as other PGM-free catalysts.

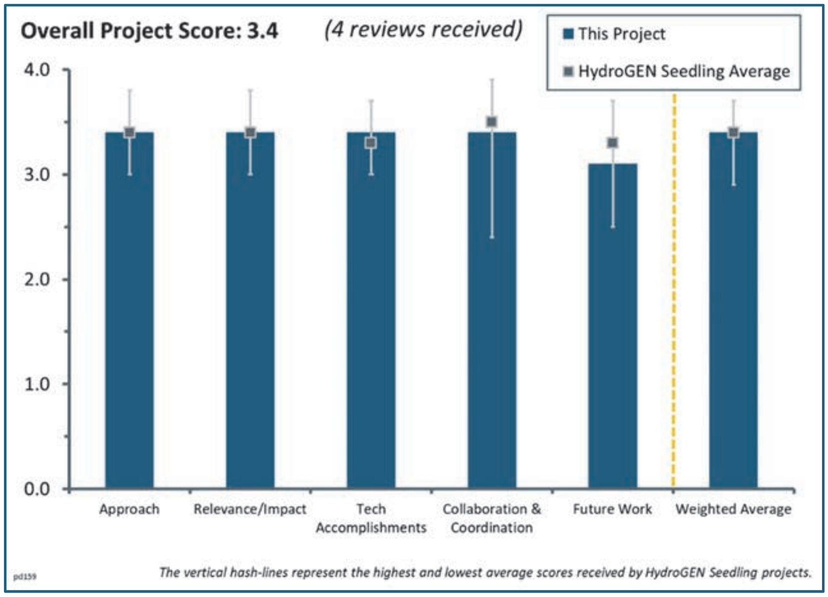
Recommendations for additions/deletions to project scope:

- It is recommended that the team include catalyst stability data of at least tens of hours. It would likely be worthwhile to peruse the expanding Energy Materials Network and HydroGEN consortium data hub prior to executing second-year activities. There is a potential that some relevant data already exists, which would enable this activity to redirect resources to the team's advantage.
- In situ durability evaluations should be performed on PGM-free catalysts, such as inductively coupled plasma mass spectrometry (ICP-MS).
- It is recommended that the team form a better description of the proposed future work and define a clearer path toward catalyst optimization.

Project PD-159: Scalable Elastomeric Membranes for Alkaline Water Electrolysis

Yu Seung Kim; Los Alamos National Laboratory

Brief Summary of Project:



The objective of this project is to develop stable, high-performance, and economically affordable alkaline anion-exchange membranes for water electrolysis operation. A low-cost synthetic method based on acid-catalyzed condensation reaction (Friedel–Crafts alkylation) will be developed to fabricate the styrene-based triblock copolymers based on polystyrene-b-poly(ethylene-co-butylene)-b-polystyrene (SEBS) to replace the prohibitively expensive metal-catalyzed reaction route. The project team, which also includes Rensselaer Polytechnic Institute and Proton OnSite, aims to develop economically viable elastomeric ionomers having conductivity at least equivalent to polyaromatic electrolytes, with much-improved mechanical properties.

Question 1: Approach to performing the work

This project was rated **3.4** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- This project will develop economically viable alkaline-conducting materials and demonstrate their performance in alkaline water electrolyzer systems. This is a good approach because membrane durability and performance has direct impacts on overall catalytic water splitting. Poor membrane performance and/or degradation will hinder the performance of even the best catalyst, so this is an important aspect to achieving low-cost hydrogen production from water splitting. The approach is reasonable for the one-year performance period and project cost. Key barriers to membrane performance include alkaline stability, hydroxide conductivity, and mechanical properties. The team has previously demonstrated more than 2000 hours of operation in an alkaline electolyzer using polyaromatic electrolytes. This work aims to develop cheaper, more mechanically robust elastomeric isomer materials with equivalent performance.
- This project focuses all effort onto one problem (alkaline membrane applicability) and methodically evaluates a number of potential options. The inclusion of both the catalyst–ionomer interface and the ionomer–membrane interface increases the potential for a successful outcome.
- This project’s straightforward approach benefits from an Energy Materials Network (EMN) node, as well as academic and industrial partners.
- Lowering the cost of alkaline electrolyzer materials is a primary goal for the project. However, it is not obvious why or how the new membrane materials or synthesis approach is expected to be less expensive. It is unclear whether the absence of a platinum-group-metal (PGM) catalyst alone makes the materials affordable or if there will be a need for other improvements. The project needs to describe the approaches on how to achieve lower cost.

Question 2: Relevance/potential impact

This project was rated **3.4** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- Membrane stability and performance are crucial components of electrolyzer performance. Improving state-of-the-art performance metrics, including hydroxide conductivity, stability, and mechanical robustness, is key to realizing cost-effective hydrogen production devices. This project directly addresses these issues. Better membrane performance will lower capital cost by removing the high noble metal loading requirements. It will also allow use of less expensive cell components because of alkaline conditions. Less permeation across the membrane will allow operation at higher pressures and current densities. Success will help mature alkaline membrane-based water electrolysis technology and ultimately achieve the goal of $2/kg H_2.
- Leveraging the polymeric tuning from previous work has resulted in a series of viable membrane options for continued development. The project is on schedule and pursues the path forward with a high probability of success.
- Exploring and demonstrating a stable and low-cost alkaline hydroxide conducting material is fully aligned with the EMN's low-temperature electrolysis development goals.
- This effort has the potential to lead to significant cost reduction of hydrogen production.

Question 3: Accomplishments and progress

This project was rated **3.4** for its accomplishments and progress toward overall project and DOE goals.

- The project has successfully synthesized and characterized semi-crystalline and cross-linked SEBS membranes. The membranes show good hydroxide (OH^-) conductivity and excellent stability during a 500-hour test in 1 M NaOH at 80ºC. First- and second-quarter (Q1 and Q2) milestones have been met, and Q3 and Q4 milestones are 50% or greater toward completion. Three materials have met the go/no-go decision criteria. Future testing will validate the performance of additional membrane materials. Modeling electrolysis performance based on membrane properties has been initiated. The findings predict thinner, more conductive membranes will improve overall cell performance. X-ray scattering experiments have characterized the membrane crystallinity as a function of water uptake. These results will help guide membrane optimization for improved performance and mechanical strength.
- Leveraging the polymeric tuning from previous work has resulted in a series of viable membrane options for continued development. The project is on schedule and pursues the path forward with a high probability of success.
- The project has met all milestones to date.
- The conclusion that "[Alkaline exchange membrane] properties are controlled by [ion-exchange capacity] and tailoring chemical structure" is not supported by the data presented. It is suggested that the project team further explore the causes or reaction mechanisms. For example, the team should determine whether there is a correlation between water uptake and hydroxyl conductivity, at least for some of the materials.

Question 4: Collaboration effectiveness

This project was rated **3.4** for its collaboration and coordination with HydroGEN and other research entities.

- The project shows good collaboration and coordination with other institutions. For example, the project has leveraged the modeling and electrochemical characterization node, while future efforts will utilize roll-to-roll manufacturing, hydrogen generation and dispensing, and X-ray scattering nodes. The project will also partner with Proton Onsite to demonstrate the best performance and durability of alkaline electrolyzer in the second and third years. Los Alamos National Laboratory (LANL) has worked closely with Sandia National Laboratories to improve chemical stability of benchmark anion-exchange membranes. LANL has discussed a possible collaboration with the National Renewable Energy Laboratory team for testing membrane-based water electrolyzers.

- This activity has delivered a potentially viable membrane for use in an alkaline system. This suggests the inclusion of the correct participants collaborating at an appropriate level to succeed.
- The collaborative plan among materials discovery, verification, and characterization teams looks sound.
- There are good interactions with the synthesis team and EMN nodes.

Question 5: Proposed future work

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

- Future work includes optimization of membrane chemical structures to balance OH- conductivity and mechanical durability, as well as completion of stability testing. The project will down-select alkaline ionomer materials based on micro-electrode testing and in situ area-specific resistance measurement and stability testing in electrolyzer cells. Future work with Proton Onsite will validate membrane performance in electrolyzer cells and test real-world stability.
- This is a good plan to address critical issues.
- The integration of the cross-linked alkaline-exchange membrane polymer will not be integrated into an electrolyzer for evaluation in a completed membrane electrode assembly until the second fiscal year. Until then, the component fundamental properties will be characterized, and a more durable balance between ion-exchange capacity and polymer durability will be sought.
- The proposed future work (assumed through September 30, 2018) to optimize the chemical structures is a bit vague. Some specific measures on how to achieve this would be helpful.

Project strengths:

- Empirical data suggests a viable alkaline membrane with potential for refinement to improve durability. The team has demonstrated the capability of executing this refinement.
- Controlled synthesis of modified SEBS polymers is the major strength.
- The novel lower-cost membrane synthesis approach is a project strength.
- The project scope is well defined and appropriate.

Project weaknesses:

- No substantial weaknesses have been observed.
- Lack of mechanistic discussion of the new materials' property–performance relationship is a weakness.
- The first year's work could have had more collaboration with EMN nodes.

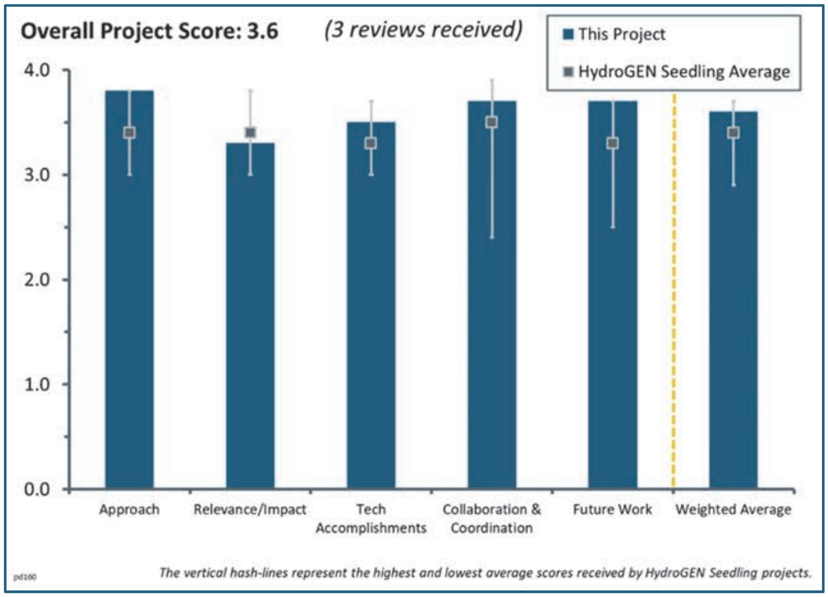
Recommendations for additions/deletions to project scope:

- The project scope is well defined and appropriate. Future work proposes adequate collaboration with EMN nodes.
- It would likely be worthwhile to peruse the expanding EMN and HydroGEN data hub. There exists the potential that some relevant data already exists that would enable this activity to advantageously redirect resources.
- It is recommended that the project team provide some explanation of why and how the new membrane materials or synthesis approach is expected to be less expensive than M-Cat or base materials.

Project #PD-160: Best-in-Class Platinum-Group-Metal-Free Catalyst Integrated Tandem Junction Photoelectrochemical Water-Splitting Devices

Charles Dismukes; Rutgers University

Brief Summary of Project:



This project will identify the best technical approaches to fabricate both high-performance (HP) and high-value (HV) platinum-group-metal-free (PGM-free) catalysts for photoelectrochemical (PEC) cells without compromising system efficiency. Next-generation devices must eliminate PGMs, even though they perform well, because of cost and sustainability limitations. Using recently developed low-cost HP catalysts, the team will examine the optimal pairing of these materials with established HP and emerging HV photoabsorbers. Cost–benefit analysis of full HP and HV devices and their individual components will enable the preparation of a hybrid product that will significantly advance the state of the art and that has the potential to deliver on all U.S. Department of Energy figures of merit: cost, performance, and stability.

Question 1: Approach to performing the work

This project was rated **3.8** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The project approach is very strong. The team clearly identified relevant barriers related to substituting PGM catalysts with earth-abundant materials that perform at the same level for solar water-splitting purposes. Other barriers related to catalyst light absorption and protection semiconductor components were identified. The research proposed is well aligned with the project objectives and is likely to result in advances toward surpassing the barriers identified. The project is well integrated with HydroGEN partners at the National Renewable Energy Laboratory (NREL), who will provide both HP III-V light absorbers and $ZnSnN_2$ HV materials. The interactions with HydroGEN partners effectively complement the expertise of the Rutgers University lead investigators in electrocatalysis.
- The team is specializing in non-PGM oxidation evolution reaction (OER) and hydrogen evolution reaction (HER) catalysts that are competitive with PGM catalysts. This is a worthy goal. The team is partnered with experts within the HydroGEN consortium, which will allow the project catalysts to be tested with the best-performing PEC materials. This project also takes a look at using these catalysts in conjunction with less expensive PEC materials to develop a HV PEC device. The approach looks likely to be very fruitful.
- The approach seems appropriate. The milestones are pretty nice. It seems as though the project team expects to meet or exceed all milestones. The team should consider the use of stretch milestones in future project phases. The purpose of these types of milestones is to give the project team an opportunity to demonstrate outstanding performance if they are achieved. If, however, the team fails to achieve a stretch milestone, the project is not punished. This is an effective way to internally standardize what the project team thinks is a big advancement versus what advancements may be expected as a matter of course.

Question 2: Relevance/potential impact

This project was rated **3.3** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- The project supports, in part, progress toward the goals of the Hydrogen and Fuel Cells Program in several ways. First, the development of inexpensive electrocatalysts could lead to lower costs when compared to state-of-the-art PGM materials, but these cost reductions are not likely to be significant enough to reach the $2/kg goal. The fraction of the cost that the electrocatalyst accounts for is small compared to other factors (e.g., efficiency and lifetime), and thus the team's focus on high-efficiency catalysts is appropriate. Implementing III-V semiconductor components could lead to the required efficiencies to achieve the desired hydrogen production cost, but these technologies suffer from significant cost drawbacks. The interactions with the NREL team are also very appropriate, as advances in III-V fabrication could help alleviate these cost disadvantages. Also, the incorporation of research on HV semiconductors is relevant, as it can also help achieve the target cost goal—as long as high efficiency is proven.
- This project supports the development of non-PGM and scalable electrocatalysts for PEC water splitting. It will lower hydrogen costs by increasing solar-to-hydrogen (STH) efficiency, decreasing production costs using lower-cost materials, and increasing lifetime. These are important to achieving the DOE target of hydrogen for <$2/kg.
- The performance is held relative to a standard. The cost tornado plot seems to suggest fairly equal opportunity for reducing costs from the center point versus getting stuck with a cost that is higher than the central point. This perhaps has better odds or opportunity than in other projects, improving the relevance and the chance for this project to stay afloat.

Question 3: Accomplishments and progress

This project was rated **3.5** for its accomplishments and progress toward overall project and DOE goals.

- The project has made significant progress toward meeting the intended performance indicators. Two milestones have been achieved related to the performance and stability of the $LiCoO_2$ OER electrocatalyst and the development of a high-efficiency III-V/Ni_5P_4 photocathode. The team has also made significant progress toward the first go/no-go criteria, and although the team has not achieved the desired current density to date, the demonstrated photocathode stability at roughly 90h at >8 mA/cm^2 is very promising. This stable behavior was achieved through the protection of the semiconductor with TiN, which protects the light absorber during the fabrication of the Ni_5P_4 layer. It is unclear whether higher current efficiencies (and STH efficiencies) will be possible with the materials system chosen, or what the strategy to improve performance is. One significant challenge is avoiding significant light losses in the electrocatalyst layer.
- Developing high-quality HERs and OERs that are stable and competitive with PGM catalysts is impressive. The team looks to be meeting project milestones and has the data to back it up.
- The accomplishments have been good. The team is advised to watch out for the oxides in the nickel phosphide film. It is unclear whether the occurrence of oxygen atoms is spatially correlated with the occurrence of Ni and/or P, and whether any spectroscopic evidence exists to justify invoking oxides in deconvolution in the x-ray photoelectron spectroscopy (XPS) spectra.

Question 4: Collaboration effectiveness

This project was rated **3.7** for its collaboration and coordination with HydroGEN and other research entities.

- The collaboration between the lead researchers and the HydroGEN Energy Materials Network has been very effective, as evidenced from successful early results achieved through joint efforts between Rutgers University and NREL.
- Good-to-excellent collaboration exists.
- The team is making good use of the expertise in the HydroGEN consortium for synthesis and benchmarking. It is unclear how the high-throughput/combinatorial synthesis node was used, other than for the fabrication of the $ZnSnN_2$, or whether the high-throughput synthesis was used to discover this material.

Question 5: Proposed future work

This project was rated **3.7** for its proposed future work.

- The proposed future work slide is perhaps the best that this reviewer has seen. It provides enough detail to shine light on the scientific line of reasoning that justifies the work, and it includes some SMiles ARbitrary Target Specification (SMARTS) characters.
- The proposed future work is consistent with the needs of the project to achieve the intended goals. Thrust 1 future work is heavily focused on stability performance, which is reasonable given the potential impact on hydrogen cost, but additional work on performance improvements will also be needed to achieve the DOE goals. More significant challenges are faced in Thrust 2, in which the HV PEC materials exhibit significant drawbacks, in terms of performance and stability. The proposed activities are reasonable efforts to improve the potential of HV materials.
- The proposed work addresses the remaining barriers. Importantly, a large focus is placed on increasing the stability in alkali conditions; this will be tested under diurnal cycles.

Project strengths:

- The strengths of the project include the following:
    - The electrocatalysts developed for the team (both for OER and HER) show very promising performances that are comparable to PGM catalysts.
    - The protection strategies have allowed the team to demonstrate enhanced stability.
    - The research approach is well designed for the project needs, and the team has been able to achieve the initial project milestones.
    - There are excellent interactions between team members and between lead researchers at Rutgers University and the HydroGEN consortium.
- The non-PGM OER and HER catalysts have performances that are on par with PGM catalysts. Another strength is the promise of TiN film for protection against corrosion.
- This project team seems technically competent and is publishing based on project results.

Project weaknesses:

- The project team should consider using stretch milestones so that one may differentiate between "business as usual" and truly remarkable advancements.
- There are two weaknesses within this project. The first is that the strategy to achieve the high performance required for PEC materials containing III-V semiconductors is unclear; light absorption losses in the electrocatalysts will be challenging to overcome. The second weakness is that the choice of HV materials is likely to result in limited performance, which would make it very difficult to achieve the $2/kg hydrogen target.

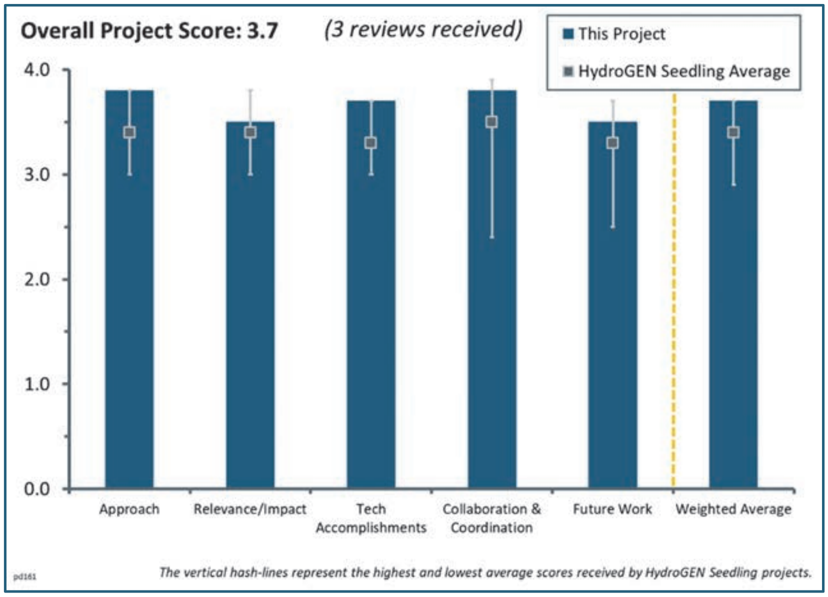
Recommendations for additions/deletions to project scope:

- Since the team has shown significant progress in the development and protection of HP III-V PEC materials, most of the benefits in the future research will likely come from advances in HV materials. Shifting efforts from Thrust I to Thrust II might make sense for the duration of the project.

Project #PD-161: Protective Catalyst Systems on III-V and Silicon-Based Semiconductors for Efficient, Durable Photoelectrochemical Water-Splitting Devices

Thomas Jaramillo; Stanford University

Brief Summary of Project:



The overall goal of this project is to develop unassisted water-splitting devices based on III-V materials, creating pathways to improve performance in terms of efficiency (>20% solar-to-hydrogen [STH]), durability (two weeks), and cost (<$200/m^2). Two distinct water-splitting schemes are being pursued: Scheme 1 (tandem III-V/III-V) aims to develop high-efficiency devices with tandem III-V photoabsorbers (e.g., $GaInP_2$/GaInAs), and Scheme 2 (III-V/Si) targets cost reduction while maintaining high efficiency by growing InGaN on crystalline Si. Both schemes will be coupled with thin-film, semi-transparent hydrogen and oxygen evolution reaction catalytic/protection layers containing reduced or zero precious metal content that can enhance durability while maintaining high efficiency and enabling low material costs.

Question 1: Approach to performing the work

This project was rated **3.8** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The research approach of this project is well designed and appropriate to achieve the goals. The barriers are clearly identified, and the team structure is composed of experts with complementary expertise that is likely to result in surpassing these barriers. Protection III-V semiconductors with electrocatalysts can lead to stable interfaces for solar water splitting, growing III-V semiconductors in Si can significantly lower the cost of production, and testing the system for long periods of time will provide insights into the possible implementation of the proposed photoelectrochemical (PEC) materials system. The activities in budget period 1 strongly support the validation of the technology being studied.
- The team identifies key barriers including stabilization, fabrication of high-quality InGaN on Si, and benchmarking on long timescales. The project proposes promising ways to overcome these barriers with the help of the National Renewable Energy Laboratory (NREL) characterization, analysis, and benchmarking nodes.
- The project seeks Si/III-V tandem solar cells to enable the technology, but the proposed target for this is a very qualitative "high-quality InGaN epitaxial growth on Si." The project claims that MoS_2 provides stability, yet the derivative of the curve representing current density versus exposure time does not equate to zero. In other words, the system is not stable in the strict sense of the word. Regarding the stabilization of Si for months using MoS_2, the J value goes to zero after about Day 65, and it does so spectacularly. It is unclear whether this is catastrophic failure and what the mechanism behind the failure is, as well as how that mechanism has been analyzed as part of a long-term strategy to overcome the failure. The technoeconomic analysis does not include error bars or other such representations of uncertainty. Uncertainty tends to compound and therefore confound the selection of winning technologies. The poster did not seem to attempt to address this. The corrosion analysis of the materials could be better structured. Metal–organic chemical vapor deposition (MOCVD) requires high vacuum. It would be beneficial to

eliminate high vacuum steps to decrease costs. Several of the project milestones contain qualitative statements or components. For example, Milestone 1.1 states, "Demonstrate >100 h stability," but "stability" is not defined as less than some absolute or less than some relative change from the starting point. Thus, if the time-based derivative of the curve representing the systems performance is not equal to zero, then the milestone is not met. Milestone 4.1 is very vague: "Demonstrate effectiveness of the operando microscopy flow cell measurement technique on a benchmark photoelectrode." This does not specify success criteria for process stability and absolute accuracy. It does, at least, specify that some benchmark will be used as a (presumably) standard material. The go/no-go states that the project will "...provide a viable pathway for achieving 20% STH efficiency..." It is unclear what this even means. "Viable" is an adjective describing the noun "pathway"; hence it would then be expected to see some pathway or timeline with SMART (specific, measurable, attainable, relevant, and time-bound) criteria flanking each and every technical point along this technology development line. It is unclear if "viable" means that no targeted performance metric should fall outside of two sigma from the existing known mean for that variable's performance. In other words, it is unknown if "viable" means that the project need not invoke any statistical long shot to achieve the overall objectives. It is unclear whether "viable" should be taken to mean that no invention must occur to achieve the objectives. Surely, pathways that must invoke invention cannot be guaranteed viable unless invention can be guaranteed. No such philosophy, schematic, analysis, or other definition of "viable" seems to be presented.

Question 2: Relevance/potential impact

This project was rated **3.5** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- The project focuses on improving the efficiency and stability of high-performing PEC materials—two of the most important factors in achieving low hydrogen production costs. This focus is highly relevant for DOE's goals and could lead to substantial advances toward the $2/kg of hydrogen target. One of the drawbacks is the implementation of expensive III-V materials, another area of focus of the project. The proposed approach of growing III-V on Si has high potential to lead to lower production costs for these high-performing light absorbers. The project fits well within the HydroGEN consortium, as it effectively leverages the fabrication, characterization, and on-sun testing tools available at NREL.
- Achieving >20% efficiency and high durability while using earth-abundant materials is necessary to achieve the DOE target. This project is likely to make progress toward those goals by improving stability through protection layers.
- This project seeks to use a (roughly) seven-layer device to effect the desired water-splitting reaction. It benefits from the use of established solar-cell manufacturing technologies. However, the complexity of the construct will ultimately limit the amount of cost reduction that can be achieved. The poster states that high-efficiency, durable, low-cost photoelectrodes are required to deliver cost-effective hydrogen. In other words, all three criteria need to achieve some measure of performance extremes for the technology to be viable. This will be challenging. The figure on slide 12, in which the known research/materials space is plotted as "Demonstrated Charge Passed" on the y-axis and "Onset potential vs [reversible hydrogen electrode]" on the x-axis, would have benefitted from shading the region that represents success. This would help visually emphasize the materials being studied in this project as potentially impactful, being on the edge of or in the success region.

Question 3: Accomplishments and progress

This project was rated **3.7** for its accomplishments and progress toward overall project and DOE goals.

- The project demonstrated a PEC photoelectrode that achieves >10 mA/cm^2 under 1 sun illumination for longer than 100 h. The team fabricated an unassisted PEC water-splitting device with a nonprecious metal hydrogen evolution reaction catalyst that achieves STH efficiencies >5% under 1 sun illumination to provide a viable pathway for achieving 20% STH efficiency, through integration strategies of the materials and interfaces under investigation.

- The team has made very significant progress toward achieving the project goals. Two go/no-go criteria have been met to date, and several of the milestones have been hit. A MoS_2 protected GaInP photoanode was demonstrated to have an efficiency >100 h, and InGaN was successfully grown on Si. Unassisted water splitting was also demonstrated for >10 h, at current densities >10 mA/cm^2 and STH >5%.
- The team has successfully grown single-crystal pn+ -$GaInP_2$, sputtered Mo onto this, and sulfidized the surface with deadly H_2S gas. Slide 11 did not explain why the MoO_3 3d peak goes away in the before/after insert. Also, the derivative of the red curve in this figure does not appear to equate to zero, suggesting that there is some change occurring in the system. The overall conclusion on slide 11, that MoS_2 is superior to PtRu, holds true. Slide 16 fails to use basic statistics to establish the precision and make claims to the accuracy of the new in situ microscopy flow cell method.

Question 4: Collaboration effectiveness

This project was rated **3.8** for its collaboration and coordination with HydroGEN and other research entities.

- The collaboration between the team members at Stanford University and the HydroGEN nodes at NREL is very strong and has allowed the team to make significant progress toward the project goals. The skills of the team members are complementary and clearly differentiated, which has led to promising results in Year 1 of the project.
- Weekly meetings with NREL and the weekly exchange of samples show a high degree of collaboration with Energy Materials Network (EMN) nodes and partners. The team has a plan in place to incorporate the project data onto the HydroGEN data hub.
- There seems to be strong interaction with NREL.

Question 5: Proposed future work

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

- The team has a clear research plan to achieve most of the milestones proposed in the project. Fabrication of Si/InGaN PEC material systems would be a promising step toward cost-effective solar hydrogen generators. Enhancing the STH efficiency to 20% from the achieved values might prove to be challenging, and a pathway for this improvement has not been clearly defined. The proposed in operando corrosion methods would be very useful in identifying failure mechanisms and will help avoid them in future materials systems.
- Future work addresses barriers, including strategies to boost performance and strategies to probe corrosive failure mechanisms.
- The proposed future work seems to be a logical extension of the prior budget period's work. However, portions of the proposed future work are vague (e.g., "implement protection scheme and catalyst") and do not convey how efficiency will be achieved in conducting the research activities—for example, "Boosting stability and catalysis on tandem photo-absorber systems by optimizing..." (a long string of inter-related parameters is then listed). It is unlikely that the project team will measure all possible parameter combinations. It is unclear how the team concludes that a better collection of parameters was not missed, if the outcome from the narrow slice of parameters actually tested was less than perfect. It is unclear why the team did not use a statistical design of experiments to lay out the entire process space and then develop science- and engineering-based arguments to focus in on portions of that space. A design-of-experiment would inform on the parameter combinations that are actually measured, as well as those neighbors around them that were not measured; efficiency of effort is realized in this way. The work exploring corrosion mechanisms of failure is poorly articulated and should receive intense scrutiny. The mechanism of failure is likely a compounded action of chemical and physical phenomena that may not be easily de-convoluted.

Project strengths:

- This project has a very competent chemical engineer as the principal investigator (PI). All project participants are well accomplished as individuals and in teams. The idea of using a transition metal to simultaneously protect the surface and to affect the desired chemical reaction stands to open a rich and fertile field of catalysis. Indeed, some of the most important catalysts of modern times are based upon transition metals. Also, nature offers many models for the manipulation of protons using transition metal complexes. This project also presents the opportunity to run a senior design course to get a better grasp of the "for sale," ready-for-deployment embodiment of this technology.

- There is strong collaboration between Stanford University team members and the HydroGEN consortium. There is also significant progress toward project goals, promising initial results on the protection of III-V semiconductors with MoS_2 electrocatalysts. The work plan is clearly defined and well aligned with project objectives. This research approach could lead to significant advances toward DOE goals.
- The approach is strong, with high levels of collaboration and coordination with EMN nodes.

Project weaknesses:

- This project needs a strong chemist if the transition metal chemistry is to be fully leveraged. Also, discussion with the PI indicated that an engineer could probably add value in devising how this technology could operate in the field.
- Significant advances are required to achieve the project's goal of 20% STH efficiency, and a clear pathway to this was not presented. Also, parasitic absorption losses in the MoS_2 may need to be investigated in further detail to avoid light losses in this layer.

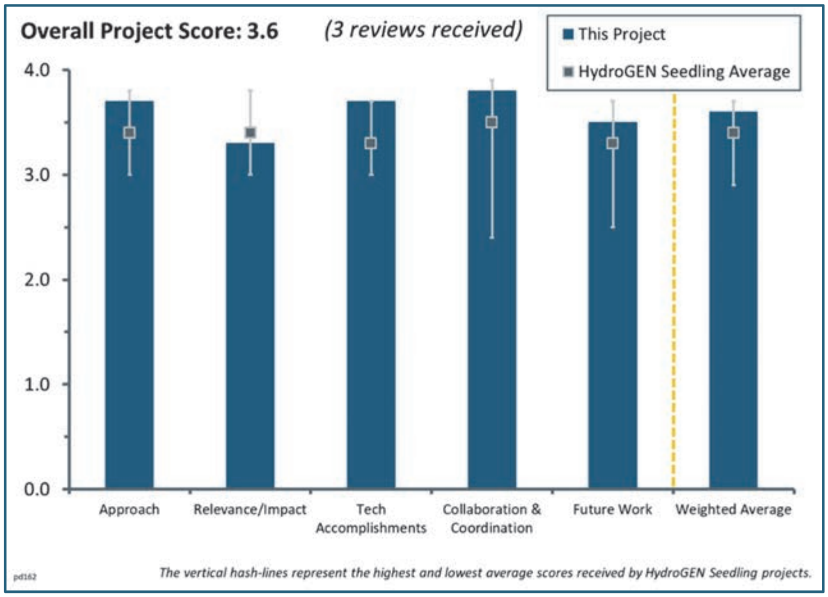
Recommendations for additions/deletions to project scope:

- No additional recommendations are provided.

Project #PD-162: Novel Chalcopyrites For Advanced Photoelectrochemical Water Splitting

Nicolas Gaillard; University of Hawaii

Brief Summary of Project:



The overarching goal of this project is to create a chalcopyrite-based, semi-monolithic, tandem hybrid photoelectrode device prototype that can operate for at least 1,000 hours with solar-to-hydrogen (STH) efficiency >10%. The performance of previously identified wide-bandgap chalcopyrite materials will be improved through alkali doping to passivate $CIGS_2$ defects, and next-generation chalcopyrites (e.g., Ga-free) will be developed. The photoelectrochemical (PEC)–electrolyte interface energetics and stability will be improved by investigating alternative buffer materials and protective layers. Also, novel fabrication methods will be developed for creating the semi-monolithic chalcopyrite-based tandem devices.

Question 1: Approach to performing the work

This project was rated **3.7** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The research approach is well defined and aligned with the goals of the project. The team will synthesize chalcopyrite materials with appropriate bandgaps using inexpensive printing methods. The researchers will also develop interfacial materials to protect the semiconductor components and enhance efficiency toward the solar water-splitting process. Lastly, the researchers will integrate materials in PEC configurations using conductive composite materials. The project effectively leverages several nodes of the HydroGEN consortium for its theory components, interfacial electrode protection, and PEC testing.
- The approach of the project is focused on overcoming the chalcopyrite manufacturing cost, non-ideal surface energetics, chalcopyrite durability, and device configuration. The project partners with theory, PEC benchmarking, and solid-state interface experts.
- This project seeks to integrate several technical thrusts. The organizing structure by which these coalesce needs to be better defined. The project would benefit from stating the critical path (in the technological sense). There does appear to be ownership of individual nodules of effort in which existing infrastructure and capabilities make this a natural phenomenon. For example, it is clear why the National Renewable Energy Laboratory owns that part which it owns, and it is clear why Stanford University owns that part which it owns. However, it could be clearer what exactly each is putting forth into the backbone of the technology's advancement.

Question 2: Relevance/potential impact

This project was rated **3.3** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- Developing high-efficiency, stable, and inexpensive PEC materials is central to achieving the DOE's cost target for hydrogen production. The team has clearly identified a pathway for the synthesis of tandem chalcopyrite light absorbers with tunable bandgaps, through the implementation of an inexpensive printing method. The use of this fabrication method has potential to lower the cost of hydrogen production, and the ability to stack multiple layers of chalcopyrite materials with synergistic light absorption properties could lead to higher performance. Despite this promising approach, the target efficiencies are well below the ones needed to achieve the DOE cost target. The project effectively leverages several nodes of the HydroGEN consortium for its theory components, interfacial electrode protection, and PEC testing.

- Overcoming the identified barriers is likely to make significant progress toward DOE's hydrogen production cost goals. The project makes good use of the Energy Materials Network (EMN) nodes.
- The project is doing a great job leveraging the Hydrogen and Fuel Cells Program's (the Program's) infrastructure. The science seems sound, and innovation is occurring on multiple fronts. There is some doubt as to the technology's ability to achieve the Program's cost targets. Low-cost flex-film photovoltaic (PV) manufacture has not achieved scale. Also, the project seeks to pass through a nine-layer intermediate prototype and then seeks to converge upon a six-layer device as the final goal. The approach will require semiconductor fab-type processing. The number of layers translates to a number of steps greater than or equal to this number of layers; each (or many) of these steps occur in a highly controlled, high-cost facility. This ultimately translates to a high-cost product. The cost sensitivity analysis does well to employ a tornado-type plot; however, what these plots reveal is that cost-reduction opportunity is actually skewed against us. That is to say, the size of the cost bands on the detrimental side of the indicated starting point are much larger than the size of the cost bands on the beneficial side. In other words, when it comes to cost reduction, there is more ground to lose than there is to gain. This asymmetry in the tornado plots probably becomes common for technologies that are reaching maturity. This technology has not reached maturity, and so a second interpretation is that achieving the cost targets is a long shot. The principal investigator (PI) seems to admit to this, stating that cost targets may only be met by achieving two extreme value events in concert: 25% STH and 10-year lifetime. The probability of this seems quite low.

Question 3: Accomplishments and progress

This project was rated **3.7** for its accomplishments and progress toward overall project and DOE goals.

- Milestones are being met, and future milestones are well on their way to being met. Efficient chalcopyrite films have been successfully printed and characterized, protection against photocorrosion with TiO_2/MoS_2 has increased the stability up to 350 hours, and innovations in transparent conductive binders have been developed to enable chalcopyrite device development. Supporting data is provided, including data from EMN nodes and partners.
- The team has shown significant progress toward achieving the proposed milestones, demonstrating the synthesis of polycrystalline chalcopyrite films and a robust nanowire-based method to create transparent binders between different PEC layers. A chalcopyrite PV device has been demonstrated with efficiency of 8.4% solar-to-electricity, placing the project close to achieving the first go/no-go criteria. Significant challenges were encountered to protect the materials under acidic conditions, but the materials seem to be stable under neutral pH.
- The PI states that the project is on track to achieve all of its milestones. This is a good outcome if the milestones were more challenging and the definition of having "met" them was less ambiguous. It may be worthwhile to consider incorporating one or more "stretch milestones," wherein the PI could demonstrate outstanding achievement if this milestone is met yet not be penalized if it is not met. This method of challenge brings into sharp contrast what is challenging for the team and what is easily met. This will vary from project to project. The milestones do appear to be Specific, Measurable, Achievable, Relevant, and Timely (SMART). The level of sophistication in how success is demonstrated could be increased. For example, there does not appear to be a widespread use of figures of merit. These mathematical formulations of success typically incorporate competing physical phenomena so as to respect the trade-offs usually encountered in developing a technology. The raw data for analyzing, to this end, are presented in the accomplishments section; it is just the synthesis of this data that could be improved upon, at least from a project management perspective. Depositing layers using a printing process instead of a vacuum process could be a big win for reducing manufacturing costs. The data show that impurity levels need to be better controlled, though, and this could re-introduce costs. When a new process is introduced, there must come a time when the stability of the process and the reproducibility of the process must be established. The

argument is oft made that the final product of the process is not known, so such endeavors are not very useful. The use of standards is one way to get around this. There is considerable concern as to the mechanism of failure for these devices. One may observe in Section 2.2c that the rate of degradation in the photocurrent (mA/cm^2) is the same in both the light and in the dark. It is not clear why the sample goes to zero in both sets of conditions in basically the same amount of time. One explanation is that light has nothing to do with the degradation phenomena—the system is just thermodynamically unstable, and the mechanism of degradation is kinetically accessible (independent of the interaction with photons, assuming dark conditions are actually dark and that there are no leaks, etc.). There exists a body of literature critiquing the value of "dark" experiments.

Question 4: Collaboration effectiveness

This project was rated **3.8** for its collaboration and coordination with HydroGEN and other research entities.

- Collaboration with EMN partners is extensive and well coordinated. The project is contributing to the HydroGEN Data Hub with information on chalcopyrite absorbers and n-type buffers.
- The collaboration between the PI and the HydroGEN consortium is effective. The different team members contribute complementary expertise to the project, which has resulted in obtaining satisfactory initial results.
- It is clear that there are multiple nodes engaged here, with each node essentially representing what could be a standalone project. The goals and impacts of each node are stated, but the researchers could benefit from a SMART approach.

Question 5: Proposed future work

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

- Future work is focused on critical barriers including improving protection layers, device integration, and printing new, higher-efficiency chalcopyrites.
- The proposed future work is a logical continuation of the Budget Period 1 work.
- The proposed future work is appropriate, as it focuses on key challenges identified on the first budget period. The discovery and integration of multiple chalcopyrite components in order to achieve >20% efficiency is well aligned with DOE goals, but a clear pathway to achieve this goal has not been identified. Stability is also very important to achieving the cost target, and the team intends to focus on surpassing the stability challenges faced in the first stage of the project. Device integration is necessary to demonstrate the potential of chalcopyrite materials in PEC devices.

Project strengths:

- This project has a very clever PI and is ambitious. The team appears to be meeting all of the milestones as they have been formulated. The researchers are attempting to select and employ/deploy significant discoveries/innovation in other fields (i.e., silver nanowires) to solve the problems in their own field. There appears to be some synergy between this project and others in the Program's portfolio.
- The project's strengths are as follows:
    - An inexpensive fabrication method for light absorbers
    - Simple assembly methods for multilayer devices through the use of hybrid polymeric materials
    - A good integration of team members and HydroGEN nodes, and the leverage of expertise from multiple team members
    - The early achievement of milestones, and a promising path to achieving the go/no-go points
- The EMN partnership and collaboration is excellent. There are solid accomplishments in materials processing, device fabrication, and stability.

Project weaknesses:

- This project is very complex and requires several substantial technical barriers to be retired simultaneously if it is to be successful. There are questions around the ability of the project to meet the cost targets. It is not clear that one could conclude that the technology does not work, even if the project proceeds through its entire planned life. Said another way: it is obvious when something works, but it is not always so obvious when it does not work.
- The project's weaknesses includes challenges for protection strategies under acidic conditions because of the roughness of semiconductor layers. Also, the initial demonstrated efficiency of the PV device is low and presents challenges for developing a 20% STH from chalcopyrite materials.

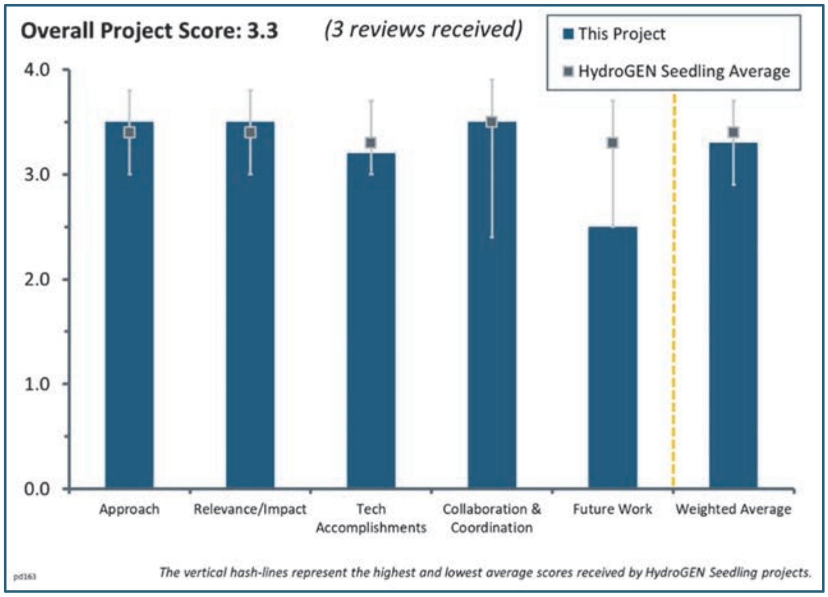
Recommendations for additions/deletions to project scope:

- Identifying a clear pathway to 20% STH is critical to achieving the project goals. Enhancing activities that lead to identifying such materials systems could enhance the potential for project success.

Project #PD-163: Monolithically Integrated Thin-Film/Silicon Tandem Photoelectrodes for High-Efficiency and Stable Photoelectrochemical Water Splitting

Zetian Mi; University of Michigan

Brief Summary of Project:



This project seeks to establish a low-cost and scalable platform for high-efficiency and stable photoelectrochemical (PEC) water-splitting devices and systems. The improved performance of the top photoelectrodes is required to realize high-efficiency, unassisted solar water splitting, and a functional wide bandgap tunnel junction that can be fabricated on a silicon platform is a critical component of a silicon-based tandem solar water-splitting device. The tandem photoelectrodes being developed in this project use silicon as the bottom light absorber and newly developed low-cost photoelectrodes made of Ta_3N_5, BCTSSe, or InGaN as the top light absorber. As silicon and gallium nitride are the two most produced semiconductors, the technology being developed will be scalable and lend itself to low-cost manufacturing.

Question 1: Approach to performing the work

This project was rated **3.5** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The project's approach is to place Ta_3N_5, BCTSSe, and InGaN top photoelectrodes on a GaN nanowire tunnel junction, and to demonstrate a double-junction photoelectrode on Si. The project utilizes resources from the Energy Materials Network (EMN) for modeling, materials diagnostics, in situ surface characterization, and catalyst deposition, as well as testing and stability analysis.
- Three barriers were identified: durability, integrated device PEC configuration, and scalable manufacturing of a monolithically integrated photoelectrode consisting of a bottom Si cell and a top light absorber with a 1.7–2 eV bandgap. The research approach is focused mainly on synthesizing multi-junction PEC materials based on Ta_3N_5, BCTSSe, and InGaN on Si and characterizing their interfacial properties. This approach is relevant to the barriers identified but lacks aspects of PEC device development and characterization. The Budget Period 1 activities related to the initial materials fabrications are designed to validate the proposed concept. While the photoelectrode development aspects of PEC technologies are important and are a key driver for the ultimate solar-to-hydrogen (STH) efficiency, equally important are device design aspects that have been overlooked in the current project.
- The approach is intellectually robust. In the hopes of keeping costs down, the project attempts to exploit the two most widely produced semiconductor materials in an innovative format(s). The project seems very academic. Budget Period 1 go/no-go decision points do have some metrics; however, the statement that "meeting these milestones will validate the concept...for the scalable production of solar hydrogen" is a leap too far to be supported by the milestones as presented. Scalability has to do with many factors not stated in the go/no-go milestones, one of which is production yield. There is no yield metric stated because there is no mature process for making these devices.

Question 2: Relevance/potential impact

This project was rated **3.5** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- The objective of the project is well aligned with DOE goals. Developing high-performing, inexpensive PEC materials that achieve efficiencies >20% is a challenging task. The team proposes to achieve this goal by synthesizing multi-junction absorbers with cost-effective semiconductors. The team leverages multiple HydroGEN nodes to complement its expertise in surface analysis, computational materials modeling, probing interfacial phenomena, and deposition of electrocatalysts and protection layers.
- The project will attempt to address DOE efficiency goals by using high-efficiency, yet inexpensive, Si light absorbers. Top photoelectrodes will be passivated by a N-rich GaN surface (which is much more stable compared to the Ga-terminated alternative), which will attempt to address the DOE stability goals.
- Overcoming corrosion would be a relevant outcome. The best approach for doing this would be one that uses a system that is thermodynamically stable toward corrosion. Perhaps N-terminated GaN achieves this. However, if it is deposited as an ultra-thin protection layer, then the project is ultimately pursuing a kinetic strategy to inhibit corrosion. Specifically, it appears to seek a mass-diffusion barrier layer to prevent corrosion. This layer is an externally deposited layer, necessitating two things: (1) it must be absolutely defect-free, and (2) it must last for the entire lifetime of the device. These are objectives that, quite frankly, all projects pursuing such non-dynamically responsive kinetic barriers to corrosion are unlikely to meet. Much more sophisticated strategies have been developed for dynamically responsive, self-healing corrosion-preventative layers in the traditional metal alloy space. Perhaps learning could be gleaned from there. For example, the so-called alumina-forming alloys rely upon a reservoir of aluminum in their composition where that aluminum diffuses to the alloy surface and oxidizes, forming a dense, adherent, mass-transport (kinetic) barrier to further oxidation. If that kinetic barrier fails (spalls off), new aluminum atoms are there, ready to heal the site of failure before component failure can take off.

Question 3: Accomplishments and progress

This project was rated **3.2** for its accomplishments and progress toward overall project and DOE goals.

- The team has demonstrated significant progress during Budget Period 1. Computational models have been developed to demonstrate the role of N-rich surfaces in the improved stability of GaN. Top absorber films have been successfully grown, a method for Pt deposition has been developed, InGaN nanostructures with varying composition and bandgaps have been achieved, and InGaN photocathodes were fabricated on top of Si, demonstrating current densities as high as 12 mA/cm^2. These developments suggest a high probability of achieving the initial go/no-go criteria.
- The project has successfully synthesized and characterized various top photoelectrodes on Si wafers. It was unclear if/how the N-rich surface would be prepared to achieve stability.
- Theoretical modeling around N-terminated GaN stability toward corrosion is underway but still in its infancy. The presentation states that "theoretical models and interpretations will be validated by experiment," yet it does not offer up which specific parameters will be stated by the model and measured by experiment, nor does it offer the quantitative level of agreement these two will achieve. Nice work was done on the atomically ordered InGaN deposition. When a non-aqueous method of catalyst deposition has to be developed to "reduce complications caused by catalyst growth in aqueous systems" but knowing the catalyst must ultimately operate in an aqueous environment, one becomes uneasy. It would seem that the synthesis of crystalline Ta_3N_5 thin films is an achievement; however, it is unclear whether these films have ever been synthesized before (by others). Also, the method of observing crystallinity (X-ray diffraction [XRD]) cannot quantify the degree of crystallinity. That is to say, if there are amorphous regions in the film, XRD will not do well to see it. Therefore, one may conclude that, yes, there is crystalline Ta_3N_5, but one may not conclude how much. It would not be good to risk visual/optical/scanning electron microscope images of grain pattern as a quantitative indication of the degree of crystallinity.

Question 4: Collaboration effectiveness

This project was rated **3.5** for its collaboration and coordination with HydroGEN and other research entities.

- Collaboration with the EMN looks extensive. This project has a specific plan to contribute to and benefit from the HydroGEN Data Hub.
- The collaboration in the team is strong, and the integration of HydroGEN resources in the project has benefited the outcomes of the project.
- The collaborations seem to be proceeding pretty much as expected; however, perhaps for reasons out of the control of the project team, data from the advanced light source has not yet been obtained. This makes it difficult to tie all the GaN-based phenomena together.

Question 5: Proposed future work

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

- The objective of the proposed future efforts is the development of multi-junction photoelectrodes with >15% STH efficiencies and long-term stability. While these are worthy objectives, a clear pathway to achieve these goals is lacking. The initial performance achieved by the team suggests that a much lower efficiency should be anticipated.
- The proposed future work provides a general outline to overcoming barriers, although it is a little light on details.
- The articulation of proposed future work was inadequate, amounting to some high-level metrics and a budget number. The proposed future work slide does not make clear which scientific hypotheses have been tested and retained, versus which have been tested and retired. Scientific hypotheses may be retired for one of two reasons: either they are accepted by the broader community as the new null (baseline condition), or they are deemed “inadequate.” It is hard to track progress if hypotheses and the changes to hypotheses are not presented in the context of budget and time.

Project strengths:

- This project features strong collaboration between team members and effective leveraging of HydroGEN nodes, as well as significant progress on the synthesis of proposed materials and characterization of performance.
- The project excels at materials synthesis and characterization. The presenter showed a video of stable, unassisted water splitting, although it would have been beneficial to see data specifically relating to the performance and durability of that demonstration.
- This project has good materials science.

Project weaknesses:

- The initial performance is low and does not warrant development of a 15% STH device.
- More detail on the future work is necessary.

Recommendations for additions/deletions to project scope:

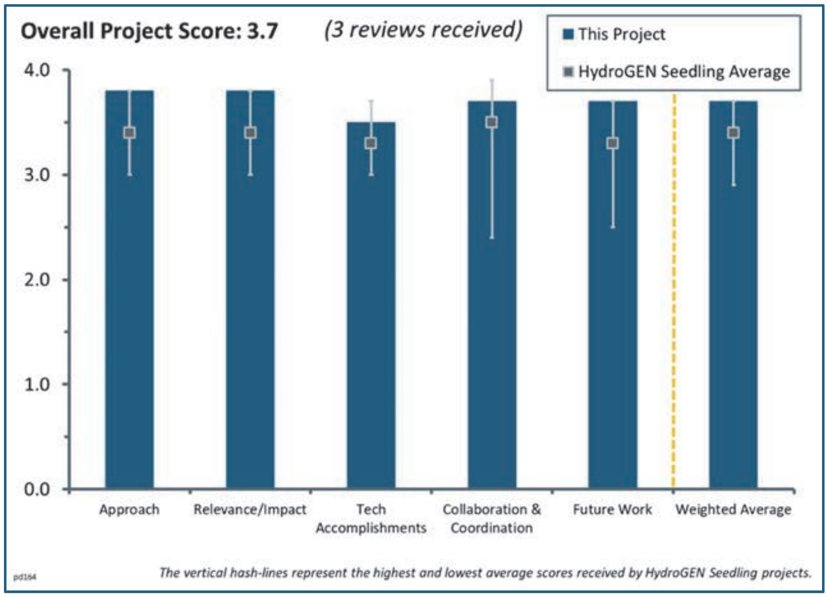
- Additional efforts should be devoted to developing high-performing photoabsorbers to develop a pathway toward the 15% STH goal.

Project #PD-164: Efficient Solar Water Splitting with 5,000-Hour Stability Using Earth-Abundant Catalysts and Durable Layered Two-Dimensional Perovskites

Aditya Mohite; Los Alamos National Laboratory

Brief Summary of Project:

This project builds on recent breakthroughs in high-efficiency perovskite solar cells and seminal work on using low-cost, earth-abundant materials for hydrogen evolution reaction (HER) and oxygen evolution reaction (OER) catalysts. Combining these two approaches enables the development of a disruptive low-cost photoelectrochemical (PEC) platform that would be a paradigm shift from the current state-of-the-art technology. The project goal is to develop a PEC device with solar-to-hydrogen (STH) efficiency of greater than 15% with 5,000 hours' stability.



Question 1: Approach to performing the work

This project was rated **3.8** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- This project is interesting, with some of the best "pure science" to which this reviewer was exposed. The "pure science" is being brought to bear upon a much applied problem: manufacturing processes do not produce perfect crystals. This project is one of the few that seem capable of portaging basic energy science over the saddle point into the applied realm. That is to say, this project/principal investigator (PI)/team seems capable of overcoming whatever the activation energy is that keeps so many of the discoveries in the basic sciences from finding their way into the applied. For example, the observed light-induced lattice defect correction (which may occur through photon–lepton–phonon "spooling") is a dynamic that is not some curiosity that lasts picoseconds or nanoseconds in a particle beam. No, this phenomenon is of a time constant (in minutes) that matters in the targeted application's timeframe. The approach was well done.
- The project has an intriguing approach and device design. It uses high-efficiency perovskite solar cells where the bandgaps are easily tuned. Anode and cathode will be connected in series in such a way that the two electrodes do not compete for light absorption. Since perovskites are notoriously unstable in water, the challenge will be adding a sufficient tunnel barrier that does not significantly increase recombination and resistance. This barrier/issue could have been emphasized a bit more. This is especially true since the goal is 5,000 hours of stability, which more stable materials have difficulty achieving. The project is well suited to be integrated into the HydroGEN network.
- The research approach relies on the incorporation of perovskite photovoltaics (PV) in PEC devices to obtain high STH efficiency with earth-abundant and inexpensive components at high efficiency. The team clearly defined a pathway to achieve this objective, and the project is divided into each of the components required for the development of the materials system. The go/no-go criteria for Year 1 was designed to evaluate the potential performance of the materials proposed for achieving high-efficiency solar water splitting.

Question 2: Relevance/potential impact

This project was rated **3.8** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- This project is very relevant and has potential for (long-term) high impact. Higher-yielding devices will occur if the defects in these devices can be photo-blotted out. Earth-abundant catalysts will be more cost-effective than those that are not—this is a classical approach to cost reduction. Cheap means adoption, which means impact. Since surface-based catalysis typically occurs at defects, lattice edges, or other surface protrusions (features), it seems that developing the capability to handle lattice defects can grow into an ability to focus phonons into coordinatively exposed transition metal centers, so that they may affect the desired reactions wherein the lattice is used as a reservoir of driving energy.
- The project is highly relevant to the DOE goals, and the incorporation of inexpensive, high-efficiency light absorbers such as perovskites can lead to significant cost reductions in hydrogen production from PEC devices. The project makes effective use of various nodes of the HydroGEN consortium to complement the PI's expertise in perovskite solar cells.
- The project uses high-efficiency, low-cost materials that have the potential to meet DOE cost and efficiency targets, assuming stability is achieved. The project is significantly leveraging Energy Materials Network resources.

Question 3: Accomplishments and progress

This project was rated **3.5** for its accomplishments and progress toward overall project and DOE goals.

- The team has made very significant progress toward the project's objective. A perovskite solar cell with solar-to-electricity efficiency >20% with a lifetime >1,500 hours was demonstrated. Effective protection strategies for the perovskite materials are being developed using graphene sheets, and MoS_2 layers were deposited via atomic layer deposition. Both MoS_2 and NbS HER and NiFe OER catalysts show sufficient performance for efficient solar water splitting. An integration strategy of the perovskite cells with electrocatalysts is being developed. The initial performance of the components developed provides high confidence for the achievement of the project's goals.
- The project is making good progress toward its milestones. Good fundamental improvements in perovskites were accomplished.
- Good progress has been made. The integration of the scientific innovation into the device (in ways that respect/preserve the physics) could occur more rapidly.

Question 4: Collaboration effectiveness

This project was rated **3.7** for its collaboration and coordination with HydroGEN and other research entities.

- The project is making good use of HydroGEN nodes and collaborating effectively in benchmarking, in situ characterization, modeling, and technoeconomic analysis. The team is also providing information on perovskites to the Data Hub.
- The collaboration between team members is strong and is reflected by the positive results from the first year of the project. The skillsets of all the investigators involved are complementary and are required for project success.
- Good collaboration is occurring, but it is sort of the "expected" kind of collaboration. The collaboration is typical.

Question 5: Proposed future work

This project was rated **3.7** for its proposed future work.

- The proposed future work is reasonable and is likely to lead to the achievement of project goals. The materials integration strategies being sought are appropriate for the development of the intended high-efficiency device. Characterization activities can provide insights into charge transport and degradation mechanisms that may limit the performance of the stability of the devices. Ultimately, characterizing STH efficiencies of the perovskite devices will be important to assessing the potential of the materials explored.

- The proposed future work is a logical extension of the prior year's work. The use of perovskite-/Pt- and Ir-based catalyst systems should probably be only a short burst of effort to create a benchmark with sufficient character and measurement count so as to enable future innovations to be held in statistically significant comparison. Understanding charge transport and degradation mechanisms will be very important; they are likely complex phenomena with multiple contributors and multiple space and timescales of action. Additional clarification on the strategies to be employed here would have been useful.
- The proposed future work addresses the remaining barriers.

Project strengths:

- This project has strong team integration and leverages HydroGEN nodes well. There is promising performance of the light absorber material with >20% PV efficiency, as well as demonstrated performance of HER and OER components. The light absorber also has long-term stability.
- This is a great project with a very talented PI and a strong project team. There are some fascinating innovations developing here, and the collaborations across the national laboratories seem to be building out upon these.
- This project, if successful, would provide a new paradigm for solving PEC challenges. This goes beyond incremental change.

Project weaknesses:

- Device design aspects are missing from the proposal. Mainly, the advantage of integrating perovskites with electrocatalysts, as opposed to developing PV electrolysis cells, is not clear.
- The involvement of students could have been better emphasized.

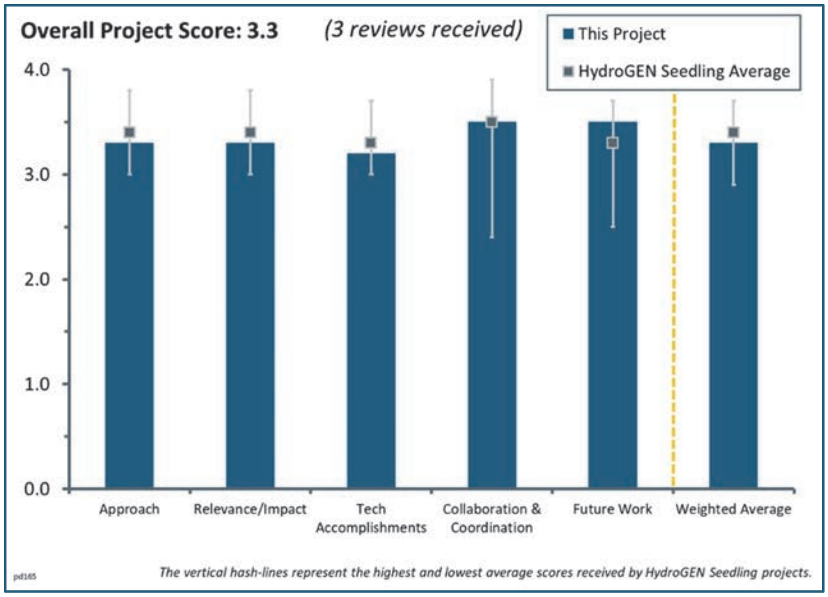
Recommendations for additions/deletions to project scope:

- Given the high performance of the light absorber, it may be important to increase the emphasis of the device design and development activities, to guarantee that the potential gains in the PV are not lost as a result of device-related factors (e.g., electrolyte resistance or poor light management).

Project #PD-165: Accelerated Discovery of Solar Thermochemical Hydrogen Production Materials via High-Throughput Computational and Experimental Methods

Ryan O'Hayre; Colorado School of Mines

Brief Summary of Project:



The current state-of-the-art solar thermochemical hydrogen (STCH) material efficiency is approximately 2%, but development of an optimal STCH material could increase the efficiency beyond 60%. This project aims to integrate combinatorial synthesis methods with combinatorial theoretical calculations to rapidly discover new potential materials for use in two-step metal oxide cycles for STCH. The effort builds on prior collaboration between the project partners, which resulted in the discovery of two novel perovskite-based STCH candidates, and leverages the Energy Materials Network (EMN) model of merging high-throughput computational and experimental techniques to accelerate new materials discovery.

Question 1: Approach to performing the work

This project was rated **3.3** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The outlined approach is well-thought-out and innovative and may lead to success. The project has leveraged multiple appropriate HydroGEN nodes in pursuit of the project goals and is well integrated into the HydroGEN consortium. The project has strongly demonstrated the collaboration and the use of these HydroGEN nodes in achieving goals. The work in Budget Period 1 has achieved the goal of demonstrating the computational approach, and as such, the ability to identify a new material, "Material X," which has achieved the first go/no-go criteria. The experimental work has also demonstrated the use of optical analysis for the presence of reduction/oxidation. It is here where further approach refinement is necessary; in rapid screening of materials, it is necessary to know the extent of reduction/oxidation, in addition to a binary analysis of whether it occurred. This could be achieved through additional non-optical spectroscopic techniques such as X-ray photoelectron spectroscopy (XPS). The project should also identify a workaround to the high-temperature diffusion and thus substrate/thin film mixing. This newly identified barrier needs to be overcome for project success.
- The idea of using combinatorial thin film deposition of up to four targets seems to be a very interesting method for identifying more reactive materials for STCH processes. The integration with excellent theoretical capabilities forms a strong partnership to deliver important results. However, the color change seems to be an easy way to find good candidates, but the reality is more complicated. The link to other groups working in the same field in HydroGEN could be improved.
- Project barriers are understood, and some new approaches are proposed. The combinatorial approach is promising if the results can be trusted. It was not clear from the slides what the innovation is in the density functional theory (DFT) screening part.

Question 2: Relevance/potential impact

This project was rated **3.3** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- This project is well aligned with the DOE project goals of identifying new materials. The rapid screening, both experimental and computational, has the potential to identify new materials if the barrier can be overcome. The team has excellent collaboration and leveraging of DOE resources. This is aided by the team's close proximity to National Renewable Energy Laboratory, which has enabled personnel exchange and training. Additionally, long-standing collaboration with Dr. McDaniel at Sandia National Laboratories (SNL) and modifications to allow for remote accessing of equipment has enabled the team to make excellent use of the stagnation flow apparatus. However, the team should start considering technoeconomic analysis to achieve the DOE cost targets.
- The combination of combinatorial chemistry and high-class simulation forms a strong synergy in this project. The impact on the goals has the potential to be very high. A better connection to the other projects in the field in HydroGEN would make this even better.
- Materials discovery is the key challenge for pure thermochemical cycles. Perovskites are the most prospective material to do better than ceria, but the number of permutations is bewildering. Any effective and, most importantly, reliable screening method could lead to a big step forward.

Question 3: Accomplishments and progress

This project was rated **3.2** for its accomplishments and progress toward overall project and DOE goals.

- Significant progress has been made toward the DOE goals. The project has completed preliminary assessment of materials and has identified an interesting new material. The team has also demonstrated the project's computational approach and some of the experimental approaches. The team has demonstrated excellent collaboration with the consortium. However, the team does need to demonstrate combinatorial deposition and quantitative measurements of reactivity, as this is critical to project success. This likely depends on new developments to prevent interaction with the substrate, as well as more concrete analysis techniques.
- The project seems to be on a very good path. There is confidence that the team will meet the go/no-go criteria. The presented work makes this statement probable.
- Slide 14 suggests that a promising material has been identified, though the performance drops significantly at realistic water-to-hydrogen ratios. Slide 13 indicates that the reduction is done with hydrogen well below the target reduction temperature (1000°C versus 1350°C). It is unclear whether this is representative of high-temperature reduction (and at what temperature). It is also unclear whether this optical test is repeatable with sequential reduction and oxidation cycles. It was also observed that the substrate (not specified) may also be interacting with the material at these temperatures (which is presumably why hydrogen is used as a reductant to reduced peak temperature). Slide 12 suggests that the DFT work is reasonably well understood, but it is unclear whether the project is using innovation or just leveraging existing expertise.

Question 4: Collaboration effectiveness

This project was rated **3.5** for its collaboration and coordination with HydroGEN and other research entities.

- The collaboration in the project is excellent. A strong practical and theoretical team has formed, one that seems to work efficiently together. The connection within HydroGEN could be improved by closer cooperation with the other projects working in the same field.
- The group has made outstanding use of the HydroGEN nodes and shows a highly integrated approach to the project. The group is utilizing the Data Hub. The project team needs to be more explicit about their Task 2b activities in regard to protocol development, as these were not directly obvious.
- The project seems to be effectively leveraging expertise at other institutions.

Question 5: Proposed future work

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

- The project has a well-thought-out plan and approach for future work, given the milestones/deliverables as stated. To meet DOE's goals, it is recommended that the team amend this plan to include further research into how to accomplish, in a quantitative way, the experimental combinatorial analysis, particularly reduction/oxidation activity quantification and film stability. This will then require amendment of the plan, but based on performance so far, it seems that the team will be able to successfully do this and plan for it.
- The proposed future work has clear targets that would have a strong impact on development in the field of active perovskites for STCH processes. In particular, the transfer into a Hydrogen Analysis (H2A) effort promises a close connection to application toward the end of the project.
- Year 2 appears reasonably well scoped, but Year 3 is not very explicitly explained. This is not a major issue, as "Full characterization and advanced study of excellent candidate, including H2A" is a reasonably significant undertaking.

Project strengths:

- The project strongly leverages DOE resources. The team has not only used DOE facilities but learned new techniques and brought them into their laboratories, and has contributed likewise by helping to exercise remote access capabilities at the SNL stagnation flow reactor facility. The team has identified a new material based on project screening and has ideas of the desired material's properties.
- The project seems to be effectively leveraging the expertise in the EMN network and has some good ideas and outcomes so far.
- The combination of combinatorial chemistry with advanced simulation tools is a project strength.

Project weaknesses:

- As with all the HydroGEN projects, there has been limited interaction between the projects, as opposed to with the EMN network resources. It is not clear where some of the innovation lies and how robust/useful the optical characterization is.
- The greatest project weakness is the experimental rapid screening. This needs to become quantitative rather than qualitative, and diffusional/thin film stability issues need to be resolved.
- Color changes seems to be a rather simple method to determine the right materials. The project could use a better connection within HydroGEN.

Recommendations for additions/deletions to project scope:

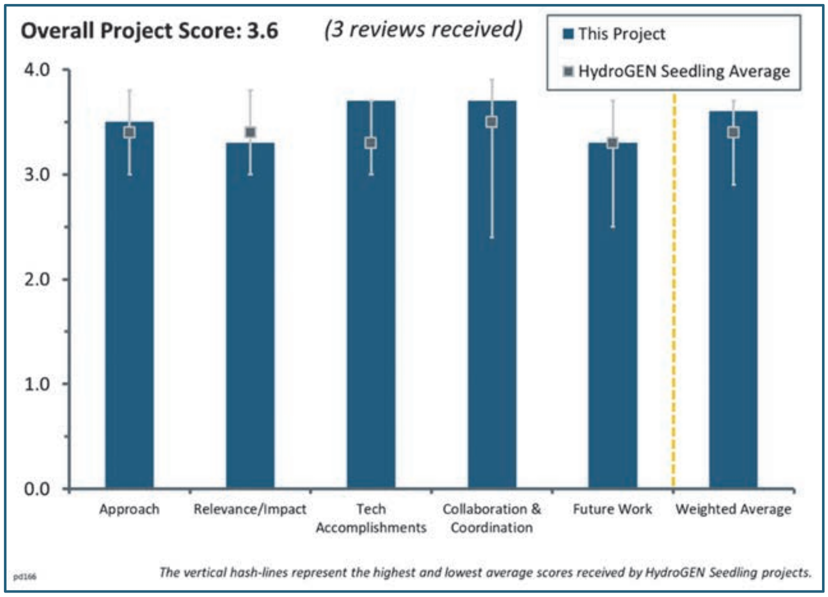
- The project scope is excellent and should be followed as is.
- The project scope should be modified to make the experimental screening of materials quantitative rather than qualitative. Additionally, the kinetic aspects of materials should be further assessed. Lastly, in-depth thermodynamic characterization of materials is needed (i.e., the determination of partial molar enthalpies and entropies). This characterization will enable thermodynamic modeling of the system and, thus, system optimization and materials efficiency comparisons.
- It is not clear how the combinatorial thin film screening really fits in, as the samples shown have rather uniform color. It would have been more interesting to see whether the color shows gradations with changing stoichiometry.

Project #PD-166: Computationally Accelerated Discovery and Experimental Demonstration of High-Performance Materials for Advanced Solar Thermochemical Hydrogen Production

Charles Musgrave; University of Colorado Boulder

Brief Summary of Project:

The project objective is to utilize machine-learned models coupled with ab initio thermodynamic and kinetic screening calculations to accelerate the research, development, and demonstration of new solar thermochemical hydrogen (STCH) materials. The approach will rapidly screen a vast number of new candidate metal oxides materials for stability, thermodynamic viability, and kinetics. The project will utilize experimental techniques to evaluate thermodynamic and kinetic properties of new materials to provide feedback to the computational thermodynamic and kinetic screening effort.



Question 1: Approach to performing the work

This project was rated **3.5** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The project is using a very appropriate way to tackle the problem: searching materials beginning with machine-learning thermodynamic basics and conducting thermodynamic and kinetic screening validated by experiments.
- Given the enormous challenges in identifying practical STCH materials, the use of machine-learning may be appropriate and timely for screening the vast combinations of multi-metal oxides.
- The barrier identified is just the large number of compounds that need to be screened. Thus far, no materials exhibit the required activity at a reasonable temperature. Aside from this, the project seems well scoped. It is interesting to see some attempt made to compute kinetics, which has always been difficult without experiment. The target reduction temperature is still a bit on the high side.

Question 2: Relevance/potential impact

This project was rated **3.3** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- This approach is very relevant and its impact probably very high. However, there are alternatives in HydroGEN that should be linked more closely to this work. The combined work of all projects on materials in HydroGEN would be outstanding.
- Finding a suitable redox material is the key challenge for this technology. Therefore, the project is appropriately targeted (providing a good system efficiency can be obtained at such a high temperature).
- The project goals are relevant and in alignment with the Energy Materials Network (EMN) and Advanced Water Splitting Materials consortium objectives. However, given the novelty of this approach, the potential impact is difficult to assess at this point.

Question 3: Accomplishments and progress

This project was rated **3.7** for its accomplishments and progress toward overall project and DOE goals.

- The project already presents outstanding results. These results promise that in the next phase of the project, important contributions to HydroGEN will be provided.
- The project seems to have made very good progress in machine learning to improve prediction of stability. There are still many candidates. Slide 13 of the presentation indicates a number of candidate materials with good performance, although the steam-to-hydrogen ratio (and variation in performance with same) is not mentioned.
- The project team has accomplished a good deal so far. However, since this project is likely to set standards for other similar materials screening efforts, it would be helpful to discuss and share the reasons for choosing the particular machine-learning algorithm over others.

Question 4: Collaboration effectiveness

This project was rated **3.7** for its collaboration and coordination with HydroGEN and other research entities.

- The project has an outstanding collaboration concept. It includes partners from outside the HydroGEN team and even international partners. This enables the project to harvest a wide and world-class contribution to the development of efficient materials for the STCH processes.
- The current collaboration effort looks appropriate, but it is suggested that the project team keep the option for other collaborators as needed, given the novelty of this approach. One such option is to seek partners with combinatorial materials synthesis or characterization capabilities to accelerate experimental efforts.
- The project is collaborating effectively both within and outside the EMN network.

Question 5: Proposed future work

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

- The proposed future work is excellent; it contains all aspects to achieve the project goals.
- The project team seems clear on objectives and activities for subsequent years.
- The proposed work looks reasonable, especially the focus on time-based hydrogen production rates. However, it is hard to understand the basis for the stated milestones. For example, it would be helpful also to provide current thinking on commercially acceptable hydrogen production rates from such systems. In addition, the team should start incorporating cost factors into their screening tools.

Project strengths:

- This is a sound combination of basic theoretical work and experiments. The team shows excellent collaboration, even with international partners.
- The project team seems to have a good handle on and understanding of computational thermodynamics and kinetic screening of these STCH materials.
- Machine learning seems to have improved model predictions of stability. The team has good collaboration with other experts in the field, both in and outside the EMN network.

Project weaknesses:

- This is a relatively high target reduction temperature, and it is unclear how the materials would perform at moderate water-to-hydrogen ratios. This will be critical for overall cycle performance.
- The collaboration within HydroGEN could be improved. Similar to other projects in the Hydrogen and Fuel Cells Program, the lack of collaboration with HydroGEN is due to limited time.
- There is an apparent weak mismatch of feedback loops between computational and experimental efforts.

Recommendations for additions/deletions to project scope:

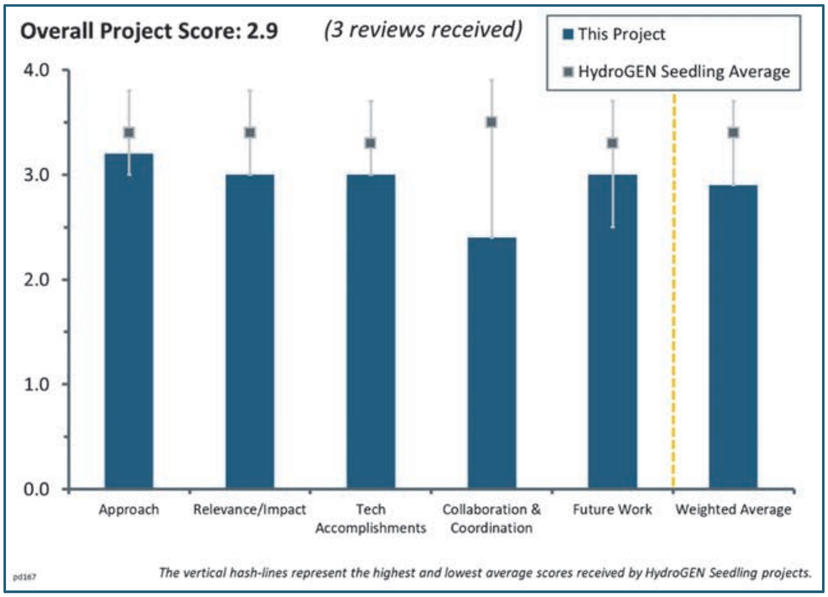
- In addition to thermodynamic and kinetic parameters, the team should consider material cost as a parameter for screening the STCH materials. Having some idea of system cost based on promising materials is also desirable, although it may not be in scope for this project phase. If possible, the project team should also consider acquiring combinatorial synthesis and/or testing equipment to accelerate the experimental work or collaborate with a laboratory that has such equipment.
- The work plan seems to be appropriate. Additions or deletions do not seem to be necessary. An exchange between the other projects on the materials topic seems to be valuable.
- As with all the HydroGEN projects, there is not enough interaction with the others. The four materials discovery projects in oxides should get together in a facilitated environment to get the most out of the work at the various institutes.

Project #PD-167: Transformative Materials for High-Efficiency Thermochemical Production of Solar Fuels

Chris Wolverton; Northwestern University

Brief Summary of Project:

The project objective is to utilize a computational–experimental approach, combined with materials design strategies to quickly discover and demonstrate novel thermochemical materials with properties superior to the state of the art. The project will investigate what is an enormous compositional space of materials utilizing high-throughput computational and experimental methods to identify promising compounds that show (1) ground state stability/ synthesizeability, (2) thermodynamics favorable for <1400°C reduction, and (3) thermodynamics favorable for facile water splitting.



Question 1: Approach to performing the work

This project was rated **3.2** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The work itself is excellent. The combination of experiments and theoretical work to create a database of possible active and stable perovskites points exactly in the right direction of HydroGEN. The work is already very advanced; it even shows some of the limits, as the number of elements determines the possible materials. The connection within HydroGEN does not seem to be as good as it could be. There are a number of other groups working in the same direction that are not linked to this work. Together, the work would be even stronger.
- The approach as demonstrated in the slides and poster represents marginal improvements to materials development, with the only seeming major advancement coming from examination of double perovskites. The methodological approach seems to be no different from the currently taken approach to materials identification. Computational methods use traditional density functional theory methods to screen materials, generate all possible structures, calculate them, identify stable materials, and then calculate oxygen vacancy formation energies. This process lacks a method for accelerating screening other than a brute force method, which is likely intractable as the phase space expands. The brute force method has identified novel double perovskites that have not been suggested before; however, there is no indication, experimentally or computationally, that these materials will facilitate water splitting. Selection of materials for analysis is based on materials that fall within a wide range of oxygen vacancy formation energies and, seemingly to an equal extent, chemical intuition. It seems that just computational identification of any material that can split water is the desired computational end goal, rather than materials that do so efficiently or cost-effectively. Experimental work to date has relied on traditional thermogravimetric analysis (TGA). The principal investigator (PI) indicates that more rapid techniques will be used in the future, but there is no demonstration of this or validation of the approach. There is little to no interaction or integration into the HydroGEN consortium. The use of nodes seems to be an afterthought at best. Rather than selecting complementary capabilities, the PI seems to have selected nodes with capabilities similar to the project team's own expertise.

- As with the four other projects in this space, this work relies heavily on high-throughput DFT modeling. This project has some slightly different approaches, but it is hard to judge whether this project is using a better approach than the others. A forum of the teams should be convened to discuss pros and cons of the various approaches. The recognition that phase change may improve performance is notable, as the challenge with non-stoichiometric reduction is low production rates. Naturally, long-term cyclability is the question.

Question 2: Relevance/potential impact

This project was rated **3.0** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- The potential for this project is very high. The project has a very interesting strategy for down-selecting possible materials as candidates for efficient STCH processes. The probability of finding the right candidate materials is rather high. However, the link to work that is more focused on the application is missing. This would add criteria that could be used in the selection matrix to narrow the candidate materials further down.
- The project is relevant and is working toward developing materials that split water. The development of new materials, such as the double perovskites investigated here, would represent a major advance in the STCH community. However, the project seems to focus on expensive rare earths for use in the materials. This will likely pose a challenge in terms of meeting the price goals and scale-up goals. To date, the project does not leverage HydroGEN consortium resources.
- Finding a suitable redox material is the critical challenge in this area. The team nominates 1400°C as the reduction temperature, which is still quite high for achieving good cycle efficiency. However, the overall cycle configuration, including recuperation, kinetics, and reactor design, will be vital also.

Question 3: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and DOE goals.

- The work is very advanced, and the results are already very impressive. The methodology is well described. The theoretical results are validated by experiments. The work is a very valuable addition to the HydroGEN program.
- Overall, there is good correlation between computational prediction of reduction and experimental measurements. Correlation between perovskite distortion and vacancy formation energy is useful.
- The project has made progress. The team has shown that some materials predicted to be stable are stable. The project has also shown, via a feedback loop, that if the calculations are done correctly (i.e., simulate the correct phases), the oxygen vacancy formation energy can be predicted with reasonable accuracy. Without careful consideration of the active phases, calculated formation energies do not match. This has informed the project leaders of the importance of careful phase determination. The go/no-go metrics are very weak. The project easily met the stability criteria. The project met the correlation between the experiment and theory through an iterative computational approach.

Question 4: Collaboration effectiveness

This project was rated **2.3** for its collaboration and coordination with HydroGEN and other research entities.

- There is good collaboration with Energy Materials Network node resources, as per the others. In the next stage, collaboration between the HydroGEN projects should be a key focus to really accelerate the work.
- All of the work to date has been completed at Northwestern University. Use of nodes or HydroGEN capabilities is an afterthought and considered at all only because it is a project requirement. The project proposes use of three nodes, one of which has significant overlap in capabilities with Northwestern University.
- The collaboration within HydroGEN is satisfactory. There is a cooperation with three complementary groups, but the link to other projects working in the field of innovative methods for materials selection seems to be rather weak. Both the project and HydroGEN would benefit if the different projects were able to work more closely.

Question 5: Proposed future work

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

- The proposed work for Years 2 and 3 is excellent. This project has the potential to produce very valuable results. However, the potential could be improved if the link to the other groups working in the field were strengthened.
- The future work is planned within the project scope. Computational work will continue and focus on calculating oxygen vacancy energies in a logical and thought-out manner. The experimental future work plan is either weak or poorly communicated within the team and, therefore, poorly communicated externally. This makes it difficult to fully evaluate. If future experimental work is merely TGA, then this work is insufficient for assessing materials. If it includes other methods of materials assessment, that is completely missing from the documentation or Year 1 validation of the plan. The project needs more concrete goals/materials identification criteria to aid in materials selection such that DOE efficiency and cost goals are achievable. Additionally, future work should include closer collaboration within the existing team and between the team and HydroGEN partners.
- This project is very much focused on the discovery side, with limited performance characterization in terms of hydrogen evolution.

Project strengths:

- This project's strength is the consideration of double perovskites; these materials have not yet been reported on within the STCH community. The project has identified many new stable double perovskites.
- This project has an innovative methodology that is able to provide a down-selection of materials based on theoretical high-throughput DFT calculations.
- This project has done nice work to date and shows good progress.

Project weaknesses:

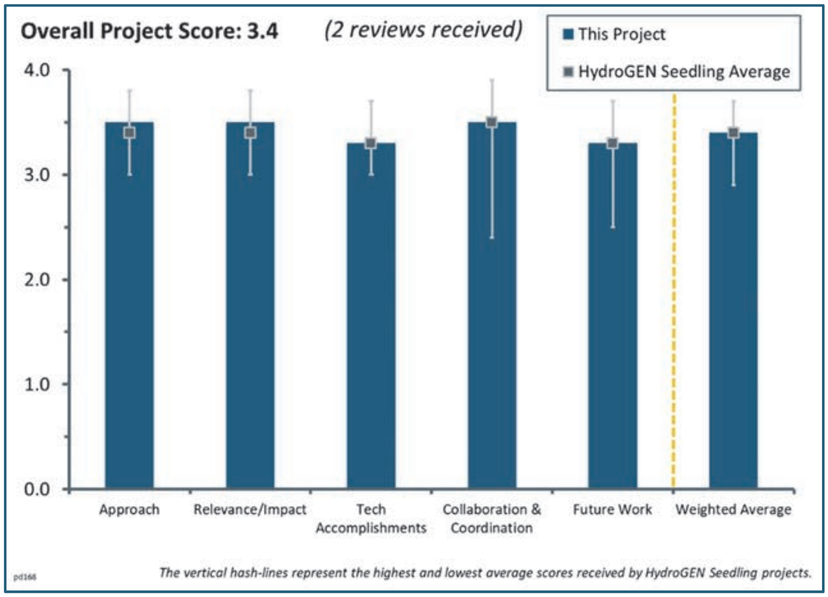
- This project has multiple areas for improvement. The largest is the use of the national laboratory nodes. This seems to be completely lacking, and it seems that the project would rather not have to interface with teams outside of Northwestern University. The communication between the computational and experimental work also needs to be improved, both in terms of collaborative goals and understanding of each other's methods and tasks. Also, the team should refine the desirable materials characteristics that will not only enable materials identification but also provide a chance of production at scale and cost targets. While massive time does not have to be devoted to the cost of hydrogen production in a materials identification project, it should at a minimum be a consideration of the team. The team should also consider methods for accelerating materials screening and analysis beyond brute force methods for both computation and experiment.
- The project seems to be not very well connected in the HydroGEN program. Perhaps this is a problem of the tight schedule of the program.
- It is unclear whether the project has considered relative abundance of the elements in the screening group.

Recommendations for additions/deletions to project scope:

- The scope of work is excellent. The only thing to add would be a better exchange with other projects.
- As with the other computational projects, a forum of the different projects should be held where the researchers can debate which techniques have more general relevance to try, accelerating progress across all groups. This project could also benefit from adding some performance evaluation (e.g., with A. McDaniel at Sandia), as with other projects.
- The project scope should consider the kinetics of the materials as well as some tasks on hydrogen production costs. Additionally, explicit tasks incorporating national laboratory nodes should be included.

Project #PD-168: Mixed Ionic Electronic Conducting Quaternary Perovskites: Materials by Design for Solar Thermochemical Hydrogen

Ellen Stechel; Arizona State University



Brief Summary of Project:

The project objectives are (1) to contribute to improved solar thermochemical hydrogen (STCH) materials discovery by providing strategies to boost solar-to-hydrogen thermal efficiency and (2) to provide experimentalists with crucial input to synthesize, validate, and perform further testing on promising candidates. The project will apply first principles computational materials design capability to calculate and validate chemical potentials for complex off-stoichiometric redox-active mixed ionic electronic conducting perovskite metal oxides. The end goal is to determine design principles for optimal and discoverable materials that have the potential to perform better than ceria, meet the target efficiency (solar-to-hydrogen thermal efficiency >30%), and approach the ultimate production cost goal of < $2/kg H_2.

Question 1: Approach to performing the work

This project was rated **3.5** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The project seems well scoped, with an interesting new way of looking at the problem. The key impact areas remain a significant challenge, in terms of how to get to a sufficiently high delta (>0.15 per cation, cf. ceria at 0.03) at a realistic temperature (1450°C is still very challenging).
- The project is a mainly theoretical attempt to find suitable materials for STCH processes. The theoretical work is linked to practical evaluation. However, the goal seems too realistic to reduce the necessity of extensive material synthesis.

Question 2: Relevance/potential impact

This project was rated **3.5** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- The project's models will be a very valuable contribution to HydroGEN. However, there are competing attempts that could jointly form a real outstanding network.
- Materials discovery is the critical challenge for this team, as with the other four projects looking at oxide cycles. Redox cycles are not realizable without a significant improvement in active material performance.

Question 3: Accomplishments and progress

This project was rated **3.3** for its accomplishments and progress toward overall project and DOE goals.

- There appears to be a significant improvement in understanding, though the reviewer is not an expert in the intricacies of density functional theory. The project's milestones are on track.

- The achieved results are very important. However, there seems to be a slight delay in the project. This is not severe, and it is probable that the project will achieve its milestones.

Question 4: Collaboration effectiveness

This project was rated **3.5** for its collaboration and coordination with HydroGEN and other research entities.

- There is excellent collaboration between the partners, as well as very efficient and honest coordination that anticipates the developments within the project.
- Collaboration seems to be well coordinated and taps into key expertise (slide 9). This is not as clearly articulated as in some other projects, which explicitly draw out Energy Materials Network (EMN) node interactions, but the collaboration seems to be appropriate and effective.

Question 5: Proposed future work

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

- The future work is very well defined and seems to be absolutely appropriate to achieve the project's goals. The work is important, and the possible impact is very high.
- Slide 19 lists many planned activities, but it is not clear from the presentation how they are to be structured over Years 2 and 3. This is likely to be explained elsewhere (e.g., the DOE project plan), but it is not in the reviewer materials.

Project strengths:

- The project team appears to have gained some clever insights into how to better use computational methods to identify the goldilocks material. There is a good understanding of fundamentals.
- The project team has excellent theoretical capabilities and project management. There is a strong link within the project consortium.

Project weaknesses:

- This may not be an actual project weakness, but the reviewer material was not that focused on addressing reviewer questions in terms of EMN collaboration and detailed project planning. Technically, the project seems to be excellent work, although (as with others) it could benefit from peer discussion between the HydroGEN projects.
- This project lacks a link to other projects on the same topics within the HydroGEN Consortium.

Recommendations for additions/deletions to project scope:

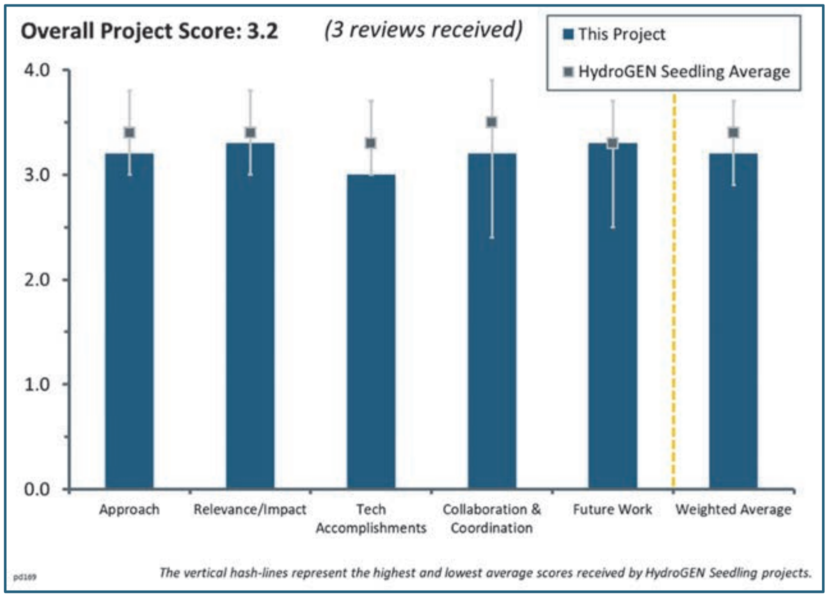
- The project is excellently defined, and no additions or deletions seem to be appropriate. As with all projects in the HydroGEN Consortium, a closer collaboration between the projects would strengthen the joint results.
- As per slide 9, the proposed future work does not include experiments at Sandia National Laboratories (slide 19), which indicates the stagnation flow reactor will be used. This should be clarified.

Project #PD-169: High-Temperature Reactor Catalyst Material Development for Low-Cost and Efficient Solar-Driven Sulfur-Based Processes

Claudio Corgnale; Greenway Energy

Brief Summary of Project:

The project objective is to develop an efficient and low-cost solar thermochemical process. In particular, this project is focused on the solar-driven hybrid sulfur (HyS) cycle and the development of catalytic materials to decompose sulfuric acid, a critical step in this two-step water-splitting cycle. The project will (1) develop a new catalyst material using the team's demonstrated surface free energy and electro-less deposition technique; (2) design a novel, integrated, direct solar reactor–receiver, based on a demonstrated cavity solar reactor, and (3) perform system and cost analysis on an effective new solar–thermochemical plant process.



Question 1: Approach to performing the work

This project was rated **3.2** for identifying barriers and addressing them through project innovation, project design, feasibility, and integration with the HydroGEN Consortium network.

- The project is unique in the field of solar thermochemical hydrogen (STCH), as the team is not only developing innovative ways to identify promising materials for water splitting but also looking at how such materials will be used in real applications. The sulfur-based processes are different from the metal–oxide cycles, as the materials necessary are catalysts and not reaction partners in the redox cycles. The work in this project is mainly on the stability and efficiency of the catalyst materials. However, this seems to be rather straightforward. Sulfur chemistry is a major topic in the chemical industry. A joint development with industry seems to be appropriate to accelerating the catalyst development. The strength of the project clearly lies in the integration attempt to describe how the materials could be efficiently used in even very large-scale applications on the several-100-MW scale. The link from materials development to how these materials are used is unique in the HydroGEN Seedling sub-category. The proposed receiver reactors seem to be very promising, and the design of different-size solar towers for hydrogen production is the key to successfully getting STCH into application. For the short time the project has been running, excellent results were achieved concerning the system, but the material development seems to be not as advanced.
- The project has identified the key areas for resolution of the solar thermal component of the HyS process. No mention is made of the electrolyzer, apart from slide 16, but this is not a major omission. In fact, the scope is already much broader than other HydroGEN projects and is much more applied.
- This project has several components, all integrating into a single overall reactor design to achieve U.S. Department of Energy (DOE) targets. Each individual piece of the project seems to be moving forward and seems to be on pace to accomplish the stated objectives. The team's work so far for budget period 1 is good and mostly demonstrates feasibility. In particular, the novel fin-based reactor concept stood out. This could be widely expanded into other solar technologies. Similarly, the novel HyS flow sheet represents a significant step forward in terms of economic and energetic efficiency. However, the main innovation seems to be coupled with inventions from a separate DOE project rather than results/innovations from this project. The weakest piece seems to be in terms of catalyst development. The catalytic material is still

dependent on expensive Pt group metals, and there is little fundamental development on how/why the catalysts are deactivating. It seems that the innovation is in the deposition method and just hoping for limited deactivation of the materials. It would be helpful to see the behavior of “baseline” materials. Lastly, more direct integration of the team and cross-level interaction would be desirable.

Question 2: Relevance/potential impact

This project was rated **3.3** for its relevance to/potential impact on U.S. Department of Energy (DOE) Hydrogen and Fuel Cells Program goals and the HydroGEN Consortium mission.

- The systems work is excellent and unique within the HydroGEN consortium projects. The other projects, and the development in general, will strongly benefit from this work. In this respect, this is dominant to the materials development—which is not much connected to other work under this framework because it deals with different substance classes.
- This project has some high-potential-impact components and is appropriately placed in the HydroGEN consortium. It looks like the systems analysis, manufacturing, and integration will potentially enable the achievement of DOE goals. The project lacks novel catalyst materials development and instead focuses on catalyst fabrication techniques. Not only does this project have the potential to leverage DOE resources at the national laboratory, but it could also be very interesting to other HydroGEN consortium partners.
- The HyS process is highly prospective for STCH production, as it is likely to achieve the highest solar-to-chemical conversion of any cycle and requires a much more moderate temperature than metal–oxide cycles.

Question 3: Accomplishments and progress

This project was rated **3.0** for its accomplishments and progress toward overall project and DOE goals.

- The project has already achieved substantial results. It seems to be on a good track to stay within the foreseen project plan. Levels 2 and 3 are unique within the project and are, therefore, outstanding in this respect. Level 1 is sound catalyst work, but it lacks uniqueness.
- It is not clear whether all the work in the three areas was able to be done in the few months available and how much was possible by tapping into work that was partially complete. All the same, the achievements for the Level 2 and 3 areas are impressive and represent good progress. The catalyst work described in Level 1 is, in contrast, rather unimpressive and in sharp contrast to some of the other HydroGEN projects, which are true materials discovery efforts. The catalyst work in this project seems rather linear and does not really include any innovation in terms of materials screening or experimental design.
- Large strides have been made in some areas of the work. Level 2 and 3 technologies seem to be moving forward very well and are on pace to achieve the targets. Level 1 catalysis development is behind. Any results of the fabricated Pt/Ir/TiO_2, in terms of reaction, should have been shown to allow assessment of whether these materials are capable of catalyzing H_2SO_4 decomposition. Similarly, it is unclear whether the Pt deposits on the Ir/Ru as a sheet (as desired) or in islands. Lastly, the go/no-go criteria and the relevant progress should be clearer.

Question 4: Collaboration effectiveness

This project was rated **3.2** for its collaboration and coordination with HydroGEN and other research entities.

- The coordination and collaboration in the project seem to be excellent. The organization of online and in-person meetings are appropriate for the collaboration. The input into DataHub is of high relevance, especially as data are provided for the technology development, which could be a starting point for other projects to scale up their technologies.
- It is not clear whether the collaborations in this project have been facilitated by the establishment of the Energy Materials Network (EMN) or represent existing ties between the researchers. Despite this, the collaborations are appropriate (in terms of tapping into key resources) and appear very effective.
- Collaboration within each level shows very good interactions between the nodes and the recipients. These interactions seem separate rather than fully integrative. More interaction between the levels should be

sought, both with the recipients and between the recipients and the nodes. This will help the project act more like a single work rather than three separate projects under one umbrella. The team also uses the Data Hub.

Question 5: Proposed future work

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

- The project team presented a good plan for moving forward and achieving project goals. The plan follows the logical next steps toward DOE targets. More aggressive catalysts/materials goals should be set, however, and more attention should be paid to this progress.
- The proposed future work is straightforward. It will definitely improve the results and is absolutely necessary for a development closer to application. In this respect, topics such as the long-term stability of catalysts, testing of receiver reactors, optimization of flow sheets, and assessment of alternative solutions are typical future work packages. However, the proposed work lacks the potential for breakthroughs.
- The biggest issue with the proposed future work—which would provide some valuable information—is clearly the very broad scope and the fact that much of this work appears to be beyond the available budget (slide 22). The catalyst work that fits most closely with the other projects in HydroGEN probably should be a bit better explained in terms of the rationale and methodology. It is not clear how the catalysts are selected and optimized and whether this is from prior work.

Project strengths:

- The project team has achieved very interesting results in the new catalytic reactor design and system integration; this has led to the patenting of new technologies. These improvements go a long way to achieving the overall goals. The team also has strong intra-level collaboration and is using nodes to achieve the project goals.
- The strengths of this project are clearly in the development of receiver reactors and the systems. These are unique throughout the whole consortium, and the result will be very important to other groups, helping them to develop their technologies on the next level. Also, the project management and communication of the project seem to be excellent.
- The HyS process is extremely promising for solar hydrogen production. This project seeks to improve knowledge in three key areas. There is excellent collaboration with national laboratories that taps into an enormous competency for this process, which would be a shame to let languish and fade away.

Project weaknesses:

- The catalyst development seems to be rather straightforward. The chemical reactions were under research for a long time; the proposed catalysts (mainly TiO_2 doped with precious metals) seem to be appropriate, but as they contain precious metals, there should be a search for alternatives.
- The project has an enormous scope and is not sufficiently funded to achieve all its objectives. The materials discovery work in Level 1 seems rather simplistic in terms of identifying new options, but perhaps that is just because the methodology and prior work are not adequately covered. While not necessarily a weakness, the project is quite different from other HydroGEN consortium projects, which consist of far more fundamental materials discovery/screening activities.
- The project should strive for more inter-level interaction and integration; the project seems a bit disparate. One of the major advances, the process flowsheet innovations, is dependent on external technology—it might be good to fold that into the project. The project needs to further examine and focus on catalyst development; the team should at least be considering non-Pt materials for catalysts. Further work on characterizing the fabricated material morphology before and after implementation is necessary.

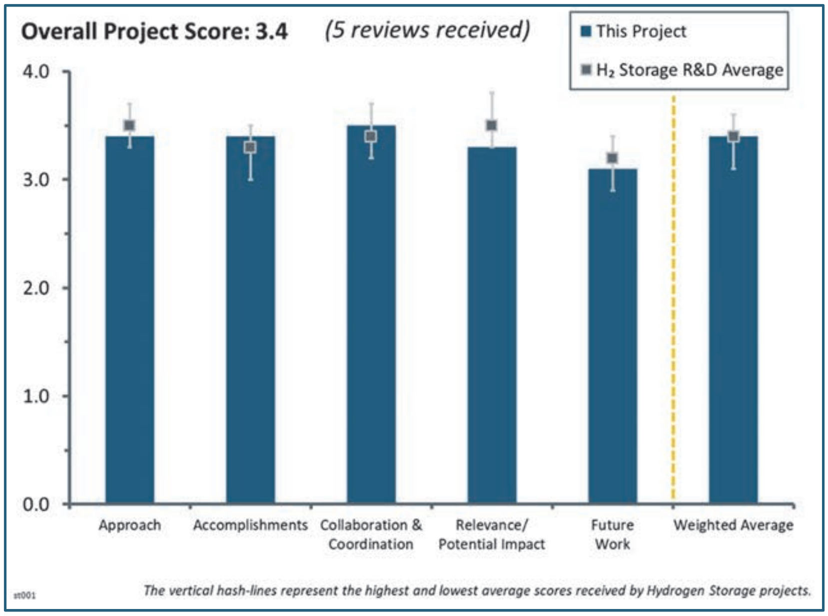
Recommendations for additions/deletions to project scope:

- If this has not already been done, the catalyst discovery work should perhaps be expanded to explore a wider suite of materials. This would provide a better fit with HydroGEN/EMN. The other activities should continue, but additional funding will need to be scrutinized on a cost–benefit basis.
- The project/scope should be expanded to include a more direct interface with the electrolyzer development. This seems critical, as it effects the overall system, and changes in operating conditions enable large changes in process flow. The catalyst scope should also be expanded to include a more fundamental understanding of catalytic behavior and the identification of a novel, less Pt-intensive catalyst.
- The project team should take into account catalysts without precious metals. A closer link with other projects that follow a more theoretical approach toward material description might also add value to the work.

Project #ST-001: System-Level Analysis of Hydrogen Storage Options

Rajesh Ahluwalia; Argonne National Laboratory

Brief Summary of Project:



The main objective of this project is to develop and use models to analyze the onboard and off-board performance of physical and materials-based automotive hydrogen storage systems. Specific goals include (1) conducting independent systems analysis for the U.S. Department of Energy (DOE) to gauge the performance of hydrogen storage systems, (2) providing results to materials developers for assessment against system performance targets and goals and for guidance in focusing on areas requiring improvements, (3) providing inputs for independent analysis of onboard system costs, (4) identifying interface issues and opportunities and data needs for technology development, and (5) performing reverse engineering to define material properties needed to meet the system-level targets.

Question 1: Approach to performing the work

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

- This is an ongoing project that DOE has relied upon to provide a detailed and meaningful systems-level analysis of hydrogen storage needs and options. The approach is focused on a critical analysis of off-board and onboard targets as well as requirements for materials properties and system configurations. The approach in fiscal year (FY) 2017 and 2018 addresses system-level issues affecting the development of compressed storage tanks (including cryo-compressed hydrogen [CcH_2] storage), hydrogen storage in sorbent materials (including sorbents containing metal cations capable of binding multiple hydrogen molecules on a single site), and (new) hydrogen carriers for hydrogen distribution and transport to the forecourt. The approach is rational and straightforward, and it has provided a means of rapidly and effectively assessing material requirements and properties as well as overall system performance.
- The development of thermodynamic and kinetic models of processes in physical, complex metal hydride, sorbent, and chemical hydrogen storage systems is a very broad and complex approach that touches on key modeling needs. It was not addressed in the presentation, but the finite element analysis for compressed tanks should (and likely does) include properties of materials at relevant cryogenic temperatures.
- The Argonne National Laboratory (ANL) systems analysis project continues to serve a valuable role by independently assessing design variations and engineering features for diverse hydrogen storage systems and materials. This year's assessments of 500 bar CcH_2 storage for buses and light-duty vehicles clearly demonstrate the feasibility of this method for these applications. The evaluation of requirements for room-temperature (RT) adsorption candidates also indicates that there are no known systems that can reach even 50% of these properties. Hence, there seems to be little reason for further exploration of these materials.
- The project has a good history of focusing on the key barriers for hydrogen storage systems using a systematic performance analysis. This year, the project review seemed to include several slides that focused on cost rather than performance; this was confusing, since cost has not been within the scope of this project

in the past. The project has a very good approach to conducting analytical simulations, although it lacks discussion about the validation of the results.

- The project covers a large area of investigation and has presented a large amount of data; however, in all cases, an indication of a path to meet DOE targets is unclear.

Question 2: Accomplishments and progress

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

- Excellent progress was achieved in FY 2017 and 2018 in all four focus areas: cryo-compressed systems, hydrogen storage in RT sorbents, compressed hydrogen storage, and hydrogen carriers for distribution and delivery. The analyses provide a quantitative and detailed assessment of requirements and performance needs in all FY 2017 and 2018 focus areas. This information assists DOE in planning for the Hydrogen and Fuel Cells Program (the Program) and provides a solid foundation for establishing the efficacy of new storage system technologies. The work on cryo-compression systems extends the studies conducted in the last two reporting periods, and it shows that CcH_2 provides clearly superior performance compared to Type 4, 700 bar RT compressed tank storage for fuel cell buses and light-duty vehicles. A particularly noteworthy and timely result concerns the requirements for a sorbent material to meet the DOE storage targets (slide 14). The results indicate that even if four H_2 molecules per metal cation are adsorbed in a (yet to be synthesized) uranium iodide-oxygen (UiO) metal–organic framework (MOF), the absolute uptake is *still* a factor of two lower than the DOE target. The work in the new area of hydrogen carrier development was also impressive because it provides a solid and quantitative systems-level foundation for the technical effort on hydrogen carrier materials efforts that are under way in the consolidated Hydrogen Materials Advanced Research Consortium (HyMARC).
- The project has shown accomplishments in various areas in hydrogen storage systems analysis. The most significant accomplishment was related to the reverse engineering of the material properties for the RT adsorbents. This work was very helpful in aligning the materials research to the DOE system targets. It is also helpful for this project to continue to pursue opportunities to reduce the cost of the 700 bar compressed tank design.
- ANL analyses indicate that the performance levels of existing storage approaches have now reached a maturity level at which only minimal enhancements look likely, with nearly zero-sum tradeoffs between performance and costs.
- The project team needs to assess the impact the modified end cap may have on the overall lay-up, fiber-winding process, and associated costs in tank manufacturing. The project may require changes in the winding pattern that affect cost. It is uncertain why the project team did not assess the potential for 700 bar ambient temperature tanks for use in buses. When asked, the presenter made a reference to Toyota's use of 700 bar tanks, but an analysis of these tanks in buses (if it has indeed not been done previously) is still important. In the "H_2 Carrier Study," it was good to see this particular activity, approach, and related accomplishments. An analysis of the scalability of the three options (C_cH_2 at 350 bar, C_cH_2 at 500 bar, and compressed H_2 at 350 bar) is important for understanding the influence they have on versions of infrastructure and renewable versus non-renewable sources; the analysis should be added to this activity. In addition, glass microspheres were studied for large-scale transport of hydrogen at about 11% gravimetric density, but at about 20 kg_{H2}/m^3 volumetric density. They can be used as one-way or two-way RT carriers. The project received positive Program Annual Merit Review results in the mid-1990s, but it was halted. The project team may be worthy of addition to this study.
- This project covers different methods for onboard hydrogen storage, but it is mostly being compared to 700 bar compressed hydrogen storage. The project does not make clear reference to DOE goals or how the project is expected to progress toward those goals. Compressed hydrogen vessel analysis with boss-reinforced dome finite elements shows the boss plastically deforming under normal operating conditions. It is unclear whether there was an analysis performed to show fatigue durability of the boss component.

Question 3: Collaboration and coordination

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

- The ANL team continues to interact closely with a variety of other organizations in both government and industry on key aspects of hydrogen production, storage, and delivery. The team remains effective in consolidating the relevant technical inputs and communicating outputs to project partners.
- Extensive collaborations are in place with multiple national laboratories and other research and development (R&D) centers. The collaborations are well managed and coordinated, providing important input to the analyses being conducted within this project. It will be important to enhance the collaborations with the consolidated HyMARC team. The HyMARC project and associated seedling activities have become the focal point for understanding sorption properties and behavior in the most promising storage candidates. It will be essential to augment collaborations between this system-level effort with HyMARC to provide DOE with a complete and fully transparent assessment of the hydrogen storage system status, as well as any challenges.
- The data shared appear to come from a variety of sources, including industry and other national laboratories.
- It looks like this project contains a very good mix of collaborators.
- The project appears to have a high level of collaboration and has been open to reviewing the analysis assumptions. The collaboration list includes several national laboratories and DOE tools, although the industry connection is limited and could be expanded to confirm the results and research direction. The cost analysis collaboration should be further explained, especially in relation to the hydrogen carriers.

Question 4: Relevance/potential impact

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

- This project continues to be critical to the success of the Program. It is an ongoing activity that builds upon prior solid successes. This project serves as the definitive source for systems-level analyses and projections for the Program.
- While it is also the greatest challenge, the potential impact of sorbents (especially RT sorbents) is huge if it is successful. This is an iconic example of what government does best that industry and markets cannot do.
- The project includes a mix of relevance and impact based on various applications. The RT adsorbent reverse engineering is highly important for this materials-based storage approach. The impact of the hydrogen carriers was not clearly explained and may have lower relevance.
- The ANL team confirmed that CcH_2 storage systems do very well meeting the onboard targets; nevertheless, there remain significant issues with the necessary liquid hydrogen infrastructure. As the ANL project showed this year with the assessment of the RT adsorption materials, there are no known solid storage media candidates that can simultaneously satisfy the updated 2020 DOE targets, let alone the ultimate values. All candidate storage options exhibit compromises of contradictory requirements and behavior for physical or chemical storage systems. The latest assessment reveals that minor improvements appear to be possible.
- Data within this project is focused on comparison to compressed hydrogen in most areas. There is no clear path to improvement for current technology in some areas of the presentation. Identified barriers such as life-cycle assessment and charge/discharge rate are not addressed in all areas of investigation.

Question 5: Proposed future work

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

- The proposed future work appears to be a continuation of the past year's activities with limited new areas of analysis. It is good that the future work includes verification of the RT sorbent with Lawrence Berkeley

National Laboratory, along with other sensitivity analysis. The work associated with expanding compressed hydrogen to medium- and heavy-duty fuel cell electric vehicles is an excellent focus.
- The proposed future work follows naturally and directly from prior studies. Work planned in all topic areas is clearly delineated. The future work will establish definitive system requirements and storage/carrier scenarios that will provide important benchmarks for the materials development efforts.
- For the "hydrogen carriers for delivery" section, it is recommended that the team add specific activities for analyzing hydrogen carriers that favor the transition to large-scale renewable resources. Atmospheric nitrogen for ammonia is a renewable resource.
- The tasks outlined by the ANL team are all reasonable and focus on the continuation and extension of the team's current efforts. The scope of work will be useful; however, it seems unlikely (in light of progress and behavior of currently identified materials) that breakthrough discoveries will be made as a direct consequence of this project.
- Future work does not give a clear indication of how progress toward DOE goals will be made.

Project strengths:

- Over the past decade, the ANL team has developed and adapted a diverse variety of models to predict both attributes and limitations of nearly all types of hydrogen storage systems. The team continues to provide useful insights on the constraints required from various storage media in order to meet DOE targets. The team's in-depth analyses are always performed systematically and include inputs and critiques from other organizations.
- This project provides system analyses that are vital for assessing the current state of hydrogen storage technologies and materials development. The results generated in this project provide a valuable "reality check" for materials and system developers.
- Individuals involved in this project have extensive backgrounds in analyzing hydrogen storage systems. In addition, the project analysis often includes a sensitivity assessment to determine the preferred operating conditions.
- This project addresses multiple storage technologies, both onboard and off-board, and identifies base cost and volumetric and gravimetric densities for each technology.
- This project consists of solid teams, solid collaboration, and a strong mix of timely and relevant R&D for hydrogen storage.

Project weaknesses:

- This is a very strong project with few deficiencies or weaknesses. The only (minor) concern is that a more robust collaboration with HyMARC is necessary. The results of the system analyses should be coupled more strongly to the work on materials development, characterization, and foundational understanding to ensure that the large parameter space for those efforts can be narrowed and distilled in a rational way.
- The project could be improved by reaching out to industry for confirmation and verification of results. The analysis performed in this project is often strong, while the confirmation and cross-reference of results could be further explained. It is assumed that the project individuals are confirming their results in the background, which should be mentioned during a review of the results.
- While the ANL team carefully evaluated detailed aspects and variations of hydrogen storage systems and supporting infrastructure, there have been limited design and materials advancements on hydrogen storage technology in recent years. Hence, the prospects for meeting the major improvements necessary for achieving the DOE targets are being affected. Furthermore, the ANL team neither possesses nor has direct access to a means of validating findings. If outside researchers or industry does not provide these necessary measurements, the refinements or modifications to the ANL findings will not be critically tested; thus, recommendations would not implemented.
- Hydrogen carrier pathways involve hydrogen during the production of the carriers, carrier transport, and hydrogen evolution from carriers, with very little information regarding hydrogen storage. The hydrogen carrier efforts, while critical to hydrogen and fuel cell success, should be moved to a different or separate activity.
- The next step to make progress toward DOE targets is not clearly defined.

Recommendations for additions/deletions to project scope:

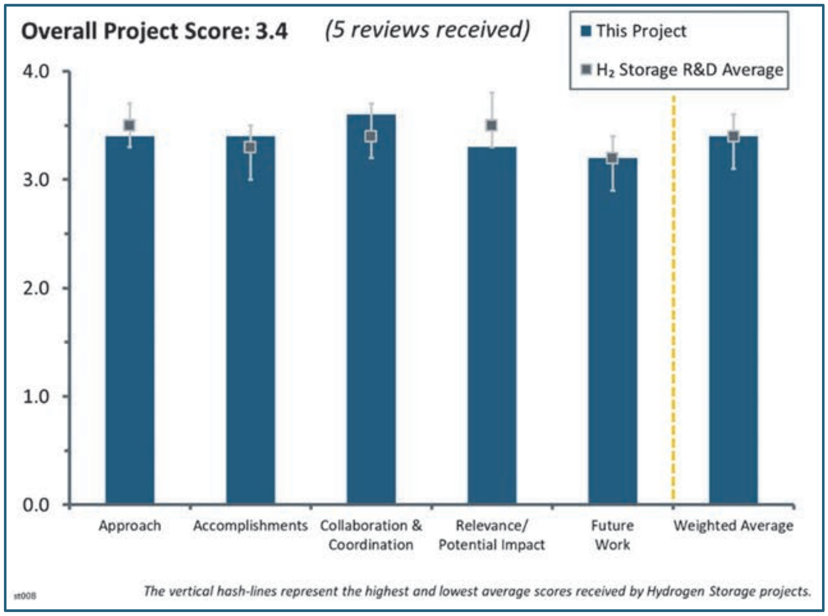
- There are no recommendations for changes to the project scope. However, at some point in the not-too-distant future, it is anticipated that the work on tanks and cryo-compressed storage will reach a point at which the emphasis can be shifted more strongly to other candidates that have the potential to supplant the compressed hydrogen approaches.
- One recommendation is for the team to complete reverse engineering for all materials-based storage systems and publish a complete summary table of these targets. It would be helpful to include additional industry feedback regarding certain analysis concepts. For example, bus manufacturers are focused strictly on the cost and robustness of the storage technology; they are not concerned with volumetric density, so cryo-compressed storage may be less interesting, especially if the technology has a high cost penalty for initial low-volume market entry.
- The project team needs to conduct an evaluation of solid storage as a possible carrier for delivery. For various liquid hydrogen carriers, it is unclear what the environmental effects (off-gas) are for each of the evaluated methods. The comparison of Type 3 tanks versus Type 4 tanks at different storage pressures has already been conducted in the past; it is unclear how this will help with progress toward DOE targets.
- It is recommended that the team consider exploring the use of high ZT or other materials for the thermoelectric cooling of the cryogenic-pressure hydrogen tanks. When first explored in 1993, there were no reasonable costing choices, but recent improvements for thermoelectric cryogen applications may dramatically extend dormancy.
- It is recommended that the ANL team be supported to perform all of the tasks summarized in the future plans.

Project #ST-008: Hydrogen Storage System Modeling: Public Access, Maintenance, and Enhancements

Matt Thornton; National Renewable Energy Laboratory

Brief Summary of Project:

The ultimate goal of this project is to provide and enhance publicly available materials-based hydrogen storage system models that will accept direct material property inputs from materials developers to accurately predict materials-based hydrogen storage system performance. In support of that goal, this project maintains, enhances, and updates the Hydrogen Storage Engineering Center of Excellence (HSECoE) hydrogen storage system modeling framework and model dissemination web page.



Question 1: Approach to performing the work

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

- This project is an extension of the analysis and modeling efforts conducted during the former HSECoE. In particular, this project addresses materials and system issues for alternative hydrogen storage media (e.g., metal hydrides, chemical hydrogen, and adsorbents). Analysis tools were developed and made available to allow outside users in the international hydrogen research and development community to make comparisons over a range of parameters and operating scenarios against reference materials. The objective is to assist material researchers to identify viable candidates with the potential to meet the U.S. Department of Energy (DOE) vehicle performance targets. The project also provides a level of technical support to the model website to assist outside users.
- The approach focuses on the development, validation, and dissemination of modeling tools that can be used to evaluate performance of new hydrogen storage materials in practical storage systems. The focus of the approach is to provide a straightforward and rational way to transfer the engineering development knowledge derived from the HSECoE consortium effort to materials researchers. The project is providing the hydrogen materials development community with the ability to input material properties and to evaluate the impact on system characteristics. Overall, through development and application of a wide range of simulation/modeling tools, the project is ensuring that the HSECoE models are managed, documented, and disseminated effectively.
- The goal of this project is to increase storage materials researchers' ability to use available modeling where there is an impedance match between materials research data and the technical targets for vehicle hydrogen storage systems. The approach was to improve the framework modeling with improved utilities so that research data are directly used to provide evaluations for the materials used in vehicle applications. In the process of improving the modeling, the user interface and website were also improved.
- The tools on the website provide excellent resources to evaluate various storage technologies and have added functionality, allowing users to evaluate most materials based on desired editable inputs.
- The project has a very good approach in developing and making available models for materials-based hydrogen storage researchers, although the project should also utilize its own models to assist in evaluating materials research. For example, the project could conduct a reverse engineering evaluation of the materials research targets using the system models.

Question 2: Accomplishments and progress

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

- Solid progress has been achieved in several areas:
    - Access to/support for the website has been improved.
    - Stand-alone system design tools that facilitate user input on material characteristics in a straightforward way have been developed—specific design tools and system estimators have been developed for metal hydride systems, sorbents, and chemical hydrogen materials.
    - A generic user interface (GUI) for a hydrogen vehicle simulation framework, and the integration of the design tools within that framework, has been demonstrated. Website analytics are being applied to evaluate user access.
- Substantial progress was made during the past year or so in updating, refining, and maintaining the HSECoE model dissemination website. While such simulations are helpful tools in understanding behavior for generic storage media and systems, they do not necessarily hasten discovery or development of the targets. The team spent considerable effort in improving support documentation and making other changes to the formatting and approach to enhance the website's usefulness. The development and implementation of stand-alone system design tools, an isotherm fitting tool, and a new GUI/framework are commendable. These activities make this website a better and more attractive resource for independent research groups.
- The models available cover the range of storage materials and technologies for relevant hydrogen storage systems. A few are in progress, but it appears that they will be completed by the end of the project. The improved website and GUI should help hydrogen storage materials researchers use this valuable utility in growing numbers over time.
- The completion and updates of the stand-alone system design tool models are very useful and an excellent accomplishment. The improved website access and support also improve the modeling effort.
- The project team met the intent of the project, providing tools for evaluation of various storage technologies.

Question 3: Collaboration and coordination

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

- The technical interchanges among the team members from the different national laboratories, as well as outside members, remains very efficient and highly coordinated throughout this project. The progress made on both the adsorption and metal hydride storage models indicates excellent cooperation. The team reached out to model users via a survey with some follow-up and revisions.
- Beneficial collaborations and cooperation among team members from Savannah River National Laboratory, Pacific Northwest National Laboratory, and National Renewable Energy Laboratory are evident. Additional collaborations with universities, industries, and a private consultant have accelerated progress on the project. The project is well managed, and effective coordination among team members and other collaborators is apparent.
- The collaborators in this project have the key capabilities needed to meet their respective responsibilities. The coordination and management of the collaborations seem to be effective enough to allow the accomplishments over the last three years.
- The project partners are highly collaborative and coordinated in their efforts to develop models. The validation of the models needs to be further evaluated with external researchers. The idea of conducting a survey was good, although the use of the results to further collaboration with the users is uncertain.
- There is good collaboration with other national laboratories. More verification of results from industry is suggested.

Question 4: Relevance/potential impact

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

- This project ensures that models developed and tested in the HSECoE consortium and elsewhere for evaluating storage materials relative to DOE targets are codified and disseminated to the materials community in a user-friendly and capable format. This project is extending and augmenting the important work that was conducted in the HSECoE. It is an important part of the overall DOE energy research, development, and demonstration strategy and is generally well aligned with the goals and objectives of the Hydrogen and Fuel Cells Program.
- The accomplishments in this project align very well with DOE objectives and should significantly aid in moving toward those objectives. The utility of the modeling system should significantly increase the development rate for materials-based and other hydrogen storage systems for vehicles.
- These good tools facilitate the ability to evaluate different storage technologies and understand the benefits and limitations.
- The project team continues making very good progress in providing very accessible enhanced numerical models that had originally been developed during the HSECoE but have undergone extensive revision and refinement over the past couple of years. Their recent work has made these models more accessible to the general hydrogen storage community. Nevertheless, it remains unclear just how much other research groups are willing or able to fully utilize these tools for assessing progress and determining limitations on meeting the DOE performance targets.
- The project has high relevance as a bridge between materials research and DOE system technical targets. However, the impact of the project is uncertain, since the project is developing the tools rather than using the tools to guide and assist the research directly.

Question 5: Proposed future work

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

- The planned expansion of the model to other near-term vehicle platforms is very important. The optimization of a given vehicle platform for a particular application does not necessarily need the same storage system as a light-duty vehicle for highway use. Adding the vehicle-side refueling model and relating to refueling infrastructure needs would be a valuable addition. All the “Next Steps” proposed plans are important to accelerating vehicular hydrogen storage development and, in turn, helping in the growth of hydrogen as a transportation fuel.
- The team appears well positioned to complete all of the prescribed tasks by the end of fiscal year 2018 (FY 2018). It would be very valuable for the team to maintain the model websites and also provide technical support to outside users. Finally, it is recommended that this project receive continued support to expand the models by applying them to other storage materials, theoretical formulations, and vehicle class options.
- The project ends this year on September 30, 2018. Completing the proposed future work outlined on slide 31 is an ambitious undertaking. The more reasonable work schedule described on slide 30 (i.e., milestones and deliverables) seems straightforward and includes reasonable extensions that could be accomplished by the project completion date.
- The proposed future work is very interesting, especially in the area of expanding to fueling and other vehicle platforms. The plan to accomplish this future work needs further explanation to ensure the effort is focused and containable.
- Validation of the model is critical. Correlation between the model and actual data is key to the acceptance of the results of the models. As a design tool, results will be taken with a level of skepticism until the correlation data is presented. The evaluation of variables that change depending on vehicle class would expand the usefulness of the tools with minimal additional effort.

Project strengths:

- As described in prior reviews, the core team members have extensive knowledge and expertise on all of the hydrogen storage media, as well as the appropriate software analytical packages to develop and execute the modeling codes for the website. This team includes a collection of experienced individuals to continue and extend the storage system parameters.
- The members involved were part of the HSECoE and understand materials-based systems along with materials research attributes. The models are very useful tools for materials researchers and are relatively straightforward to utilize, based on GUI screens and other instructions.
- This is a well-coordinated effort that is ensuring that the results and knowledge gained in the HSECoE effort can be used effectively by the hydrogen storage materials research community. This is an important legacy of the HSECoE.
- This project's organization and project execution are solid, as is the relevance to the DOE goals and the needs for the growth of hydrogen vehicles in the marketplace.
- The project team created good evaluation tools with clear definitions and flexibility to allow users to evaluate various materials and storage technologies.

Project weaknesses:

- This is a strong technical team with diverse expertise; hence, there are few weaknesses associated with the team's ability to develop and deploy the analytical tools. The primary limitation is the absence of validation results and feedback from outside users.
- There are many incomplete tasks ahead with only three to four months of the three-year project time and 14% of the budget left. If expansion of materials models expects researchers to provide specific technical data on their materials, it is unclear what will motivate them to provide that information.
- The main project weakness is that the models are being developed but are not being utilized to make projections or develop strategies for closing the gap to the DOE hydrogen storage system targets. Also, the validation of the models should be further explained to allow researchers to gain confidence in the results.
- Validation of the models continues to remain a dominant issue. Greater emphasis is needed on a more detailed description of the approach for validating the models and design tools.
- Correlation data between models and real life are desired to validate the models. Having more data on the website showing this correlation would be desirable.

Recommendations for additions/deletions to project scope:

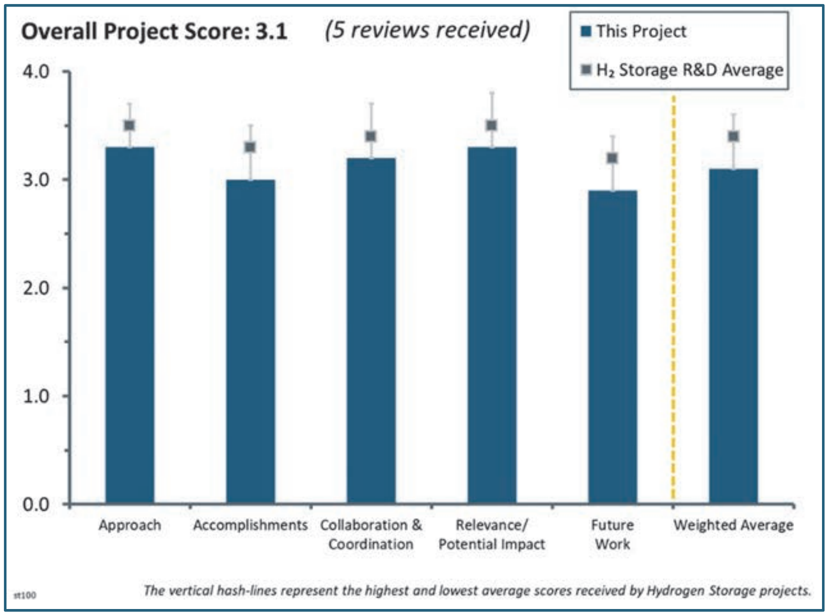
- Since the project is apparently essentially complete with respect to the planned analytical models, the planned final additions are satisfactory. However, funds should be made available for at least another year or longer to maintain the models on the Internet and also support appropriate team members to respond to user inquires. It is recommended that this project be extended to allow the team to expand the model platforms to other fuel-cell-powered vehicle types (e.g., medium and heavy-duty trucks, forklifts, buses, etc.).
- The project should include the scope of using the models to develop strategies and provide sensitivity analysis of key materials attributes to achieve the DOE system targets. The project team should assess the level of interest and downloads for certain models and develop a plan regarding the deletion of support for certain models based on the interest level. Overall, the project team should attempt to increase the usage of the models through various communication approaches to connect with materials researchers.
- Use of the website by industry has been quite limited; this may be due to industry's not knowing of it. Links to this website from other locations (e.g., DOE, California Fuel Cell Partnership) could help people find the site and increase the benefits of the work completed.
- This project is concluding this year on September 30, 2018. There are no additional recommendations for changes in the project.

Project #ST-100: Hydrogen Storage Cost Analysis

Brian James; Strategic Analysis, Inc.

Brief Summary of Project:

The goals of this project are to (1) conduct independent Design for Manufacture and Assembly (DFMA) cost analysis for multiple onboard hydrogen storage systems and (2) assess/evaluate cost-reduction strategies to meet the U.S. Department of Energy (DOE) cost targets for onboard hydrogen storage for light-duty fuel cell electric vehicles.



Question 1: Approach to performing the work

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

- The project team's DFMA-based cost analysis approach provides rational and meaningful component and system cost analysis predictions and is useful for identifying optimum cost design and manufacturing pathways. The approach has been proven effective in prior studies, and optimization of the approach provides important new cost analyses that are helping to inform decisions for the DOE Hydrogen and Fuel Cells Program (the Program). The focus in fiscal year (FY) 2017 and 2018 was on the analysis of light-duty vehicles (LDVs), fuel cell electric buses (FCEBs), and Type IV natural gas storage systems. These analyses directly complement those conducted in the companion systems analysis project led by Argonne National Laboratory (ANL) (ST-001). The reverse engineering of metal hydride (MH) systems was an important addition to the overall project scope for this reporting period.
- The project work is focused on the cost of hydrogen storage systems, which is the most critical barrier for the commercialization of these systems. The activities within the past year include a very good balance between LDVs, bus applications, and compressed natural gas (CNG) analysis. It was excellent that the project team utilized its cost estimation tools to conduct a reverse engineering approach with MH. This approach should be extended to other hydrogen storage systems. The approach of extending to CNG examples is helpful in order to evaluate a similar technology.
- Strategic Analysis, Inc. (SA) uses well-established analytical tools and detailed descriptions of the various hydrogen storage systems to explore the impact of design choices and materials selection on the cost of representative configurations. Systematic assessments were made to ascertain relative roles of specific components on both performance parameters and costs. One limitation is the absence of commercial validation of projections and findings.
- The project team looked at five different storage systems and derived potential costs and cost sensitivities. The use of DFMA as the primary method for defining manufacturing costs should lead to reliable predictions of costs and sensitivities. The reversible MH system design and analysis were straightforward and reasonable, and the results are probably realistic. It is a useful foundation for sensitivity analysis of potentially practical vehicle hydride storage system designs and MH material choices. For composite pressure vessels, a comparison between wet lay-up fiber winding and advanced tape placement (ATP) was done to look at cost drivers for both methods for composite overwrapped pressure vessels. It is uncertain that the collaboration with DuPont provided reliable data to develop a reasonable cost comparison. It seems to be a little ambiguous at this point. It is uncertain that this activity shows much future programmatic value.

- This project consists of a general coverage of current technologies and reasonable estimates of technology costs. ATP costs and speed are the same as those for wet lay-up. Typically, ATP is slower in speed and its materials more expensive than wet winding; it is necessary to verify those numbers.

Question 2: Accomplishments and progress

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

- Solid progress was made in FY 2017 and 2018. Most notably, the comprehensive MH reverse engineering work successfully identified parameters that could be altered/optimized to meet cost targets. A manufacturing process flow comprising elements unique to MH storage systems was developed, and an analysis of thermodynamic constraints on cost projects was performed. This analysis is necessary for a rational and sensible assessment of the MH system and its manufacturing cost. A similar process flow analysis was performed for a CNG storage system. In this case, the analysis focused principally on the manufacturing approach. This analysis facilitated a formulation of tank cost comparisons and the evaluation of primary cost-critical components and processes. Cost analysis for storage options in FCEBs was focused on a 500 bar cryo-compressed system. Useful results and conclusions concerning costs of specific steps and components in the process flow and comparisons with conventional compressed gas storage were presented. As an aside, on slide 10, it seems that the "ship in a bottle" approach to the assembly of the MH heat exchanger begs the following concern: if the heat exchanger fails, it is unclear whether it can be swapped out or the entire assembly needs to be replaced. That could be a costly and time-consuming proposition.
- A key accomplishment of the project is the reverse engineering effort, since it concluded that the DOE target cost is not attainable with the MH concept. This important result can be used to influence the MH research strategy and targets. The 700 bar system baseline updates were also useful in evaluating attribute tradeoffs to improve the cost.
- With the exception of the wet lay-up versus ATP study, the project's accomplishments this year all represent progress toward the Program's Hydrogen Storage R&D category goals. The 700 bar baseline updates provide interesting insight into a number of things, in particular, the regulator pressure drop decrease and relaxed fuel cell system requirements. The latter implies the need for an additionally interesting trade analysis iterating different fuel cell/regulator configurations versus drive cycles. The FCEB cryo-compressed versus compressed hydrogen analysis is valuable in showing the best current-application benefit from transitioning to cryogenic fuel infrastructure and cryo-compressed vehicular storage.
- During the past year, the project addressed four primary topics: (1) revised costs for 700 bar Type IV hydrogen gas storage in LDVs; (2) reverse engineering analysis of hybrid 350 bar reversible MH systems based upon a Type IV vessel; (3) cost analysis of the 500 bar (60–80 K), cryogen-compressed hydrogen (CcH_2) storage systems for FCEB applications that had been previously conceived by Lawrence Livermore National Laboratory (LLNL) and evaluated by ANL; and (4) Type IV tanks for a 3600 psi CNG storage system. The purpose or rationale for evaluating a CNG system within the Program is not clear, other than to provide some independent validation of the assumptions for commercial materials and manufacturing costs on the SA analysis methodology. Furthermore, the selection of a Type IV hybrid MH tank is very ill advised because the polymer liner is highly vulnerable to damage from the expanding and contracting hydride particles; this would generate excessive hydrogen permeation and leakage via generated defects, and the outgassing of volatile organic species from the polymers over time and during temperature excursions would likely contaminate the hydride material. This, in turn, would seriously impede kinetics and storage capacities and would form impurities in the delivered hydrogen gas. Only metal inner liners should be considered for vessels containing MHs.
- This project successfully evaluated the costs of leading technologies and identified issues and possible solutions for technology shortcomings. The cryo-compressed data references ANL with no new data presented, thus not providing independent analysis for this technology.

Question 3: Collaboration and coordination

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

- Collaborations with national laboratories (Pacific Northwest National Laboratory, LLNL, and ANL) facilitated and enhanced the technical effort within this project. The collaboration with ANL is especially noteworthy because the ANL systems analysis efforts provide important and timely input to the cost analyses in this project. In addition to the national laboratory collaborations, more active participation of industry partners is being sought and encouraged.
- The project team has direct collaboration with national laboratories and industry to assist in confirming assumptions and reviewing the results of the team's analysis. The project's proactive communication with technology stakeholders to direct and review the team's analysis is a key strength of this project.
- The collaborators and their roles are defined and appear to be well managed for the goals of this project.
- The presentation lists a wide group of national laboratories and industry that have collaborated on this project.
- SA has cooperated closely with ANL on many aspects of the analyses and also gathered information and input from a number of independent organizations on various aspects of hydrogen storage media and methods. However, the team apparently had only limited contact with organizations that had hands-on experience with intermetallic and complex hydrides prior to assessing the high-pressure hybrid storage vessels.

Question 4: Relevance/potential impact

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

- This project, in tandem with the ANL systems analysis project, provides DOE with quantitative and comprehensive technoeconomic analyses and technical status and projections for hydrogen storage systems. The DFMA analysis employed in this project has been shown to be effective for predicting the costs of manufacturing processes, materials, and components. DOE uses this information to inform system and manufacturing development decisions. Overall, the project is well aligned with the objectives and goals of the Program.
- The project has high relevance, especially since cost is a key barrier to the commercialization of hydrogen storage systems. The project not only identifies the cost but also attempts to identify solutions—although further work could be conducted to cascade cost goals and opportunities for certain system elements in order to achieve the targets.
- Excepting the ATP versus wet lay-up, all the projects in this effort meet the needs of the Hydrogen Storage R&D category and are helpful in moving the bar for hydrogen vehicle storage systems.
- This project contains the cost analysis of leading hydrogen storage technologies and helps to identify areas that prevent these technologies from reaching DOE targets.
- The independent, yet coordinated, assessments by SA are valuable to the development of cost-efficient hydrogen storage systems. The concern is that without validation from actual vehicles and fabricated systems, these analyses will emphasize minor (rather than unanticipated major) issues.

Question 5: Proposed future work

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

- The proposed future work is a reasonable and rational extension of the technical direction established in the prior year. The addition of metal–organic framework (MOF) reverse engineering analysis (complementary to ANL systems analysis) is important.
- The proposed future work appears to be a continuation of the past year's activities, rather than new activities to advance the technology. The only new item was the MOF reverse engineering analysis, which would be very useful. The project team should review the future work plan and modify it to include higher-

impact analysis of options to improve the 700 bar baseline system for the tank, rather than the regulator, since the tank has the highest cost contribution. The cryo-compressed cost estimates should be validated by original equipment manufacturers that have produced these systems, since the cost projections do not seem to align with the current actual costs.

- Analyses of the high-pressure MHs should consider Type III vessels rather than Type IV designs. Since adsorption systems have considerable temperature excursions during operations, analyses should consider the impact of cryogenic temperature on the mechanical properties of carbon and polymers. Unless stronger justification can be provided by SA, there seems to be only limited purpose for continuing the CNG cost and design analyses.
- The extrapolation of ATP translation efficiency from 3600 psi containers to 700 bar containers needs to be verified for this data to be usable. The recyclability of a vessel is a desired benefit for society, but it is not typically a cost-saving measure for vehicle manufacturers or primary customers, as recycled fibers cannot be used in vessels at this time.
- It is important to continue all the LDV and FCEB plans as listed. The FCEB analysis should coordinate with some fueling infrastructure analysis. Continuing the Type IV CNG analysis does not appear to seriously move toward DOE storage goals.

Project strengths:

- The focus of this project and the individuals involved in the analysis are both strengths of this work. The focus of the project deals with cost, which is the key barrier to hydrogen storage commercialization; this is the only project in the portfolio dealing with cost. The individuals conducting this analysis have excellent capabilities and are transparent with the assumptions and results.
- A well-formulated approach, conducted by a highly capable engineering team, is being used to generate important information concerning the optimization of the manufacturing process flow and the cost of materials and components. The information has important impact on storage system development decisions.
- The SA staff have an excellent grasp of the analytical methodology and computation models on assessing costs for a wide array of hydrogen storage systems. The team clearly communicates the findings and implications of these evaluations.
- This work has a good use of DFMA techniques. The management and collaboration appear to be project strengths. The results of current and future FCEB analysis could strongly influence the advent of better hydrogen bus fleets.
- This project's strengths lie in its independent cost analysis of technologies and the identification of cost barriers that need to be overcome.

Project weaknesses:

- A minor weakness of this project is that a "Summary Slide" would have been helpful; one should be included in future presentations. In addition, the dominant challenges, obstacles, and risks to achieving the project objectives (and meeting DOE requirements) should be stated clearly and candidly. Without that information, it is difficult to fully assess the overall status and future direction. Finally, closer ties to the consolidated Hydrogen Materials Advanced Research Consortium (HyMARC) should be established. Although the cost analysis activity does not fall within the purview of "foundational understanding," it is important for the principal investigator and his team to be fully aware of new and recent developments in the hydrogen storage arena.
- Without actual in-house hardware engineering and materials characterization experience, unrealistic and risky selections have been made on candidate configurations and manufacturing approaches for hydrogen storage systems. This issue could be addressed by more extensive interactions and consultations with organizations and individuals with the appropriate expertise.
- Cryogen-compressed data appears not to be independent. The evaluation of this technology is weak. Technology improvements discussed in the new baseline are not evaluated for further reduction for compressed hydrogen by compounding possible reductions.
- The project team's weakness is the inability to verify the results. These results may appear optimistic in comparison to actual industry costs owing to comparing price versus cost information.

- The ATP versus wet lay-up activity is well executed, but the value versus cost of the activity is unclear.

Recommendations for additions/deletions to project scope:

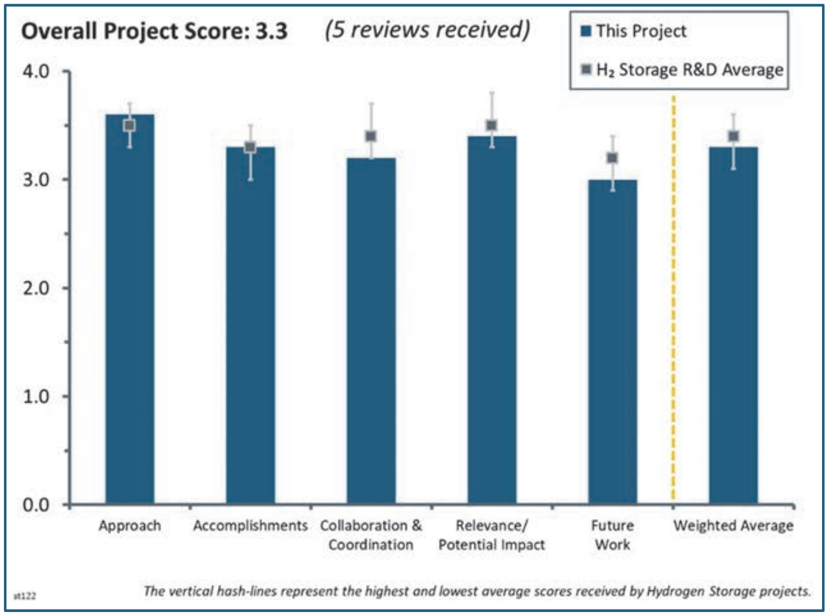
- It is recommended that the team continue and expand the reverse engineering effort to establish goals for materials costs and other system elements. The project scope should be modified to focus on the highest-cost contributors (such as the 700 bar tank), rather than smaller items (such as the regulator). Additional low-volume cost estimates for the storage system would be useful. The consistency of the cryo-compressed cost analysis should be confirmed for low volume to ensure the initial introduction of these technologies could be implemented without a significant cost penalty.
- If the FCEB CcH2 versus compressed hydrogen sensitivity analysis is to be finalized, it should be accompanied by the initiation of a cost analysis for the fueling infrastructure differences between 350 bar compressed and 350/500 bar CcH_2. This is because bus fleet applications, like materials-handling operations, are fleet-like, self-contained, and require concurrent installation.
- Revised analyses of the 350 bar MH should be done on Type III vessels instead of completing assessments of the Type IV configurations. It is recommended that the team cease work on the Type IV CNG analyses and devote more time to other topics or decrease the scope of this project.
- The balance of plant (BOP) evaluation was done only for the regulator. It is suggested that the team possibly evaluate other BOP components for additional cost reductions.

Project #ST-122: Hydrogen Adsorbents with High Volumetric Density: New Materials and System Projections

Don Siegel; University of Michigan

Brief Summary of Project:

A high-capacity, low-cost method for storing hydrogen remains one of the primary barriers to the widespread commercialization of fuel cell vehicles. Storage via adsorption is a promising approach, but high gravimetric densities typically come at the expense of volumetric density. This project's goal is to demonstrate best-in-class metal–organic frameworks (MOFs) that achieve high volumetric and gravimetric hydrogen densities simultaneously, while maintaining reversibility and fast kinetics. The approach entails high-throughput screening coupled with experimental synthesis, activation, and characterization.



Question 1: Approach to performing the work

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

- High-throughput screening combined with synthesis, activation, characterization, and system-level projections is used to identify and test promising MOF candidates for hydrogen storage. The high-throughput screening approach is novel and powerful. It has provided a way to explore a huge parameter space that would be experimentally intractable. It provides useful information for identifying MOFs capable of high gravimetric and volumetric hydrogen densities under cryogenic conditions.
- The principal investigator (PI) screened most of the 500,000-MOF database of databases. The screening is to identify any MOFs with gravimetric and volumetric hydrogen densities greater than the reference, MOF-5. Volumetric density is critical since it is closely linked to packaging issues in the design of vehicles. The 700 bar tank volumetric densities are about as low as tolerable for many light-duty vehicle designs. Once identified, the best MOF candidate properties are used to define relevant storage system designs or characteristics.
- The project has a very good approach by combining (1) a theoretical basis to screen candidate MOF sorbents based upon critical properties that were complemented by synthesis and (2) empirical characterization of the most promising materials.
- The project made important progress between August 1, 2015, and July 31, 2018. The computational screening performed showed good success. Improvements over MOF-5 performance were demonstrated for three cases. The system-level tests were performed, and challenges with increasing usable gravimetric capacity were clearly identified.
- The project goal is to outperform MOF-5 at 700 bar and to pay further attention to volumetric capacity (without sacrificing kinetics). The approaches to meet these goals are clearly laid out as (1) systematic modeling and (2) system modeling.

Question 2: Accomplishments and progress

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

- The identification and synthesis of MOFs with usable capacities exceeding MOF-5, the incumbent material, is a particularly noteworthy result. The scope of the computational effort is stunning. The PI and his team have done a superb job of surveying such a vast parameter space and down-selecting promising storage candidates. These results will serve as an important research benchmark for future work on physisorption materials. However, the obstacles to synthesis of the hypothetical structures remain problematic (desolvation, framework collapse, etc.). The "synthesis bottleneck" is a serious challenge. It may be beyond the scope of this study, but only limited information is provided concerning MOFs that may support metal cations that facilitate binding of multiple hydrogen molecules. This has become the preferred approach and "holy grail" for achieving the highest capacities. In addition, optimal adsorption enthalpies are equally important but received only minimal emphasis (slide 23). It would be valuable to know whether the screening approach can be adapted to include predictions of heats of adsorption.
- Three interesting MOFs were identified and studied. They had significantly higher gravimetric hydrogen densities than MOF-5, but only one exceeded the volumetric density of MOF-5 by almost 15%. Supercritical carbon dioxide (SC CO_2) activation of MOFs is better than vacuum activation of MOFs, but in the magnesium-based Mg-MOF, it resulted in a structure collapse. Perhaps the extraction rate of solvent or SC CO_2 is an issue. Supercritical solvent extraction was necessary for the production of aerogels, and it is possible the extraction rate was an issue in the final structural integrity. The team developed a useful database for MOFs that will be used by others. It is hoped that this project can continue so the lessons learned for things like supercritical activation, system-level optimization, and packing fraction optimization methods will not be lost and instead continue improving the likelihood of identifying the storage material and system that brings hydrogen vehicles to the next commercialization stage. Using the UMCM-9 MOF in Figure 29 (or any other good MOF), it is unclear whether there is enough pore volume when it is compacted to 0.2 g/cc so that changing the pressure from 100 bar to a higher bar (e.g., 200 or 300) has a significant enough change in volumetric capacity to be significantly better than the 700 bar system, or as good as the cryo-compressed hydrogen vessel at 350–500 bar.
- The research has identified IRMOF-20 as one that outperforms MOF-5. Additionally, SNU-70 and NU-100 have been discovered to outperform MOF-5 by 14.1%.
- This project's accomplishments are commendable. The progress toward the goal, however, is intertwined with basic assumptions made in the project. Scientific progress made will be valuable to future researchers. The system-level challenges remain a major road block. Computations led to many more suggestions than can be synthesized or tested. It may be time to rethink high-throughput capabilities for synthesizing/testing such unexplored suggestions.
- Over the past three years, good progress was made that led to viable candidates being identified and examined by volumetric measurements; however, it must be pointed out that none met or exceeded the criteria necessary to achieve the 2020 DOE targets.

Question 3: Collaboration and coordination

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

- There were excellent interactions within the project among the partners. The most significant external interaction was with the Savannah River National Laboratory on modeling. However, apparently little characterization with either the Hydrogen Storage Characterization Optimization Research Effort (HySCORE) or Hydrogen Materials—Advanced Research Consortium (HyMARC) was solicited or performed; such work would have enhanced this project.
- Solid and beneficial collaborations with Ford Motor Company and the Hydrogen Storage Engineering Center of Excellence (HSECoE) have augmented the core project team's technical efforts. Closer cooperation with investigators in HySCORE (especially J. Long, Lawrence Berkeley National Laboratory) might have accelerated progress.

- This project showed very good and well-coordinated collaboration. There was a great use of organizational capabilities with the University of Michigan, Ford Motor Company, and HSECoE.
- Collaboration with Ford Motor Company was important; however, the rest of the collaboration inside the University of Michigan and with other groups could have increased the net impact of the effort and funding.
- The team collaborates with Ford Motor Company and the HSECoE.

Question 4: Relevance/potential impact

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

- The project addresses what the potential is of identifying and discovering adsorbent materials with the highest volumetric and gravimetric capacities for reversible hydrogen storage. Several potentially promising candidates were found and prepared for characterization. These studies are valuable for establishing whether cryogenic adsorption offers viable solutions for vehicle hydrogen storage. However, no candidates were found that allowed ambient temperature storage that exceeds conventional 700 bar compressed gas capacities or would be capable of meeting the DOE targets.
- This is a unique project in the Hydrogen and Fuel Cells Program portfolio. The extensive survey of MOF candidates has provided information that will serve as a useful resource for future research. Although there are many unanswered questions and obstacles, particularly with regard to actually synthesizing the most promising candidates identified in the survey, these results nonetheless provide DOE with important and meaningful new information. The project is well aligned with DOE research and development goals and objectives.
- The potential is high for materials-based hydrogen storage systems with weights and volumes important to using hydrogen as a transportation fuel. Seeking out the right adsorbent materials among likely candidates is critical to long-term hydrogen success.
- New MOFs as sorbents that outperform MOF-5 are being pursued. The shift in idea to pursue high volumetric capacity, rather than gravimetric capacity, forms a unique approach for this research project.
- The relevance of this project is high for the field. The methods developed, if made available, and collaborations established with the broader community of researchers can have a high impact. However, at this point, the impact is limited.

Question 5: Proposed future work

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

- The project is ending July 31, 2018. The proposed future work on evaluating performance characteristics of selected MOFs identified in the modeling work, archiving computational data, and submitting manuscripts for publication seems reasonable in the short time remaining in the project.
- Since this project ends within a few weeks of the 2018 Annual Merit Review, there can be little practical guidance on future work. However, it is recommended that the project's final report fully document the properties of the most attractive candidates, as well as identify any promising screened candidates that either could not be synthesized or had hydrogen adsorption well below the predictions.
- The project is due to end July 31, 2018. If it continues beyond that, the Potential Future Work slide and Challenges and Barriers slide should guide the project.
- The project should provide a summary of factors that enhance volumetric capacity. Systems modeling will be undertaken in the future (in collaboration with the HSECoE).
- No new ideas were proposed to address the main challenges. The ideas listed seem to be some more of the same approach. To be competitive in future rounds of competition, new ideas and new collaborations will be necessary.

Project strengths:

- The project had a strong effort in handling and classifying real and predicted MOFs for their hydrogen storage potential. The approach is solid, and the theory level is commensurate with the goal of screening a large number of potential candidates. The team met some of the goals; some intractable ones remain a challenge in this field.
- The project shifts thought to the pursuit of volumetric capacity for sorbent materials. This has proven successful for this project and will likely spawn additional research in this topic area.
- An impressive survey of the overall MOF landscape and parameter space volumetric and gravimetric capacity performance has been conducted. The vast scope of the survey has provided information that will undoubtedly motivate future work.
- This project is a great foundation for high-throughput screening of hydrogen storage material candidates and for translating data to system-level modeling.
- Extensive screening with the MOF materials group was combined with supporting experimental work on the more promising candidates.

Project weaknesses:

- The “holy grail” for hydrogen storage in MOFs is the identification and synthesis of physisorption materials capable of supporting metal cations that bind multiple hydrogen molecules. This was apparently beyond the scope of the present study. However, it remains the most important and challenging area of study for hydrogen storage in MOFs. It is unfortunate greater emphasis was not given to that issue. In addition, it would have been helpful if the survey could have been linked to an analysis that included predictions of heats of adsorption.
- Perhaps calculations could include a packing fraction estimate subroutine coefficient that can quickly and reasonably determine packability levels of each proposed structure. If it is possible, it would be interesting to know if that could aid in the high-throughput calculations using pelletization or powder packing rather than using crystal densities.
- Assessments were based upon compiled information on only MOF materials; hence, it is highly likely that there were promising candidates overlooked or inadequately considered if data were not available or incorrectly cited. This project appeared to make little use of the potential insights possible from the HySCORE and HyMARC consortia and obtained greater understanding on promising candidates identified during the screening process.
- One project weakness is the very linear attempts at organizing and running this project. Some of the challenges are foreseeable, but no new mitigation approach was developed to address the system-level concerns and underperformance of the gravimetric storage density.
- The researchers are urged to summarize the factors that contribute to high volumetric capacity.

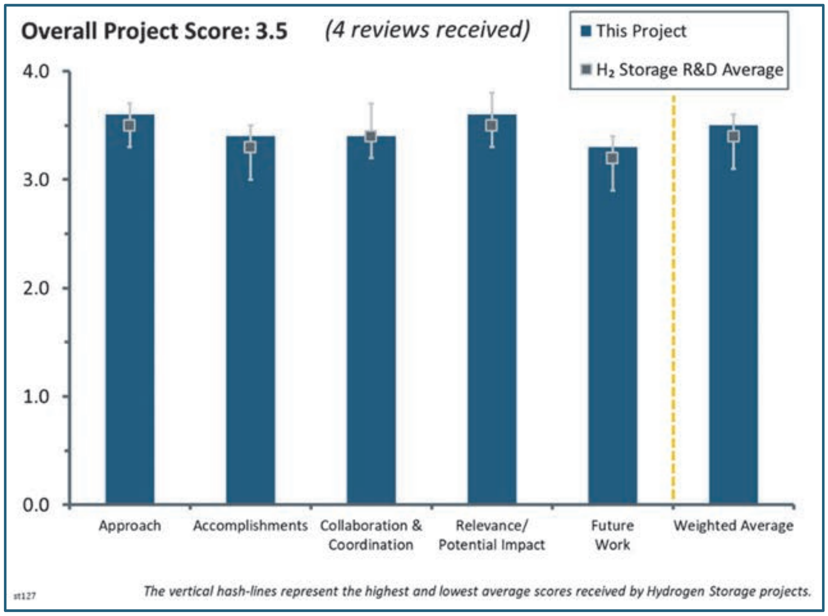
Recommendations for additions/deletions to project scope:

- This project should continue.
- The project is ending July 31, 2018. Therefore, no recommendations are provided concerning revisions to project scope.
- The project is ending. Recommendations for additions/deletions to project scope are not applicable.
- Since this project is now effectively completed, there is nothing more to say.

Project #ST-127: Hydrogen Materials—Advanced Research Consortium (HyMARC): A Consortium for Advancing Hydrogen Storage Materials

Mark Allendorf; Sandia National Laboratories

Brief Summary of Project:



Critical scientific roadblocks must be overcome to accelerate materials discovery for vehicular hydrogen storage. The project objective is to accelerate discovery of breakthrough storage materials by providing capabilities and foundational understanding. Capabilities will include computational models and databases, new characterization tools and methods, and customizable synthetic platforms. Foundational understanding is needed for phenomena governing the thermodynamics and kinetics-limiting development of solid-state hydrogen storage materials.

Question 1: Approach to performing the work

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

- Hydrogen Materials—Advanced Research Consortium (HyMARC): The barriers and challenges to onboard hydrogen storage are clearly understood. The approach that has been adopted by the core HyMARC team is comprehensive and focused keenly on the thermodynamics and kinetics issues that limit the performance of reversible hydrogen storage media. An impressive combination of theory and modeling across multiple time and length scales, synthesis and unconventional processing (e.g., very high pressure), and sophisticated in situ and ex situ characterization techniques is facilitating a deeper understanding of fundamental processes that control hydrogen sorption behavior in storage materials. In addition, the HyMARC team is providing excellent support (mainly theory/modeling and characterization) to multiple new seedling projects that are now part of the overall HyMARC effort.
  Hydrogen Storage Characterization Optimization Research Effort (HySCORE): The approach adopted by the HySCORE core team comprises two major elements: (1) development of improved validation measurements and protocols and advanced characterization techniques, and (2) development of improved hydrogen storage materials and creation of a more in-depth understanding of thermodynamics and kinetics operative during sorption reactions in candidate materials. The approach in part (1) is comprehensive and impressive, spanning a wide range of existing and new characterization techniques that have direct relevance to understanding the properties and behavior of hydrogen storage materials. The techniques directly complement the new in situ methods being applied in the companion HyMARC effort. The approach in part (2) focuses on adsorbent materials, mainly carbon-based and metal–organic frameworks (MOFs) and, to a lesser extent, reversible metal borohydrides. The work on development of models and fundamental understanding of thermodynamics and kinetics in those systems is very similar to the work under way in the companion HyMARC. The consolidation of the two consortia will undoubtedly result in more clearly defined roles and objectives and refinement of candidates selected for further study.
- HyMARC has established the analytical tools, models, and teaming methods to quickly increase the information base needed to identify solid-state materials and their behaviors that will be needed to enable

improved hydrogen storage so critically needed for the growth of hydrogen use in transportation. The HyMARC team is clearly going to add to those abilities with this consortium makeup and strategy.

- The overall approach is strong. The combination of theory, modeling, experiments, and characterization is appropriate for achieving HyMARC goals.
- The team is a newly combined HySCORE and HyMARC group. As such, overlapping objectives still exist. Before the next review cycle, it is recommended that the teams define a set of objectives that considers the overlap (as well as the status of the field).

Question 2: Accomplishments and progress

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

- HyMARC: Excellent progress was made in 2017–2018 in all core project areas and seedling support activities. Especially noteworthy was the integration of in situ, high-resolution characterization tools (e.g., X-ray photoelectron spectroscopy [XPS], X-ray absorption spectroscopy [XAS], transmission X-ray microscopy [TXM], scanning transmission X-ray microscopy [STXM], inelastic neutron scattering [INS], low-energy ion scattering [LEIS]) into the overall effort. This is providing entirely new and valuable insight in fundamental processes that is being used to directly validate and support the thermodynamic and kinetic models being developed. Likewise, understanding how emerging nanoscale encapsulation structures can be used to effectively confine hydride molecules and clusters is having an important impact on improving hydrogen-sorption kinetics. The results from the theory and modeling efforts are impressive and are providing useful guidance and support for the experimental efforts. The focus of experimental and theory efforts on understanding and improving the performance of metal borohydride systems is providing new insight and paying big dividends. This is a critical area for continued emphasis. The development of an improved understanding of how additives and catalysts alter sorption reaction kinetics and complex hydride systems remains a serious challenge. Greater attention to this important—and admittedly complicated and multifaceted issue—is needed.
  HySCORE: Solid accomplishments and progress toward DOE goals were made in 2017–2018. The extensive effort on development and application of characterization techniques is paying big dividends, and it is providing DOE with important resources that can be used in seedling work and by other collaborators. Impressive progress is being made on the challenging problem of multiple metal site incorporation in MOFs for enhanced hydrogen storage capacity. The work on C_2N and Ca-oxalate is producing intriguing results, but the ultimate payoff remains questionable. The team should seriously consider whether this work should be continued in the expanded consortium activity. The underlying causes of hydrogenation enhancement in boron- and nitrogen-doped carbon remain as outstanding research issues. It is not yet clear if enhancement is caused by actual chemical doping or by defect-mediated processes. Further work is needed to elucidate the detailed mechanism(s). The foundational work on hydrogen carriers conducted within HySCORE will be important in the consolidated consortium with HyMARC. The HySCORE team, especially at Pacific Northwest National Laboratory, provides important chemical insight that will definitely be needed in the future work.
- The accomplishments relative to DOE's goals are extensive throughout the consortium project.
- The accomplishments are being met, and the pace of progress since the last review is impressive.
- The goal to establish in operando probing is within reach but subject to availability of beam time at the facilities. It is an external factor but a predictable challenge. Currently, Li_3N and $LiNH_2$+LiH explored using STXM seem to provide promising results. In general, the STXM method is promising but needs further work to establish this as an established capability. Since not much of it is under the current team's control, some rethinking on making mesoscale imaging capabilities more available to a broader class of materials needs to be at the forefront of future planning.

Question 3: Collaboration and coordination

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

- HyMARC: Wide-ranging and fruitful collaborations have been established with other national laboratories and DOE Building Energy Sciences (BES) user facilities, universities in the United States and overseas, and seedling projects. These collaborations and interactions have greatly benefited the consortium and will undoubtedly be important in advancing this effort. Effective collaboration with HySCORE researchers is evident, and complementary research and development (R&D) activities are under way. A critical issue going forward is how to effectively manage and coordinate the multiple and diverse projects that will comprise the new HyMARC/HySCORE/seedling/new project enterprise. This is not an easy task. It will require an innovative management approach.
  HySCORE: The HySCORE core team has established extensive and valuable collaborations with other national laboratories and universities and research institutions in the United States and worldwide. These collaborations directly complement those in the current HyMARC activity, and they provide the foundation for an impressive and comprehensive capability in the consolidated consortium. The activities of the core team and project partners and collaborators are well coordinated and managed. The core team efforts are strongly leveraged by those collaborations, and they are greatly enhancing the progress being achieved in the overall project.
- The collaboration for this consortium and its sub-project is extensive and seems to be well coordinated. All key areas for the project's objectives are covered. The seedling projects enhance collaboration by providing a low-energy step to generate new tasks, collaborations, or approaches. It is also valuable to DOE's goals to have the vigorous collaborations with external research groups such as BES, University of Michigan, and non-U.S. organizations.
- One of the activities for the HyMARC team involves supporting the seedling projects. The mechanisms (i.e., meetings, workshops, etc.) by which seedlings interface with the HyMARC team members should be better articulated in the review documents and clearly spelled out for the seedling investigators.
- A broad set of partners was identified through seedling projects. The rest of the interactions with the broader community seem limited. Some of the relationships are only in discussion stages, such as hydrogen sponges with the University of California, Berkeley, so the list includes some members who are in the early, exploratory phase.

Question 4: Relevance/potential impact

This project was rated **3.6** for its relevance to/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan (MYRDD).

- HyMARC: HyMARC is the centerpiece of the current DOE hydrogen storage R&D effort. The reviewer acknowledges and thanks DOE for initiating and sustaining a broad-based consortium of this kind that focuses on a foundational understanding and on tools/capabilities for modeling and characterization. It has become increasingly apparent that a deeper understanding of fundamental processes operative during hydrogen sorption reactions in complex hydrides is needed to facilitate discovery of storage materials that meet the challenging DOE targets. This project provides a direct and focused approach to developing improved materials in a systematic, controlled, and rational way. The current project focuses keenly on critical storage problems and issues. The new expanded consortium (HyMARC–HySCORE) will provide even more powerful capabilities and potential synergies, but it may be encumbered by the attendant management difficulties that invariably accompany a large endeavor of its kind. Successfully addressing that problem will pose a significant challenge for both the DOE and HyMARC management teams.
  HySCORE: The project directly supports the goals of the Hydrogen and Fuel Cells Program. The new expanded activity (merger of HySCORE, HyMARC, and associated seedling projects) will provide DOE with a truly world-class R&D capability that will be able to effectively address the challenging search for a hydrogen storage material capable of meeting DOE system goals in a rational and effective way.
- The HyMARC team is undertaking a major part of DOE's hydrogen storage research activities. The team members are additionally interfacing with international partners. The impact and visibility, nationally and

internationally, of HyMARC are impressive. At the point where such material(s) are developed, cost-effective, and producible, the challenges that 700 bar tanks pose to vehicle design and delivery infrastructure will fade away, and the growth curves in hydrogen vehicle markets will steepen.

- Each sub-project within HyMARC is playing a role to increase the likelihood of timely development of breakthrough materials that can have a major impact on hydrogen storage for transportation.
- As a whole, some important milestones have been achieved. Some important publications made a strong case for the team and the seedling interactions.

Question 5: Proposed future work

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

- HyMARC: The future work follows directly from prior progress and results. Extending the knowledge gained on model media to more complex systems will be an important, if not vital, aspect of the future work. This will include the role of surface oxides, additives/catalysts, and phase formation—especially in metal borohydrides—and reaction kinetics and mechanisms in encapsulated systems. The addition of hydrogen carrier R&D (H2@Scale activity) to the expanded HyMARC–HySCORE consortium is important, but it will divert resources from a core team that is already heavily committed. Moreover, that effort may require expertise that is currently missing from the core team. Discussions with the Chemical Hydrogen Center of Excellence (COE) team are strongly recommended to develop a clear pathway to meeting hydrogen carrier goals.
  HySCORE: The future work will clearly evolve as discussions between the current HyMARC and HySCORE teams proceed. It will be crucial to avoid simply merging the existing efforts within the two consortia without careful consideration of overlapping or duplicative efforts, "stovepiping" issues, and the possible elimination of sub-projects that have limited potential going forward. Candid discussions are needed. This is undoubtedly recognized by the management of the core teams, as well as DOE, but it cannot be emphasized strongly enough. As stated in the HyMARC review, this consolidation provides a comprehensive, wide-ranging, and most impressive array of capabilities and resources for DOE. However, a careful and creative approach to managing the new activity and establishing the proper framework for effectively conducting the multitude of R&D sub-activities is needed. This is a major challenge that requires input from the entire team.
- Initiating the data management plan is important to ensure the availability of important experimental and theoretical data to researchers and developers.
- The planned future work is a logical extension from the already ambitious goals of prior HyMARC and HySCORE activities.
- The proposed future work listed in the slide is somewhat uninspiring. It does not connect the accomplishments and challenges well and propose a vision to take the activities to the next level of maturity and technical complexity. A better focus on technical challenges and building strong, cross-cutting teams for the Phase 2 proposal with some key forward-looking milestones will make HyMARC a truly unique program in the DOE complex.

Project strengths:

- HyMARC: This is a comprehensive and well-coordinated R&D effort that is successfully providing in-depth understanding of the thermodynamics and kinetics of hydrogen sorption reactions that is facilitating the development of candidate storage materials. The consortium is also providing extensive theory support and experimental resources to assist seedling R&D projects. Solid progress was made in theory, synthesis, and characterization tasks in 2017–2018. Extensive and beneficial collaborations with other national laboratories and universities are in place. The work has set the stage for an expanded, and even more comprehensive, consortium in 2018. A strong and very capable R&D team has been assembled to conduct the work on this project. With only a minor exception (lack of in-depth chemistry expertise—especially for hydrogen carrier R&D), the team has experience and background in the areas relevant to achieving the project goals and objectives.

HySCORE: There is a very strong and capable R&D team in place. The characterization and diagnostic efforts provide a unique and powerful capability. The work on MOFs is important, and assuming further progress, it offers an opportunity for sorbent materials to make a serious and positive contribution.

- The consortium has developed, and hopefully will continue to develop, valuable diagnostic tools and analytical techniques necessary to develop hydrogen storage materials critical for the use of hydrogen as a transportation fuel. The seedling projects are a great way to appropriately initiate new activities and/or collaborations and effectively enable new physical and intellectual resources for hydrogen storage objectives. The ability to model materials and interactions over a continuum of relevant length scales is a very significant accomplishment. It is important to understand the discontinuities from different methods as models transition between scale ranges.
- The project has made a visible impact on hydrogen storage. Numerous publications have resulted from the work. This project represents the overview and management of several research projects. As such, the management team's strengths are also evaluated here. The leadership from Mark Allendorf and Tom Gennett represents two national laboratories with experience in leading a HyMARC or HySCORE team. The leaders have clearly defined objectives, and the projects have major scientific outputs.
- Overall, both teams' efforts were a strong strength. The model development made strong progress. The advanced characterization capabilities and methods made new capabilities available to the team. The surface chemistry instrumentation part was not adequately discussed, but the directions seem promising. The new synthetic methods component had some interesting results that need further exploration for realizing their potential. Overall, this is a strong team of experts making progress in multiple areas.

Project weaknesses:

- The consortium has developed, and hopefully will continue to develop, valuable diagnostic tools and analytical techniques necessary to develop hydrogen storage materials critical to the use of hydrogen as a transportation fuel. The seedling projects are a great way to appropriately initiate new activities and/or collaborations and effectively enable new physical and intellectual resources for hydrogen storage objectives. The ability to model materials and interactions over a continuum of relevant length scales is a very significant accomplishment. It is important to understand the discontinuities from different methods as models transition between scale ranges.
- HyMARC: The question as to whether detailed knowledge and understanding gained from "simple" model systems can be extended straightforwardly to more complex materials remains largely unanswered. The team must avoid diverting resources to understanding problems in systems that may not necessarily be relevant to those encountered in more promising materials. This is a difficult and enigmatic issue that has existed since the inception of the consortium. Of course, it is important to validate the models using well-understood systems, but a thoughtful and creative approach is needed to rapidly extend the work to the most appropriate and relevant candidate systems. It seems that the resources and core personnel are spread thin over a wide range and diverse set of research problems. This was especially apparent in 2017 with the infusion of a large number of seedling projects requiring large amounts of time and operational support from the core team. This problem will be compounded by the addition of the HySCORE team to the consortium. Time and resources must be allocated carefully.

  HySCORE: Although good progress has been made on understanding hydrogen sorption reactions in the C_2N and Ca oxalate systems, the fact remains that these materials have limited potential (at best) to meet the storage goals. Likewise, it is not entirely clear whether they serve effectively as model systems. It is therefore questionable whether continued work is justified.
- This project represents the overview and management of several research projects. As such, the weaknesses of the management team are also evaluated here. Each leadership group has clearly defined objectives. However, these objectives are not yet seamlessly integrated from the prior HyMARC and HySCORE ones. There does not seem to be a clearly articulated methodology for interacting with the seedling projects. Although the interactions are indeed occurring, there does not appear to be a systematic way for the seedling projects to feed in to the HyMARC team, and vice versa. This should be established and clearly articulated across all parties, if it has not been already.
- A few areas of concern are common for a project of this size. These comments are primarily about the overall organization of domains and management. Obviously, the project has a complex task of managing

and chaperoning resources for a multi-institutional, multi-disciplinary team. The individual performers and groups are, in general, working well. The team may consider streamlining project management. For example, a strategy for seedling integration into the activities and identifying people with more availability of time or technical background may benefit the overall progress. The resource planning and execution calendars should be maintained. It is unclear whether this is already the case, because it was not highlighted in the presentations. It is not clear if the teams share their individual plans and adjust the different moving parts adequately. Issues with access to the beamlines and contingency plans for achieving the objectives if resources cannot be secured is one example where more effort could be appropriated for reducing the project's execution challenges and risks.

Recommendations for additions/deletions to project scope:

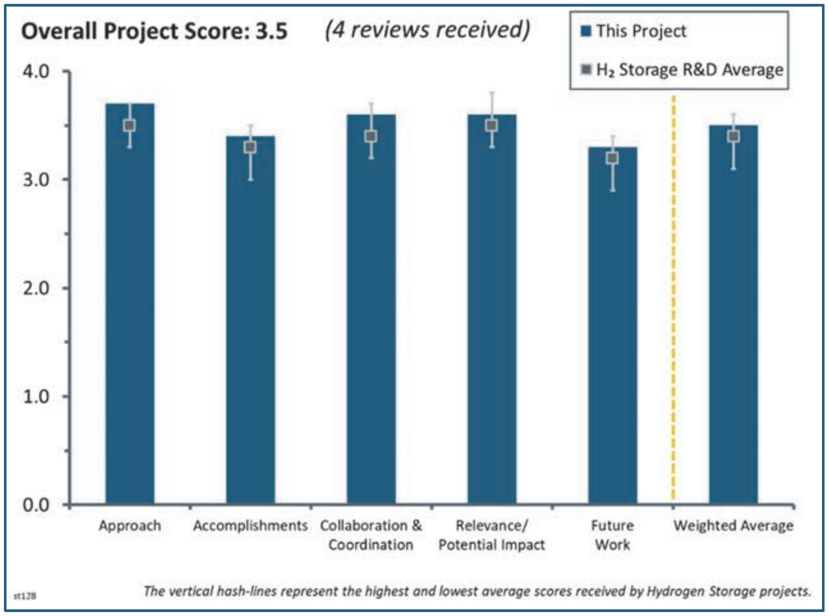
- HyMARC: Integration and coordination of complementary research efforts in the expanded HyMARC consortium will be a serious and challenging issue going forward. It will obviously require detailed planning by the management and DOE teams. "Stovepiping" and unneeded duplication of research efforts will undermine the positive aspects of a HyMARC–HySCORE merger. Although this problem is undoubtedly recognized by the HyMARC and HySCORE management team, a creative and dedicated approach to management of the expanded project will be needed to ensure success. Work on hydrogen carriers (part of the H2@Scale effort) is proposed for inclusion into the consolidated consortium. This places additional challenges on time and resources for the core team. It will be important to add additional chemistry expertise to the team (discussions with Chemical Hydrogen COE personnel are recommended for guidance and possible assistance).

  HySCORE:
    - *C_2N and Ca-oxalate*: Careful consideration concerning the efficacy and potential of the C_2N and Ca-oxalate work is needed. This should be done as part of the planning of the consolidated HyMARC effort.
    - *Hydrogen storage in MOFs*: Recent system analysis projections provided by the Argonne National Laboratory team (project ST-001) suggests that even with four hydrogen molecules per metal cation, the overall capacity in MOFs is still approximately two times lower than the DOE target. In view of this result, the HySCORE project team should conduct a candid assessment of its future work on MOF systems.

  Additional Comments for DOE and HyMARC/HySCORE management): Prior to the start of the HyMARC Phase 2 consolidated activity effort, it might make sense to convene a "Hydrogen Summit" with attendees from the consolidated HyMARC core team, DOE Hydrogen and Fuel Cells Program managers, and possibly some external experts to conduct a candid, frank, but informal assessment of the project's status and where it is headed. Some topics might include a discussion of impactful insights that have emerged in the last three years, what storage material candidates (if any) have risen to the top, which ones should be taken off the list, what dominant "foundational understanding" issues remain outstanding, what else DOE needs to know or do to move the bar, and what new resources and/or expertise are needed. That discussion and assessment might help the HyMARC management get on "the same page" in planning and implementing the Phase 2 effort.
- The project made progress. The overall recommendation is for a stronger and more streamlined management structure. This is crucial for improving the transparency of resource availability, planning, and progress by different members of the team. In general, phone calls and many such means are not adequate. Often, areas of concerns are not identified through such means. A serious attempt at handling the complexity of a large team, allocation of calendars, and tracking of progress needs to be modernized and made more seamless to improve productivity. This can help all team members have an aggregated view of the different activities and provide more opportunities for innovation and cross-cutting work across the board.

Project #ST-128: Hydrogen Materials—Advanced Research Consortium (HyMARC): Sandia National Laboratories Technical Activities

Mark Allendorf; Sandia National Laboratories



Brief Summary of Project:

This project addresses a lack of knowledge about hydrogen physisorption and chemisorption. Researchers will develop foundational understanding of phenomena governing the thermodynamics and kinetics of hydrogen release and uptake in all classes of hydrogen storage materials. Sandia National Laboratories (SNL) will (1) provide data required to develop and validate thermodynamic models of sorbents and metal hydrides, (2) identify the structure, composition, and reactivity of gas–surface and solid–solid hydride surfaces contributing to rate-limiting desorption and uptake, (3) synthesize metal hydrides and sorbents in a variety of formats and develop in situ techniques for their characterization, and (4) apply multiscale codes to discover new materials and new mechanisms of storing hydrogen.

Question 1: Approach to performing the work

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

- A systematic approach has been adopted to explore the energetics/thermodynamics, kinetics, and reversibility/cycling in sorbents and metal hydrides. The approach comprises theory/modeling, synthesis and processing, and detailed characterization methods to elucidate reaction mechanisms and probe the salient features of hydrogen sorption reactions in those materials. Collaborations with other Hydrogen Materials—Advanced Research Consortium (HyMARC) core team members and partners have contributed notably to the progress achieved in this reporting period. Overall, the approach is rational and is leading to progress that is having an impact on project goals and objectives.
- The research topics are well organized into a classification scheme, with the researchers responsible for each topic clearly listed and identified. This appears on slide 3, “Relevance and Impact.” The research strategies are clearly identified. The work is done in the area of sorbents (e.g., gate-opening metal–organic frameworks [MOFs]), borohydrides, and phase equilibria modeling.
- The approach is very well defined around energetics, kinetics, and reversibility. For energetic considerations, it is crucial that the nanostructuring/destabilization and tuning of enthalpy and entropic contributions be balanced against the challenges to reversibility. The kinetic considerations are identified, and challenges with optimizing all three of these areas are within the team’s capabilities.
- The project has a very good and comprehensive approach to understanding fundamental aspects of hydrogen storage materials. Most critical aspects are identified. In the examples, the project team could, and probably should, try to understand the materials with low melting temperatures that are currently the most promising.

Question 2: Accomplishments and progress

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

- The project team achieved solid accomplishments and progress in numerous important areas. The most notable results include establishing a validated phase diagram for $Mg(BH_4)_2$, which is providing a framework for understanding materials properties and transport processes; developing a new understanding of the role of surface oxide layers in the dehydrogenation of $NaAlH_4$; providing a deeper understanding of the rate-limiting step(s) for MgB_2 (bulk) dehydrogenation; developing an improved understanding of nucleation and growth events and evolution in Mg-B-H materials using sophisticated scanning transmission x-ray microscopy (STXM) diagnostics; and identifying a new high-pressure method for infiltrating $Mg(BH_4)_2$ into a nanostructured host, which is important for probing kinetics and thermodynamics changes that may accrue by incorporating the metal hydride in a nanostructured template. (Data on hydrogen sorption behavior in the latter area are apparently not available at this time—those results will be important for guiding future experiments.) Progress in all of these areas is noteworthy and is providing a firm basis for future work.
- The work the team has done on sorbent materials indicates nickel nanoparticles form from aggregation of nickel atoms from the metal sites in MOFs after 700 bar or post-1000 cycles. The high-pressure work shows melting of $Mg(BH_4)_2$ occurs at 355°C (at 1000 bar H_2). Amorphization, occurring first, is considered a strategy to thermodynamically tune the material. The Mg-B-H phase diagram has been validated (along with Wood, ST-129).
- The progress made toward technical goals and DOE targets is satisfactory. The accomplishments presented provided some very interesting insights. For example, the role of Ti in the dehydrogenation of $NaAlH_4$ was revisited. The Delmelle et al. paper from 2014 was interesting, and the work by the current team provided better supporting evidence on the role of oxide phases of Ti with a strong link to the mobility of oxide ions as known for other supported catalytic systems.
- The project team has achieved outstanding accomplishments, both experimentally and computationally. This project is a great step forward.

Question 3: Collaboration and coordination

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

- Extensive collaborations are evident. The fruitful collaborations with numerous investigators in the HySCORE program are especially noteworthy because they are providing a straightforward way to facilitate joint work that will be conducted in the consolidated HyMARC–HySCORE consortium. In addition, numerous collaborations with investigators at other national laboratories, universities, and research institutions are augmenting the work by the core SNL team. The project also supports several seedling projects, providing a mutually beneficial way to explore new topics and project areas. The critical challenge in the future will be to find the best ways to manage, organize, and coordinate the collaborative efforts among multiple institutions and projects within the much larger consolidated consortium.
- The SNL technical team has collaborative research with other HyMARC teams, including in computational modeling (with ST-129) and seedling projects.
- The project team has initiated extremely good internal (within HyMARC) and external collaborations.
- The collaboration is reasonable, exchanging samples with seedling projects at Argonne National Laboratory, University of Missouri–St. Louis, Liox Power/HRL Labs, National Renewable Energy Laboratory, etc. The team can benefit from more university connections to leverage the enthusiasm of graduate students and bring them into the community for training the future workforce in this domain.

Question 4: Relevance/potential impact

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

- The technical effort at SNL is an integral element of the overall HyMARC project and, as such, directly supports the goals and objectives of the Hydrogen and Fuel Cells Program (the Program). The principal investigator and the SNL team are conducting a research and development (R&D) activity that is closely aligned with the mission of the consortium and the DOE goals for the overall Program.
- The impact of the work is well documented. The characterization methods developed and results discussed have clarified some key issues. Overall, the work presented made strong progress in integrating capabilities and demonstrating results that are important for exploration of materials that have been known for a while. This is important proof that the new methods are better capable of providing detailed understanding or bringing the community closer to the ground truth. If there is any chance to increase either volumetric or gravimetric storage capacities of existing systems, it would be by using hydrogen storage materials such as this.
- So far, nobody understands why there is a gap of high-capacity hydrides with ambient working temperatures. Unfortunately, many important aspects in the reaction mechanisms of such materials are not understood. This project hints in the right direction. By advancing the understanding of these materials, this project lays the basis for the design and discovery of these sought-after materials with the right properties.
- The objectives are clearly laid out and are in line with objectives of the HyMARC program.

Question 5: Proposed future work

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

- The future work is ambitious and intends to employ machine learning to define structure–property relationships. In addition to theory work, experimental work that examines amorphization as a strategy for improving storage properties will be undertaken. The team also plans to make contributions to in situ measurements.
- There is an urgent need for the characterization and better understanding of materials based on complex hydrides with low melting temperatures that alone show high reversible hydrogen uptake and release rates at low temperatures.
- The proposed future efforts are a straightforward extension of the prior work. The future work should provide a reasonable framework to develop an improved understanding of sorption kinetics and mechanisms, especially in metal borohydrides. However, it is surprising that the remaining challenges and future work (slide 20) contains no mention of the role of oxide layers in the reactions and transport of hydrogen in metal borohydrides. Specifically, it is unclear whether the $NaAlH_4$-oxide results can be extended to other complex metal hydrides (especially $Mg(BH_4)_2$). Additionally, it is unclear what the optimum oxide thickness is and what impact the oxide has on reversibility, etc. The "oxide issue" is clearly a serious one because it affects the performance (either positive or negative) in hydrogen sorption reactions in virtually every metal hydride under conventional operating conditions. Also, the motivation for achieving "pure nanoparticles of MgB_2 and $Mg(BH_4)_2$" is not readily obvious, especially in view of the agglomeration/clustering problem that often accompanies sorption cycling. Likewise, an understanding of the size distribution of metal borohydride particles in an encapsulated state is important (also, whether the particles reside in an amorphous, crystalline, or sub-crystalline state). Finally, it would be good to know the capacity "overhead" imposed by the nanostructured hosts that are being planned for use for future studies. All of these issues are important, but they are not addressed adequately in the future work statement.
- The work identified is reasonable. The modeling, synthesis, and characterization work proposed continues the overall approach. The encapsulation approach proposed for synthesis will need more creativity in improving the uniformity of performance for large-volume storage. Therefore, adequate emphasis on such promising areas needs to be coupled to the other parts of the work. There are some minor areas of concern. The team needs to identify the value chain better by dividing the future work among priority areas and leave some for the collaborators and other academic research groups for handling research on things

that might be considered low-impact activities. For example, the characterization team still plans to explore thermal degradation of MOFs using X-ray and neutron diffraction. This seems to be a good candidate that can be handled by the larger MOF community and may not provide the return on investment for the hydrogen storage community until there are very good candidates that can meet DOE targets. Owing to the diversity of MOFs, this seems like opening a large area of research without a good sense of targets and solutions needed to make them more thermally stable.

Project strengths:

- A very strong and capable core R&D team, including external collaborators, has been assembled to conduct this project. The project addresses a large number of critical issues that affect the eventual discovery and development of storage materials that meet DOE targets. The project comprises theory/modeling, synthesis, and advanced characterization efforts within a highly collaborative framework. The project is well coordinated and managed. The expansion of the project to include the HySCORE team, plus seedling activities and associated collaborators, will provide a powerful capability, but the management and coordination challenges will be daunting.
- The project is making true progress in the quest for hydrogen storage materials that hold promise for high capacity and reversibility. Fundamental research (e.g., amorphization) is being tied to material performance (e.g., utilizing amorphous phases to tune materials thermodynamically). This is the sort of connection that is necessary to have breakthroughs in hydrides and sorbent materials.
- The strength of the approach and the team is quite evident. Some important results and publications came out of the work. The progress is satisfactory, and capabilities are getting well integrated into the workflow. The partnerships and leveraging activities are moderate.
- This project has a very comprehensive approach and, so far, impressive results.

Project weaknesses:

- The weaknesses are minor. The project needs to develop a list of priorities and provide a strong technical rationale for ranking of such priorities. It is often hard to identify the strategic vision behind a large set of activities handled under the project. Therefore, a stronger vision and targets need to be identified in Phase 2. Another area of importance is shifting the focus from identifying the challenge of making nanocrystalline materials to developing a stronger plan for predicting, testing, and observing their behavior during cycling. The team needs to focus effort on classifying mechanisms and predicting materials behavior. This will save costs and research expenditures. Additionally, the research team needs to include a pipeline for materials that are more likely to achieve targets into the thought process. It is not clear if part of the time is spent with partners and collaborators to identify new and promising systems. Overall, the team is fully capable of improving the prioritization and allocation of resource problems identified above. Therefore, some are identified to improve focus and efficiency in an otherwise very exciting set of projects and progress reported. Additionally, when pursuing amorphization as a strategy in materials that must be thermally cycled for performance, the team should give careful thought to the recrystallization of amorphous structures during service.
- The results on the involvement of the oxide layer on reaction kinetics in $NaAlH_4$ are certainly intriguing; however, a clear pathway to understanding the relevance or translation of those results to other metal hydrides (especially metal borohydrides) is not evident. To this reviewer’s knowledge, the fact remains that no experiments have been performed using pristine, oxide-free surfaces to establish a definitive baseline. There are challenges in conducting such experiments (ultra-high vacuum, in situ oxide removal/surface cleaning, detailed characterization during hydrogen exposure, etc.), but without that information, it is difficult to fully assess and understand the impact of the oxide on the reaction processes.
- The project team must give careful thought to whether amorphization plays a major role in thermodynamics (i.e., entropic terms), as the HyMARC team asserts, or whether amorphization more so affects kinetics (i.e., faster diffusion because of more grain boundaries and low electron density regions in the lattice).
- There is no focus on multinary composite systems with high capacities, low working temperatures, or high complexity.

Recommendations for additions/deletions to project scope:

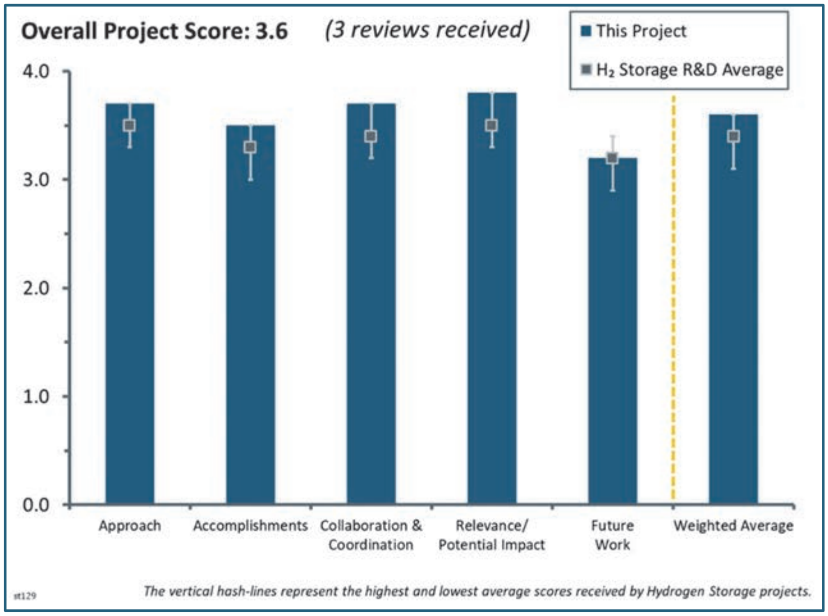
- Apart from addressing the issues raised in the "Future Work" and "Project Weaknesses" sections, by far the overarching issue for the HyMARC team will be how to coordinate and manage the consolidated consortium in a way that enhances inclusion and cooperation without duplication and "stovepiping" of efforts. Although this is a comment that is mainly relevant to the entire HyMARC effort, it nonetheless affects almost all aspects of the SNL work. This is an issue that the principal investigator and entire SNL team acknowledge and are actively engaged in solving, but the importance of focusing clearly and "getting it right" should be reinforced.
- Some more effort on mesoscale imaging and connections to mesoscale modeling needs to be allocated. For reduced scope, it is not clear that MOF characterization work is the best use of resources.
- The group should take into account the more complex systems of multinary composite systems with high capacities, low working temperatures, and high complexity.

Project #ST-129: Hydrogen Materials—Advanced Research Consortium (HyMARC): Lawrence Livermore National Laboratory Technical Activities

Brandon Wood; Lawrence Livermore National Laboratory

Brief Summary of Project:

The Hydrogen Materials—Advanced Research Consortium (HyMARC) is providing community tools and foundational understanding of phenomena governing thermodynamics and kinetics to enable development of solid-phase hydrogen storage materials. HyMARC team member Lawrence Livermore National Laboratory (LLNL) is conducting porous carbon synthesis; X-ray absorption/emission spectroscopy (XAS/XES); and multiscale modeling including density functional theory (DFT), ab initio molecular dynamics, phase-field mesoscale kinetic modeling, and kinetic and quantum Monte Carlo (QMC).



Question 1: Approach to performing the work

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

- The approach on this theory/modeling project addresses a wide range of critical phenomena and issues and obstacles that have an impact on our understanding of hydrogen sorption reactions in storage materials. Initial work on model systems is being extended to more promising (and complex) materials. As noted by the principal investigator, the focus is three-fold: (1) bridging scales via multiscale integration, (2) getting past model systems and moving to "real" materials, and (3) leveraging the interaction between experiments and theory. The approach is comprehensive, rational, and strongly coupled to the experimental work being conducted by the HyMARC core team and seedling project partners. This effort is a critical element of the entire HyMARC project. The project team is addressing important issues in an innovative and impressive way. Moreover, the team has been willing and able to make mid-course corrections to the approach and project focus as needed.
- The project's focus on theory and modeling to remove barriers to these areas for the particular case of hydrogen storage systems is an excellent one to tackle. As such, progress is being made in understanding the modeling inaccuracies to thermodynamic terms such as entropy. The Mg-B-H phase diagram has been developed. These efforts are poised to have lasting impacts on gas/solid models (even those outside of the scope of hydrogen storage).
- The LLNL team's approach is focused on the theory about handling the multiscale challenges involved in the hydrogen storage phenomena, coupled to validation activities. A multiscale scheme is proposed. The team may be well advised to realize that the traditional multiscale approach will meet a range of challenges in scale bridging. This review will not focus on the multiscale challenges when judging the approach. The main strength of the approach is in the atomic-scale modeling. The multiscaling part is somewhat unremarkable and will be hard to accomplish at the level of clarity provided by the atomic-scale results.

Question 2: Accomplishments and progress

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

- Solid progress has been made in numerous areas. The collaborations with seedling partners are especially noteworthy, and the results from those joint studies are leading to improved understanding of important phenomena and processes. Some especially notable accomplishments include:
  - Elucidating the decomposition mechanism of MgB_2; predicting the Mg-B-H phase diagram (a collaboration with Sandia National Laboratories [SNL] and Pacific Northwest Laboratory [PNNL] with a focus on a high-pressure regime).
  - Understanding and predicting changes in entropy due to surface anharmonicity from molecular rotations, and showing how a confining medium can "freeze" anharmonic rotations and destabilize the hydride.
  - Demonstrating that confinement stress dramatically affects thermodynamics and kinetics in nanoconfined metal hydrides.
  - Indicating that surface oxide facilitates the dehydrogenation of $NaAlH_4$—resulting in important implications for other complex hydrides.
  - Using simulated interactions of ethers and metal hydrides, showing that etherates destabilize the surface B and generate structural defects (collaboration with the University of Hawaii seedling project).
- The strong set of accomplishments presented demonstrate the excellent value the team is bringing to HyMARC. It is evident that multiscale schemes are always hard to match as a whole. The accomplishments are primarily atomic-scale in nature. The work on multiple systems demonstrates a strong capability in data-driven corrections of DFT enthalpy from previous computations and very good, plausible mechanistic explorations of MgB_2 and $NaAlH_4$ systems.
- The project is making excellent progress. The role of oxides at the surface of hydrides has been elucidated as important. New mechanisms such as B-B bond breaking for the step on adsorption of hydrogen in MgB_2-THF (etherates) have been put forth from theory and modeling leading to new (and effective) "design rules." Further experimental validation of the B-B bond-breaking mechanism is necessary.

Question 3: Collaboration and coordination

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

- Collaboration with seedling teams and with other HyMARC (and prior Hydrogen Storage Characterization Optimization Research Effort [HySCORE]) teams is visible and is a major strength in terms of validating this theory and computational work.
- Collaborative efforts are critical to the success of this project. The project team has done an excellent job of collaborating in an effective and timely way with experimentalists and other theorists within the HyMARC, HySCORE, and seedling projects. The collaborations have been valuable and are definitely leading to enhanced understanding and insight. The only concern is that the extensive collaborations could divert the time and resources of the project team and limit the ability to conduct work on the "core" problems of the consortium. That potential problem will be compounded as more sub-projects requiring theory/modeling resources are introduced into the consolidated HyMARC consortium.
- The team contributed by improving the overall intellectual quality of the discussions and results by providing strong predictive results on multiple systems. The phase field model from Michigan State University seems to be a good direction, but not much regarding the results was discussed. LLNL has an Arbitrary Lagrangian–Eulerian 3D (ALE3D) for large-length-scale work. It might be worth discussing some work with the ALE3D team. Idaho National Laboratory's Multiphysics Object Oriented Simulation Environment (MOOSE) framework has a very good phase-field module. It might be worth exploring such connections. Symmetry-adapted perturbation theory (SAPT) potential work is very useful and can potentially help with many other systems if it can be standardized and tested for thermodynamic accuracy.

Question 4: Relevance/potential impact

This project was rated **3.8** for its relevance to/potential impact on supporting and advancing progress toward the Hydrogen and Fuel Cells Program goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- The project is pushing forward on the boundaries of some computational areas that have not been worked out. For example, the anharmonic contribution to entropy improves model fits to experimental data. These effects are able to explain the role of nanoconfinement and amorphization of the lattice on hydrogen desorption and uptake kinetics.
- This project is a critical component of the HyMARC effort and directly supports the HyMARC core mission of providing foundational understanding of important thermodynamic and kinetic phenomena and processes in hydrogen storage reactions. The project is closely aligned with HyMARC goals and objectives and, as such, is closely aligned with the goals and objectives of the Hydrogen and Fuel Cells Program.
- The impact is strong for the domains where theoretical work was performed. It is hard to quantify whether the multiscale scheme proposed is successful as a whole. Much work is needed if HyMARC decides to invest in codes that can explore mesoscale chemistry. The atomic-scale work has definitely made a significant impact and will continue to do so.

Question 5: Proposed future work

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

- The proposed future work is a direct and sensible extension of prior investigations. There are many intriguing topics that are especially important. A few examples requiring more detailed study include:
    - Developing an improved understanding of the role of surface oxide layers in dehydrogenation reactions in complex hydrides. Specifically, it is important to discover whether the results obtained on $NaAlH_4$ are extendable to other metal hydrides, especially metal borohydrides. Another question is about the optimum oxide thickness needed to facilitate dehydrogenation and whether a sub-monolayer oxide is effective.
    - Gaining a better fundamental understanding of how catalysts and additives alter the hydrogen sorption reaction kinetics in complex metal hydrides.
    - Developing a better understanding of the effects of B- and N- doping in C-containing materials (in collaboration with PNNL). The change in isosteric heat has been ascribed to a defect-mediated process, not chemical doping. If correct, the nature of the defect and whether this is a general phenomenon in carbon materials would be interesting questions to explore. The addition of projects from the HySCORE effort (i.e., core and seedling projects) will undoubtedly have an impact on future work on this project. A careful prioritization of project needs and impact must be made to ensure that the most important issues and problems are addressed in the most effective and timely way.
- The proposed future work is in line with prior findings and is a logical extension. In fact, the prior findings have led to opportunities to explore new ideas (such as a study of amorphous materials). As another positive impact, much of the future work will hinge on interfacing with seedling projects.
- The future work proposed on sorbents includes the completion of calculations on hydrogen physisorption and the stability of functionalized covalent organic frameworks (with a seedling from the National Renewable Energy Laboratory), as well as the establishment of a "best practice" for DFT calculations of hydrogen physisorption on MOF-74, in collaboration with the Lawrence Berkeley National Laboratory and SNL. This seems reasonable; the addition of anharmonic effects on the thermodynamics of metal hydrides is promising and can include MOFs. The publication of the anharmonic free energy database for model complex hydrides is important. However, pressure/temperature effects still need some work. It is hoped that the team will be able to dedicate resources to bring these capabilities forward through the streamlining of workflows. In the chemistry of metal hydrides, hydride intermediates within the high-temperature ab initio molecular dynamics (AIMD) of $NaAlH_4$ and $Mg(BH_4)_2$, with and without Ti, are very complex, and development of reactive molecular dynamics approaches is needed. It was not clear whether AIMD or tight-binding density functional theory (TB-DFT) dynamics can achieve the timescales, even after using

large-scale computing resources. The objective of identifying and validating pathways for closo-borane formations from MgB_2 is interesting. It is not clear why this is a capability that is preventing the materials community from succeeding in achieving their proposed goals. More details on the LLNL multiscale modeling framework for hydriding kinetics is definitely of interest. Finally, comparing the nucleation model with scanning transmission x-ray microscopy (STXM) microstructures is hugely important. The hydrogen storage community may find some work currently pursued by the battery research community (e.g., the study of charged/expensed battery material imaging has made a lot of progress in the past five years). The additive work will require a stronger connection to multiscaling and traditional materials modeling efforts of phenomena at the grain boundaries. This part of the team may need some reinforcements. In standards and tools, some work in Mg-B-H and pairwise potential was indicated. In DFT work, the use of materials genome machinery, databases available through the efforts of the National Institute of Standards and Technology, and many other free (but still requiring full-time equivalent resources) will be able to aggregate the future work into an organized framework. It is recommended that the team discuss the overall progress and gaps with the materials project team, as well, to encourage others in the community to contribute to screening hydrogen storage materials using DFT. In other words, there are many options for expanding activities and attracting/leveraging funding from other sources.

Project strengths:

- This is an impressive project being conducted by a highly qualified and experienced team. Extensive collaborations have augmented the overall impact of the project. It is a critical element of the overall HyMARC effort and is providing critical foundational understanding and guidance for experimental work.
- The atomic-scale work and DFT calculations, in particular, are the main strengths of the effort. The progress made in improving predictive capabilities and correcting long-help errors in both experiments and theory provide great proof for the impact the project had in the previous performance period.
- Overall, the project is excellent/outstanding. The theory and modeling work done is an essential cornerstone to other HyMARC and seedling endeavors.

Project weaknesses:

- The theory and modeling team should continue to make every effort to experimentally validate the findings. Particularly, the B-B bond breaking by etherates, as a mechanism for the uptake of hydrogen by MgB_2 adducted with tetrahydrofuran (THF), would be validated by relevant spectroscopic techniques. Those techniques appear to be available within the HyMARC group.
- This is not necessarily a weakness but a comment/observation: It is not entirely clear how specific topics were selected for detailed study in this project. It would be useful to understand how projects are prioritized and selected. The main reason for raising this issue is that there is some concern that the project could become a victim of its own success—i.e., time and resources of the project team could be stretched so thin by multiple needs that the impact in any specific area becomes diluted. This will be especially important as the consortium grows with the addition of HySCORE and associated seedlings.
- The weakness of this project is in other length scales compared to atomic scale. The areas of reactive dynamics need more work. The phase-field and grain-boundary phenomena are not at the forefront of this project. Some weaknesses do not necessarily need to be corrected just because a multiscale scheme was proposed. Therefore, the team is encouraged to evaluate the plan and continue the good work.

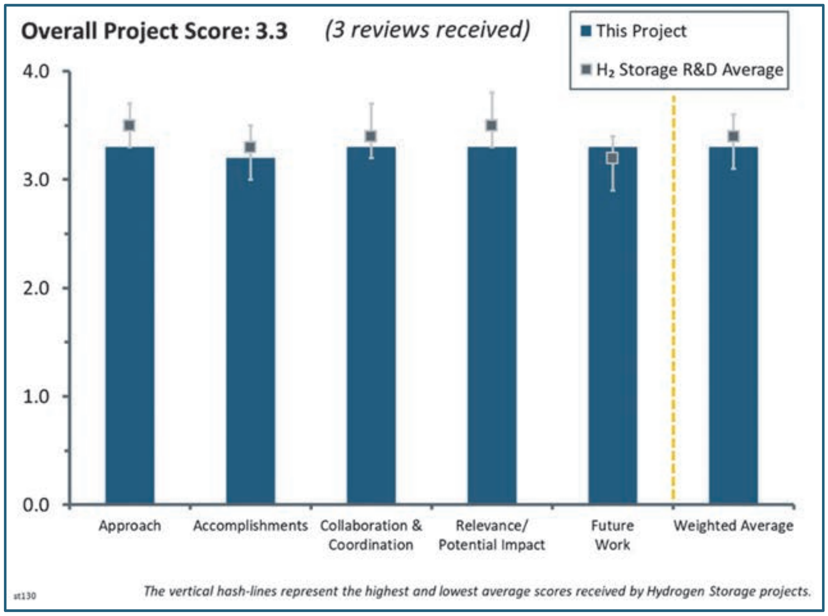
Recommendations for additions/deletions to project scope:

- It is recommended that the team evaluate progress of the multiscaling strategy and efforts. The addition of reactive dynamics in the mesoscale is recommended to match STXM and similar progress in experimental mesoscale domains. The effort with MgB_2 and other borohydrides seems less exciting compared to making progress with reactive dynamics at the grain level in mesoscale for helping with resolution of stress and reaction progress. For example, some of the contrasts in STXM imaging can be evaluated using DFT approaches developed for the scanning tunneling microscope (STM) in the past. Additional input from the phase-field models needs to be evaluated for the actual strength in predicting physically meaningful insights. Otherwise, much of the phase-field models suffers from unphysical assumptions regarding the phase-field parameters and a lack of mechanics in their models. Therefore, an objective analysis of the multiscale strategy will help to consolidate the progress from more predictive techniques such as DFT at an appropriate level.

- There are no specific recommendations for changes to the project scope. Thoughtful and candid discussions with the entire HyMARC team need to occur so that the most critical needs are addressed and the proper focus is achieved in this project going forward.

Project #ST-130: Hydrogen Materials—Advanced Research Consortium (HyMARC): Lawrence Berkeley National Laboratory Technical Activities

Jeffrey Urban; Lawrence Berkeley National Laboratory



Brief Summary of Project:

The Hydrogen Materials—Advanced Research Consortium (HyMARC) is providing community tools and foundational understanding of phenomena governing thermodynamics and kinetics to enable development of solid-phase hydrogen storage materials. Lawrence Berkeley National Laboratory (LBNL) will (1) focus on light materials and synthesis strategies with fine control of nanoscale dimensions to meet weight and volume requirements, (2) design interfaces with chemical specificity for control of hydrogen storage/sorption and selective transport, (3) explore storage concepts, (4) develop in situ/in operando soft X-ray characterization capabilities in combination with first principles simulations to extract details of functional materials and interfaces, and (5) refine chemical synthesis strategies based on atomic-/molecular-scale insight from characterization/theory.

Question 1: Approach to performing the work

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

- The project team has implemented an approach comprising theory/modeling, synthesis, and characterization of metal hydrides, sorbents, and hybrid nanoscale systems. The project facilitates access by HyMARC investigators to the extensive capabilities of the Molecular Foundry and Advanced Light Source (ALS) at LBNL. The approach is evolving to address important problems and obstacles to hydrogen sorption and reversibility in more promising storage material candidates.
- The LBNL research team is focused on both computational and modeling work and experimental work, including facilitating interactions with the X-ray spectroscopy team at the ALS. Both areas (computational and experimental) are functioning collaboratively with other groups in the HyMARC team. Also, the group is interacting with the seedling projects, particularly seedling projects on graphene-wrapped borohydrides and etherate–MgB_2 rehydrogenation.
- The approach is fundamentally strong, with a good connection between theory and experiments. The novelty of the approach and accuracy of the experiments are well suited for the problem.

Question 2: Accomplishments and progress

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

- The progress made to date is impressive. The research on the fundamental side has elucidated the phase formation (alpha, beta, or gamma) in graphene-wrapped nano-$MgBH_4$. Other progress has been made in the important issue of examining surface and bulk spectroscopy in a single sample via total electron yield

(TEY) and total fluorescence yield (TFY), respectively. Finally, the successful design and demonstration of an in situ X-ray absorption spectroscopy (XAS) flow cell (1 bar and 400°C) is a major accomplishment.

- The project team achieved progress in several areas. The most notable were controlling the $Mg(BH_4)_2$ phases in a reduced graphene oxide (rGO) host; incorporating a highly active hydrogen dissociation catalyst to functionalized graphite nanoribbons; developing in situ XAS for hydrogen sorption measurements; and modeling high-pressure hydrogen storage in metal–organic frameworks (MOFs), with results validated by experiments. Overall, compared to prior reporting periods, the technical effort in 2017–2018 was more suitably focused on materials that at least have potential to be viable storage candidates. However, it is not entirely clear what criteria the principal investigator and his team have used to select specific topics for study. The team presented very little information concerning the potential of the different materials to meet DOE goals or to serve as model systems that might inspire work on more relevant materials. Some clarification and elaboration would be helpful. Likewise, the presenter should state the actual impact and/or importance of the accomplishments listed in the slides. An experimental result in a review of this kind is useful only if it is shown to be meaningful or to have relevant impact on the understanding of an important issue.
- The accomplishments listed are well coordinated. The team made some important advancements in graphene nanoribbon (GNR) materials. Extending this to other hybrid materials is quite promising. The team also performed theory work with a great degree of detail and innovation. It is unclear why Grand Canonical Monte Carlo (GCMC) and Quantum Monte Carlo (QMC) indicated for MOFs show only GCMC. It is also not clear why QMC was performed (multiple hydrogens in QMC calculations can be interesting, as can high oxidation state metal centers) and what the finding was.

Question 3: Collaboration and coordination

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

- Collaborations are in place with investigators in the HyMARC and Hydrogen Storage Characterization and Optimization Research Effort (HySCORE) core teams and seedling projects. It is anticipated that collaborations will expand as the technical efforts in the new consolidated consortium are coordinated and come up to speed.
- As mentioned before, the LBNL team is collaborating with both other HyMARC teams and seedlings.
- Interactions with collaborators is somewhat minimal. The team can expand connections and increase the impact of both theory and experimental capabilities developed.

Question 4: Relevance/potential impact

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

- The project generally aligns with the goals and objectives of the Hydrogen and Fuel Cells Program. A significant aspect is that the project facilitates access to advanced diagnostic and synthetic capabilities at the LBNL ALS and Molecular Foundry. These capabilities are critically important for the overall HyMARC activity and will be of great benefit to the consolidated HyMARC–HySCORE consortium and associated seedling projects.
- The contributions of this team are important. They are responsible for the beamline programs and gaining access to beamlines at DOE laboratories and international facilities (when needed). This is a clear and unique contribution made by the team (in addition to the above-noted collaborative accomplishments).
- The project team made some major advances in this time period. The effort is headed in a strong technical domain with the addition of increasingly relevant systems. Thanks to strong mechanistic understanding, the team has the capability and sufficient experience to expand the effort and provide simpler diagnostic tools for scaled volumes of samples in conditions closer to the realistic storage environment. In particular, higher-pressure environments will need further maturation of the current XAS techniques. The target of extending to 1 bar may not achieve the proposed objective.

Question 5: Proposed future work

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

- The proposed future work follows from major accomplishments (including developing a high-pressure, 10 bar gas cell that operates at 600°C).
- The future work scope is quite large and includes some of the harder milestones. The team is capable of handling this but needs to clearly articulate a prioritization of possible areas listed.
- The future work extends the work conducted in 2017 and 2018. The future work plan is rational and consistent with the overall HyMARC goals. One question concerning the systematic MOF study (last bullet) is that it seems that several other studies have been done on the relation between structure and hydrogen adsorption in MOFs. It is unclear in what way the present study differs from the prior work and what new information is expected. Also, as stated later in this review, careful consideration of the capacity "overhead" imposed by encapsulation matrices should be considered and reported.

Project strengths:

- The project is well balanced and developed a well-integrated approach for synthesis, characterization, and theory/modeling of the selected systems. The reversibility of $Mg(BH_4)_2$/rGO was explored. The challenges are not surprising, but the approach is well grounded in good experimental approach. In situ XAS measurements at 1 bar of hydrogen detected MgH_2. The fundamental insights are important outcomes.
- A capable project team with extensive experience in materials synthesis, advanced characterization, and modeling has been assembled. The project provides direct access to unique characterization tools that are of great benefit to the overall HyMARC effort.
- The project has numerous publications and presentations. The research is making large contributions to the HyMARC team through discoveries (at LBNL) and in collaboration with HyMARC team members at other laboratories, as well as the seedling projects.

Project weaknesses:

- The overall weakness is lack of a systematic management of complexity and scale to address realistic pressure–temperature conditions under which hydrogen storage materials and systems will operate. The in situ work is promising, but the challenge will always be the high-pressure and high-temperature cells. The theory effort is relatively modest. A stronger integration of theory in the design of experiments is recommended. The remaining challenges are identified, but the future work is clearly aligned as a response to challenges. The strategic intent in the planning of future work needs to follow the objectives, and organization of theory efforts around some of the more intractable problems are needed; examples include more reliable screening of MOFs beyond GCMC, exploring innovative approaches to materials in rGO reversibility, and better control over synthesis and scale-up problems. As stated in the objective, the connection between theory and synthesis is important. This connection needs to be more direct. Also, the sharing of codes and data seems to have taken a back seat. Better management of ancillary tasks that are important for the community requires a stronger project management framework and assignment of efforts.
- It was very difficult to ascertain either from the slides or from the presentation what the real impact is of each accomplishment. For example, it is unclear why "pure phase control" in slide 8 is important. In slide 9, "Reversibility in…" could easily be re-stated as "Poor Reversibility in…" This is hardly a stellar example of reversibility. Also, in slides 11 through 15, it is not clear what important information is conveyed by the results of in situ and ex situ characterization. Likewise, in some of the "Accomplishment" slides (e.g., slides 8 and 14), the phrase "First example of …" or "First achievement of …" is used. A first demonstration of something may be important, but a statement about why it is important is far more useful. The project team is urged to clarify and augment future presentations with statements about the importance and impact of the results. An important addition to future presentations should be a statement of what volumetric and gravimetric capacities are expected from the new materials, assuming complete success. For example, in the $Mg(BH_4)_2$ nanoparticles wrapped by rGO, the gravimetric penalty imposed by the rGO host is unclear. Likewise, the overhead imposed by GNRphenIRCp*OH2 is unclear. If it is onerous, then it is time to rethink the encapsulation approach.

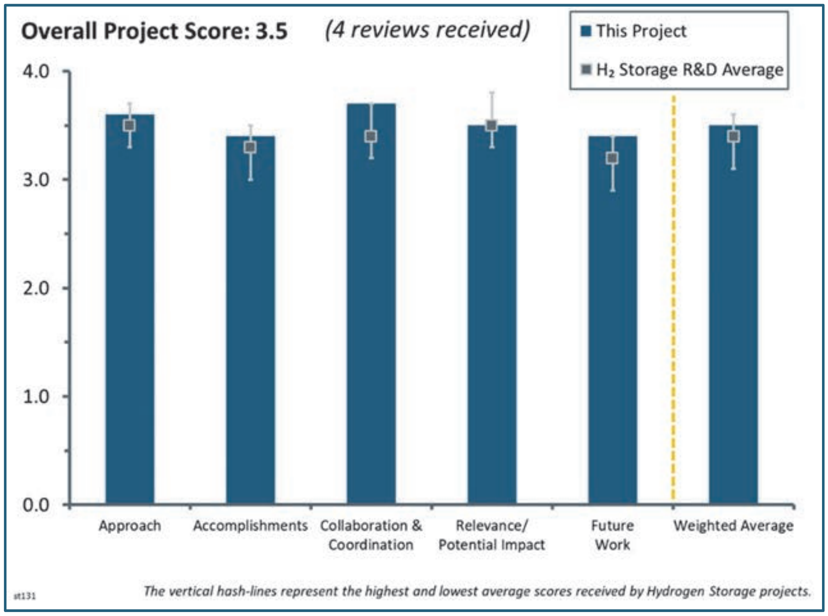
- There was no data management plan found in the materials submitted. There were two new patents mentioned, but there was no indication as to what the patent was for or what the patent number was (so that it could be easily looked up). Presumably, the in situ gas cells were patent opportunities; however, after reading the slides and listening to the presentations, it remains unclear whether this is the case.

Recommendations for additions/deletions to project scope:

- The team should give additional thought to experiments that probe the role of surface oxides on hydrogen sorption behavior in complex hydrides. Based on recent results from the HyMARC team, the oxide could be a blessing or a curse, depending upon the system being studied. This is clearly an important topic and is one that the LBNL team seems qualified to address in collaboration with other HyMARC investigators.
- The team should develop better mapping between challenges and future work proposed. The project has very well-defined objectives; however, not all these connections are explored with equal emphasis. The project team may need to carry out some rescoping to establish greater cross-cutting.

Project #ST-131: Hydrogen Materials—Advanced Research Consortium (HyMARC): National Renewable Energy Laboratory Technical Activities

Thomas Gennett; National Renewable Energy Laboratory



Brief Summary of Project:

This project represents a collaboration between national laboratories to investigate the properties of promising new hydrogen storage materials, and works in coordination with the Hydrogen Materials—Advanced Research Consortium (HyMARC) core team. The National Renewable Energy Laboratory (NREL) leads the collaboration, which includes Lawrence Berkeley National Laboratory, Pacific Northwest National Laboratory, and the National Institute of Standards and Technology. The objectives are to (1) develop new characterization capabilities such as nuclear magnetic resonance (NMR) spectroscopy, diffuse reflectance Fourier-transform infrared spectroscopy (DRIFTS), calorimetry, diffraction, and scattering, and (2) validate performance claims and theories critical to the design of new hydrogen storage materials.

Question 1: Approach to performing the work

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

- The approach comprises two distinct elements: (1) development and enhancement of characterization/ diagnostic capabilities and use of those methods for validation of hydrogen storage claims and concepts, and (2) rational design of selected hydrogen storage materials and advancing insight into thermodynamic and kinetic obstacles to achieving optimum storage performance. The first aspect builds on core capabilities, expertise, and experience at NREL and partner organizations. It provides the U.S. Department of Energy (DOE) with a unique set of capabilities; NREL has become DOE's "go to" organization for sample and concept validation and measurement protocol implementation. The second part of the approach is less compelling as a stand-alone NREL effort. In fact, it could easily be argued that the second aspect falls more naturally within the purview of the HyMARC activity. With the pending consolidation of the legacy HyMARC and Hydrogen Storage Characterization Optimization Research Effort (HySCORE) projects, that confusion will hopefully be resolved, and a candid assessment and evaluation of the NREL materials development effort will hopefully be a high-priority topic.
- The approach of the NREL team is to expand core capabilities and also to aid in materials development. One extremely valuable aspect of this effort is in the leading of round robin sample measurement in order to baseline measurement metrics for hydrogen storage. This effort should be extended to other areas for which variability of results in the literature makes the underlying materials phenomena intractable.
- This project has a very comprehensive approach. In particular, the analysis and results from the inter-laboratory comparison are very impressive and are a big step forward for the scientific community.
- This is well-organized work handling some of the major issues that have plagued the community during the hydrogen storage research done in the past decade.

Question 2: Accomplishments and progress

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

- Good progress was made on development and application of measurement and validation methods. Particularly noteworthy accomplishments include (1) completion of inter-laboratory comparisons of volumetric capacity and implementation of standard measurement protocols, (2) characterization of the importance of packing density (and measurement methods) on volumetric capacity, (3) development of advanced variable-temperature pressure-composition isotherm and thermal conductivity measurement apparatus and methods, and (4) extensive characterization and diagnostic support for several seedling projects. The materials work focused on characterizing the behavior of small pore materials (e.g., calcium [Ca] oxalate), two-dimensional C2N framework materials, and effects of boron (B) and nitrogen (N) doping on the isosteric heats of adsorption for hydrogen in carbon materials. Intriguing results were obtained in all areas. However, based upon the results obtained in prior years and the results reported in this review, it is not obvious that the Ca oxalate and C_2N work should be continued. In both cases, the future prospects for these materials as viable storage candidates are dim. Moreover, a compelling case has not been made concerning either a pathway to achieving higher performance or how these materials might serve as model systems that could inspire work in related materials. The N- and B-doping effects on heats of adsorption seem to be more complex than originally thought (i.e., a defect-mediated process rather than chemical doping may in fact be operative). Careful experiments that definitively test the defect mediation versus doping ideas and identify of the relevant defect type(s) need to be formulated.
- In addition to the round robin efforts and paper that disseminates findings of the round robin so that measurements are performed more consistently across many laboratories, other activities include (1) building a unique thermal conductivity apparatus, which will enable assessment of cracking and strain during hydride cycling, and (2) discovery of phonon effects for hydrogen uptake in sorbent materials.
- The development of new techniques to analyze the material properties are highly appreciated. The inter-laboratory comparison study, along with its analysis, is a big achievement of this project.
- The primary objectives were mostly accomplished. Support of HyMARC and seedling teams is commendable. This team is working well.

Question 3: Collaboration and coordination

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

- Extensive collaborations between the NREL investigators with the HySCORE and HyMARC core teams, other national laboratories, universities, private companies, and seedling projects are extending and advancing the impact of the NREL project. The most beneficial collaborations are focused on measurement and validation, protocol formulation, and new technique development. The collaborations are well coordinated, and there is close cooperation among all partner organizations.
- The team is collaboratively interacting with other HyMARC groups. Collaboration with seedling projects is not apparent (if it is ongoing). Given the massive collaborative effort required for completion of a round robin, this comment is less of a criticism and more of an observation.
- Project collaboration with other institutions is enormous.
- No deficiencies were identified in this project. The team is working well with others.

Question 4: Relevance/potential impact

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

- The technical activities at NREL are an important component of the overall DOE Hydrogen and Fuel Cells Program portfolio of projects. NREL has become the DOE focal point for measurement and validation of hydrogen storage material properties, comparison, and verification of results obtained at other laboratories, and development of definitive measurement protocols. The NREL materials work is a less critical adjunct to the characterization effort. The consolidation of the HySCORE and HyMARC activities should clarify and improve the nature of the NREL involvement in materials development, as well as efforts directed toward developing a foundational understanding of thermodynamics and kinetics in storage materials.

- This project is serving an important need. It is, in fact, helping HyMARC achieve some of its targets and goals.
- The project is aimed in the right direction. The final material to solve all problems has not been found yet. Nevertheless, methodologies and instruments developed in this project can be used to promote research on the most interesting materials.
- Unique and impactful areas are being pursued with the developments in this project.

Question 5: Proposed future work

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

- Work on kinetic control through manipulation of pores is of great importance, both for the direct storage of hydrogen and for the purification of hydrogen, and should be continued.
- The proposed future work follows directly from the results obtained in 2017 and 2018. At some point in the characterization/diagnostics effort (relatively near term), it seems that the focus will shift from technique development and formulation of measurement protocols to more routine application of the techniques to provide additional direct support for collaborating partners and seedling projects. Given the concerns about the viability of C_2N and Ca oxalate, either as candidate materials in their own right or as model systems for development of more suitable materials, the proposed future work on those materials is questionable. Thoughtful and candid discussions concerning future work, if any, on these systems should be a priority in the newly consolidated HyMARC project.
- The project's proposed future work builds upon past successes.
- The future plan has more emphasis on C_2N materials. It is not clear why any other core capability development or improvements in the efficiency of measurements or some of the outstanding challenges are not identified as future work.

Project strengths:

- The project is extremely meritorious. The leading of the round robin, as well as the development of unique capabilities (e.g., thermal conductivity apparatus) and fundamentally new ideas (e.g., phonon effects), results in meaningful contributions to the HyMARC group.
- The NREL team is very capable, with expertise and experience in all areas of the project. The characterization and diagnostic work is first-rate and is providing DOE with a vital resource for evaluating materials and storage processes.
- The project strength is in the service provided to different participants in making appropriate measurements and development of core capabilities. It has made a difference and has kept HyMARC from becoming a fundamental science effort.
- This is a very successful project. In particular, the outcome of the inter-laboratory comparison test and the conclusions drawn thereof are impressive.

Project weaknesses:

- The NREL materials development work, especially the C_2N and Ca oxalate efforts, are only marginally valuable. Without major advances, those materials are simply non-starters, serving neither as viable storage candidates themselves nor as model systems that might inspire work in related materials. A careful review of these projects should be performed within the consolidated HyMARC project, and a rational decision should be made concerning the path forward (if any) for these materials.
- The project plan and future scope seem to lack excitement and new ideas. The team can propose faster and more accurate measurements. The team identified better communication with theorists as a challenge but did not propose the future work to keep growing the activities to have an impact on the broader community.

- It would be desirable to gain a deeper understanding of the phenomena and even closer collaboration with simulation groups.

Recommendations for additions/deletions to project scope:

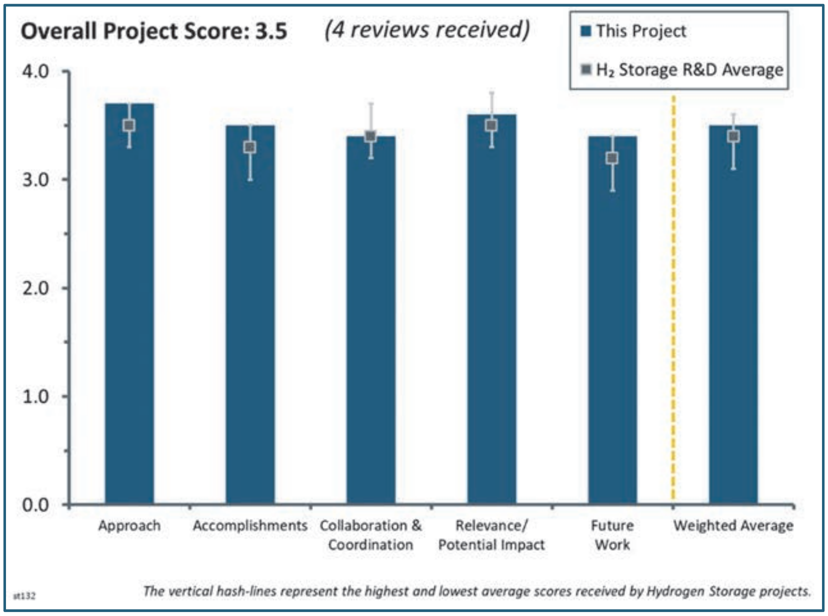
- Rethinking the scope with the identification of systems other than C_2N will be helpful. Many of the HyMARC team will need help with scale-up. Engagement of a broader community of researchers and creating a more open system for access and project data sharing can also be part of the work going forward.
- It is anticipated that the new HyMARC framework will provide NREL with a more natural home for more relevant materials work.
- Stronger collaboration with groups doing computer simulations, and especially molecular dynamics simulations, could be beneficial.

Project #ST-132: Hydrogen Materials—Advanced Research Consortium (HyMARC): Pacific Northwest National Laboratory Technical Activities

Tom Autrey; Pacific Northwest National Laboratory

Brief Summary of Project:

This project is part of a collaboration between national laboratories to develop new characterization capabilities to investigate the properties of promising new hydrogen storage materials. The project works in coordination with the Hydrogen Materials—Advanced Research Consortium (HyMARC) core team. Pacific Northwest National Laboratory (PNNL) will focus on nuclear magnetic resonance (NMR) spectroscopy and calorimetry to complement parallel efforts at other national laboratories. The project will also work toward validating claims and theories critical to the design of new hydrogen storage materials that show promise.



Question 1: Approach to performing the work

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

- The approach comprises both experimental and theoretical elements. The principal focus is on predicting and characterizing reactions in boron-doped carbon and $Mg(BH_4)_2$ using in situ, variable-temperature NMR to identify intermediates and products, as well as variable-pressure reaction calorimetry to measure hydrogen sorption enthalpies. The approach also uses theory to predict intermediates, products, and binding energies. The two aspects of experimental and theoretical elements are closely coupled, and the approach is being successfully employed to understand the increased binding energy of hydrogen in boron-doped carbon, as well as the enhanced reactivity of $Mg(BH_4)_2$ in the presence of Lewis base adducts. In addition, the PNNL team is exploring new ways to tune the thermodynamics in liquid-phase hydrogen carriers by altering the electron density. The overall approach is well focused on addressing important fundamental questions in these systems and optimizing the hydrogen sorption characteristics.
- The approach taken by PNNL is robust and poised to make significant contributions to the HyMARC teams (and seedling projects) by assisting materials developers with solid-state variable-temperature NMR and providing high-pressure and varied-pressure calorimetry.
- The project has demonstrated successful development, implementation, and a combination of advanced computational and experimental (in situ NMR) equipment to allow world-class research.
- This is a strong approach complementary to other work performed in HyMARC. The synthesis, NMR, and theory efforts are well suited for the systems under investigation.

Question 2: Accomplishments and progress

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

- Good progress has been made toward all project objectives in 2017 and 2018. Especially intriguing were the results obtained in the boron-doped carbon studies, where it was shown that the binding energy for hydrogen increases significantly in the boron-doped samples. Although the enhanced reversible physisorption capacities in heteroatom-substituted carbon scaffolds of heteroatoms have been demonstrated in prior work (e.g., Rice University, the National Renewable Energy Laboratory, the California Institute of Technology [Caltech], *Journal of the American Chemical Society* 132 [43] 2010), the present study provides new insight into the enhancement mechanism. Surprisingly, the action of the heteroatom is to facilitate the defect-mediated process that alters the hydrogen binding energies in unusual ways. Clearly, additional work is needed to identify the nature of the defect(s), find ways to controllably generate additional defects, and fully elucidate the mechanism. However, the results obtained thus far are provocative, and they could provide new insight into possibilities for tuning the hydrogen binding energy in carbon materials.
- The project's research has the following accomplishments:
  - It provides an understanding of shuttle boranes and their impact on kinetics for seedling teams.
  - It supports computational work with a post-doctoral researcher who examined boron-doped coronene.
  - It provides an understanding of the addition of tetrahydrofuran (THF) in lowering the melting temperature for $Mg(BH_4)_2$.
  - It provides an understanding of the formation of $Mg(B_3H_8)_2$ in support of seedling projects.
- There is an outstanding advancement in the understanding of hydrogen physisorption on doped carbon, the effect of THF and sorption properties of $Mg(BH_4)_2$, and hydrogen uptake and release from borohydrides, as well as the development of methods to predict hydrogenation enthalpies of liquid carriers.
- A good list of accomplishments was provided. The project team followed stated goals. The liquid-phase hydrogen carrier work is also headed in a positive direction.

Question 3: Collaboration and coordination

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

- Extensive collaborations are positively affecting all research and development (R&D) areas in this project. The scope of the technical effort is augmented significantly by the experience, expertise, and resources offered by those collaborations. The collaborative work is well coordinated and managed and is accelerating progress in all aspects of the project.
- The PNNL team has demonstrated collaborations with other HyMARC teams; however, there appears to be limited collaboration with seedling projects.
- The consortium collaborates with high-ranking and world-leading national and international research groups.
- The team collaborated well with other laboratories and peers.

Question 4: Relevance/potential impact

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

- This project is focused on issues that are directly aligned with the goals and objectives of the DOE Hydrogen and Fuel Cells Program. The importance of bringing additional chemistry expertise to the consolidated HyMARC/Hydrogen Storage Characterization Optimization Research Effort (HySCORE) consortium cannot be overstated. The PNNL team will undoubtedly provide the consortium with valuable chemistry expertise and insight that will be needed in the expanded program.

- The computational and experimental work has significant impacts on the HyMARC portfolio. The usefulness of the contributions of PNNL, as one of the core groups providing solid-state NMR capabilities, is clear.
- The impact of using NMR and theoretical approaches, along with standard analytical capabilities, has been the strength of the team. They have identified important fundamental questions and provided well-organized results.
- The team has developed and leveraged unique capabilities to assist materials developers.

Question 5: Proposed future work

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

- The proposed future work is a straightforward and sensible extension of prior work. It is hoped that the boron-doped carbon effort will include a more detailed investigation of the nature of the defect responsible for enhancing hydrogen sorption rates—it is unknown whether this is the same process operative in N- and P-doped carbon. This may involve the participation of other members of the consortium or seedling projects with expertise in solid-state defect generation and characterization. The future work on adduct-enhanced reactions in complex hydrides will, one hopes, include a description of the dependence of reaction kinetics on adduct concentration. This should provide useful information to aid in the elucidation of the enhancement mechanism. Overall, the proposed future work generally addresses important questions. This project will be a valuable addition to the consolidated HyMARC effort.
- Boron-doped carbon, complex hydrides, and hydrogen carriers are in the future scope. Good empirical ideas are backed by some good theory work, making this a strong effort in adding a systematic thought process to advancing state-of-the-art technology.
- The project maintains its future aim of understanding, predicting, and improving physisorption materials as well as hydrogen carriers. In the field of complex hydrides, the new field of liquid-phase complex hydrides will be investigated.
- The proposed future work builds on the past success. Including at least one other interaction with seedling projects would be useful for future work developments. There is nothing to add; the project is heading in the right direction.

Project strengths:

- The project is well organized and uses the strengths of the technique without trying too many experimental methods to answer the questions. It has a very meticulous approach, and the addition of theory made the fundamental connections strong. A lot of interesting mechanistic insights are involved in the hydrogen cycling in these materials. A good baseline understanding will be available from the team. A good collaboration strategy and links to other activities by peers are notable.
- The project addresses fundamental research, which aids in greatly enhancing the current understanding of hydrogen storage systems. For example, the researchers used 50% C-B-N and demonstrated that defects form and that hydrogen associates with these defects. Likewise, the project has addressed the formation of $B_{10}H_{10}$ phases. Another valuable contribution is the idea that the adducts act as a shuttle for boranes and impact kinetics (with seedling project collaborators).
- A strong R&D team using sophisticated diagnostic capabilities is conducting this project. Extensive collaborations are supporting the core effort. The team brings valuable chemistry expertise and insight to the consolidated HyMARC activity. The work on hydrogen carriers will be especially important as the overall hydrogen carrier initiative receives greater attention in the new consortium.
- Within the project, world-class instrumentation and capabilities are developed and successfully utilized to gain a better understanding of and improve hydrogen sorption properties of the most promising material classes of today.

Project weaknesses:

- There are no project weaknesses.
- No major weaknesses are apparent. A minor issue continues to be the same as the one pointed out last year concerning the Mg-borane–etherate work. It was suggested in the prior review that measurements of reaction rate as a function of ether concentration (especially at sub-stoichiometric levels) might provide important insight into the rate enhancement mechanism and the changes in reaction products. The team stated that it was important to do that work, but it is not apparent that the study has been done. A renewed effort to explore the etherate concentration dependence, and to determine how the results affect the understanding of the reaction mechanism, is recommended.
- The weakness is minor. The work proposed seems to be playing it safe and lacks the excitement of discovery. A little bit higher risk-taking and the inclusion of new ideas could improve the impact of the work and explore more uncharted territory. For example, the team is not utilizing the full strength of NMR. Much greater insights are available through 2D NMR and cross-polarization experiments coupled to theory. The theory work is mostly used for thermo-kinetics, and not in designing better experiments. Better integration of the parts will make the project more novel and impactful. These steps are within the team's expertise and experience.
- Perhaps additional interactions with (at least one) other seedling project(s) would prove beneficial.

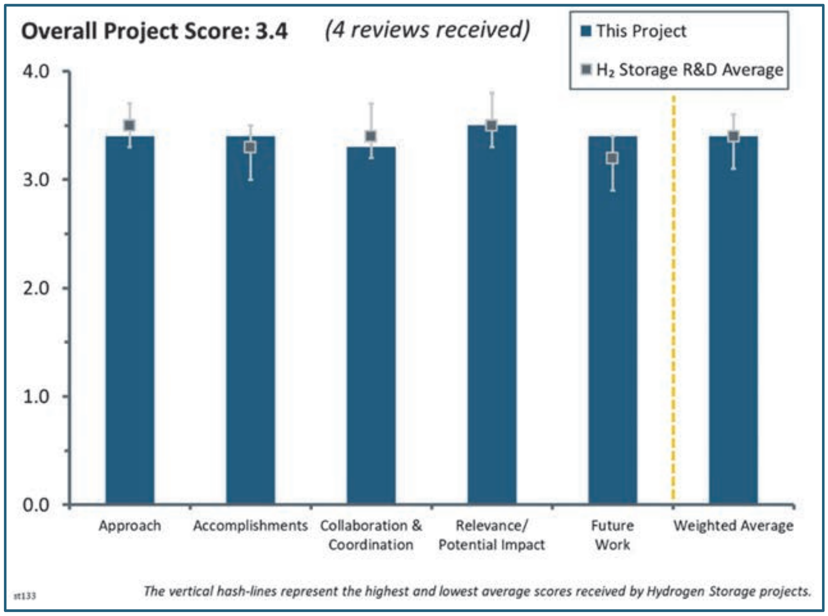
Recommendations for additions/deletions to project scope:

- This is a very good project. The team should go forward in combining excellent experimental and computational capabilities to help materials developers understand and improve their materials' performances.
- The scope is appropriate. No addition to or deletion from this project is requested, only better integration and higher-risk ideas that need to permeate across the board to improve the impact of the work and generate potential breakthroughs.
- The work on boron-doped carbon might be extended to include N- and P-doped materials. It is unclear whether the same defect-mediated processes are operative in those cases. Understanding the roles played by those "dopants" might allow a more general description and model to be formulated. Also (this may be a crazy idea), the team might consider ways to introduce active defects in a controlled way without the possible confusion arising from chemical doping (e.g., inert gas, carbon ion implantation, or something else entirely). That might help to clarify the mechanism.

Project #ST-133: Hydrogen Materials—Advanced Research Consortium (HyMARC): Lawrence Berkeley National Laboratory Technical Activities

Jeffrey Long; Lawrence Berkeley National Laboratory

Brief Summary of Project:



This project is part of a collaboration between national laboratories to develop new characterization capabilities to investigate the properties of promising new hydrogen storage materials. The project works in coordination with the Hydrogen Materials—Advanced Research Consortium (HyMARC) core team. Researchers will also validate new concepts for hydrogen storage mechanisms in adsorbents and provide accurate computational modeling for hydrogen adsorbed in porous materials. Specifically, Lawrence Berkeley National Laboratory (LBNL) is developing in situ infrared (IR) spectroscopy as a tool for characterizing emerging hydrogen storage materials, as well as metal–organic framework (MOF) materials that will allow for more than one hydrogen molecule per open metal site, which will increase hydrogen capacities for sorbent materials.

Question 1: Approach to performing the work

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

- The principal investigator (PI) and his team have formulated a coherent, rational, and creative approach to addressing the formidable barriers to producing MOFs that adsorb hydrogen at high capacities (multiple hydrogen binding sites) and with enthalpies in the optimal range for hydrogen storage applications. Building upon the successes obtained in prior work (most notably, the synthesis of an MOF with two hydrogen molecules per metal cation and record-high adsorbent uptake at ambient temperature and 100 bar pressure), the approach in 2017–2018 involved the search for an MOF system capable of binding more than two hydrogen molecules per cation site and a hydrogen adsorption enthalpy in the optimal range (-15 to -25 kJ/mol) for hydrogen storage applications.
- The LBNL technical approach is meritorious. The goal is to double storage capacity at 100 bar fill pressure in sorbent materials by tuning binding energies and adding more than one hydrogen atom per metal site. Another feature of the work is the development of in situ IR as a major tool (which will also be available to the seedling projects and other HyMARC laboratories).
- The project has a very well organized objective and approach. The polarization approach with metal cations increases binding energy. A new adsorbant design can help with incremental improvements. The in situ IR is a valuable tool that can operate in an important pressure–temperature range. The experimental results are well connected to the Co/Ni systems. The project sets the enthalpy target and provides good mechanistic insight.
- This is an interesting approach. However, not only is the uptake per liter important (page 5), but so is weight. Ni is a heavy element. Therefore, it is questionable whether the gravimetric storage capacity would be acceptable for such a system.

Question 2: Accomplishments and progress

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

- Impressive and noteworthy results were obtained on synthesis of a promising MOF–metal cation structure capable of adsorbing hydrogen with optimal enthalpy, as well as on identifying and synthesizing promising structures capable of binding more than two hydrogen molecules per metal site. Specifically, the team was able to synthesize and perform structure determination on the first MOF with open V^{2+} sites. The high-surface-area MOF has an adsorption enthalpy in the optimal range. The team also synthesized other structures (e.g., Fe- and Co-CPF-5) with the potential to bind three hydrogen molecules per metal site. In addition, in ongoing theory work, density functionals to model hydrogen adsorption and storage capacities were developed. Results in these areas are advancing state-of-the-art physisorption materials for hydrogen storage; the project is on pace to meet the overall project objectives.
- The project's accomplishments are listed and are in line with the proposed activities and milestones. The V^{2+} MOF with reasonable binding energy is quite promising. The theory seems to be lagging in providing insights on diffuse reflectance infrared fourier transform spectroscopy (DRIFTS) and other observables.
- The development of the DRIFTS instrument is a good achievement. Successful synthesis of V-MOF is very good. Demonstrating two MOFs with high adsorption enthalpy values is a very important result and should be validated.
- The in situ IR data are unique and valuable. The potential found in this approach is clearly demonstrated.

Question 3: Collaboration and coordination

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

- Collaborations with researchers in the Hydrogen Storage Characterization Optimization Research Effort (HySCORE) and HyMARC are enhancing the core project. Most notable support from collaborations is evident in materials and process characterization and theory. The collaborative efforts are well coordinated with the core project work and are facilitating rapid progress in all areas of the project.
- Unique tools are being developed and will be made available for other HyMARC teams and seedling project researchers.
- The team worked with other laboratories and partners.
- The number of additional collaborations should still be increased. Important findings of high adsorption enthalpy should be confirmed experimentally by other research groups outside the project team, as well.

Question 4: Relevance/potential impact

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

- The project directly supports the goals and objectives of the DOE Hydrogen and Fuel Cells Program (the Program). The project is closely aligned with Program needs and is an important element of the overall HySCORE/HyMARC research and development (R&D) activity.
- The results are promising. Some of the open questions need to be answered, and the theory effort needs to approach the high-pressure hydrogenation question before the impact of the work can be fully realized. The doubling of hydrogen storage capacity will be impactful and will bring MOFs back into serious consideration. Nevertheless, the current results are exciting and will provide fundamental insights for new types of MOFs. The metalation and metal exchange effort also shows strong progress. However, some key challenges remain. The theory effort was successful in benchmarking density functional theory (DFT) functionals. The identification of B97-D3 with Becke-Johnson's approximation is a nice result. It will save time that would otherwise be spent exploring more expensive methods.
- The IR instrument has potential to have major impacts. The discovery phase of high-surface-area sorbent materials with two hydrogens per metal site is also promising.

- This project fits with the Program's goals and objectives.

Question 5: Proposed future work

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

- This project is scheduled to end on September 30, 2018. The future work is clearly stated and builds upon the solid results obtained earlier in the project. The success demonstrated in prior work inspires confidence that the future work will also be successful and impactful. Finding conditions that promote complete activation of the V-MOF, and the successful search for frameworks with sites capable of multiple hydrogen binding, are critical aspects that will require special emphasis. In addition, a careful examination of the potential of MOF-based systems to meet overall DOE objectives needs to be conducted.
- The plans are consistent with previous work. The utilization of lattice Boltzmann simulations to model the storage performances is an important development that should receive focus.
- The planned work is excellent and a good continuation of the effort in more exciting directions. Theory for multiple hydrogen adsorption and polarization effects will be important. The group seems to ignore dynamics of MOFs and their changes with temperature. Overall, the project is moving into a more exciting phase.
- The proposed future work is well in line with prior discoveries.

Project strengths:

- The PI and the core project team are working at the forefront of research on advanced physisorption materials for hydrogen storage. The LBNL core team and collaborators within HySCORE and HyMARC, as well as associated universities, have experience and expertise in all relevant areas of the synthesis, characterization, hydrogen sorption measurements, and theory needed for this ambitious project. A coherent and well-formulated approach is in place, and solid results in all areas have been obtained. This project is a vital element of the HySCORE/HyMARC activity and is an important component of the overall Program R&D portfolio.
- The overall strength is in the very well-defined objectives and approach. This project has strong fundamentals and a good synergy with theory. The new MOFs and DRIFTS results are encouraging. The synthesis effort is also working well to make good candidates.
- The project provides two clear contributions: in situ IR and the discovery of MOFs that contain more than one hydrogen per metal site.
- The combined experimental–computational approach is praiseworthy.

Project weaknesses:

- In the Argonne National Laboratory systems analysis project presentation (ST-001, Ahluwalia), the PI showed (on slide 14) that even in the most optimistic case of four hydrogen molecules per metal cation, the theoretical uptake of hydrogen (at 25°C, 100 bar) is still a factor of two lower than the value needed to reach the system target. Even though excellent progress has been made in the present project, it seems the Ahluwalia predictions may suggest that the MOF approach is essentially a non-starter. A candid and thoughtful assessment needs to be made by the PI and consolidated HyMARC team.
- The extraction of thermodynamic variables from the in situ IR data could be a stretch. Particularly, the data on slide 12 of the presentation show that enthalpy and entropy appear to vary widely with the selected temperature range. Careful baselining work (on systems with known thermodynamics) should be undertaken to establish the use of IR intensity to extract thermodynamic data.
- The theory effort has not supported the experimental work closely. Specifically, much of the work on predicting candidates and variations could use a more high-throughput DFT using the reasonable performance of DFT-D2(BJ) functional. Also, it remains to be seen whether this functional has such a good performance across the board. More effort needs to be devoted to multiple hydrogen molecules and understanding bonding, polarization, and dynamics. More complex calculations of phonons, the effects of anharmonicity, and the predictions of the spectra could be helpful in the future to build the capabilities in a synchronous fashion.

- The group should also aim for a critical evaluation of the potential of characterized and developed MOFs. Important findings should be confirmed by external groups.

Recommendations for additions/deletions to project scope:

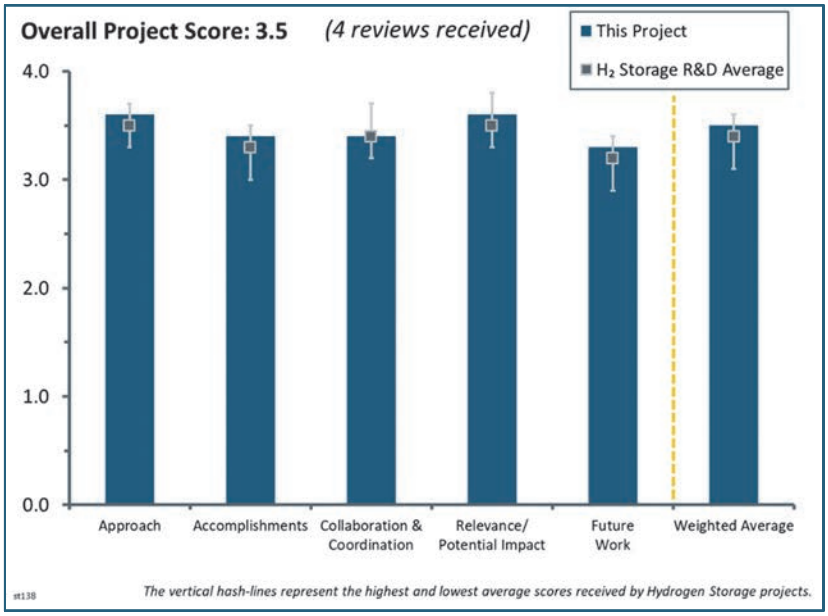
- The scope is well defined. The addition of ab initio molecular dynamics could be instructive in how to stabilize these MOFs in higher-temperature/-pressure conditions and with multiple hydrogens bound to a single site.
- A critical analysis of the materials under investigation for their potential to fulfill the DOE targets and goals should be part of the project. High adsorption enthalpy values should be confirmed by external groups.
- The project is scheduled to conclude on September 30, 2018. There are no recommendations for changes in the project scope for the remainder of the activity.

Project #ST-138: Hydrogen Materials—Advanced Research Consortium (HyMARC) Seedling: Development of Magnesium Boride Etherates as Hydrogen Storage Materials

Godwin Severa; University of Hawaii

Brief Summary of Project:

The objective of this project is to synthesize and characterize magnesium boride (MgB_2) etherate hydrogen storage materials that are capable of meeting the U.S. Department of Energy's performance targets. The project will synthesize MgB_2 etherates using ball milling and heat treatment techniques, study hydrogenation of the materials using variable pressure and time, study and optimize hydrogen cycling of the materials, and develop theoretical models.



Question 1: Approach to performing the work

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

- This project is innovative and important. It demonstrates that the hydrogenation of MgB_2 is enhanced significantly by the presence of etherate adducts. The approach in fiscal year 2017 and 2018 extends the initial study to include the synthesis of MgB_2 etherates by ball milling and heating from the reaction of MgB_2 with ethers and other additives, the comprehensive characterization of the MgB_2–ether composite, and the demonstration of dramatically improved hydrogen uptake in the modified MgB_2 materials. In addition, in collaboration with the Hydrogen Materials Advanced Research Consortium (HyMARC), at the Lawrence Livermore National Laboratory (LLNL), ab initio molecular dynamics (MD) simulations were used to identify how the coordinating species perturb the MgB_2 structure and lead to enhanced hydrogenation rates. The overall approach is straightforward and is keenly focused on overcoming the kinetic and thermodynamic barriers for hydrogen sorption reactions that exist in this promising material.
- This is an interesting and innovative project. The $Mg(BH_4)_2$ ammoniates and etherates are promising systems with kinetic challenges. The research is driven by the hypothesis that MgB_2 is destabilized by the ether coordination. The team attempted to lower the MgB_2 hydrogenation pressure from 900 bar to 700 bar. The project is in the initial stages. The plan is well organized and commensurate with the team's capabilities and background.
- The approach is clearly defined: to examine etherate–MgB_2 for hydrogen uptake and to extend the mechanism to other modified MgB_2 materials.
- This project has very good results. However, the check to see whether at least some reversibility is given should have been done at the beginning. Long-term cycling is scheduled for the end of the project—there is no doubt, but at least two cycles should have been tried already, since the start date was October 2016.

Question 2: Accomplishments and progress

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

- Good progress was made in several areas. Most notable was the observation that the kinetics for hydrogenation were greatly improved in the MgB_2–tetrahydrofuran (THF) composite system compared to bulk MgB_2 (hydrogen uptake at 300°C and 700 bar versus 400°C and 900 bar). Moreover, it was shown that the effect was not limited to etherates. Proprietary work has shown that other additives also produce "THF-like" effects. The MD simulations suggest that the additives may destabilize the surface boron structure, thereby creating structural defects that could facilitate hydrogenation. As pointed out in the DOE Hydrogen and Fuel Cells Program (the Program) Annual Merit Review last year, it would be helpful to experimentally determine the dependence of the rate enhancement on the concentration of the etherate (and/or proprietary additive), especially at sub-stoichiometric concentrations. Those findings could have important implications on the mechanism for enhanced hydrogen uptake.
- The progress made so far is reasonable. The project team claimed to have overcome a major barrier. This data was not shared, nor were the caveats presented. Overall, the key hurdles are next in the plan for Year 2. The progress is reasonable or very good, depending on the high-pressure hydrogenation results.
- This project has excellent results and achievements. It would have been outstanding if the project team could have shown that the effect of the X or THF is maintained after the first cycle without new ball milling.
- The interactions with other HyMARC projects are clear (both computational and experimental).

Question 3: Collaboration and coordination

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

- Collaboration with the LLNL theory group was important for providing an initial understanding of the structural changes that could occur in MgB_2 in the presence of etherate adducts or other additives. Likewise, the collaborations with HyMARC/Sandia National Laboratories and the National Renewable Energy Laboratory and Pacific Northwest National Laboratory on high-pressure hydrogenation and the characterization of reaction intermediates and products, as well as the identification of desorbed gas species, have significantly accelerated project progress.
- It is via collaboration (computationally) that other materials that imitate the behavior of the etherate adducts for MgB_2 can be evaluated based on their enhanced hydrogen uptake. X-ray absorption spectroscopy (XAS) will be tremendously useful in validating the suggested mechanism of B-B bond-breaking.
- Collaboration should be easy for the project team because of the members' well-known background in this domain. Some collaboration on theory and characterization was reported. This seems reasonable for the current stage of the project.
- This project consists of very good collaborations; the LLNL collaboration concerning the MD simulations is especially straightforward. Unlike the other projects, there is no mention of international collaborations.

Question 4: Relevance/potential impact

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

- The $MgB_2/Mg(BH_4)_2$ system is one of the most promising candidates for a practical hydrogen storage system. However, significant barriers to kinetics/rates and reversibility/cycling issues have made the early adoption of this material especially problematic. Through the creative use of additives and adducts to modify reaction rates and pathways, this project is improving the prospects for the Mg–borohydride system in practice storage applications. Overall, this project is well aligned with Program goals and objectives.

- Solution variants of this problem have been explored. However, for solid-state conditions, the role of sub-stoichiometric presence of ethers could open up new possibilities for improving kinetics or lowering hydrogenation pressure. This project could be very impactful.
- Borohydrides are one of the most promising classes of materials for hydrogen storage. Reversibility remains an issue. The pursuit of the adduct–MgB_2 to understand how to drive the reverse reaction is a clever approach and is very impactful.
- If successful, the project could have an enormous impact on hydrogen storage materials development and technologies.

Question 5: Proposed future work

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

- The future work capitalizes on past successes and is reasonable.
- The proposed future work is reasonable and on track.
- The future work follows naturally and directly from previous results and the research and development directions established in prior work on the project. However, the statements concerning specific areas are very general/vague, for example, "synthesis of modified Mg-boride materials"; "optimize MgB_2-X-THF system"; in terms of computations: "effect of additives on hydrogenation." Elaboration and clarification of what will actually be done in each of these areas would have been helpful and would inspire confidence that the future work has been carefully formulated. In addition, as pointed out last year, measurements showing the dependence of the hydrogenation rates on the additive concentration (especially sub-stoichiometric concentrations) are necessary. This information could provide important insight into the overall mechanism.
- The first hydrogen cycling experiments are urgently needed to evaluate the real potential of project outcome. This is currently the most important aspect and should be performed even before the long-term cycling experiments.

Project strengths:

- This is an innovative and novel project that addresses a critical DOE need. The core team at the University of Hawaii and collaborators in the HyMARC consortium bring considerable expertise, experience, and resources to the challenging problems addressed in this work. More generally, the principal investigator and his coworkers are providing the HyMARC team with much-needed chemistry expertise. This will be increasingly important as the technical effort in the consolidated HyMARC consortium expands into new topical areas requiring more extensive chemistry knowledge and intuition.
- The project's goals are noteworthy. The fact that progress is being made in understanding the mechanisms for hydrogen uptake by the etherate–MgB_2, and that these ideas are being used to optimize and develop new material systems, means that the overall project is moving the bar forward for hydrogen storage materials.
- The project is hypothesis-driven and innovative. The team is engaged in exploring modeling for explanations and other experimental capabilities to strengthen the mechanistic insights. This is definitely a good seedling project and supports innovation in storage research.
- This project utilizes a new approach to alter the thermodynamic properties of complex metal hydrides, which is very good.

Project weaknesses:

- The weakness of this project would be minimal if the lowering of pressure with reasonable hydrogenation kinetics was demonstrated. Some further characterization could be helpful, such as engaging in mesoscale imaging (a HyMARC capability) to observe partially hydrogenated MgB_2 grains. It will also be important to map the morphology in the starting materials and through different stages of cycling. This will be explored later in the year and hopefully will be clearer in next year's review.
- Although the modified MgB_2 system has been vastly improved, a wide gap remains between the current state of the art and the DOE targets. It is not obvious that incremental improvements (e.g., different

additives/catalysts, more detailed characterization) will be effective in bridging that gap. At some point in the not-too-distant future, a candid and thoughtful assessment of the prospects for this system vis-a-vis DOE targets will need to be made.
- The whole project currently hangs on the possibility of cycling.

Recommendations for additions/deletions to project scope:

- Changes to the scope are not necessary, since the project is making progress and is close to an important milestone.
- A study demonstrating the dependence of the hydrogenation rate on etherate (and/or other additive) concentration (especially at sub-stoichiometric levels) is necessary.
- Cycling of the materials should now be the top priority.