Hydrogen Technologies – 2021

Subprogram Overview

INTRODUCTION

The Hydrogen Technologies subprogram focuses on research, development, and demonstration (RD&D) to reduce the cost and improve the reliability of technologies used to produce, deliver, and store hydrogen from diverse domestic feedstocks and energy resources. In support of RD&D needs identified through the U.S. Department of Energy's (DOE's) H2@Scale efforts, the Hydrogen Technologies subprogram is developing a set of hydrogen production, delivery, and storage technology pathways. The subprogram addresses technical challenges through a portfolio of projects in three RD&D categories:

- **Hydrogen Production** addresses low-cost, highly efficient hydrogen production technologies that use diverse domestic sources of energy. RD&D activities include advanced water splitting and innovative concepts such as biological hydrogen production. The former is predominantly coordinated through the HydroGEN Advanced Water Splitting Materials consortium (HydroGEN) and the Hydrogen from Next-generation Electrolysis of Water consortium (H2NEW) to accelerate RD&D of advanced water-splitting technologies for clean, sustainable hydrogen production.
- **Hydrogen Infrastructure** addresses low-cost, high-efficiency technologies to move hydrogen from the point of production to the point of use. RD&D activities investigate liquefaction, pipelines, chemical carriers, and tube trailers to transport hydrogen over long distances, as well as compressors, pumps, dispensers, and stationary storage to support the development of hydrogen stations serving fuel cell electric vehicles. The Hydrogen Materials Compatibility Consortium (H-Mat) coordinates RD&D on accelerated test methods and novel, low-cost, durable metals and polymers for use in hydrogen infrastructure.
- **Hydrogen Storage** addresses cost-effective onboard and off-board hydrogen storage technologies with improved energy density and lower costs. RD&D activities investigate high-pressure compressed storage, materials-based storage, and hydrogen carriers. Activities in the latter two topic areas are coordinated through the Hydrogen Materials–Advanced Research Consortium (HyMARC) to accelerate the discovery and development of breakthrough hydrogen storage materials.

In fiscal year (FY) 2020 and FY 2021,[1] projects in the Hydrogen Production category focused primarily on RD&D for advanced water-splitting materials and systems through HydroGEN, which is part of the DOE Energy Materials Network, and the H2NEW consortium. Production pathways under investigation include four advanced water-splitting technologies: high- and low-temperature electrolysis, direct solar thermochemical hydrogen production, and photoelectrochemical water splitting. Additional work outside of HydroGEN included RD&D on microbial-based processes using biomass and waste-stream feedstocks and efforts leveraging electrolysis technology for carbon dioxide (CO_2) reduction to useful chemicals and fuels. Hydrogen Infrastructure projects included (1) low-cost, high-efficiency liquefaction, pipelines, chemical carriers, and tube trailers; (2) low-cost and reliable compressors, pumps, dispensers, and stationary storage; and (3) hydrogen delivery technologies analysis. Hydrogen Storage projects in FY 2020 and FY 2021 focused on materials-based hydrogen storage RD&D through HyMARC, advanced tanks through innovative approaches to develop low-cost carbon fiber precursors, large-scale hydrogen storage through hydrogen carriers, and storage technologies for medium- and heavy-duty transportation.

GOAL

The overarching goal of the Hydrogen Technologies subprogram is to enable commercialization of sustainable and efficient hydrogen technologies that are competitive with incumbent technologies in terms of cost and performance, across diverse applications. The subprogram pursues this goal by developing solutions for all aspects of the

[1] This subprogram overview covers the period since the publication of the last annual progress report in April 2020 (see U.S. Department of Energy, *DOE Hydrogen and Fuel Cells Program 2019 Annual Progress Report*, https://www.hydrogen.energy.gov/annual_progress19.html).

hydrogen pathway—from hydrogen production, leveraging the nation's diverse renewable resources, to all aspects of hydrogen infrastructure, including the storage, transmission, distribution, delivery, and dispensing of hydrogen for various delivery pathways and end uses.

KEY MILESTONES

- Reduce the cost of clean hydrogen production from diverse domestic resources to <$2/kg by 2025 and to $1/kg by 2030.
- Reduce the cost of delivering and dispensing hydrogen to a fuel cell electric vehicle to <$2/kg by 2025. This cost is independent of the technology pathway and takes into consideration a range of assumptions for fuel cell electric vehicles to be competitive.
- Develop onboard hydrogen storage systems for Class 8 long-haul tractor-trailers, achieving a cost of $9/kWh by 2030 and the capability to withstand 11,000 pressure cycles.

FISCAL YEAR 2021 TECHNOLOGY STATUS AND ACCOMPLISHMENTS

The Hydrogen Technologies subprogram actively monitors technical progress achieved through the Hydrogen Production, Hydrogen Infrastructure, and Hydrogen Storage RD&D project portfolios. That progress is incorporated into the technology status with respect to performance metrics such as cost, efficiency, and energy density. The status of various technologies being funded by the subprogram is described below.

Hydrogen Production

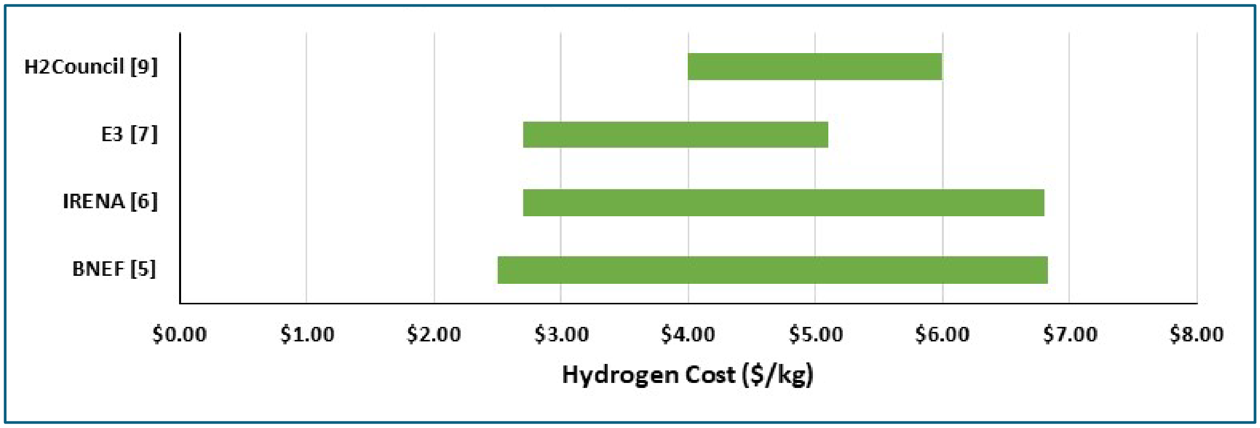
RD&D is focused on reducing the cost and improving the efficiency of producing clean hydrogen from domestic, renewable feedstocks and energy resources. Feedstocks include water, biomass, and organic waste such as wastewater and food waste. Clean hydrogen can be produced at either large centralized or smaller distributed production facilities.

- **Near- to Mid-Term Electrolysis Options:** These approaches for clean hydrogen production use different electrolyzer technologies—with some operating at low temperatures (using polymer membranes limited to <100°C) and others at high temperatures (using solid oxide membranes that operate efficiently at >600°C). Low-temperature polymer electrolyte membrane (PEM) electrolyzers are commercially available at the megawatt scale but currently provide a relatively small portion of the hydrogen in the United States. High-temperature solid oxide electrolyzers offer higher electrical conversion efficiencies but are at an earlier stage of development, with only small-scale manufacturing under way. There is a growing interest in integrating electrolyzers with renewable energy sources, which could lead to greater manufacturing demand for both low- and high-temperature electrolysis. Higher volumes of electrolyzer manufacturing, coupled with further RD&D advances, are needed to achieve cost parity with hydrogen produced from natural gas.

  The figure below shows published costs of hydrogen production from currently available PEM electrolyzers, collected from several external sources.[2] Overall, these data show that hydrogen can be produced today within a cost range of ~$2.50–$6.80/kg from a mix of renewable and grid feedstocks. This is in good alignment with the DOE analysis, which shows that hydrogen can be produced via PEM electrolysis at a cost of ~$4–$6/kg for specific conditions.

[2] U.S. Department of Energy, "Cost of Electrolytic Hydrogen Production with Existing Technology," Program Record #20004, September 22, 2020.

Existing PEM Hydrogen Production Costs



- **Longer-Term Innovative Hydrogen Production – Beyond Electrolysis:** Emerging longer-term options for hydrogen production from renewable resources are at varying stages of development. While there are currently some plant-scale operations in service to extract hydrogen from biomass and waste-stream feedstocks using thermal and catalytic processes (such as biomass gasification), newer and more promising approaches using these feedstocks are in earlier stages of development. These include microbial processes such as fermentation and microbial electrolysis. Other promising renewable approaches in their early stages include photoelectrochemical and solar thermochemical processes, which use direct solar and solar thermal energy, respectively, to split water without the efficiency losses associated with converting the energy source into electricity.

Hydrogen Infrastructure

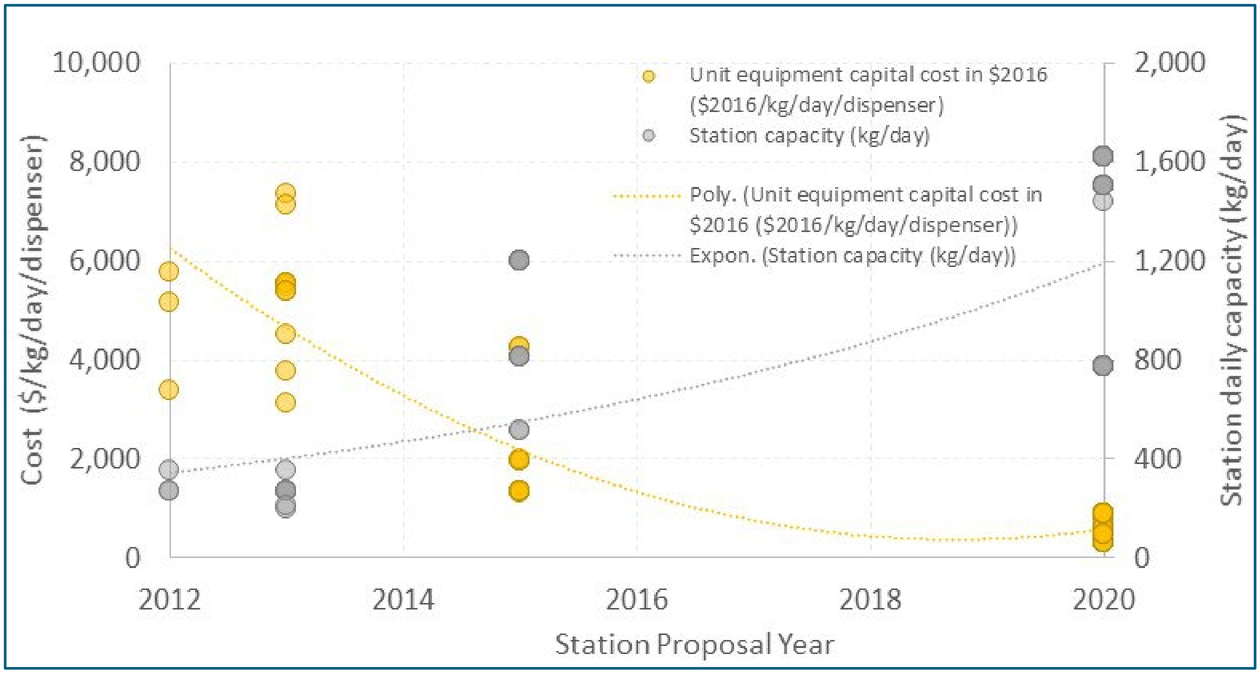
Hydrogen can be transported and distributed as a compressed gas, as a cryogenic liquid, or bound within a chemical hydrogen-carrier material. Each of the delivery methodologies requires a range of technologies, such as compressors, liquefiers, and dispensing technologies (which may need to meet specific needs for the particular application). The Hydrogen Infrastructure activity focuses on developing these technologies to meet targets for dispensed hydrogen. It also works to identify materials for hydrogen delivery technologies (e.g., pipelines) that are compatible with hydrogen under various operating conditions. Hydrogen delivery options in the subprogram's RD&D portfolio are discussed below.

- **Near- to Mid-Term Hydrogen Transport and Dispensing for Heavy-Duty (and Other Near-Term) Applications:** In the near- to mid-term, it is expected that most of the hydrogen for these applications will be delivered as a compressed gas—either via pipeline or using high-pressure tanks—or as a cryogenic liquid. Most hydrogen is supplied via pipeline today for use in petroleum refining. For other current end uses in the transportation, industrial, and chemical sectors, hydrogen is commonly transported and stored as either a high-pressure compressed gas (via pipeline or truck) or cryogenic liquid (via tanker). Near- and mid-term RD&D efforts are focused on reducing the cost of these technologies and minimizing the losses of hydrogen that occur during transport and dispensing.

  Over the past decade, hydrogen fueling stations have declined in cost as a result of both RD&D and economies of scale. The evolution of costs and capacities of stations proposed for construction in California is depicted below.[3] Stations for heavy-duty fueling are expected to be several-fold larger in capacity than those proposed to date for light-duty markets in California, and to also require fueling at about 5X faster flow rates than light vehicles. These deployments will require designs for components such as compressors, dispensers, and chillers. In support of this market, in FY 2021, the DOE issued multiple solicitations around development of high-flow fueling equipment, including a funding opportunity announcement and two Small Business Innovation Research (SBIR) topics.

[3] U.S. Department of Energy, "Hydrogen Fueling Stations Cost," Program Record #21002, February 11, 2021.

Costs of Hydrogen Fueling Station Equipment in Proposals to the California Energy Commission



- **Longer-Term Advanced Hydrogen Liquefaction and Carrier Distribution Approaches:** Over the longer term, innovative approaches to liquid hydrogen production, storage, transport, and dispensing are expected to enable new, expanded opportunities for large-scale liquid hydrogen use. Key challenges include minimizing hydrogen losses and reducing the cost of liquefaction technologies. Another emerging option is the use of chemical hydrogen carriers, which are solid or liquid materials to which hydrogen temporarily bonds, enabling low-pressure, high-capacity transport and storage. Carriers have been deployed in prototype demonstrations to supply hydrogen to industrial applications (such as thermal power generation) and are currently being explored for use in bulk transport and storage of hydrogen, including on marine vessels for a potential export market.

Hydrogen Storage

Hydrogen can be stored either physically, as a compressed gas or cryogenic liquid, or as a material within which the hydrogen is physi- or chemisorbed. Each storage methodology has advantages and disadvantages, making it more or less suitable for specific applications. Key challenges for all are reducing the cost of storage and improving the overall performance of the technology.

- **Near- to Mid-Term High-Pressure Tanks and Other Physical Hydrogen Storage Options:** RD&D to improve physical storage methods is focused primarily on reducing cost and minimizing losses from tanks and other technologies in use today for compressed gaseous and liquid hydrogen storage. Compressed gaseous hydrogen can also be contained in bulk in caverns (i.e., underground rock-lined or salt caverns) for long-duration storage applications; however, this approach is limited to specific geographical areas, and further research and optimization are needed to address cost and safety issues. Activities also look at development of other large-scale bulk storage technologies for practical applications.
- **Longer-Term Material-Based Hydrogen Storage Options:** Longer-term RD&D is focused on material-based options such as adsorbents, metal hydrides, and chemical hydrogen carriers to open new opportunities in affordable hydrogen storage. These approaches offer the potential to achieve comparable hydrogen storage densities, but at near-ambient operating conditions, without the need for high pressures (as in compressed hydrogen storage) or very low temperatures (as in cryogenic liquid hydrogen storage), both of which add significant energy expenditures and costs to the entire pathway.

SUBPROGRAM-LEVEL ACCOMPLISHMENTS

The Hydrogen Technologies subprogram made significant progress during FY 2020 and FY 2021. The subprogram released two funding opportunity announcements (FOAs), resulting in the award of five new electrolyzer manufacturing projects; two microbial electrolysis projects to produce hydrogen from biomass waste streams; two analysis projects to assess the cost and performance of hydrogen production and hydrogen storage pathways, respectively; three RD&D projects to build the domestic supply chain for high-flow hydrogen fueling stations for heavy-duty applications; four RD&D projects to develop advanced carbon fiber for compressed hydrogen and natural gas storage tanks; and one project on developing and validating the engineering design of ultra-large-scale liquid hydrogen storage tanks. In addition, eight Small Business Innovation and Research (SBIR) Phase I projects were awarded: three to address hydrogen RD&D challenges and advance progress in hydrogen production from wind power, three to develop low-cost scalable hydrogen pre-cooling and filter technologies for heavy-duty stations, and two to mitigate boil-off losses in liquid hydrogen storage systems. The subprogram also awarded one Phase II SBIR project on the evaluation of micrometer-scale flaws in pressure vessels and six Cooperative Research and Development Agreement (CRADA) projects.

Another notable development in the Hydrogen Technologies subprogram is the merger of the U.S. DRIVE Partnership's Hydrogen Delivery and Hydrogen Storage Tech Teams into one technical team: the Hydrogen Delivery and Storage Tech Team (HDSTT). The HDSTT works to ensure close communications across delivery and storage pathways to focus efforts on viable, long-term solutions that are compatible and complementary, while also identifying technology gaps impeding commercialization.

Accomplishments in each of the subprogram's RD&D areas are described below.

Hydrogen Production

- The HydroGEN Benchmarking Project team held the 2nd and 3rd Annual Advanced Water-Splitting Technology Pathways Benchmarking and Protocols Workshops.
  - The second annual workshop for the HydroGEN Benchmarking Project was held October 29–30, 2019, at the Scottsdale campus of Arizona State University. The workshop, which was open to international participants, was attended by about 90 people, with representation evenly distributed across the water-splitting technology areas. Effort was made to engage at least one international representative for each technology to summarize related initiatives in Europe and encourage communication and awareness.
  - The third annual workshop for the HydroGEN benchmarking project, originally scheduled for fall 2020, was rescheduled to March 2021 and held as a virtual workshop. Approximately 200 people attended, with 20–50 participants across each of 29 breakout sessions. Participation was evenly distributed across the water-splitting technology areas. At least three countries were represented.
  - The HydroGEN Benchmarking Project drafted four advanced water splitting pathway roadmaps and drafted and reviewed 45 test protocols. The team is currently drafting 25 additional protocols. The protocols are to be published in an upcoming issue of *Frontiers in Energy*.
- The subprogram awarded the HydroGEN Energy Materials Network Phase 2 project (HydroGEN 2.0), a three-year project comprising five new lab projects and five supernode projects.[4] The lab projects will focus on low-technology-readiness-level advanced water-splitting materials for alkaline exchange membrane electrolysis, metal-supported solid oxide electrolysis cells (SOECs), proton-conducting SOECs, photoelectrochemical hydrogen production, and solar thermochemical hydrogen production.
- The H2NEW consortium was launched in October 2020. This consortium consists of nine national laboratories, led by the National Renewable Energy Laboratory and Idaho National Laboratory. H2NEW will conduct RD&D to enable large-scale manufacturing of affordable electrolyzers that use electricity to split water into hydrogen and oxygen. The work will focus on materials and component integration,

[4] In 2019, DOE established five new "supernodes" within the HydroGEN consortium through which multiple lab capability nodes and experts work synergistically to address specific water-splitting materials problems or research needs.

manufacturing, and scale-up to help support large industry deployment of durable, efficient, and low-cost electrolyzers for hydrogen production.

- The subprogram developed four program records (Records 19009, 20004, 20006, and 20009) documenting the 2019 hydrogen production cost from PEM electrolysis, the cost of electrolytic hydrogen production with existing technology, the hydrogen production cost from high-temperature electrolysis, and electrolyzer capacity installations in the United States.

Hydrogen Infrastructure

- The National Renewable Energy Laboratory, in partnership with Air Liquide, Honda, Shell, and Toyota under the Innovating Hydrogen Stations CRADA, is currently in the process of commissioning a test facility with first-of-its-kind, experimental research capability for 10 kg/min, 60+ kg hydrogen fueling for heavy-duty applications. This facility will be used to conduct experimentation that informs the laboratory's computational models of high-throughput fueling and may be used in future RD&D to develop and test new fueling technologies.
- The subprogram developed three program records (Records 19001, 21002, and 20007) documenting the current cost status of hydrogen liquefaction, hydrogen delivery and dispensing, and hydrogen fueling stations.

Hydrogen Storage

- The subprogram hosted the Novel Pathways for Optimized Hydrogen Transport & Storage Workshop on November 13–14, 2019, to engage stakeholders from industry, academia, and DOE national laboratories in determining the suitability and requirements of hydrogen carrier materials as a novel pathway for hydrogen transport and stationary storage. The workshop was attended by approximately 75 participants.
- The subprogram hosted the Compressed Gas Storage for Medium- and Heavy-Duty Transportation Workshop on January 21, 2020. Participants identified performance gaps and technology metrics (e.g., weight, volume, cost, and durability) that can enable competitiveness of compressed gas storage technologies in medium- and heavy-duty transportation. The workshop was attended by 63 representatives of government, laboratory, academia, and industry.
- The subprogram published a program record (Record 19008) documenting the cost and performance status of onboard Type IV compressed hydrogen storage systems.

NEW PROJECT SELECTIONS

In FY 2020 and FY 2021, the Hydrogen Technologies subprogram added a number of new projects to the portfolio: 19 projects selected from the competitive FOAs, 9 SBIR projects, 6 CRADA projects, and 7 projects selected from the annual competitive Lab Calls. The projects are listed below.

Funding Opportunity Announcements

Hydrogen Production

- 3M Company – Advanced Manufacturing Processes for Gigawatt-Scale Proton Exchange Membrane Water Electrolyzer Oxygen Evolution Reaction Catalysts and Electrodes
- Plug Power (formerly Giner ELX, Inc.) – Integrated Membrane Anode Assembly and Scale-up
- Proton Energy Systems, Inc. – Enabling Low-Cost PEM Electrolysis at Scale through Optimization of Transport Components and Electrode Interfaces
- Cummins, Inc. – Automation of Solid Oxide Electrolyzer Cell and Stack Assembly
- Nextech Materials, Ltd. – Low-Cost Manufacturing of High-Temperature Electrolysis Stacks
- The Pennsylvania State University – Novel Microbial Electrolysis Cell Design for Efficient Hydrogen Generation from Wastewaters

- Southern Company Services, Inc. – Novel Microbial Electrolysis System for Conversion of Biowastes into Low-Cost Renewable Hydrogen
- Strategic Analysis, Inc. – Hydrogen Production Analysis

Hydrogen Infrastructure

- Czero, Inc. – Advanced High-Throughput Compression System for Medium- and Heavy-Duty Transportation
- Gas Technology Institute – Cost-Effective Pre-Cooling for High-Flow Hydrogen Fueling
- Nikola Corporation – Autonomous Fueling System for Heavy-Duty Fuel Cell Electric Trucks

Hydrogen Storage

- Collaborative Composite Solutions Corporation – Melt Spun Polyacrylonitrile (PAN) Precursor for Cost-Effective Carbon Fiber in High-Pressure Compressed Gas Tankage
- Hexagon R&D LLC – Carbon Composite Optimization Reducing Tank Cost
- University of Kentucky – Low-Cost, High-Strength Hollow Carbon Fiber for Compressed Gas Storage Tanks
- University of Virginia – Low-Cost, High-Performance Carbon Fiber for Compressed Natural Gas Storage Tanks
- Strategic Analysis, Inc. – Hydrogen Storage Analysis
- Shell – First Demonstration of a Commercial-Scale Liquid Hydrogen Storage Tank Design for International Trade Applications

Small Business Innovation and Research Phase I

- Alchemr – Hydrogen from Wind
- Giner, Inc. – Cost Model and Design Requirements for a Wind-to-Hydrogen Generation System
- Greenway Energy – New Low-Cost and Efficient Electrolysis System That Can Be Directly Coupled with Wind Turbine Power
- Skyhaven Systems – Hydrogen Pre-Cooling System Using Phase Change Materials, for Use in Medium- and Heavy-Duty Fueling
- NanoSonic, Inc. – Electrospun In-Line Filters for Heavy-Duty Hydrogen Fueling Stations
- Global Research and Development, Inc. – Ceramic In-Line Particulate Filter for Heavy-Duty Hydrogen Stations
- NuMat Technologies – Evaporative Emission Control for Hydrogen: Boil-Off Mitigation with an Adsorbent-Based System
- The Protium Company – Tank-Integrated Heat Exchanger for Boil-Off Reduction

Small Business Innovation and Research Phase II

- Luna Innovation – Use of Wave Mixing for Non-Destructive Evaluation of Micrometer-Scale Flaws in Pressure Vessels

H2@Scale CRADA Projects

- The HyBlend project – Launched to address the technical barriers to blending hydrogen in natural gas pipelines – National Renewable Energy Laboratory, Sandia National Laboratories, Pacific Northwest National Laboratory, Oak Ridge National Laboratory, Argonne National Laboratory, and the National Energy Technology Laboratory, and more than 20 participants from industry and academia

- Analytic Framework for Optimal Sizing of Hydrogen Fueling Stations for Heavy-Duty Vehicles at Ports – CRADA between Pacific Northwest National Laboratory, Seattle City Light, Port of Seattle, Northwest Seaport Alliance, and PACCAR/Kenworth
- Heavy-Duty Reference Station Design, Test Device Development, and Capacity Modeling –CRADA between National Renewable Energy Laboratory, Sandia National Laboratories, California Governor's Office of Business and Economic Development, California Energy Commission, California Air Resources Board, and South Coast Air Quality and Management District
- Optimization of Pre-Cooling at Heavy-Duty Stations and Cyber Vulnerability Analysis – CRADA between Savannah River National Laboratory, Sandia National Laboratories, Argonne National Laboratory, and Nikola
- Development of High-Flow 350 bar Hydrogen Fueling Method – CRADA between National Renewable Energy Laboratory and Frontier Energy
- Assessment of Heavy-Duty Fueling Methods and Components – CRADA between National Renewable Energy Laboratory, Argonne National Laboratory, NextEnergy, and Chevron

Lab Call

- Argonne National Laboratory – Cost Assessment and Evaluation of Liquid Hydrogen Storage for Medium- and Heavy-Duty Transportation Applications
- Lawrence Berkeley National Laboratory – Integrated Onsite Waste-Heat-Driven Hydrogen Carrier System for Steel and Renewables Coupling
- Savannah River National Laboratory – Determining the Value Proposition of Materials-Based Hydrogen Storage for Stationary Bulk Storage of Hydrogen
- Oak Ridge National Laboratory – Cost-Optimized Structural Carbon Fiber for Hydrogen Storage Tanks
- Argonne National Laboratory – Hydrogen Carriers for Renewable Energy Farm Application
- National Renewable Energy Laboratory, Lawrence Berkeley National Laboratory, Sandia National Laboratories, Lawrence Livermore National Laboratory, and Idaho National Laboratory – HydroGEN 2.0: A multi-lab approach, utilizing and integrating national laboratory capabilities to address critical research gaps in each of the advanced water splitting pathways
- National Renewable Energy Laboratory, Idaho National Laboratory, Lawrence Berkeley National Laboratory, Argonne National Laboratory, Los Alamos National Laboratory, Oak Ridge National Laboratory, Pacific Northwest National Laboratory, Lawrence Livermore National Laboratory, and National Energy Technology Laboratory – H2NEW (H2 from the Next-generation of Electrolyzers of Water): A multi-lab consortia addressing components, materials integration, and manufacturing research and development to enable affordable, reliable, and efficient electrolyzers

PROJECT-LEVEL ACCOMPLISHMENTS

During FY 2020 and FY 2021, projects in the Hydrogen Technologies R&D portfolio made important progress in several key areas, as highlighted below.

Hydrogen Production

- Demonstrated improved anion exchange membrane electrolysis durability over ~750 hours at relevant current density. (Georgia Institute of Technology)
- Established a promising interconnect protective coating for the air/O_2 side of high-temperature electrolysis stacks. (Nexceris)
- Aided by high-performance computing, discovered a new water-splitting material family, $(Ca,Ce)(X,Y)O_3$. (Arizona State University)
- Achieved a 2.5 times higher solar-to-hydrogen efficiency than state-of-the-art perovskite cells in an integrated 3D-printed photoelectrochemical reactor. (Rice University)

- Doubled hydrogen production at 60 g/L crystalline cellulose loading via fed-batch operation scheme. (National Renewable Energy Laboratory, Pacific Northwest National Laboratory, Lawrence Berkeley National Laboratory, and Argonne National Laboratory)
- Increased hydrogen production 33% from the original baseline via better hemicellulose and cellulose co-utilization. (National Renewable Energy Laboratory, Pacific Northwest National Laboratory, Lawrence Berkeley National Laboratory, and Argonne National Laboratory)
- Increased hydrogen production rate 100% over the state of the art using brewery wastewater. (Oregon State University)
- Described the chemical mechanisms behind curiously resilient photocathodes made from silicon and gallium nitride (Si/GaN). (University of Michigan, Lawrence Livermore National Laboratory, and Lawrence Berkeley National Laboratory)

Hydrogen Infrastructure

- The H-Mat consortium is focused on RD&D to enhance the durability of materials in hydrogen service. Key recent accomplishments are described below.
    - The H-Mat polymers team has identified mechanisms by which rubber materials fail in hydrogen, as well as likely locations of failure. Using advanced imaging capabilities (e.g., helium ion microscopy) and chemical analysis of materials stressed in hydrogen, the team identified that plasticizers used in commercial rubbers can separate from the material and migrate when stressed in hydrogen. The researchers additionally found that zinc oxide activators (used in rubber manufacturing to expedite the curing process) are likely locations of void formation in hydrogen, which can result in material failure. These results are informing efforts in FY 2022 to synthesize model materials with increased resistance to hydrogen effects. (Pacific Northwest National Laboratory)
    - The H-Mat metals team has identified that austenitic stainless steels saturated in hydrogen at cryogenic temperatures (20 K) experience approximately 50% reduction in ductility (characterized via reduction of area measurements) relative to testing in hydrogen at ambient temperatures. Previously available reports had indicated that hydrogen effects at cryogenic temperatures would be minimal because of limitations in the kinetics of hydrogen at these temperatures. Results generated by the H-Mat team may inform future RD&D to understand and enhance the long-term durability of materials used in liquid hydrogen infrastructure. (Sandia National Laboratories)
- The National Renewable Energy Laboratory completed design and construction of a high-flow hydrogen fueling system at the lab's Energy Systems Integration Facility, through a CRADA project co-funded by the Hydrogen and Fuel Cell Technologies Office, Shell, Air Liquide, Toyota Motor Company, and Honda R&D Americas. The facility will be capable of fueling at 10 kg/min. when commissioning is complete (anticipated by the end of 2021), such that it can be used for future experimentation on high-flow fueling components.
- Sandia National Laboratories and the National Renewable Energy Laboratory collaborated with the DOE Office of Fossil Energy and Carbon Management and Southern Company to complete an evaluation of codes and standards relevant to hydrogen blending in pipelines, identifying current gaps. The analysis is currently in press.
- A team led by the National Renewable Energy Laboratory developed the publicly accessible H2FillS model. H2FillS allows users to generate 1-dimensional simulations of the impact of varying fueling methods on the thermodynamics of fueling equipment and hydrogen storage onboard. The National Renewable Energy Laboratory is now expanding H2FillS to be capable of 3-dimensional simulations of high-throughput fueling of medium- and heavy-duty vehicles. The other team members were Kyushu University, Frontier Energy, Ford Motor Company, General Motors, LLC, Honda R&D Americas, Hyundai, IVYS Energy Solutions, Shell, Toyota, Sandia National Laboratories, and Argonne National Laboratory. H2FillS is available here: https://www.nrel.gov/hydrogen/h2fills.html

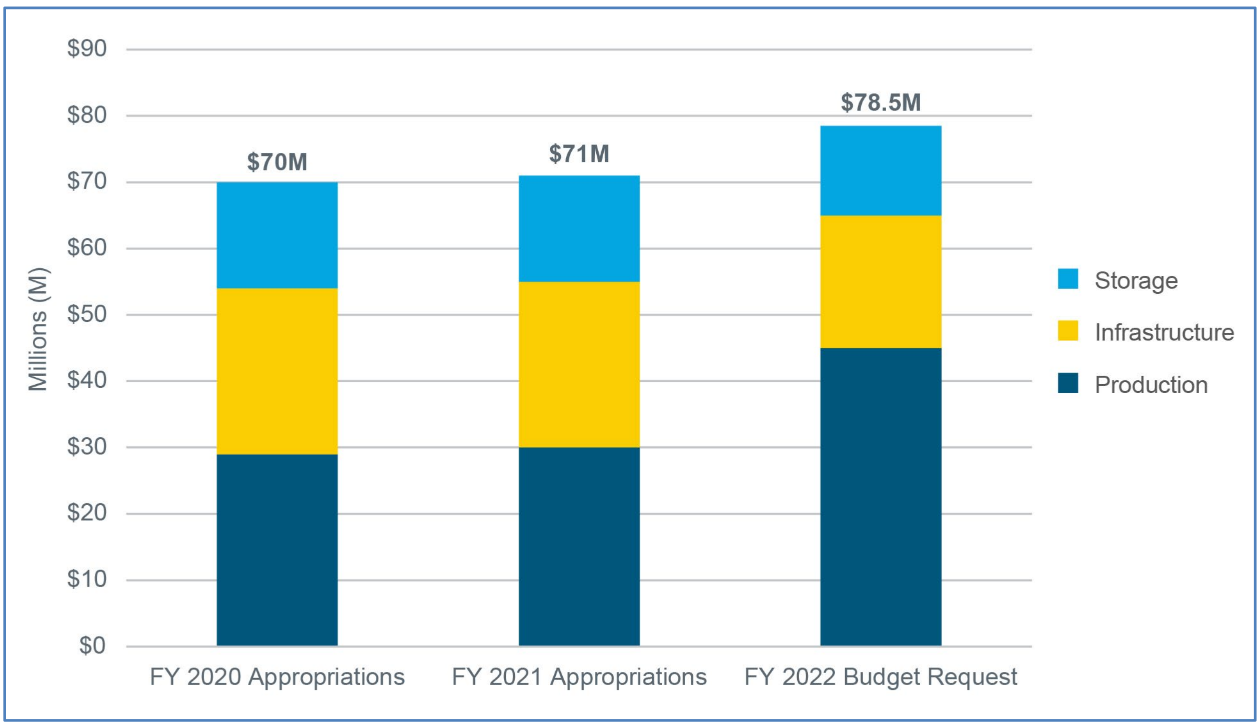
Hydrogen Storage

- Synthesized $V_2Cl_{2.8}$(btdd), the best-performing metal–organic framework (MOF) for room-temperature hydrogen adsorption and the first MOF with a binding enthalpy in the optimal range for ambient temperature storage (-15 to -25 kJ/mol). (Lawrence Berkeley National Laboratory)
- Improved MgB_2-$Mg(BH_4)_2$ hydrogenation conditions by 100°C and 200 bar over the state of the art. (University of Hawaii/HyMARC)
- Demonstrated 2x (de)hydrogenation rates for high-capacity Li/Mg-amides through nanoconfinement in carbons. (Sandia National Laboratory)
- Applied machine learning and modeling to identify thousands of MOFs with potential to exceed state-of-the-art hydrogen volumetric capacities. (University of Michigan and Northwestern University)

BUDGET

The appropriations for the Hydrogen Technologies subprogram totaled $70 million in FY 2020 and $71 million in FY 2021. Of these appropriations, $29 million and $30 million were allocated for hydrogen production research in FY 2020 and FY 2021, respectively. In both FY 2020 and FY 2021, $25 million was allocated for infrastructure research and $16 million for hydrogen storage research. These allocations are shown in the graph below. Projects funded in the Hydrogen Technologies portfolio are expected to accelerate development of low- and high-temperature electrolyzers, materials for advanced water-splitting technologies, electrolyzer manufacturing technologies, microbial hydrogen production, and hydrogen infrastructure and storage technologies. Specific emphasis is focused on meeting the DOE Hydrogen Shot goal of producing clean hydrogen for $1/kg by 2030. This emphasis is expected to continue into FY 2022.

Hydrogen Technologies RD&D Funding



Annual Merit Review of the Hydrogen Technologies Subprogram

SUMMARY OF HYDROGEN TECHNOLOGIES SUBPROGRAM REVIEWER COMMENTS

Reviewers commended the Program for achieving a hydrogen production cost of $5–$6/kg and expressed support for the Hydrogen Energy Earthshot hydrogen production target of $1/kg by 2030. Reviewers stated that the new target will require re-balancing the RD&D activities to prioritize end-use demonstrations of hydrogen production and scale-up of mature electrolysis technologies, as well as new ideas with potential for transformational changes. More active engagement of the Hydrogen Program with other DOE offices, other federal agencies, and state and regional stakeholders was recommended. In particular, reviewers recommended increased engagement with the Office of Fossil Energy (to advance carbon capture and storage and "blue hydrogen" production technologies from steam methane reforming plants or other fossil fuel sources), the Office of Basic Energy Sciences, and the Advanced Manufacturing Office, as well as the National Science Foundation.

Reviewers of the Hydrogen Production category commented that the projects were relevant and potentially impactful and, if successful, would contribute to achievement of DOE's cost and performance targets. The reviewers responded favorably to the approaches of these R&D efforts, describing them as reasonable and effective, and were particularly supportive of integrated approaches that include both experimental and computational methods. Review panels welcomed the addition of the H2NEW consortium and praised the HydroGEN, H2NEW, and BioH2 consortium projects for their coordination between the national laboratories. However, reviewers encouraged adding industrial partners, both to obtain their input and to enhance and encourage technology transfer. Adding international partners and increasing engagement with academia were also recommended. Projects were commended for making use of the HydroGEN Benchmarking Project and for their contributions of resources to the HydroGEN Data Hub. Reviewers commented that they would like to see increased attention to technoeconomic assessments of the technologies and validation of results. Reviewers also suggested increased focus on stability, degradation, and mechanistic understanding of the processes involved for some of the HydroGEN seedlings.

Reviewers of the Hydrogen Infrastructure category applauded the projects' novel and sound scientific approaches. The infrastructure RD&D projects were deemed to be relevant to meeting the Program's goals and, if successful, potentially impactful in enabling hydrogen infrastructure implementation. Reviewers praised the Innovating Hydrogen Station at the National Renewable Energy Laboratory for addressing critical information needs for hydrogen fueling for heavy-duty vehicles, as well as providing significant and reliable demonstrations of equipment performance and capabilities in line with industry needs, combined with valuable multiscale modeling. Review panels noted the importance of Argonne National Laboratory's hydrogen delivery technologies analysis in providing input to guide RD&D funding decisions, as well as informing industry investment decisions. Reviewers commented favorably on the progress that many projects have made in spite of the COVID-19-related restrictions that have been in place for the last year but noted that the level of collaboration on the Hydrogen Infrastructure projects should be improved as restrictions are lifted. Recommendations included increased focus on obtaining input from industry, technologies with potential to be broadly applicable and scalable, leveraging ongoing hydrogen infrastructure developments in California, characterization efforts, and validation of results. The Program was commended for its increased focus on infrastructure to support medium- and heavy-duty vehicles, but reviewers cautioned that light-duty transportation applications still require data, research, and demonstration. An explanation of the reasons for discontinuing work on electrochemical compression was requested. Coordination with Canada on hydrogen blending with natural gas was recommended.

Reviewers of the Hydrogen Storage category commended the projects for their novel and innovative approaches, their solid progress toward meeting project objectives during the COVID-19 pandemic, and their relevance and alignment with the Program's goals and objectives. The project teams were described as competent and experienced, and the projects were deemed well-managed and -coordinated. Reviewers commented that some projects, if successful, could lead to major breakthroughs in hydrogen storage technology. Reviewers found the storage analysis projects to be complementary to each other and to the materials development efforts and to have thorough and systematic approaches. Reviewers recommended that the analysis projects seek additional industrial input and partners to validate cost assumptions. Reviewers commented favorably on HyMARC's use of "push projects" that complement the core HyMARC efforts and expand the technical scope and depth of the consortium, as well as seedling projects pursuing high-risk research. The hydrogen carrier projects were particularly praised for integrating analysis, computation, experimental kinetics, and catalysis into their approaches. Some projects included machine learning, which was considered a valuable addition. Reviewers emphasized the importance of continuing research on hydrogen storage and noted that progress needs to be accelerated, observing that materials-based approaches have made only incremental advances in recent years.

Six Hydrogen Production projects (not including the HydroGEN Seedling projects) were reviewed, with overall favorable scores ranging from 2.9 to 3.7, with 3.3 as the average score. Eleven HydroGEN Seedling projects were reviewed, with scores ranging from 2.8 to 3.8, with 3.2 as the average score. Fourteen Hydrogen Infrastructure projects were reviewed; their scores ranged from 2.3 to 3.8, with the average score being 3.2. Fourteen Hydrogen Storage projects were reviewed; the highest, lowest, and average scores were 3.7, 2.9, and 3.4, respectively.

Following this subprogram introduction are individual project reports for each of the projects reviewed. Each report contains a project summary, the project's overall score and average scores for each question, and the project-level reviewer comments.

Project Reviews

PRODUCTION

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

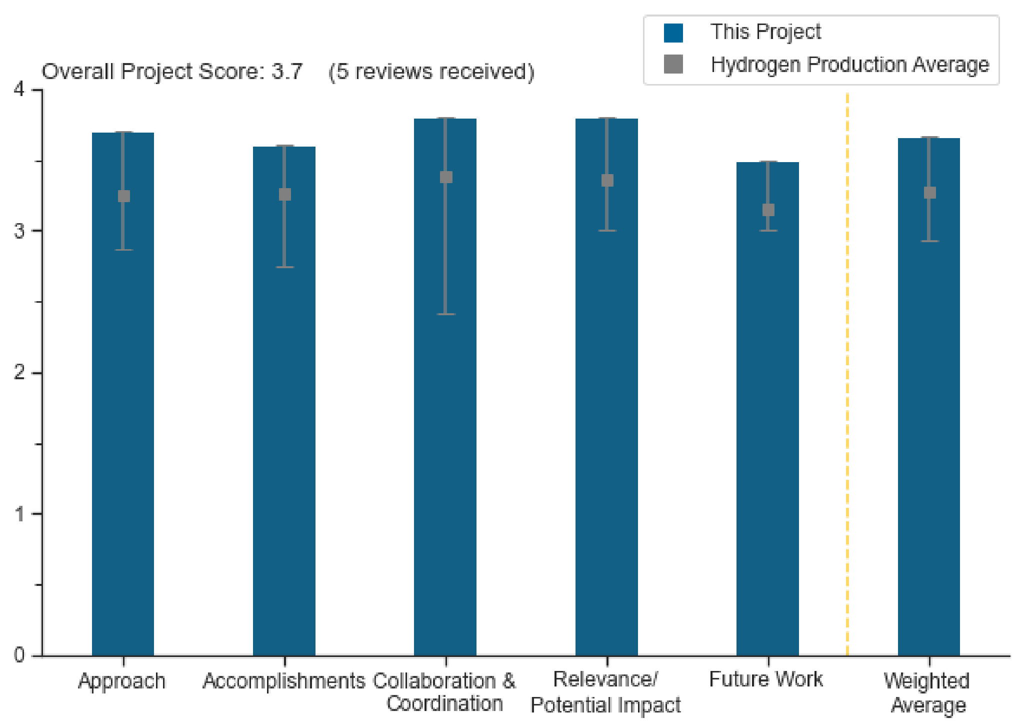
Huyen Dinh, National Renewable Energy Laboratory

DOE Contract #	WBS 2.7.0.518 and 2.7.0.513
Start and End Dates	6/1/2016
Partners/Collaborators	HydroGEN Consortium
Barriers Addressed	• Cost • Efficiency • Durability

Project Goal and Brief Summary

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 (WS) technology pathways supported by HydroGEN include (1) photoelectrochemical (PEC), (2) solar thermochemical (STCH), (3) low-temperature electrolysis (LTE), and (4) high-temperature electrolysis (HTE). In addition to collaborating with industry and academia, HydroGEN uses a synergetic, multi-laboratory approach, utilizing and integrating the labs' world-class capabilities to address the critical research gaps identified by the lab teams and HydroGEN Benchmarking and Protocol workshops in each of the advanced water-splitting (AWS) technologies.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Production projects.

Question 1: Approach to performing the work

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

- The HydroGEN Advanced Water Splitting Materials Consortium (HydroGEN) makes up a good blend of AWS technologies and ensures some coordination within the community. The coordination among the laboratories is a much-improved model versus previous history, when the laboratories had to compete against each other for funding. This approach allows much higher effectiveness and teamwork. The capabilities within the laboratories are also impressive, and if directed properly, can help advance U.S. research and product development at an accelerated pace to compete globally. HydroGEN 1.0 started out with funded projects that leveraged the nodes, which was somewhat constraining for the laboratories in terms of being able to work on new ideas. The super nodes addressed this to an extent, with laboratory-only projects addressing broader challenges. HydroGEN 2.0 is early in its lifetime, but initial projects seem to represent more individual laboratory projects. The team should be cautious in maintaining a balance and not going too far the other way versus maintaining the consortium model.
- The HydroGEN 1.0 consortium approach adopts a highly interactive model for collaboration across national laboratory members, extramural community (including both academia and industrial laboratories), and other agencies (e.g., National Science Foundation, National Institute of Standards and Technology). It features multiple mechanisms for providing exchange of information, benchmarking, and sharing of samples and data. It represents an outstanding approach to accelerating learning and achieving technical objectives. Dr. Dinh, who manages this consortium, is highly motivated to coordinate and facilitate the numerous efforts.
- The demarcation of HydroGEN 2.0 and Hydrogen from Next-generation Electrolyzers of Water (H2NEW) based on current technology readiness levels (TRLs) is smart and practical to move the two consortiums forward. It is a good idea to focus HydroGEN 2.0 on low-TRL areas of AWS R&D, since low-temperature polymer electrolyte membrane (PEM) electrolyzers are far more advanced compared to other AWS routes. Although the HydroGEN 2.0 scope seems to specifically exclude PEM-based LTE technologies (slide 5), it is not clear why PEM electrolysis projects are still in the portfolio, as shown in slide 14. The goals of STCH approaches to develop a theory-guided material-design strategy for optimizing the capacity/yield tradeoff and use machine learning to find new STCH WS materials are reasonable. However, given the basic state of current STCH concepts, it is not clear if the implied progress in the label STCH 2.0 is justified.
- This is an excellent approach to supporting lower-TLR hydrogen production technologies.

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 funded projects are addressing key fundamental challenges and appear to be making good progress. Since this was an overview of the full consortium, there were not sufficient details to evaluate the specific projects, which are presumably being reviewed separately, but the portfolio seems generally robust.
- HydroGEN 1.0 had many successful outcomes, both technical and in terms of people working together from diverse sectors. The cooperative spirit was a major outcome and led to accelerating the pace of discovery and validation. HydroGEN 1.0 evolved to HydroGEN 2.0 in 2021. Information was not shared on how projects were deemed successful or not successful. It is unclear how the selection was done, how the most promising technical advances were selected for further development in HydroGEN 2.0, and what other factors contributed to selection (e.g., public communication, number of graduating students, etc.). More information on this topic would help in the future.
- The highlighted accomplishments are impressive; however, it would be beneficial to understand the likely tradeoffs between efficiency, durability, and cost, which historically has been the key challenge in most WS approaches. It is possible that this performance tradeoff concern is addressed at the individual project level.
- More effort on technology transfer, beyond filing patents, is recommended.

Question 3: Collaboration and coordination

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

- The collaboration between the five core laboratories and U.S. academia is strong. Even if not formally, the HydroGEN consortium is highly encouraged to continue to engage with the international research community through the annual workshops.
- This is definitely a strong component of the consortium. The external collaboration has greatly improved the productivity of the national laboratory system and energized the academic and industrial partners. This model should be continued and stressed.
- The current form of the consortium is extremely laboratory-heavy. HydroGEN 1.0 had 30 funding opportunity announcement projects, and HydroGEN 2.0 appears to have five to date. While the laboratories within the consortium are collaborating very well together, HydroGEN 2.0 should ensure that similar outreach and external interaction to HydroGEN is achieved. In addition, STCH is very modeling-heavy. While this is important work, and the focus of the Energy Materials Network is materials, there are cases where infrastructure and equipment are necessary to really understand material performance, such as some of the reactor facilities in Europe. The strategy should include evaluating the need for a domestic resource for these development efforts.
- The project is driving laboratory interactions.

Question 4: Relevance/potential impact

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

- The project topics represent highly relevant and multidisciplinary challenges within the WS field. By design, projects directly address the DOE Hydrogen and Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan targets and DOE Hydrogen Program (the Program) objectives.
- The goals of HydroGEN are very much relevant and consistent with the goals of the Program in advancing knowledge at all levels—theory, synthesis, characterization, and analysis—toward low-carbon and low-cost hydrogen generation.
- The benchmarking and standards component is a highlight; however, the setting of future goals for technical metrics and cost metrics is one of the more obscure processes that DOE management controls. This apparent absence of discussion within the extramural (and possibly the intramural) community could be improved so that one could recognize what is the basis for these expectations. Without such information exchange, participants will eventually come to believe that there is an arbitrary selection. The net outcome could then be disengagement.
- The community would benefit from more thought around how research in each of the different technology areas can support success in the others. The obvious mechanism is modeling, but it would be good to see this fleshed out more. The "roadmap for cross-cutting modeling" is a great way to start.

Question 5: Proposed future work

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

- The proposed future work is appropriate and builds on past progress in most areas.
- The proposed work for HydroGEN 2.0 is reasonable given this is the first year of the new funding period. Preparing a roadmap for cross-cutting modeling is worth doing; however, there could be a bit more specific and quantifiable tasks and goals. For example, future work could include growing the HydroGEN Data Hub usership by xx% or something similar.
- Fiscal year (FY) 2021 funding is down from FY 2020. It would be good to understand what research is not being continued.
- The Annual Merit Review (AMR) has been an effective means to share future plans for both technical follow-up and future funding opportunities.

Project strengths:

- The HydroGEN consortium's strength is its collaborative approach in facilitating knowledge sharing of old and new AWS concepts and projects among core laboratories and the research community, including the Data Hub approach. The consortium's cross-cutting effort to develop multiscale modeling capability to simulate performance, durability, and material properties among various AWS concepts is a good example. The consortium's effort in seeking broad input from the research/user community in developing performance benchmarks is also an important feature.
- The laboratory capabilities are some of the best in the world. Key barriers are being investigated across a wide range of technologies. The coordinated projects and tasks lead to much more effective utilization of laboratory scientists than previous models.
- HydroGEN 1.0 has been a great success in technical advances, introducing new ideas, and creating new partnerships.
- Overall, this is a great initiative to bring together all fundamental R&D related to hydrogen. Also, the work makes good use of national laboratory resources.
- The decision to focus fundamental studies on selected early-TRL areas is a strength.

Project weaknesses:

- A potential weakness is the project's lack of a defined pathway to transfer laboratory knowledge to other researchers. For example, while the super nodes were formed to address key challenges, there is no connection to the balance of the community. It is unclear how the advancements in IrO_2 roll-to-roll coating will get to the U.S.-based companies, such as Plug Power and Nel Hydrogen, or a U.S.-based membrane electrode assembly manufacturer. It is unclear how the improved solid oxide electrolysis cell (SOEC) electrode will get utilized by Oxeon or other SOEC companies. In addition, the benchmarking effort appears to disappear from HydroGEN 2.0. It is unclear how the workshops and other activities will be maintained.
- Whether this will be sustained by the newly launched HydroGEN 2.0 consortium, which focuses on national laboratory projects with no extramural component, remains to be seen. Leadership by the consortium's director will be a key in guiding future success.
- Given the commercial state of LTE PEM technology, it is not clear that the consortium should continue to invest in PEM electrolyzer component integration. The project should consider moving PEM electrolysis work to H2NEW instead. It is not compatible with the rest of the low-TRL efforts.
- It is understood that HydroGEN still supports separately funded projects via the node structure, but it is not clear how this effort is different from or synergistic to the efforts that HydroGEN funds at the laboratories.
- This project has no major weaknesses at this point.

Recommendations for additions/deletions to project scope:

- Overall, this project is great. It is recommended to include a bit more emphasis on alkaline electrolyte membrane (AEM) water electrolysis and SOEC electrolysis, given their potential for future impact on bringing the cost of hydrogen down to the $1/kg range.
- HydroGEN 2.0 and H2NEW are very valuable consortia that are needed in this competitive and rapidly growing technology space. However, with the split, it is not clear where some aspects fall. For example, benchmarking is not called out in either consortium and spans technologies in both. Also, HydroGEN 2.0 has a stated goal of materials research for AEM water electrolysis, proton-conducting SOEC, metal-supported SOEC, PEC, and STCH, while H2NEW focuses on integration for PEM and SOEC. There are still material improvements required for PEM and it isn't clear where these are covered. Materials such as membranes, gas diffusion layers, and porous transport layers still need development. These should be explicitly included in either HydroGEN or H2NEW.
- It is a good idea to focus HydroGEN 2.0 on low-TRL areas of AWS R&D and not on PEM electrolyzer technology, since it is far more advanced and commercial compared to other AWS routes. It is understood that the HydroGEN consortium, through its community benchmarking activities, has prioritized some attributes (such as material durability for PEC) more than others; however, the go/no-go milestones should

try to incorporate some form of all three key barriers that HydroGEN aims to address (i.e., efficiency, durability, and cost). Considering the basic research nature of the consortium's activities, the cost estimates may not be as firm or meaningful as efficiency or durability estimates at this early stage of technology development. Nevertheless, the consortium should still attempt to establish some sort of guardrails against potential cost-tradeoff efforts (intentional or not) by projects to achieve their efficiency and/or durability targets. For instance, if not dollars per unit hydrogen, there could be an upper limit of platinum or other precious metal loadings per unit hydrogen produced. That way, real and meaningful advances can be realized. The consortium should continue to build robust benchmarking protocols through the annual community discussions or other means on all portfolio areas (LTE, HTE, PEC, and STCH) and ensure projects frequently update their performance against the benchmark. Projects/concepts that seem to be challenged to meet the minimum benchmark performance expectations should be encouraged to quickly adjust or move on.

- The partnership between national laboratories and academic research and engineering programs has been abandoned in HydroGEN 2.0 in favor of national laboratory funding only or national laboratory and industry funding. The notion that DOE can turn on/off academic research, development, and engineering as needed is likely to create poor engagement in the long term.
- The philosophy for allocating funds among the four technology areas (AEM, SOEC, PEC, STCH) is not clear, but a strong recommendation would be to overweight on the electrolyzer technologies.

Project #P-179: BioHydrogen (BioH2) Consortium to Advance Fermentative Hydrogen Production

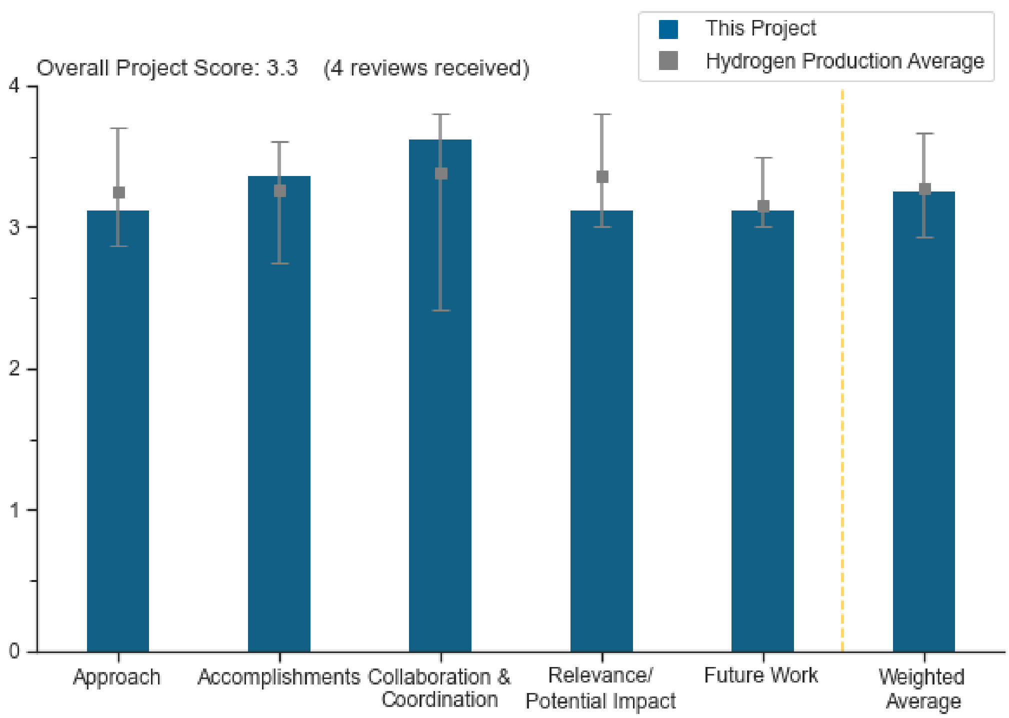
Katherine Chou, National Renewable Energy Laboratory

DOE Contract #	WBS 2.4.0.516
Start and End Dates	10/1/2018
Partners/Collaborators	Lawrence Berkeley National Laboratory, Pacific Northwest National Laboratory, Argonne National Laboratory
Barriers Addressed	• Hydrogen molar yield • Feedstock cost • System engineering

Project Goal and Brief Summary

The goal of the BioHydrogen Consortium is to develop a direct, high-solids-loading microbial fermentation technology integrated with a microbial electrolysis cell (MEC) to convert renewable lignocellulosic biomass resources into low-cost hydrogen. This collaborative team of national laboratory scientists aims to (1) improve the rates and molar yields of hydrogen production (moles of hydrogen/moles of sugar) via metabolic engineering of the cellulose degrader, Clostridium thermocellum, (2) optimize the bioreactor for high solids loading to reduce reactor cost, (3) develop an integrated MEC system to improve hydrogen molar yield and reduce fermentation waste product, and (4) conduct a technoeconomic analysis (TEA) and lifecycle analysis (LCA) with data generated by team partners to identify major cost drivers and guide integration efforts.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Production projects.

Question 1: Approach to performing the work

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

- The project is exploring several different approaches to maximize hydrogen yield. Tasks 1 and 2 are highly synergistic, and Task 3 complements by reducing some of the waste. In tandem, this makes for a nice research consortium. With hydrogen as a goal, the two-stage approach makes sense, utilizing organisms that produce hydrogen concurrently with acids and other volatile chemicals that are later converted in the MEC system.
- The team has followed a well-defined and effective approach in meeting the project goals. The members have complementary expertise and work on tackling different aspects of the challenges to develop the hybrid process to convert high-solids lignocellulosic biomass to hydrogen with a cost goal of $2/kg hydrogen. The approach is reasonable and well-executed.
- The main barriers are identified and presented, and an approach to overcoming most of them has been proposed. There are a few remaining issues that need to be addressed in the near future:
    - It is not clear if the purity of the cultures can be maintained over one/multiple cycles during fermentation.
    - It is not clear why the fermentation is operated for 30+ hours even though the maximum production rate typically occurs in the first 5–10 hours or what the reason is for the decrease in hydrogen productivity over time.
    - The approach of using a pure co-culture in the MEC is questionable. Maintaining a pure culture over several consecutive cycles and for more than 200 hours can be challenging, and whether or when an MEC was contaminated and the relative impact on the performance can be complicated to investigate.
    - The impact of the electrode catalyst on the MEC performance (slide 13) is misleading. Several different parameters, ranging from the electrode spacing and solution conductivity (solution resistance) to the anode material (carbon felt, brush, and cloth) to the solution buffer capacity (50 mM, 200 mM phosphate or carbonate buffer), can have drastic impacts on the current density in a bioelectrochemical system. It is not clear whether the current densities reported were obtained from a singular study using only different cathode materials or from different MEC configurations and setups. This can be misleading, as the TEA will then suggest to the public a focus on a specific catalyst (Mo in this case), even though the performance improvement in that specific study can be due to factors other than the catalyst. For example, a typical current density of an MEC with a brush anode and a Pt/C cathode is 12 A/m^2 in 50 mM phosphate buffer. In a different study, Ni is used, a higher-buffer-capacity solution is used (200 mM), and the current density is increased up to 25 A/m^2. However, the increase in current density will be due to the Ni catalyst and the higher buffer capacity, and it will not be correct to report that Ni is a better catalyst than Pt. A fair evaluation of the impact of the catalyst on the performance can be obtained only in a standardized MEC configuration. Moreover, there is no literature on MECs achieving up to 180 A/m^2 in peer-reviewed journals; such a high current density is around six times larger than that typically reported for MECs (around 30 A/m^2), and it is not clear how or in which conditions it was obtained. A few studies in the literature claim high current densities by normalizing the current produced by arbitrary areas and typically small areas to inflate the performance and try to make the results appear better; it is not clear from the presentation if this is the case for the high current density reported. Several years ago, the International Society for Microbial Electrochemistry and Technology community agreed to use primarily the cross-sectional area or the largest electrode area to normalize the current and power density in bioelectrochemical systems for a system with one anode and one cathode (stacked systems will have higher projected areas). Thus, it is suggested that the project use a standard configuration for the catalyst comparison to determine how the current density was obtained and whether the reported method is correct.
- Overall, the team has taken a reasonable approach to converting plant biomass to hydrogen. An initial focus on xylose utilization to increase yield was a good choice, but devoting more effort to engineering Clostridium thermocellum fermentation pathways will likely be important for the future progress. The

complexity of the solids that are left over after fermentation is concerning. It is unclear how many recalcitrant sugar linkages remain, what happens with the lignin, how these components interface with the MEC system, and how many of the electrons can be captured by the MEC. The plausibility of reaching feedstock loadings of 175 g/L is also concerning. If high loading is critical to the current TEA, the team may want to consider other processing modalities that are more feasible.

Question 2: Accomplishments and progress

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

- The project has made significant accomplishments overall in the past years and met the performance milestones. Improvements in the rate of hydrogen production through fermentation and hemicellulose utilization are impressive, and the system scale-up is well on the way. The TEA estimates provided clear guidance on major barriers to be overcome, and the team has identified clear steps in addressing the challenges and meeting the milestones. The connections between different aspects of the project in achieving the overall goals could be better elaborated.
- Better utilization of the biomass for increased hydrogen production represents an outstanding achievement. The progress in the MEC appears to be less relevant. The TEA will have a tremendous impact on the MEC community—the reviewer is not aware of any study of this kind in the literature—however, for this reason, more attention should be paid in the selection of the studies for the MEC performance in the TEA.
- The project has made significant strides in sugar conversion and hydrogen production, with a >10% increase in hydrogen production rate. The hemicellulose utilization work demonstrates impressive five-carbon sugar consumption improvements. The team has achieved long-duration demonstration of the MEC cells (>200 hours). While improvements have been made for the solids loading, significant work remains. In particular, there are concerns about how the organism will handle and be able to hydrolyze the solids at these higher levels. Task 4 details on the LCA are lacking.
- The team has generally made sufficient progress to date, but the project has a long way to go to get a functional system. The hydrogen yield's not being part of the cost sensitivity analysis is surprising, as this is presumably a major cost driver. Similarly, it is unclear what productivity is needed to be economically competitive. The project is at 2.75 L of hydrogen per day, but there needs to be clarity as to whether, for instance, a 20% increase or an order-of-magnitude increase is needed.

Question 3: Collaboration and coordination

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

- The research activity seems to be perfectly coordinated, and each participant is working on a specific task to advance the overall project goals.
- This project has a robust collaboration between four national laboratories, leveraging expertise from each team to be greater than the sum of the individual parts.
- The national laboratory teams assembled bring expertise across the value chain. National Renewable Energy Laboratory has demonstrated expertise in cellulose/hemicellulose conversion, Lawrence Berkeley National Laboratory is a leader in fermentation and strain development, Pacific Northwest National Laboratory brings experience in MEC development, and Argonne National Laboratory has core capabilities in LCA development. The consortium could benefit from industrial partners, if nothing else, to comment on the industrial relevance and viability of the approach.
- The project is carried out by a very capable team coming from four national laboratories. The consortium has a clear goal, and members are addressing different barriers to meeting the Hydrogen and Fuel Cell Technologies Office hydrogen production cost goal. Each member has made good progress individually, but more coordination could further help identify the weak links of the project, such as the relationships between fermentation effluents, compatibility with the designed pure cultures in MECs, and the overall hydrogen production yield and rates from all systems.

Question 4: Relevance/potential impact

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

- The project provides a unique approach for biological hydrogen production from renewable biomass. It complements other hydrogen production technologies and helps accomplish the DOE Hydrogen Program goals. While it is understandable that cost is still high compared to abiotic systems, the project has identified the barriers and made good progress in addressing the challenges.
- The potential impact is excellent; a few remaining points (mentioned in the approach section) will need to be addressed to further increase the project's impact on the scientific community.
- The work is very relevant for the development of technologies for hydrogen production from plant biomass. The impact will really depend on whether high titer/rate/yield, high solids loadings, and robust/cheap MECs are plausible. The MEC work, in particular, could be very impactful, but the impact of actual lignocellulose on MEC function is concerning. The team should consider prioritizing evaluation of real biomass effluent rather than Avicel effluent.
- Based on the TEA provided, it is difficult to see a line of sight to \$2/kg H_2. The overall feasibility of hydrogen from cellulosic materials seems difficult without comparison of the current state of the art (hydrogen productivity, yield, recovery, etc.) versus the ultimate technoeconomic targets. The supply chain aspects of the project are not described. Lignocellulosic materials such as corn stover are located in rural areas, but it seems unlikely that there would be the hydrogen product demand in close proximity.

Question 5: Proposed future work

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

- The team has identified many areas that need to be targeted for improvement. Improving hemicellulose utilization would certainly be beneficial, but so would increasing yield and rate. Also, it is surprising that the team is trying to eliminate "inhibitors" such as protein and alcohols (and possibly sugars) since these are going to be present in the system and they represent electrons that are not being captured as hydrogen. This could be an opportunity to increase the utility of MECs and capture more electrons.
- The project clearly identifies critical barriers to the project goals and proposes an actionable plan overall on the next steps.
- The proposed future work will address most of the remaining challenges of the project.
  - The assumption that improving the MEC cathode catalyst alone will substantially improve the performance is not convincing. The project should invest in other factors that can potentially affect the MEC performance, such as the reactor configuration. The H-cell reactors are not the best option for improving MEC performance, even though they are the most-used configuration for pure culture MECs. Using smaller spacing MECs can potentially reduce the solution resistance and improve the overall maximum current density. Increasing the electrode area/volume will also increase the performance; unfortunately, it will be more complicated to maintain the purity of the cultures.
  - The project should standardize the TEA to a single/similar MEC configuration before further implementing the current model.
- Limited time was allocated to discussing future work activities.
  - Task 1: Integration of the genes is an important step. The utility of the hemicellulose degradation process is less certain, given the strides already made.
  - Task 2: Further fermentation engineering work is relevant and necessary as the system increases solids loadings. Loadings of 175 g/L would be approaching pilot readiness.
  - Task 3: There are not enough details on what current density, lifetime, yields, and other targets are proposed for this task.
  - Task 4: Ongoing TEA will be refined, which is appropriate, as well as considering particular incentives.

Project strengths:

- The project brings DOE experts of different areas together to advance biological hydrogen production and overcome the technological barriers to meet the DOE's cost goal for hydrogen. This hybrid approach with dark fermentation and microbial electrolysis is considered the most feasible approach for achieving hydrogen goals biologically, and it targets a unique feedstock that complements other abiotic approaches. The researchers have made good progress, and they know what barriers are ahead. The TEA findings are valuable and bring insights for technology development, and the high hydrogen rates from actual biomass demonstrate the potential of applicability.
- The accomplishments with regard to hemicellulose utilization are very significant and notable and have led to significant increases in hydrogen production rate. Feeding strategies and modifications are starting to yield modest improvements in hydrogen yield. The project is well-coordinated across the member laboratories and uses appropriate steps to maximize system-level conversion of carbohydrates to hydrogen.
- The project addresses several outstanding questions in biohydrogen generation. Each member of the team appears to contribute perfectly to the overall project goal with that member's own technical capabilities. The team seems to be collaborating effectively to advance to the project goals.
- This is a strong team with expertise that spans all of the areas needed for this project. The team is focusing on some of the major challenges that need to be addressed to further develop this technology.

Project weaknesses:

- The overall concept of producing hydrogen (with no co-products) from cellulosic materials seems daunting from a technoeconomic perspective. Based on the data provided, it is difficult to see how hydrogen could approach $2/kg from this approach, even if every metric and target were achieved. The project lacks industrial partners to inform project targets or other risks and barriers. The presentation does not lay out clear technical targets for key performance parameters such as carbon conversion in the MEC and hydrogen production rate in the dark fermentation step. LCA was not presented.
- The project could better elaborate and carry out coordination between different tasks, such as:
    - How to better design an overall system that can convert cellulosic biomass more efficiently to hydrogen (and CO_2) with defined microbial cultures. The current MEC co-culture can be optimized to convert fermentation effluent more effectively.
    - How the two systems can be scaled together to take advantage of the liquid flow connection, as well as hydrogen production.
    - How to make sure the two reactors are compatible in terms of flow rate, size, product reconciliation, etc.
- A better understanding is needed of the importance of yield and productivity in the TEA. The impact of real lignocellulose effluents on the MEC will also be critical to determine.
- The main weaknesses to address are in the approach, as has been reported in that section. There are no other apparent weaknesses.

Recommendations for additions/deletions to project scope:

- There could be value in performing reactor modeling, especially for gas delivery, which could become a significant mass transfer issue at larger scales. Rheology will become an issue at higher solids, and alternative reactor designs should be considered (e.g., paddle reactors). The team might consider scaling some of the steps with industrial partners (e.g., at existing corn ethanol refineries or breweries) to further scale and de-risk particular unit operations. Market assessment and viability should be considered. It is unclear what distribution/transportation costs would need to be considered for the hydrogen produced.
- The project should investigate TEA parameters and prioritize testing of lignocellulose effluents in the MEC.
- No addition or deletion is recommended.

Project #P-182: Binary Chloride Salts as Catalysts for Methane to Hydrogen and Graphitic Powder

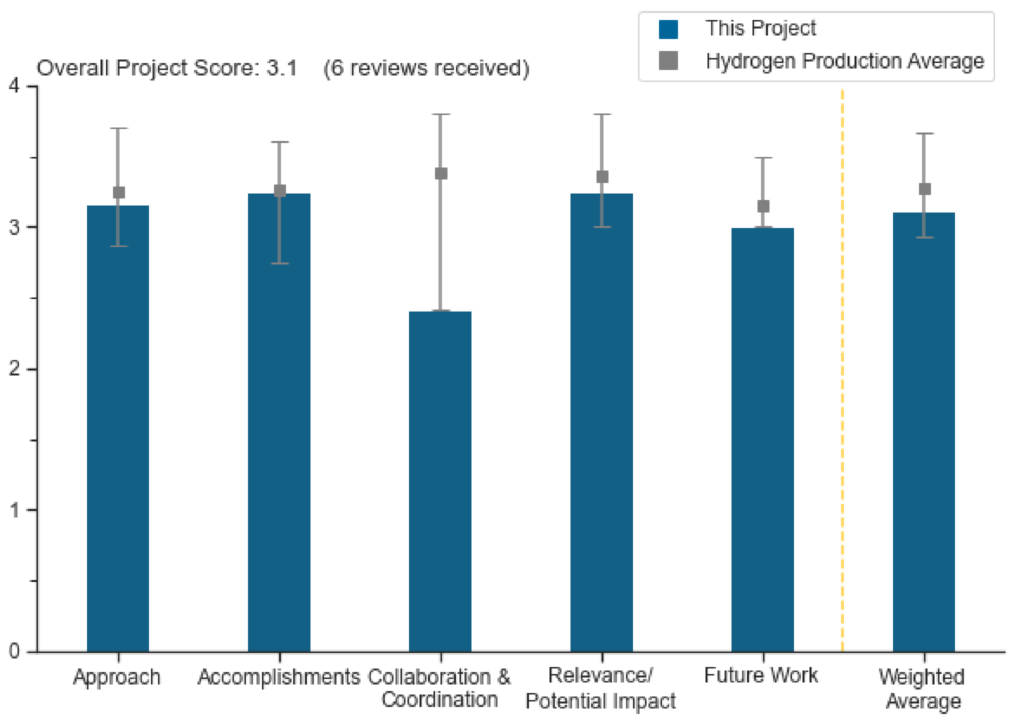
Eric McFarland, C-Zero, LLC

DOE Contract #	DE-EE0008845
Start and End Dates	12/1/2019–5/31/2022
Partners/Collaborators	University of California, Santa Barbara
Barriers Addressed	• High-temperature robust materials • Material and catalyst development • Chemical reactor development and capital costs

Project Goal and Brief Summary

This project aims to develop a scalable methane pyrolysis process that produces inexpensive low-emission hydrogen from natural gas. Ideally, the process will also result in a useful byproduct: graphitic carbon with properties favorable for battery anodes and additives. If successful, this project could reduce the carbon dioxide (CO_2) emissions associated with hydrogen production from natural gas and facilitate CO_2 removal in areas not amenable to CO_2 sequestration.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Production projects.

Question 1: Approach to performing the work

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

- A simple and accurate identification of the project objectives and critical barriers was presented. It would be very helpful to identify significant elements that underlie each barrier to more clearly demonstrate the comprehensive nature of C-Zero, LLC's (C-Zero's) efforts. The carbon product is defined as battery anodes and additives. The correlation between barriers, tasks, and approaches to achieving U.S. Department of Energy goals would be helpful.

- The project develops a molten salt pyrolysis process that decomposes natural gas to produce hydrogen and solid carbon. Because less hydrogen is produced from the decomposition process than from steam methane reforming (SMR), where part of the produced hydrogen comes from water, product carbon needs to be valorized to be cost-competitive with SMR. The team realizes this and is looking to produce graphitic carbon, which can be used in batteries. There should be closer correlation between the salt development task and the carbon properties task. The salts should be selected based not only on the catalytic activity toward methane decomposition but also on the morphology and cleanliness of the produced carbon, which should be important criteria.
- Although natural gas pyrolysis concepts are not new, the use of binary salt catalysts is an interesting approach that aims to reduce energy input and CO_2 emissions per unit of hydrogen produced. However, the approach on how to achieve the second objective of making a "graphitic carbon product that has properties favorable for battery anodes and additives" is vague. It is unclear whether the bimetallic test runs were aimed at optimizing hydrogen yield, graphite carbon quality, or both. The project seems to aim its process as an alternative to traditional CO_2 sequestration. It may be wise to settle quickly on what to do with the carbon product. The research approach for simple carbon sequestration or disposal is vastly different from targeting revenues from battery electrodes that require a high-quality carbon product with well-controlled metallic impurities or even for the proposed future work as cement additives.
- The binary/ternary salt carbon decomposition has been demonstrated and will work. The team should test real natural gas in addition to methane to better understand the impact on the process. The quality of the carbon produced will likely be low. A task on potential carbon uses and/or on how to improve the carbon produced would be useful. The carbon separation will be a tremendous problem. The research team is targeting 75% reduction in CO_2 emissions compared to SMR. The researchers are planning on using renewably powered electric heating. It is unclear what CO_2 emissions are being generated by the project so that it is not at a higher CO_2 emissions reduction.
- The approach is proprietary and unlikely to contribute to general knowledge.

Question 2: Accomplishments and progress

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

- The accomplishments and progress toward the overall project and DOE goals were impressive and include the following:
    - The melt system design was completed.
    - The catalytic action and mechanism were identified.
    - Catalytic activity was determined to be greater than the milestone goals (<250 kJ/mol).
    - The project operated a molten salt system at a high temperature and pressure.
    - Carbon analog was removed from the aqueous system (~500 g/hour).
    - The technology was successfully demonstrated in a high-temperature molten salt system.
    - The centrifugal carbon removal system demonstrated less wet salt in the carbon product.
    - The carbon produced was tested for both battery anode use and for cement additives.
    - The project updated and refined the Aspen model with the ongoing process, with the goal of a process showing <$2/gge hydrogen production and carbon production cost of <$1/kg.
- In spite of the difficulties imposed by the COVID-19 restrictions, the project made very good progress in the first year. The team has assembled the melt system activity apparatus and started measuring binary chloride salt activity. The computational evaluation of the salt systems to guide the composition development is done in parallel. The project designed and demonstrated high-pressure operation at up to 17.7 bar pressure with a methane feed. The team also started verification of the types of carbon produced. In Task 2, the team is developing and testing multiple approaches to carbon removal. Carbon analysis of the produced carbon for electrochemical applications demonstrated that graphitic carbon has been

produced, yet still the conductivity properties are not as good as those of the commercial graphitic carbon. Good overall system integration analysis has been performed using Aspen Plus system modeling.

- The project's overall goals are consistent with DOE's goals. The identification of the binary melt systems, as well as the reactor testing at high pressure and temperature conditions, is encouraging.
- This project had good progress during its initial year, which was during the pandemic (although it is not clear whether any progress results from efforts prior to the award).
- Since this project just started, it is hard to judge the accomplishments. The project is addressing some key issues. The operation temperature of 1000°C is very high and will cause material compatibility issues. The carbon being made is not crystalline carbon. This results in very low-quality carbon that has low value. For the carbon used in the electrochemistry tests, it would have been good to know if the team cleaned the carbon to remove the salts or if the carbon was used without any processing. The way the discharge capacity is reported on slide 14 is somewhat misleading. While it is true that the initial capacity is around 279 mAh/g, the shape of the I-V (current–voltage) curve is such that the realistically usable capacity is closer to 110 mAh/g. In addition, the project reported only the first five cycles. Lithium-ion batteries typically go through five to ten cycles for a break-in period, so the actual capacity in a real system would be lower. The researchers have shown some initial concepts for the carbon separation but have a long way to go for a real solution. The amount of salt removed with the carbon was not reported. The process and economic modeling are for conditions different from those used in the experimental tests and assume much higher conversion than what is being achieved. The natural gas, landfill gas, and biogas will need a good deal of cleanup prior to use. The flowsheet does not include any cleanup prior to use, which raises the question of whether the costs are captured. It is also unclear what the "LT processing" in slide 16 is.

Question 3: Collaboration and coordination

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

- There is good work sharing between C-Zero and the University of California, Santa Barbara (UCSB). The team needs to look for industrial partners, such as Cabot, especially for carbon applications.
- The project could greatly benefit from having some input from subject matter experts in the natural gas processing and conversion sector, such as refineries, hydrogen plant operators, or industrial gas companies, as well as battery electrode manufacturers.
- It is recommended that the team make further use of national laboratory resources on high-temperature materials and process equipment. There are quite a few similarities with work done in the past on solar thermal.
- There is some collaboration between C-Zero and UCSB. It is unclear what the UCSB collaborators are doing for the project.
- The presenter did not identify or discuss significant activities in collaboration and coordination with other institutions. Collaboration could reduce the project costs and decrease DOE project risks, as well as overall C-Zero risks associated with the burn rate required to support 30 or more employees over the life of the project and beyond.
- Collaboration was discussed only briefly. Its impact on the project is not apparent.

Question 4: Relevance/potential impact

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

- The project has a potentially high impact due to the large availability of natural gas versus renewable electricity. Hence, hydrogen produced via pyrolysis could be a strong alternative to green hydrogen.
- The project is targeting production of hydrogen at <$2/kg and a 75% reduction in CO_2 emissions relative to SMR.
- The process is relevant to DOE's ultimate goals, although it is unclear what technical goals the project is trying to address. The stated potential impact of C-Zero's process as "75+% reduction in CO_2 emission from hydrogen production from natural gas" is well-supported. It looks unreasonably high, given the

nonsignificant energy input to drive the high-temperature pyrolysis process, heat recuperation, and multiple separation steps. The team should clearly show the impact of the process with high-level mass–energy balance compared to conventional SMR.

- An updated and refined Aspen model was reported as progress to the goal of a process toward <$2/gge hydrogen production and a carbon production cost of <$1/kg. The Aspen model results need to be presented and supported in greater detail. The Aspen mass and energy balance was not offered in support of progress. A defined path toward the DOE goal could be helpful.
- The reduction-of-CO_2 target of only 75% seems inconsistent with the Hydrogen and Fuel Cell Technologies Office goals of low or no CO_2 emissions. The team should consider electric heating and recycling of the unreacted natural gas to reduce CO_2 emissions.
- The project will need to target greater than 75% reduction in CO_2 versus the benchmark (unabated SMR).

Question 5: Proposed future work

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

- The project's plan to continue work on each task is logical and efficient.
- Given that the project has less than a year left, the project team may consider narrowing the focus of the remaining work on optimizing bimetallic melt catalysts and process conditions around hydrogen yield and efficient catalyst recycling (i.e., assume for now disposal of solid carbon byproduct). Going after testing for battery electrode or cement additives may be distracting from the primary DOE goal. The proposed technoeconomic analysis and process modeling tasks are reasonable.
- The team needs to develop a viable carbon separations approach. The filtration approach has some potential, but the researchers did not discuss how they will recover the carbon from the filter. The carbon will likely need further processing to make it usable, which was not discussed. It was not clear in the economic analysis how much loss of the salts was planned or is acceptable. The project proposed using the carbon in cement, but it is not clear whether trace amounts of the salts will have a negative impact on the cement.
- The technoeconomic analysis, from Task 5, may need to wait until the carbon separation problem from Tasks 2 and 3 is solved.
- This reviewer would seriously challenge the use of pyrolytic carbon for battery applications. Impurities in the produced carbon would be a bit too high for battery anodes.
- Task 3 is not defined or discussed.

Project strengths:

- The project effectively combines scientific research on molten salts and the methane pyrolysis process with an engineering development of the reactors, separation systems, and overall project integration. Proper attention is given to mass–heat balance and how it affects the carbon properties and the overall process economics.
- Overall, C-Zero is a very strong organization that has a strong team working on this project. It is not surprising that the team has produced such robust results. Whatever the challenge is, the team is capable of achieving remarkable results.
- It is appreciated that the team is already looking at pilot-scale opportunities and related process modeling. That will lead to many learnings along the way.
- The main strength of the project is the capability to test high-temperature and pressure pyrolysis reactor systems.
- This is a well-funded project with a very large team. The concept is very innovative.
- The focus on fundamentals and key hurdles/showstoppers is a strength.

Project weaknesses:

- The project team is very large and seems management-heavy (seven managers/directors/associates and six scientists/engineers). Carbon separation is a major issue. Operating at pressure will increase the difficulty of the separations. The researchers need to describe any cleanup they are doing with the carbon for additional salt recovery or other processing to improve the carbon quality. The high temperature and use of molten salts may produce material compatibility problems. The project should consider recycling the unreacted methane rather than burning it to produce heat. The team needs to test real natural gas to better understand the process. The natural gas, landfill gas, and biogas will need a good deal of cleanup prior to use. The flowsheet does not include any cleanup, so it is unclear whether the costs are included in the analysis. The carbon produced is low-quality.
- The approach is simple and elegant at first blush, so it is easy to underestimate the actual difficulty of what is being attempted. Issues such as thermal management, materials handling, media and product contamination and purification, carbon morphology and uniformity, and component durability present significant challenges. It is unclear that the team has a comprehensive plan that will carry the project all the way to a commercially viable system.
- The greatest project uncertainty and challenge remain the molten salt composition and the ability to produce the desired carbon morphology and purity. More efforts should be directed toward finalizing the salt composition, as it may affect the rest of the project tasks.
- Because of the relatively high operation temperature (1100°C) of the pyrolysis unit, the team needs to pay further attention to what materials and equipment reliably work under those conditions at scale. Finding equipment fit for this purpose may not be the easiest.
- The presentation lacked discussion of the scale mismatch between hydrogen production and the solid carbon market being targeted (battery anodes).
- The project is trying to demonstrate too many unproven early-stage concepts in such a short time. The team should note that, to date, there are no pyrolysis-based commercial processes to make hydrogen or carbon products.

Recommendations for additions/deletions to project scope:

- Future work could include thermal integration pilot system design, including pressure swing adsorption tail gas heat recovery, resolution of potential coking issues, assessment of potential toxic waste production, salt recovery and purification for reuse, and characterization of produced gas content based on various feedstock assumptions. Additionally, the team should also consider how to achieve uniform pressure, temperature, bubble size, and space velocity to produce carbon with a more consistent, precise, and higher-value morphology.
- Considering the relatively short funding period, the project needs to focus on narrower and more achievable tasks to advance the key concepts. The project could choose to address potential scale-up challenges, including the reactor system, solids separation, catalyst recirculation, and product gas purification. The project should also clearly demonstrate the energy and greenhouse gas reduction benefits of its process against conventional SMR.
- Based on the energy analysis, it seems that more of the effort should go toward solving the carbon separation step (Tasks 2/3). Optimization of the salt composition (Task 1) and assessing the carbon as a cathode material (Task 4) can come later.
- The team should look further at the materials and corrosion aspects of the design. Several pathways (hydrogen embrittlement, halide-induced corrosion, carbon deposition, etc.) might severely limit the scale-up and increase the probability of unsafe conditions.
- No changes are recommended.

Project #P-183: Extremely Durable Concrete Using Methane Decarbonization Nanofiber Co-Products with Hydrogen

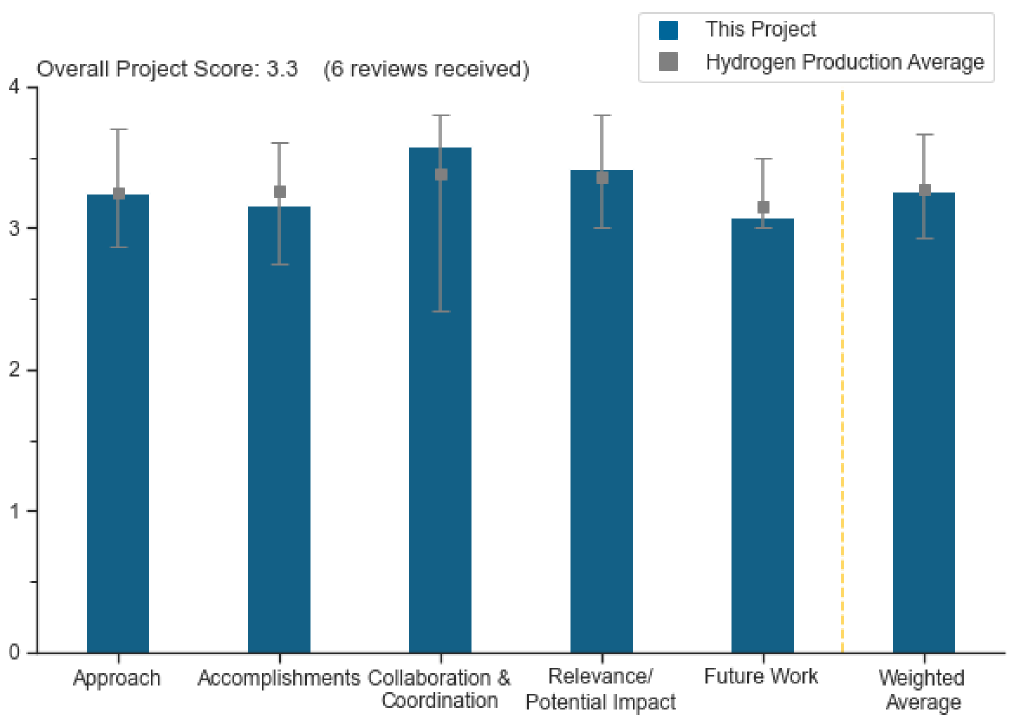
Alan W. Weimer, University of Colorado, Boulder

DOE Contract #	DE-EE0008846
Start and End Dates	5/1/2020 to 5/1/2023
Partners/Collaborators	Forge Nano, Inc., National Ready Mixed Concrete Association
Barriers Addressed	• High-temperature robust materials • Material and catalyst development • Chemical reactor development and capital costs

Project Goal and Brief Summary

The University of Colorado is developing a scalable, low-cost chemical vapor deposition (CVD) process to produce carbon nanofibers (CNFs) and hydrogen from methane using a sacrificial atomic layer deposition (ALD) catalyst deposited on a fumed silica substrate. This process offers a cleaner alternative to steam methane reforming, as it allows hydrogen to be produced without releasing carbon dioxide through the co-production of CNFs. The CNFs sequester the carbon and can be added to a concrete mix to improve durability and performance, offering a value-added byproduct. The project will design a commercial path forward for a typical hydrogen plant producing 480,000 kg H_2/day, with 70% conversion efficiency of CH_4 to H_2, and will identify potential industrial collaborators and customers.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Production projects.

Question 1: Approach to performing the work

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

- A brief, precise, and detailed identification of the project objectives and critical barriers is presented. It is a brilliant approach that is clearly described and very well-organized. Goals, barriers, approach, tasks, and milestones are clearly and precisely connected.
- The project is developing the process for growing CNFs on fumed silica, coated with sacrificial Fe (or Ni or Co) catalyst through the ALD and using this CNF to improve the performance of concrete. In situ catalyst synthesis will be conducted from a metallocene precursor. Other projects are developing similar processes for carbon nanotube (CNT) growth on stand-alone Fe nanoparticles. The project should compare the advantages and disadvantages of having Fe supported on Si rather than just Fe catalyst for the CNF growth and for the concrete stabilization. Ferrocene is a rather expensive feedstock, so other precursors and techniques to generate catalyst nanoparticles should be investigated. In the proposed process, it is likely that each Si bead will support a large number of CNFs, which are likely to entangle and may not be easy to disperse in the concrete.
- The idea is very interesting: methane pyrolysis for carbon nanofibers for cement. The use of silica seems like a reasonable approach, but the choice of nickel seems relatively expensive, especially since the project will not be recovering its catalyst. Even if the process uses only a very little catalyst, it will be producing many metric tons of carbon fibers, and it will soon add up. It is not clear why ALD is required; it might make sense to use a less expensive catalyst synthesis technique. It is not clear that ALD can be economically scaled to the level needed for this process to be commercially acceptable.
- The project's approach and goals are clear. The three tasks—namely, CNF/hydrogen production, CNF use, and technoeconomic analysis (TEA)—are well-defined and aim to address specific technical challenges. However, the ultimate feasibility of the scale-up process may be doubtful, given the inherent dependence of the CVD process on vacuum or clean-room-type environments, which tend to be rather expensive to operate and more common in high-value products compared to commodity products such as hydrogen or concrete.
- It is great that the intent to jointly evaluate a process concept with the utility of the product in a high-volume application. However, it is not clear from the presentation that the research plan will support a direct test because of a mismatch in scale.

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 presentation provides detailed documentation of significant progress achieved on challenging tasks:
  - Construction and start-up of in situ ALD/CVD reactor systems
  - Deposition of ALD catalyst onto fumed silica
  - Demonstration of in situ carbon growth
  - Completion of preliminary TEA and analysis via the Hydrogen Analysis (H2A) model.
- All Task 1 objectives for the first year have been met, with the exception of the characterization of CNF produced in the process. It is important to get to the CNF characterization as soon as possible, as these CNFs may be very different from the commercially acquired CNFs that are currently used in the concrete testing. A good set of testing has been performed on the effects on concrete properties of adding CNFs. Uncertainty remains as to how much these results could be reproduced with the actual CNF obtained from the pyrolysis process. The project is using the Aspen Plus simulation for the system integration development. The gas pre-heat heat exchanger (HX) needs to be carefully considered. It appears in the process schematic that the 900°C reactor effluent flows to that HX. This will cause coking of the incoming gas and plug the tubes.
- The project has well-defined tasks and milestones, and it has demonstrated progress toward project objectives and achievement of DOE goals.

- The methane conversion seems low at 20%. It would be helpful if the space velocity was reported. On slide 15, the presenter says "...as catalyst deactivates." The purpose is growing CNFs on the catalyst, so it does not seem like the catalyst is deactivating; it seems to be operating as expected. Also, the catalyst synthesis seems expensive. It is not clear that the ALD process is needed. The cement testing with the commercial CNFs was interesting. It was unclear why the CNF percentage in the cement changed from 0.1 wt.% on slide 17 to 1 wt.% on slide 18. The flowsheet does not include natural gas cleanup. This needs to be included in the analysis. The TEA is interesting. The cost assumed for the catalyst synthesis using ALD should be shared, as well as the percentage of the operational cost it represents. The CNF cost range was \$1.50–\$2.50/kg, and it was not clear that the improved cement characteristics support the increased cost of adding this additive. The team needs to be careful when comparing the CNF cost to the price paid for the commercial CNFs; cost and price are very different.
- It would have been good to see some evaluation of the silica particles with tethered CNTs that is the basis for the project's approach. It is suggested this be an immediate focus.

Question 3: Collaboration and coordination

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

- The project demonstrates close collaboration between participants from the chemical, biological, and civil engineering departments of the University of Colorado, Boulder. The project also collaborates with Forge Nano, Inc. (Forge Nano) for the reactor and process development, along with the National Ready Mixed Concrete Association (NRMCA) on concrete properties and testing.
- Forge Nano and NRMCA are strong partners for overcoming both technical and commercialization challenges.
- The internal collaboration is applauded, even if the activities are not synched up so far. Two external partners are mentioned as well, but it is not apparent what impact they had on the progress.
- The collaboration with NRMCA is really appreciated, as it gives strong credibility to the durable concrete workstream.
- The project has clearly defined roles. The collaboration seems to be going very well.
- The project collaborated with Forge Nano and the NRMCA. However, to better inform the TEA, the project could benefit from additional discussions with other experts on ALD/CVD scale-up challenges and related greenhouse gas (GHG) lifecycle analysis (LCA).

Question 4: Relevance/potential impact

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

- The project offers the potential to develop a large-scale hydrogen production process competitive with steam methane reforming (SMR), while drastically decreasing the amount of carbon dioxide emissions. The market for use of co-produced carbon in concrete production is large enough so as not to be saturated by the amounts of hydrogen production consistent with H2@Scale.
- The sufficiently large scale of the concrete market and the value-add associated with the CNF/silicon product make this approach highly impactful. The estimated cost of hydrogen is low enough to make this approach economically viable. However, the feasibility of scaling the proposed process to the match the potential market is uncertain at this stage. Additionally, the economic impact of the various catalyst options is unclear. A sacrificial iron catalyst should be much more economically viable than a nickel catalyst and may be essential to commercial adoption. This issue was not sufficiently discussed.
- This process may result in a low-carbon-intensity hydrogen that captures the carbon in structural materials, effectively sequestering the carbon.
- There is potentially high impact for pyrolysis as a low-carbon-intensity alternative to green hydrogen.
- The intended use of solid carbon as a concrete additive has the scale potential to be relevant in a hydrogen economy.

- The research objectives are relevant; however, it is highly unlikely that the process will have the stated impact of the "potential to displace U.S. hydrogen production by SMR with a low-cost and scalable CVD process" any time soon, given the current state of the technology and the mere scale of the existing SMR-based hydrogen, domestic or global. When considering GHG impact, the project team should incorporate the LCA of the whole process, not just "CO_2 produced directly from CVD of CH_4," such as excess reactants associated with the sacrificial catalyst feed (which appears incredibly significant at more than 600,000 metric tonnes per year for the commercial plant).

Question 5: Proposed future work

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

- The future work should assess the economic impact of the various catalyst options on performance and production costs. It would be good to know the expected incremental impact, if the system is developed at scale, on Fe, Ni, and Co global demand and on prices. A conceptual design for a commercial-scale (10,000 ton/d) CNF/silicon production system should be developed.
- The project should focus on low-cost catalysts and avoid expensive ones such as nickel and cobalt. The researchers should not look at cobalt (slide 22 states that they will). There is already high demand for cobalt and nickel. Using cobalt and nickel to make cement does not seem like the best use of these valuable metals. The researchers should do some tests with real natural gas as part of their future work. The researchers should compare the ALD synthesized catalyst to the conventionally synthesized catalyst to see whether the ALD process is needed.
- The process needs to start generating and analyzing sufficient quantities of CNF from the ALD process as soon as possible to make sure that the CNF is consistent with the properties of the commercial CNF. More effort should be put toward a lower-cost Fe catalyst precursor and Fe delivery into the reactor.
- The project's impact would benefit from the following two efforts, which were not seen as part of the plan: (1) estimating the cost of the ALD catalyst and (2) testing, or at least modeling, the impact of the expected filler morphology (silica particles with CNT hairs) versus free CNT.
- The future work looks good, but it could include a rough estimate of an LCA of the logistics and impact of a sacrificial catalyst on the concrete performance.

Project strengths:

- The project develops a large-scale process for low-carbon-intensity hydrogen production by natural gas pyrolysis, which can be competitive with SMR and have a large enough market to be consistent with the large-scale hydrogen economy.
- This project's strengths include the following: the University of Colorado, Boulder, team and other collaboration partners; the well-organized approach and tasks; and the integration of methane pyrolysis CNF production with a potentially high-volume end-use product (ultra-strong cement) that suggests potential commercial viability.
- The project's strength is the overall project team mix of chemical engineers and subject matter experts (concrete and ALD) who contribute to the project team. The results from mixing CNFs into concrete seem promising.
- The project's strength lies in its clarity of tasks and the potential to carry out all three tasks in parallel.
- This project has a strong team and addresses a critical need.
- The combined research on process and product is this project's strength.

Project weaknesses:

- The team is using an expensive process (ALD) to make a catalyst. It is not clear that if it is successful, the ALD process could be economically scaled to the level required for cement manufacturing. The current work is using nickel, and the team is proposing using cobalt. Less expensive alternative sacrificial catalysts should be considered. The researchers need to report more details on the TEA to validate their findings. The TEA is assuming process performance that is not yet achieved.

- The scale-up pathway does not have an existing market along the scale of the cement market. Metal catalyst by ALD is not being practiced at the scale of the cement market. Cement is a bulk commodity that, as such, cannot tolerate price increases.
- The TEA assumptions are almost too hopeful. Instead of assuming ideal parameters such as DOE hydrogen targets, the team should consider a scenario with assumptions of current natural gas and CNF costs with a 20- to 30-year lifetime to come up with a more realistic hydrogen estimate. Currently, there is no commercial hydrogen manufacturing process involving pyrolysis of natural gas, so the TEA should reflect realistic challenges common to new technology platforms. A minor criticism is that there seems to be some confusion around the definition of the gas conversion milestone (and thus accomplishment) with respect to the methane-to-hydrogen volume ratio as stated in slides 5 and 11. It is not clear whether it is 20% or 80%.
- For the TEA, it is recommended that the team use economic steering values (capital cost, indirect costs, etc.), which are the same as the H2A model. Otherwise, comparisons with the $2/kg target price will be difficult.
- This project's weakness is the mismatch in material availability, specifically the amount of CNT hairy silica particles that can be made versus the amount needed to conduct meaningful evaluation in concrete.
- There is still significant uncertainty about the quality of the CNF that can be produced by the process.

Recommendations for additions/deletions to project scope:

- Assuming additional technical progress and a viable TEA update, a commercial-scale concept could be developed to attract additional funding. Even at this early stage, a line of sight to commercialization is important.
- It is not clear whether the sacrificial catalyst ALD/silica fume is separately manufactured or whether it is designed to be part of the commercial plant. The project team should consider carrying out high-level GHG LCA, including, but not limited to, the logistics of hauling a significant volume of the sacrificial catalyst, ~14 kg/kg of H_2 produced. Task 2 should separately explore the impact of the sacrificial catalyst ratio on the concrete performance. For Task 2's fiscal year 2021 go/no-go, the team should consider alternative concrete tests using simulated additives (e.g., commercial CNF + fumed silica + metal catalyst mixture) in the event that suitable quality and/or volume of CNF from Task 1 is not realized.
- The team should focus on low-cost catalysts and avoid expensive ones, such as nickel and cobalt. The researchers should not look at cobalt (slide 22 states that they will), given that there is already high demand for this material. Using it to make cement does not seem like the best use of this valuable metal. The researchers should make some catalysts using conventional high-volume catalyst synthesis techniques and compare their results to see whether the expensive ALD process is required.
- The project may benefit from collaboration with the Carbon Hub run by Rice University, where similar concepts are being developed and similar issues and challenges are considered.
- A TEA of catalyst production should be added.

Project #P-184: Scalable and Highly Efficient Microbial Electrochemical Reactor for Hydrogen Generation from Lignocellulosic Biomass and Waste

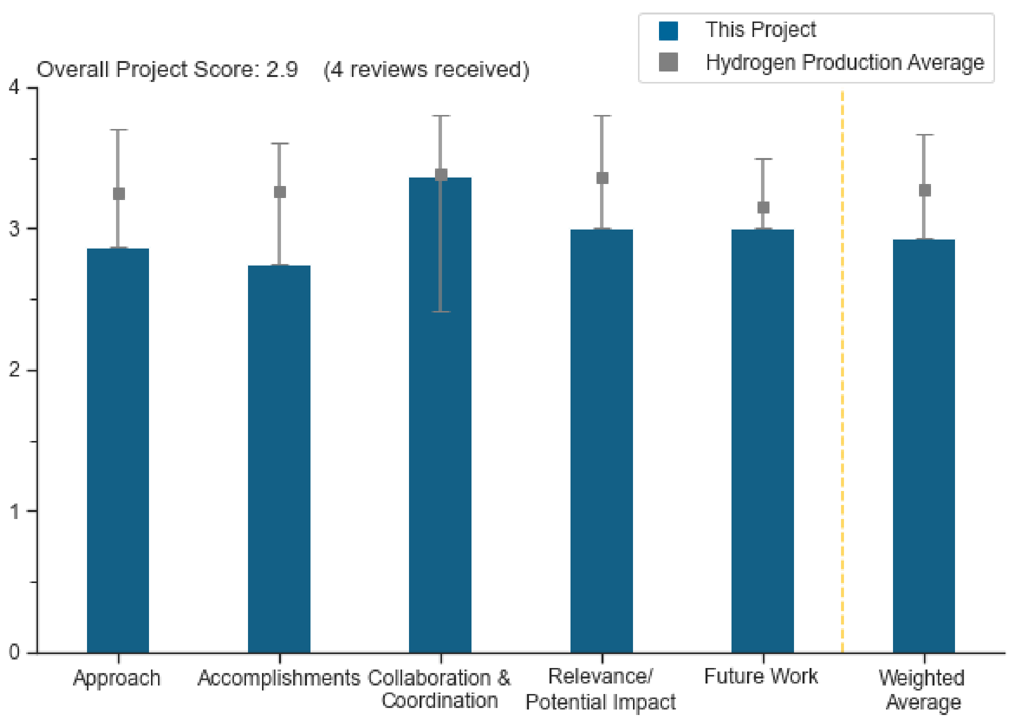
Hong Liu, Oregon State University

DOE Contract #	DE-EE0008844
Start and End Dates	1/1/2020 to 12/30/2023
Partners/Collaborators	Texas A&M University, Pacific Northwest National Laboratory
Barriers Addressed	• High electrode cost • Low hydrogen production rate

Project Goal and Brief Summary

This project is developing a scalable hybrid microbial electrochemical reactor to produce hydrogen from waste streams. The reactor design combines fermentation and microbial electrolysis cells (MECs) and includes low-cost electrodes and catalysts. Robust microbial communities will be used to optimize operating conditions, reducing the operating cost. This project will provide a method of producing hydrogen from waste streams at a cost of close to or less than $2/kg H_2 (the U.S. Department of Energy target).

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Production projects.

Question 1: Approach to performing the work

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

- If successful, the conversion of wastewater to hydrogen has the potential to be both cheap and sustainable. Printing carbon nanotubes (CNTs) on steel mesh to increase conductivity and decrease cost seems like a good approach, owing to the greatly increased surface area of CNTs as compared to carbon fiber. Because this will be expensive to build, the team modeled a 10-year lifespan for the electrode, but it is unclear whether this is plausible, given the fact that the current version loses 25% activity in 50 days. Approaches to determining the cause of the decreased activity over time, as well as approaches to solving this issue, will be critical to demonstrating improved lifespan and commercial potential.

- The project identifies that cathode material is a key barrier to MEC scale-up and focuses on addressing this issue. The team is well-qualified to perform the tasks and has made good progress overall. The MoP_X-based catalysts have gained good interest in hydrogen evolution in recent years.
- The approach sounds rational and articulated to overcome the most limiting barriers in an MEC.
    - The scalability and application of the CNT-based anode in this project seem questionable. Currently, the team can develop an electrode with an area of a few square centimeters, and the scaling up of the equipment for the CNT growth will increase the complexity and the cost for the electrode development. From the performance evaluation, it also seems that the CNT anodes do not perform statistically better than the carbon cloth anodes, and the CNT anodes' performance quickly decreases over time. Based on these considerations, the CNT approach, even though extremely interesting, might need to be redirected.
    - The polymer coating of the CNT appears to increase the mechanical strength of the material; it is suggested that the project investigate whether such modification also changes the electrical properties (conductivity) of the material.
- The work to develop a new CNT fabrication approach is novel and could have benefits beyond this project. The project does not contain lifecycle analysis to compare the hydrogen produced from this method to steam methane reforming (SMR) or other sources. Waiting until late in the project—when real wastewater is being used—is a significant weakness and risk to the overall project. The project should explore the impacts of fouling, poisoning, etc. as early as possible so that if mitigation steps are necessary, they can be considered.

Question 2: Accomplishments and progress

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

- The researchers made good progress on catalyst development, and they have compared the performance with existing materials. The CNT and MoP_X ideas are interesting and new, and they showed potential in improving hydrogen production. The current data show that the performance in terms of hydrogen production from these new materials is not significantly higher than benchmarks, so more improvements will be needed, and the team has shown the project plan to do so. The longevity of the materials soaking in the electrolyte is not clear. There have been studies reported that CNT could fall off after a certain period, which in this case may cause issues. The technoeconomic analysis (TEA) could give more details on comparing with current benchmarks so one can better understand the cost levels.
- The team has demonstrated increased conductivity at much lower costs by using CNTs on the stainless steel mesh. Other improvements have been conferred by the CNT approach, including mechanical strength and pore size control. These results are muddied somewhat by some impacts of the coatings. The data on corrosivity are useful, particularly exploring 50 to 100 cycles. In the question-and-answer session, it was explained that 50-day testing on acid whey has occurred and that there is an accelerated testing system. No data were presented, even on mock streams, about how much biological oxygen demand (BOD) reduction or waste carbon conversion was realized.
- Several excellent accomplishments were obtained toward the overall project goal. However, a few results presented do not allow for fair evaluation of the progress made, such as using high buffer capacity and substrate loading that are not representative of real wastewater feedstocks (acid whey in 200 mM phosphate buffer is not a real feedstock), and claiming excessively high current density that does not reflect the real performance of the cell.
    - The exceptional performance of the MoP_X cathode compared to Pt represents an outstanding advancement.

  - The anode performance steeply decreased over time, reaching 48 A/m^2 in 150 hours, but decreased to 35 A/m^2 after only an additional 50 hours. A similar decrease can be observed in the larger-scale system (12.5 cm^2) where the carbon cloth outperformed the CNT at 1,200 hours. The reason for such a steep decrease, particularly for the CNT electrode, could be further explicated.
  - It is not evident that testing the MEC or anode performance in 200 mM phosphate buffer will produce insightful results for evaluating the performance of the system in real wastewater, typically characterized by a low buffer capacity.
  - On slide 16, current densities ranging from 80 to 180 A/m^2 were claimed. These current densities are around five to six times larger than the highest current densities ever claimed in MECs (30–40 A/m^2). The carbon cloth used here has been used previously in MECs with current densities never exceeding 20 A/m^2, and the principal investigator reported on slide 15 that the anode current density cannot exceed a stable current density of 35 A/m^2. It is not clear how such a high number was calculated. (It is well-known that the current density cannot be calculated by normalizing the current by the smallest area in the reactor, as this inflates the current density, resulting in unreproducible results). It is also not clear whether the extremely high current density that was reported was due to a biotic electrochemical reaction or the abiotic water-splitting reaction. With a current density claimed here and an electrode packing density of 28 m^2/m^3, the theoretical hydrogen production rate can be calculated as follows: 180 A/m^2 * 28 m^2/m^3 = 5,040 A/m^3 * 86400s = 435,456,000 C/m^3 / 96485 F / 2 nH_2-e^- = 2256 mol H_2/m^3-d = > 50 L/L-d. With a packing density of 100 m^2/m^3, the hydrogen production rate will be 180 L/L-d, which seems unrealistic.
- Good progress has been made on the electrode development, but a more balanced focus—that includes work on the microbial community—is needed.

Question 3: Collaboration and coordination

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

- The team seems well-suited to performing this work, and the roles of each partner are well-defined.
- The team members have complementary areas of expertise, and they work well to advance the different tasks to achieve the common goal of the project.
- Each team member is operating on a different aspect of the project (cathode, anode, configuration, and microbial community) to advance the final project goals.
- The project team and institutions involved have experience relevant to the MEC design. The project could benefit from collaborations with wastewater treatment plants, environmental engineering firms, or other industrial entities. Likewise, hydrogen customers could be useful as well, to assess the market potential for a process if it becomes successful.

Question 4: Relevance/potential impact

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

- The project aligns well with the DOE Hydrogen Program (the Program) and will contribute to advancing the DOE research, development, and demonstration goals. The material cost is a major barrier, and the team is targeting this.
- The findings on increasing current density and cathode costs could have benefits to similar projects/approaches. The overall approach of utilizing wastewater streams for hydrogen production is a compelling value proposition. There are significant wastewater resources available, and high-strength wastewaters (acid whey, industrial wastewater, etc.) are a considerable liability to producers. It is difficult to assess the project's progress toward the ultimate objective of producing hydrogen from wastewater. No data were presented on hydrogen production rates or yields for the new materials or baseline materials. The technoeconomics rely heavily on the $10/kg hydrogen credit. It is unclear whence the assumptions for this are derived, specifically in regard to the BOD reduction credit. The project is assuming a 10-year catalyst

lifetime in the economics and has not done sensitivity analysis to explore the impact of this. In other applications, commercial catalyst lifetime is usually assumed to be in the one- to two-year range.

- The project is certainly relevant and has the potential to be impactful to the goals of the Program. However, this is dependent on the proposed technology working and the TEA actually reflecting reality. A greater focus is needed to determine whether a 10-year lifespan for the electrode is realistic.
- The project is aligned well with the progress required to advance the Program. A few reported results do not allow anyone to fairly evaluate the progress made, such as using high-buffer-capacity solutions and claiming excessively high current density that does not reflect the real performance of the cell.

Question 5: Proposed future work

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

- The team identified the work needed in the near future and developed a plan to carry out the tasks.
- The future work sounds rational and developed adequately to address the remaining challenges of the project.
- It is correct that developing strategies to maintain stable performance of the electrodes in MECs will be critical, but it is not clear that this critical issue is being addressed. Modifying the microbial community is also mentioned, but there is no real description of how the project team intends to do that, or to what extent. Also, a sensitivity analysis on each component of the TEA will be critical to determining which parameters are most critical to an economical process.
- Scale-up activities seem risky without knowing whether the materials are compatible with real wastewater streams. If there are no issues, the rate of scaling is otherwise appropriate. The source of the wastewater is not defined, and it is unclear whether any cleanup will be necessary. More specifics on the coating mitigation work would be useful.

Project strengths:

- The project has made significant strides in CNT fabrication as it relates to MECs. This will have benefits to a variety of industries that might consider MECs or a similar technology. The team has clear expertise in materials science to bring to the project. In this regard, the researchers are mindful of the performance parameters—pore size, conductivity, etc.—to track on these systems. The overall concept of using wastewater streams to produce hydrogen has significant promise. The results on current density are encouraging to date and relative to Pt-C.
- The project team is developing a unique technology to produce CNT-coated electrodes for wastewater cleanup via MECs. Electrode development seems to be making substantial progress.
- The project is carried out by experts in different fields and aimed at addressing a key challenge of cathode materials in MECs. The researchers have made good progress in cathode development, and they aim to significantly improve the performance to meet the hydrogen cost goals.
- The project advances biohydrogen production through MEC technology by addressing important challenges such as cost reduction and performance improvement. The approach is on anode, cathode, and MEC configuration improvement.

Project weaknesses:

- It is not clear that modeling a 10-year lifespan for the electrodes is realistic. More information on the microbial community and modifications to that community needs to be provided.
- The newly developed materials need to be further improved and tested to demonstrate their superiority over existing benchmark materials. TEA analysis can be more detailed to provide guidance for technological development.
- Most of the data were reported in unrealistic conditions, such as high-buffer-capacity solutions, and reported current density was also unrealistic, likely because of an arbitrary normalization of the current. This does not allow for a fair evaluation of the performance. The CNT approach to improving the performance appears to provide limited improvements.

- It is difficult to assess this project as a potential approach for producing renewable hydrogen since no data were presented to compare to other approaches. This is a major weakness. Lifecycle analysis—to compare to SMR or other hydrogen production processes—does not appear to be part of project scope. Details on the TEA are limited. The project team does not contain external partners or advisors to contextualize or guide the research to commercial relevance.

Recommendations for additions/deletions to project scope:

- The project is encouraged to start testing real wastewater as soon as possible to evaluate impacts of fouling, solids, and potential poisons such as nitrogen or sulfur. The project is encouraged to partner externally to gain access to real wastewater streams and understand the design/siting considerations at a wastewater source. The project should track BOD reduction, which is a key value proposition for the industry with which the project is trying to partner.
- A greater focus on mitigating the decrease in electrode activity over time is needed, as is an understanding of the importance of different parameters in the TEA via sensitivity analysis.
- No changes are suggested.

Project #P-196: H2NEW Consortium: Hydrogen from Next-Generation Electrolyzers of Water

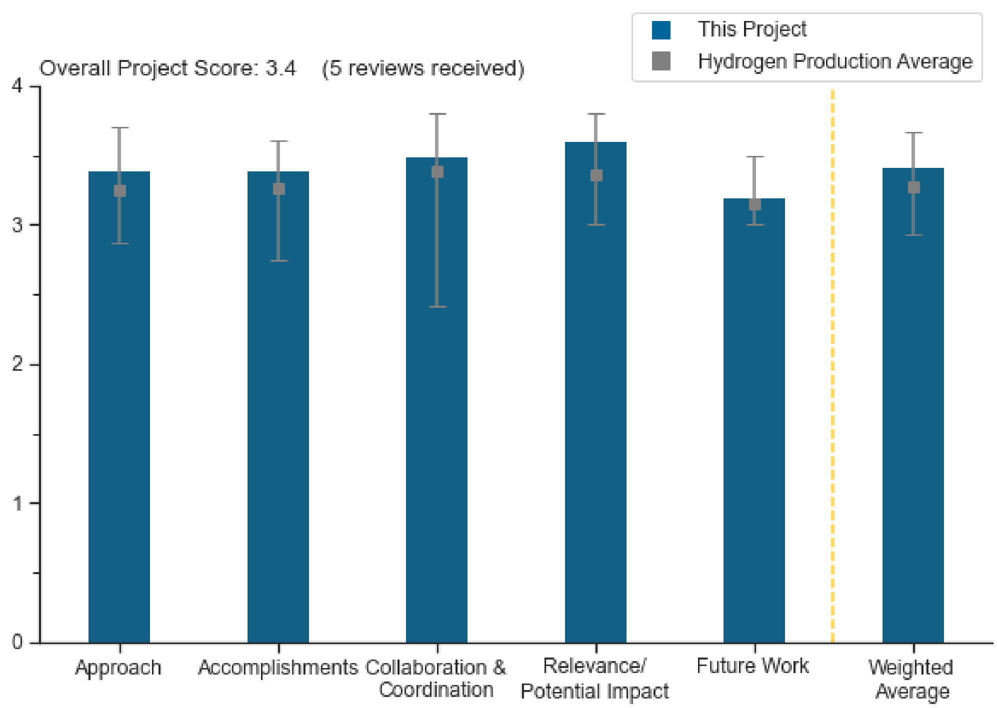
Bryan Pivovar, National Renewable Energy Laboratory, and Richard Boardman, Idaho National Laboratory

DOE Contract #	WBS 2.7.0.519 and WBS 2.7.0.1003
Start and End Dates	10/1/2020
Partners/Collaborators	National Renewable Energy Laboratory, Idaho National Laboratory, Argonne National Laboratory, Pacific Northwest National Laboratory, Lawrence Berkeley National Laboratory, Los Alamos National Laboratory, Lawrence Livermore National Laboratory, Oak Ridge National Laboratory, National Energy Technology Laboratory, National Institute of Standards and Technology
Barriers Addressed	• Durability • Cost • Efficiency

Project Goal and Brief Summary

The H2NEW (Hydrogen from Next-generation Electrolyzers of Water) consortium is a comprehensive, concerted effort focused on overcoming technical barriers to enable affordable, reliable, and efficient electrolyzers that can achieve <$2/kg H_2 by 2025. H2NEW is studying both low-temperature electrolysis (LTE), based on an acidic polymer electrolyte membrane (PEM), and high-temperature electrolysis (HTE), based on oxide-ion-conducting solid electrolyte. The core H2NEW national laboratory team is addressing components, materials integration, and manufacturing research and development. The team is working to improve scientific understanding of the performance, cost, and durability tradeoffs in electrolysis systems, including under predicted future dynamic operating modes, by using a combination of experimental, analytical, and modeling tools.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Production projects.

Question 1: Approach to performing the work

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

- A concerted, focused effort on reducing the cost of green hydrogen is both needed and an excellent aspect of the project's approach. A consortium approach is appropriate. This is a lab-only effort but with industry observation and recommendation through the LTE and HTE stakeholder advisory boards. This makes sense for core technology development to be shared by all, but extra efforts should be made to involve industry as much as possible.
- This seems like a comprehensive approach to PEM and solid oxide electrolyzer cell (SOEC) technology challenges. The project helps developers by setting industry targets.
- This project uses comprehensive advanced approaches to study PEM and SOEC performance and durability. The approaches cover cell testing, characterizations, advanced characterizations, modeling, and analysis. The project takes advantage of state-of-the-art facilities and strong capabilities in the national laboratories. This project is large and is solely undertaken by national laboratories. However, given the complexities and practical applications of electrolyzer technologies, industrial leaders should actively participate in this project. Additionally, there remains a question about how SOEC technology can reduce hydrogen cost to <\$1/kg H_2 because of the technology's lifetime constraints. The technology still has a long way to go. The scale-up of electrolysis, using automated processes, is key to reducing hydrogen cost. The project needs to put more effort on this aspect.
- This consortium aims to integrate materials, components, and manufacturing processes to advance water-splitting technologies to achieve goals in durability, cost, and efficiency. This initiative is perceived as an intermediary vehicle between fundamental research and product development.
- The approach, consortium structure, and 75/25 weighting for LTE/HTE are sound, as is the effort to go after the right barriers, at a high level. The only concern is whether the next level of scientific targets, while worthwhile, are the key ones from an industry perspective. It may be that they are, but it is not clear how hard that has been tested or whether input from the stakeholder advisory board is enough. If the board is helping to set directions, the board's voice needs to be more apparent.

Question 2: Accomplishments and progress

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

- The work done on LTE for marginal pricing, heat maps, and hydrogen cost impacts is meaningful and worthy analysis. LTE cell aging studies and characterization are very helpful. HTE development and understanding are critically important because of the potential high electrical efficiency. The detailed and methodical HTE experiment and characterization work is a very good achievement to date. The focus is appropriately placed on the identification of the HTE degradation modes. HTE multiscale modeling, validated by experiment, is a key aspect of the project.
- Very good progress has been made on identifying degradation sources for both HTE and LTE.
- Given the short time of this project in this year, this project's progress is satisfactory.
- A good start has been established.
- Results on durability and interfaces are highly anticipated. More emphasis on multi-physics modeling is suggested—in fact, for future presentations, the team should lead with this aspect as the "one ring to rule them all" for the other efforts.

Question 3: Collaboration and coordination

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

- The LTE/HTE multi-laboratory approach fosters excellent collaboration and synthesis of lab capability. The stakeholder advisory board is a very good structure for input and review from the industry.

- There is clearly a great deal of interaction at the national laboratory level. More outreach to industrial partners would be beneficial. One concern is that efforts are being made to dig deeply into issues that will not have the biggest return on investment for the industry.
- There is good collaboration between national laboratories. The stakeholder advisory board that has been set up for the LTE work is good. The advisory board for HTE does not contain any major commercial companies developing solid oxide electrolyzer products (granted, there are very few from which to choose).
- This project is a collaboration of multiple national laboratories. The principal investigator (PI) and co-PI have strong experience in a variety of areas. Unfortunately, industry participation is largely missing from this project. It involves a large number of researchers across national laboratories, and the tasks are also very broad. A more stringent coordination plan is needed to ensure the good progress of this project.
- This is a consortium involving almost all major national laboratories.

Question 4: Relevance/potential impact

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

- The project is highly relevant and directly advances the achievement of long-term DOE goals. The DOE Hydrogen Program's focus on identifying, understanding, and modeling degradation modes is critical to goal achievement. There would be a major beneficial impact on achievement of Hydrogen Program goals.
- The consortium has the potential to accelerate the commercialization of water-splitting technologies. It aligns well with DOE's overarching goals promoting hydrogen technologies.
- These are critical technologies. Maximizing our understanding of the fundamentals will help support needed cost and performance improvements—and even more so if the work is focused on industry's most pressing problems.
- This project is very relevant and will help industry develop lower-cost, more durable electrolyzers.
- This project is very relevant to DOE's H2@Scale goals. It can provide insightful information to the electrolysis community for renewable hydrogen production. It is not clear yet how the information attained from this project can be shared with the electrolysis community

Question 5: Proposed future work

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

- The LTE future work is described at a high level, but substantial further activity is indicated in each of the four main tasking areas. The HTE future work tasks are clear and appropriate.
- There are no reservations with the work plan. However, the team should seek and incorporate deep industrial perspectives on priorities and impact.
- The identified challenges are well-articulated and proposed to be addressed in the future.
- Future plans are very comprehensive.
- The future work makes sense.

Project strengths:

- Project strengths include the following:
    - The focus on reducing LTE stack and system capital cost, the Argonne National Laboratory LTE system diagram and energy balance, and LTE durability focus on operational, material, and cell-design-based mitigation techniques—all three are important to pursue
    - Leveraging across other consortia
    - LTE technoeconomic analysis
    - HTE extensive test stands and testing plans
    - HTE multi-scale modeling, when validated by experiment
    - HTE focus on identification and understanding of degradation modes

    - Having 28+ HTE test stands
    - HTE cost analysis

  HTE cost analysis should be extended to the balance of plant (BOP), and the project should examine the levelized cost of electricity sensitivity to stack and BOP cost.
- This project's strength is in the amazing power of the national laboratories being focused on a couple of specific technologies. It is bound to have a significant impact. Bryan Pivovar is a great champion and voice for this effort.
- This is a large consortium that is much needed for water-splitting technologies and that will play a key promoting role in meeting DOE goals in cost, durability, and efficiency.
- This project's strengths consist of the following points:
    - There is a strong collaboration across multiple DOE laboratories.
    - Advanced and comprehensive approaches are used in the project.
    - The PI and many co-PIs are highly technologically competent.
- There is a strong focus on understanding the problem areas in PEM and SOEC technologies.

Project weaknesses:

- The project has the following weaknesses:
    - The project is performed solely by national laboratories, so industry participation is largely missing.
    - The project needs a more stringent project coordination plan because of the project's high budget and complexity. For example, it is not clear how the attained information from this project can be used to achieve \$1/kg H_2.
    - There should be more transparency in budget spending, and project progression is also needed for such a high budget.
    - The project combines LTE PEM with HTE SOEC. They both have different materials, focuses, and applications.
    - This project focuses on testing, characterization, and analysis. Clear innovations should be identified.
- The LTE waterfall graph showing \$0.86/kg H_2 for the ultimate goal seems to require about \$0.01/kWh electricity. This assumption should be clearly stated, as it may not be realistic as an average price for intermittent electricity. Alkaline electrolysis is not addressed in the project, yet there have been substantial technology advances in recent years, and it is likely to capture a large market share in 2030 and beyond. The lack of details in validating the multiscale modeling effort is a weakness.
- The project feels a bit like everything but the kitchen sink is being thrown at understanding cell performance. This makes it hard to conceptualize the progress. It is unclear which pieces are the more scientifically interesting (long shots) and which are the technology advancement priorities. Perhaps the efforts should be organized around objectives instead of methods.
- The difference of the programmatic focus from the current industrial efforts in developing water-splitting technologies is not very clear.
- There is no mention of other emerging technologies, such as HTE proton-conducting technology or LTE anion exchange membrane technology.

Recommendations for additions/deletions to project scope:

- It needs to be clearer whether the capital cost targets are for the stack alone or for the full system. The HTE capital cost goal of \$100/kW should explicitly state this is for the stack, not the system. HTE stack performance and durability are primary factors in reducing the levelized cost of hydrogen, but BOP is reported at \$550/kW and needs to be addressed, too. Ways to reduce HTE BOP cost should be explored. There are seemingly no milestones (or a clear detailed timeline) for the HTE multiscale modeling. Modeling is insufficient without validation, and the timeline to achieve a validated model should be explicitly stated. Like the LTE advisory board, the HTE advisory board structurally draws in outside commercial electrolyzer expertise. This is good, but ways to further capture industry ideas and capabilities should be explored.

- The project should add the participation of industry leaders, and the interactions between national laboratories and industry should be strengthened. This project can be separated into two projects, as the focuses, technology readiness levels, and applications of PEM and SOEC are different.
- The project is focused on standard technologies and material sets. It would be great to see some investigation into promising alternatives that have the potential to leapfrog over the existing state of the art. The recommendation is not necessarily for an in-depth study but rather for some assessment of the potentials and problems with these promising alternatives.
- There should be even more emphasis on facilitating scale-up of electrolyzer production. The benefits of economies of scale are a huge assumption behind estimates of future low-cost electrolyzers. Manufacturability should be a key lens for the entire consortium.
- The pathways toward mitigating degradation should be clearly identified.

PRODUCTION—HydroGEN Seedling

Project #P-185: High-Performance Alkaline Electrolyte Membrane Low-Temperature Electrolysis with Advanced Membranes, Ionomers, and Platinum-Group-Metal-Free Electrodes

Paul A. Kohl, Georgia Institute of Technology

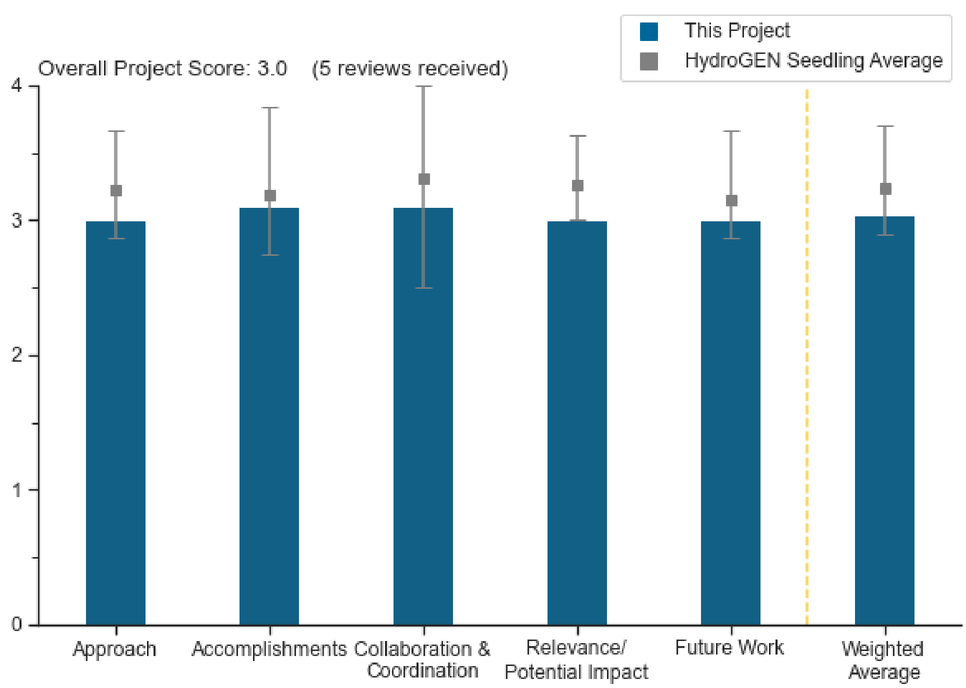
DOE Contract #	DE-EE0008833
Start and End Dates	1/1/2020 to 4/30/2023
Partners/Collaborators	Pajarito Powder, University of South Carolina, NEL Hydrogen
Barriers Addressed	• None listed

Project Goal and Brief Summary

The objective of this project is to combine state-of-the-art alkaline polymer electrolyzer components into one optimized membrane electrode assembly (MEA) system to achieve U.S. Department of Energy low-temperature electrolysis goals. The project will first implement the best materials at hand and establish best practices in each of the individual component areas (membranes, catalysts, ionomers, electrodes, and MEAs), leading to performing single-cell scale-up, as well as evaluating the electrolyzer performance and durability. This will lead to a platinum-group-metal-free (PGM-free) MEA optimization effort for operation on pure water (i.e., no added salt or base), as well as scale-up and detailed degradation modeling and mitigation studies. By the end of this project, the team expects to meet the following three metrics with a PGM-free alkaline electrolyte membrane (AEM) electrolyzer MEA operating on pure water:

- Performance: 1 A/cm^2 at 1.75 V
- Durability: <4 mV/1000 hour degradation
- H_2 production cost: <$2/kg.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by HydroGEN Seedling projects.

Question 1: Approach to performing the work

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

- This project is very well-rounded; it not only has polymer/ionomer development but also has low-loading PGM catalyst development and cost model projections. This holistic approach is rarely seen but is appreciated.
- This project is conducting interesting work on alkaline membrane/ionomer development, combined with electrode integration and understanding of the related effects on electrolysis cell performance and stability. The project's development of high-stability, low-cost hydrocarbon-based membrane and ionomer materials seems promising for advancing AEM electrolysis. The role of cost analysis in guiding the project's approach is confusing. Some other important aspects of the project approach were not clearly presented, especially regarding the use of the supporting electrolyte that appears to be used in most or all experiments, although it was not made clear whether the project vision is to develop cells running with supporting electrolyte or pure water. This is important because it seems likely that the use of the supporting electrolyte will change the relevant degradation and performance loss mechanisms, as well as optimal ionomers and electrode structures, in comparison to a pure water system. For instance, the project found good performance prioritizing adhesion/binding of the electrode over conductivity, though this may not translate to a system without the supporting electrolyte.
- The approach to developing AEM with glass transition temperature is commendable. The $PbRuO_X$ catalyst has caused some concerns. Pb is a hazard material. RuO_X is not thermodynamically stable (from the Pourbaix diagram).
- The approach slide is missing. Performance and durability improvement using membrane and ionomer development looks reasonable. The team has extensive experience synthesizing high-performance and alkaline-stable anion exchange ionomers. Therefore, leveraging such expertise in the project is low-risk and saves resources. Catalyst development seems to be separated from ionomer development. However, in many cases, the performance of electrolyzers largely depends on catalyst–ionomer interaction. No strong justification is provided to use perovskite catalysts versus PGM-free, metal-based catalysts for oxygen evolution reaction (OER).
- There is no clear approach slide in the presentation. No risk and risk mitigation strategy is outlined. There is no clear strategy to identify possible durability issues. Membrane development is, however, impressive. The project needs a clear strategy to understand the issues about local hydroxide ion concentration and its effect on OER performance.

Question 2: Accomplishments and progress

This project was rated **3.1** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals, as well as the HydroGEN consortium mission.

- There is appreciable progress on all fronts, with membrane, ionomer, and electrocatalyst development.
- There is, overall, good progress with ionomer development. There is noted accomplishment on the effect of ionomer water uptake on electrode performance. Experimental results to improve durability, including catalyst detachment, are interesting and provide some insight. The steady-state voltage plot in slide 14 is another highlight. The data is impressive, as the electrolyzer ran at 1 A/cm^2. However, the data was obtained using 0.3 M KOH and PGM catalysts. Under such conditions, ionomer stability cannot be accurately measured. Demonstrating good performance and durability is critically important. More studies on the fundamental understanding of polymer effects on electrolyzer performance and durability are desired. There is very little, if any, work on PGM-free catalysts. Slide 3 indicated that the team accomplished the go/no-go point with performance <1.75 V, PGM-free electrolysis at 500 mA/cm^2, but all data presented uses PGM catalysts. Most accomplishment slides lack information. Some slides do not have catalyst information (slides 9, 10, 11). For slide 9's left-bottom plot, the legend is missing (not clear whether that red curve meant GT18 or GT38). Some slides do not have liquid electrolyte information. Other slides have the data using different liquid electrolytes (0.1 M KOH for slides 12 and 16, 0.1 M NaOH for slide 13, 0.3 M KOH for slide 14). On page 14, the title is confusing. It noted a non-conductive binder, but the information indicated GT32 and GT69 ionomers. It is unclear whether those are non-conductive.

- The project has demonstrated reasonable progress toward its targets, met the budget period 1 (BP 1) go/no-go milestones, and produced several publications from 2020 and 2021. Some good and interesting results are presented on stabilizing performance through ionomer/electrode optimization, including the ionomer swelling and adhesion properties. Not all results are clearly supported with evidence in the presentation. For instance, there was no clear evidence that poor adhesion of the electrodes is the principal stability challenge, as stated. Also, some results appear to have significant variability (e.g., the anode catalyst loading study) without a very clear trend visible, although a simple trend is claimed, raising concerns about the reproducibility of such results.
- There are good accomplishments regarding the membrane, but the project needs more work on understanding interfacial issues and electrocatalysis on both electrodes.
- There is good progress on membrane development. All the performance is obtained using KOH, so the AEM development becomes less significant.

Question 3: Collaboration effectiveness

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

- Collaborations between the team members are excellent. Each team member has different expertise, and several results were accomplished by the collaborative work. Non-core research activities can be supported by Energy Materials Network resource nodes as the project goes further.
- There are several partnerships with various laboratories, with each lab taking on responsibilities in which that lab is expert.
- The collaborations, such as those with the National Renewable Energy Laboratory, are just beginning at this point; it must have been a challenge, considering the circumstances.
- The project seems to have demonstrated good collaboration between project partners, but it is not clear that the HydroGEN nodes have been integrated into the project effectively yet. It appears that more collaboration with HydroGEN is planned in the next year, so one hopes that this will be remedied. Use of the HydroGEN Data Hub is not addressed in the presentation.
- This project has proposed a good collaboration with the HydroGEN consortium. The data from the collaboration with the HydroGEN consortium was not reported.

Question 4: Relevance/potential impact

This project was rated **3.0** for supporting and advancing progress toward DOE Hydrogen Program goals and the HydroGEN consortium mission.

- The development of AEM electrolysis is quite significant because of the high scarcity and high price of Ir, which has been used in polymer electrolyte membrane (PEM) water electrolysis. The success of this project may enable meeting the DOE ultimate hydrogen production goal of $2/kg hydrogen.
- This project has excellent relevance, and potential impact is great. Collaborators have been carefully curated, and collaborations should bear fruit next year.
- The project aligns well with the DOE Hydrogen Program and DOE research, development, and demonstration objectives. AEM electrolyzers are a promising technology for green hydrogen production. The cost model output (slide 4) is reasonable. However, the project approach does not demonstrate how to get the target performance. For example, it would be very challenging to use a PGM-free cathode catalyst to get 1,000 mA/cm^2 at 1.85 V. Also, it has not been demonstrated that thin AEM (<50 micrometers) can produce hydrogen at 30 bars. Adding KOH solution into the system will also increase the cost, although it is unclear that the project is aiming for electrolyte-free liquid.
- The project's work on development of alkaline membranes and ionomers, as well as understanding and improving electrode stability and performance, has clear value for the field and advances the viability of AEM electrolysis. However, the project's impact is motivated with confusing cost analysis results. Several important assumptions relevant to the project are not presented clearly. It is unclear whether the analysis assumes the use of a supporting electrolyte. The support for the claim that AEM water electrolysis will have lower balance of plant costs than PEM water electrolysis is missing. The analysis assumes both very low-cost electricity ($0.02/kWh) and a 100% capacity factor; this is not clearly a realistic scenario, but it

may alter the relative priorities for lowering cost. It is not clear whether the results in the sensitivity analysis tornado chart are relevant to capital cost or levelized cost of hydrogen, and there are apparent contradictions about the importance of membrane and catalyst costs. The cost analysis seems to imply that these are not important, but then they are identified as major project focus areas.

- The progress made this year is good—but not out-of-the-ballpark good. For example, the principal investigator's (PI's) team did show stable AEM electrolysis stability, but only in the presence of a supporting electrolyte. It would have been interesting at least to hear about the AEM electrolysis durability in deionized (DI) water, especially since the PI did highlight the stability of sp^3 carbons. It looks like the electrocatalyst and ionomer durability will need to be supplemented with a supporting electrolyte. If that is not the case, there was no evident plan to overcome these issues.

Question 5: Proposed future work

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

- The planned future work focus on continued research in stability and durability is good. Longer-term durability testing is a good next step and warranted by good results demonstrated by the project so far in durability tests now spanning about 100 hours or hundreds of hours. For electrode stability tests, some kind of cycled accelerated stress test (AST) would also be useful. For the planned scale-up activities, the electrode technology readiness level (TRL) seems low, but there is broader interest in commercially available AEMs, so demonstrating membranes at commercial scale is a reasonable next step. Further catalyst investigations are also reasonable; the project approach so far of not varying the catalyst too much is good, as it is preferable to have fewer variables in the electrode development work.
- Future work has been clearly outlined and is logical.
- Future work will be focused on long-time durability. Future work should be focused on electrode design to enable pure water operations.
- The future work is well-organized. However, the scope of work is little too broad. It seems that AEM and ionomer durability reach a certain level when low-concentration KOH electrolyte is used. Maybe it is time to investigate more on pure-water-fed conditions. Moreover, as the project's go/no-go decision criterion is <1.75 V PGM-free electrolysis at 750 mA/cm^2, the focused work for PGM-free anode and cathode catalysts is required. Demonstration of the stable membrane at a commercial scale may be less important, as those efforts can be leveraged with other projects.
- In the future work, the first bullet is that "there are clear pathways toward durable ionomers/catalyst/binder-based electrodes." However, this does not seem to be the case, as DI water electrolysis durability is not presented and it was mentioned that the OER catalyst has some stability issues. The ionomer direction is toward materials that are non-conductive. If that is the case, this project will need to focus on electrolyte-supported electrolysis. Whether that will be the focus is not clear, which is why the future work is rated as good.

Project strengths:

- There is good collaborative research. There is holistic research and development of membrane, electrocatalyst, and cost model development. There is promising materials development.
- The strength of the project is teaming. Georgia Tech (membrane) and the University of South Carolina (electrode) are capable team members for these efforts. Pajarito Powder has good resources on various hydrogen evolution reactions (HER) and OER catalysts. Nel Hydrogen has extensive testing experience. The high performance of AEM fuel cells using the Georgia Tech materials has been demonstrated. Therefore, the project team has a good chance to achieve high performance and durability for AEM electrolyzers.
- Work on development of alkaline membranes and ionomers and understanding electrode integration and durability are important priorities for the AEM electrolysis field, and the project appears to be achieving progress on these topics. The membrane/ionomer technology appears promising, and improvements to cell stability have been demonstrated.

- AEM development with high glass transition temperature is a strength. The project has a great team with complementary experience.
- The project strength is mainly on the development of a new membrane, though the PI has long experience with this class of polymer materials.

Project weaknesses:

- The main weakness is the seemingly ambiguous suggestion that focus will be on electrolyte-supported electrolysis (there is no DI water electrolysis durability, nor was there any mention that the anode catalyst is not stable in DI water). The PI did mention that the polymer chemistry is scalable, but it was not clear whether the production of the crosslinked membranes could be performed via large-scale membrane manufacturing, such as roll-to-roll or tape casting. Typically, crosslinked membranes are difficult to control from batch to batch, and this is especially difficult with square-meter-sized membranes.
- The project tasks are too diversified. The scope of the project is much larger than that of other HydroGEN projects, although the size of the project is similar. The team may try several catalysts, gas diffusion layers, membranes, and ionomers in MEAs. However, systematic studies on a focused area may be difficult. For example, in anode ionomer optimization, the team tried many ionomer combinations and concluded that the OER requires an ionomer with lower water uptake. However, the comparison between GT72-5, GT72-10, and GT72-15 suggested that the GT72-15 with the lowest water uptake showed the lowest voltage during the 2.5-hour test. Showing high performance and durability is critical, but a more systematic approach to understanding electrolyzer performance and durability may be necessary. Unfortunately, the resources to carry out such experiments seem to be limited.
- The cost analysis is confusing and not clearly adding value to the project. Some results presented appear to have possible issues with variability and reproducibility, and the project should be careful about validating such results. The project's approach to the use of supporting electrolytes is unclear.
- The $PbRuO_X$ catalyst has a hazard concern. Pb is a hazard material. RuO_X is not thermodynamically stable (from the Pourbaix diagram). The focus will be on electrode design without using KOH solution. BP 1 and BP 2 go/no-go milestones contain PGM-free catalysts, but actual data was obtained using PGM catalysts.
- A potential weakness is in electrocatalysis and the lack of a clear strategy for understanding interfacial issues both from an electrocatalysis point of view and from the transport aspect.

Recommendations for additions/deletions to project scope:

- The project needs to demonstrate baseline performance under standardized conditions first. The project should clearly define AEM, AEM thickness, ionomer, HER and OER catalysts, catalyst loadings, alkaline-electrolyte-fed conditions, temperature, pressure, and voltage degradation rate at a specified current density for a specified number of hours. This can be used as a progress measure. The project should also use non-PGM HER and OER catalysts (if those are proposed). The project should use differential pressure operation. The project should delete all Ir-based catalyst work (except for baseline performance).
- The cost analysis in its current form has unclear value to the project. This analysis should either be improved to clearly provide relevant guidance to the project or be omitted from future work. Some validation work to ensure that results are reproducible would also be very useful for the project and would be a good use of HydroGEN support resources. Durability testing should possibly also include dynamic operation/cycled ASTs to test electrode stability.
- The team should decide whether to focus on DI water or electrolyte-supported electrolysis to help gauge the TRL of the technology.
- It is recommended that the project shift the electrocatalysis focus to non-PGM materials; there is no purpose to moving to alkaline pH while using PGM catalysts.
- The project should focus on the testing without the KOH solution. Without the addition of KOH, the significance of this project has been compromised.

Project #P-186: Performance and Durability Investigation of Thin, Low-Crossover Proton Exchange Membranes for Water Electrolyzers

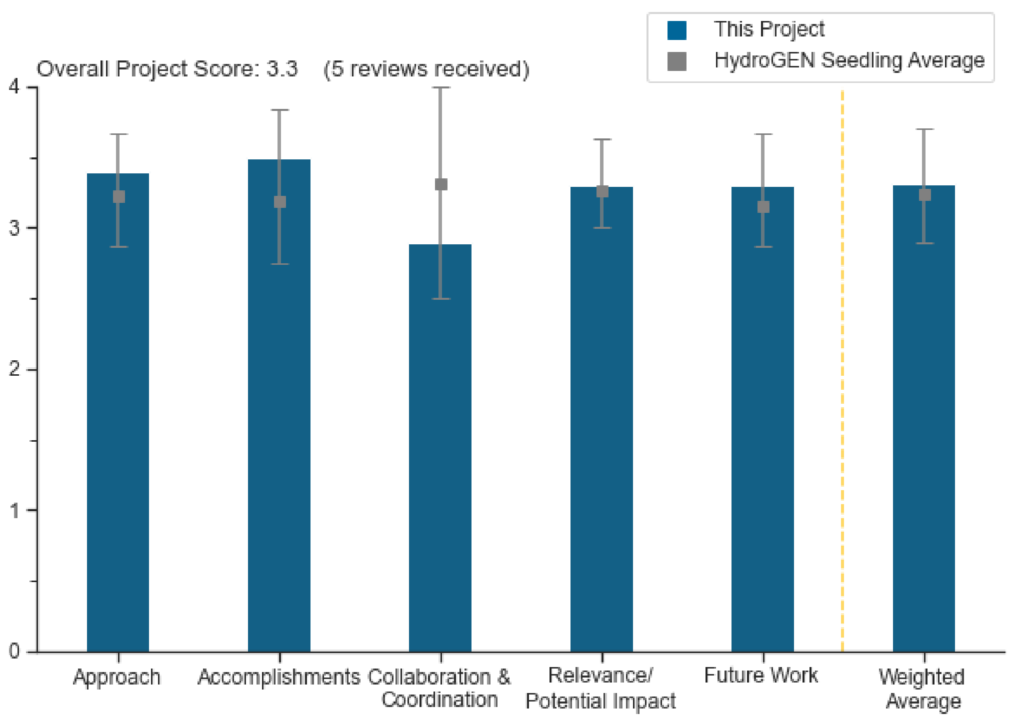
Andrew Park, The Chemours Company FC, LLC

DOE Contract #	DE-EE0008836
Start and End Dates	3/1/2020 to 2/28/2023
Partners/Collaborators	Los Alamos National Laboratory
Barriers Addressed	• Manufacturability: These membranes will be constructed on roll-to-roll systems for easy transition to the commercial scale • Durability: The additives envisioned to enable thin membranes can move, agglomerate, or leave the system entirely, which will be studied and mitigated

Project Goal and Brief Summary

The goal of this project is to develop next-generation membranes specific for polymer electrolyte membrane water electrolyzers (PEMWEs) with improved performance and durability. Thin, mechanically reinforced perfluorosulfonic acid (PFSA)-based membranes will include gas recombination catalysts (GRCs) and radical scavengers to reduce gas cross-over and increase durability, respectively. State-of-the-art roll-to-roll manufacturing technologies will be leveraged to fabricate the membranes on a commercial scale, where the placement and loading of the additives can be precisely tuned/distributed within the membrane structure. The critical factors for success include (1) integrating/optimizing additives within a thin, reinforced PFSA membrane, (2) understanding the additives' behavior (i.e., activity, migration, dissolution, and/or retention) within the membrane over a polymer electrolyte membrane (PEM)-water-electrolysis-relevant lifetime, and (3) validating membrane performance/durability using duty cycle and accelerated stress tests that are representative of dynamic electrolyzer operation.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by HydroGEN Seedling projects.

Question 1: Approach to performing the work

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

- This project has a very strong approach, including the development of multiple distinct improvements to advance membranes for PEMWEs, following from successful approaches for PEM fuel cells but optimized for the PEMWE context. These membrane improvements are all useful in isolation and address critical challenges for PEMWE membranes, but the improvements also have the potential to be combined in an advanced membrane product. The project has a good structure, beginning with isolated modifications, verifying their effectiveness, and then moving to combined product and durability work in future budget periods. The HydroGEN partners are integrated from the beginning, providing characterization and modeling capabilities that complement the membrane development at the Chemours Company (Chemours), and can provide validation and guidance as the project progresses.
- This project aims at understanding the limits of PEM thickness using various strategies such as reinforcements and other additives. With this, the researchers expect to advance the membrane resistance from the current state of the art of 0.2 ohm-cm^2 to below 0.7 ohm-cm^2. They also plan to advance the state of the art in gas recombination catalysts and radical scavenging.
- The approach to making reinforced PEMs with GRCs is meaningful in lowering overpotential caused by ohmic loss. Thin membranes can also lower the capital cost. Hydrogen crossover characterization is quite comprehensive. More mechanical property testing of the developed membranes is necessary.
- This project tackles a very important opportunity for PEMWEs.
- There are reservations about this approach since it is still employing Nafion™, which is known to be fairly expensive, but now adding the additional process of including a GRC and radical scavengers will further increase the cost of this material. Furthermore, there was no discussion on exactly what this GRC composition is; one hopes it is an inexpensive transitional metal oxide, but this was not clear. In addition, adding additives have other consequences such as changing mechanical properties, leaching, and increasing materials costs.

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, as well as the HydroGEN consortium mission.

- The project presentation demonstrates strong progress on all budget period 1 (BP 1) goals, with results and data very clearly presented and showing clear progress on critical metrics for crossover, mechanical properties, and processing of radical scavengers and the GRC. Durability progress has not yet been demonstrated, but this work is planned for BP 2. The project also demonstrated strong deployment of the characterization and modeling capabilities from the associated national laboratory HydroGEN nodes. The BP 1 milestones were met, with a short extension needed on BP 1 go/no-go milestones because of COVID-19-related delays on the lab work.
- The team made excellent progress on its initial milestones in the first year of the project. Uniformly dispersing the GRC in the membrane and demonstrating three-fold reduction in hydrogen permeation are good achievements.
- The project has made a great accomplishment in terms of reduced membrane crossover. The hydrogen crossover of the developed membrane is close to that of N117, but the membrane has a thickness of 50 µm..
- This project has made good progress with the homogeneous incorporation of both the GRC and radical scavengers. The resultant composite material had an electrolysis performance similar to N212 but hydrogen permeation around three times lower than N212. It is hard to gauge this advancement by itself; it would help if there were a cost model showing whether the cell could be run at around 1 A/cm^2 at 1.5 V in a real electrolysis system. It would also help to then see the equivalent cost savings for the current state of the art. Moreover, it would be beneficial to determine whether the higher performance would offset the higher cost of the membrane.
- The accomplishments have been very good, considering the challenges of the previous year's COVID-19 pandemic challenges.

Question 3: Collaboration effectiveness

This project was rated **2.9** for its collaboration and coordination with HydroGEN and other research entities.

- There is very strong collaboration with national laboratory partners and HydroGEN nodes, which provide characterization and modeling to complement the membrane development at Chemours. The collaborators appear well-integrated and are providing significant valuable contributions early in the project, providing validation and opportunities to guide membrane development as the project progresses. The collaborative activities are well-matched to partner capabilities and targeted to key project objectives. The use of the HydroGEN Data Hub and data sharing were not addressed in the presentation.
- There is a good workflow within the team that has addressed all important aspects of the research.
- The collaborations are just starting for this project, and they should be in good stead next year.
- There is good collaboration with key national laboratory experts, but what takes away from the feasibility of this project is that there is no original equipment manufacturer (OEM), such as Giner, Inc., or Nel Hydrogen, as a testing partner. If this project cannot get the OEM excited about testing these materials with the current progress, it is unclear whether there will ever be a viable end user.
- The only collaborator is Los Alamos National Laboratory. Collaboration with an electrolyzer company for further evaluation is highly recommended.

Question 4: Relevance/potential impact

This project was rated **3.3** for supporting and advancing progress toward DOE Hydrogen Program goals and the HydroGEN consortium mission.

- This project has very high potential impact for PEMWE devices, the H2@Scale vision, and the DOE Hydrogen Program in general. If successful, the advanced PEMWE membrane developed by this project would combine multiple improvements to advance durability, cost, performance, and operational flexibility—all critical barriers for achieving the $2/kg H_2 cost target for water electrolysis. The approaches pursued all appear highly promising based on their successful application for PEM fuel cell membranes and based on strong initial results from this project. Development and deployment of characterization and modeling tools with project partners and HydroGEN nodes will also be beneficial to the research community, as these tools will be transferrable to future projects.
- This project addresses a serious opportunity to enhance the efficiency and durability of PEMWEs.
- The development of reinforced PEMs may reduce the thickness of the membrane without much influence on the hydrogen crossover; therefore, it may bring down the overall cost of PEM electrolyzers. The cost of reinforced membranes with GRC needs to be determined.
- The project's relevance is good but not outstanding. The need to lower the membrane and interfacial resistance is well taken but is in the domain of optimization. The field of GRCs is also well-documented and not terribly new. Using Ce as a free radical scavenger is also well-known. However, success in this project will certainly offer greater functionality in a mature technology.
- This is interesting science, but it is difficult to see the breakthrough impact of lowering PEM electrolysis cost by essentially enabling use of thinner Nafion membranes. Moreover, the additives can potentially degrade and migrate out (there is already some evidence of this, seen in slide 14), and it seems that the additives open other problems with unknown consequences and seemingly little benefit.

Question 5: Proposed future work

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

- This project has strong planned future work, including integrating various membrane features developed separately into one advanced membrane and testing the resulting product. Planned testing includes durability testing to ensure that membrane additives are stable and provide expected improvements to membrane chemical durability. All future work appears reasonably planned and clearly targeted to achieve project goals and overcome critical membrane development barriers.
- The proposed future work properly builds on previous results to work toward achieving the overall goals.

- The proposed future work has been well-delineated.
- The proposed future work is detailed and reasonable.
- Testing efforts are planned in the future work, but it would make more sense if there were dialog between the project lead and OEMs, such as Giner, Inc., or Nel Hydrogen, to determine whether the performance metrics are of value to them. It is not clear whether a membrane that gives electrolysis performance similar to N212 and has three times less hydrogen permeation is a key hurdle in this technology space.

Project strengths:

- This project has a strong and clearly focused research effort to develop next-generation membranes for PEMWEs, including multiple promising pathways to improve the technology. There is high potential impact for the DOE Hydrogen Program if the project goals are met. Also, there is good integration with collaborators and effective use of support from the HydroGEN consortium.
- The project has good fundamental science. The approach includes homogeneous incorporation of additives, and the additives improved performance as predicted (lower area-specific resistance and lower hydrogen permeation).
- The reinforced PEMs with the GRC are a project strength, although this idea has already been adopted by major electrolyzer companies. Stringent hydrogen crossover measurements are commendable. The hydrogen crossover data are impressive. Also, the project has progressed well and met major milestones.
- The project has a great approach and an excellent team.
- This project aims at furthering the current state of the art of PEM electrolyzers. Some risk analysis, however, is important, considering the thinness of the membrane.

Project weaknesses:

- The project has provided little technical detail on the membrane modifications being developed. This is understandable because the project is working toward a proprietary product; however, it does limit the informational value for the broader community. One hopes that in a later stage of the project, more detail can be shared.
- The ultimate weakness of this project is that it is not clear whether the project is solving a real issue in current PEM electrolysis. The project might be introducing more issues with increased membrane cost and additive migration/degradation.
- The project's weaknesses are that the long-term mechanical testing has not been implemented and the cost of the developed membrane has not been analyzed.
- This project, as formulated, does not provide any advancement in the science of technology.

Recommendations for additions/deletions to project scope:

- There needs to be a partnership with an electrolysis OEM that can validate that the performance of this new membrane will lower system/operational cost and is a material the OEM can get behind once the lifetime testing and accelerated stress test are completed.
- The project should consider other possible failure modes that may fall outside membrane stress testing (such as anode catalyst dissolution and redeposition on the GRC) and inclusion of relevant testing for any risks identified.
- The project should complete the cost analysis of the developed membranes and add an electrolyzer company as a collaborator to validate the data.
- No recommendations are necessary at the moment.

Project #P-187: Pure Hydrogen Production through Precious-Metal-Free Membrane Electrolysis of Dirty Water

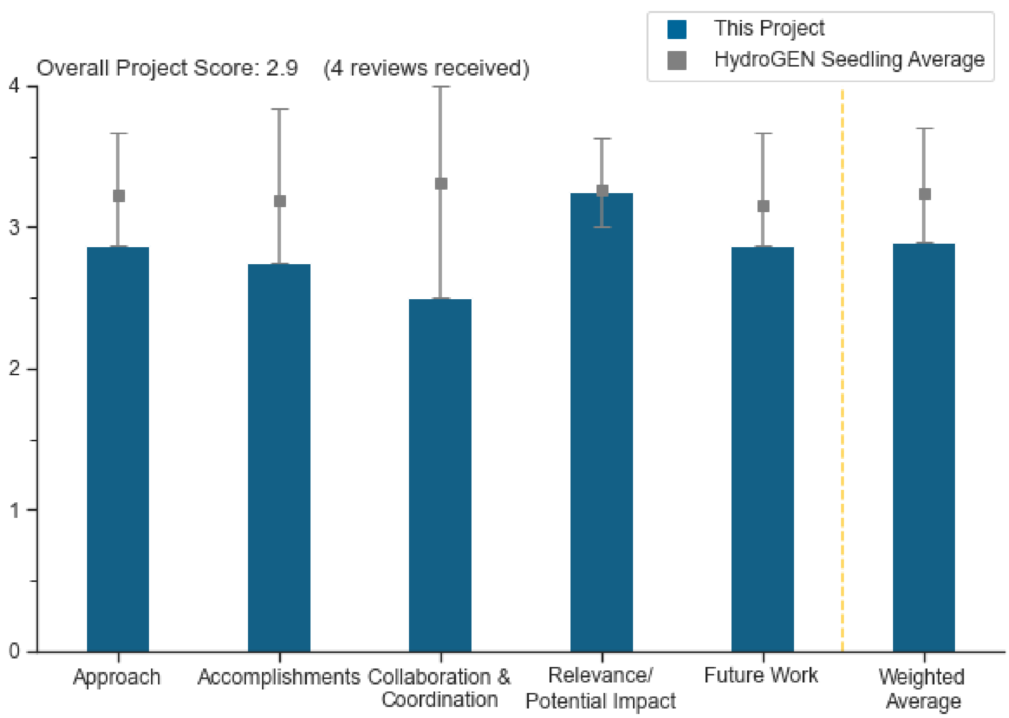
Shannon Boettcher, University of Oregon

DOE Contract #	DE-EE0008841
Start and End Dates	4/1/2020 to 3/31/2023
Partners/Collaborators	National Renewable Energy Laboratory, Lawrence Berkeley National Laboratory, Sandia National Laboratories
Barriers Addressed	• Ion exchange in membrane(s): Minimize by controlling ion flow direction • Deposition of impurities: Use high loadings of low-cost catalyst, control location and morphology of deposits • Cl-oxidation: Maintain local basic anode

Project Goal and Brief Summary

The project team will develop a technical understanding of alkaline and bipolar membrane electrolyzers, specifically how their performance degrades in both pure and dirty water. Using this knowledge, the researchers will engineer impurity-tolerant systems. This project will improve the longevity of platinum-group-metal (PGM)-free electrolysis devices, make them more tolerant of input water impurities, and lower costs. The University of Oregon is collaborating with Lawrence Berkeley National Laboratory (LBNL), the National Renewable Energy Laboratory (NREL), and Sandia National Laboratories (SNL) on this project.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by HydroGEN Seedling projects.

Question 1: Approach to performing the work

This project was rated **2.9** for identifying barriers and addressing them through project innovation, as well as for project design, feasibility, and integration with the HydroGEN consortium network.

- The project is following a good approach to begin with baselining and understanding the degradation and transport in pure-water-fed cells using PGM catalysts before proceeding to introduce added complexity from contaminants or PGM-free catalysts. The inclusion of baselining and validation work with HydroGEN to ensure reproducibility of results is a good feature of the project. This approach appears to give the project a strong position for addressing challenges for alkaline electrolyte membrane (AEM) and bipolar membrane (BPM) systems.
- The project aims to develop AEM electrolyzers using PGM-free catalysts. The project aims to deal with dirty water. The design of the reference electrode is impressive. It is unclear what the focus of the principal investigator (PI) is: catalyst development, electrode design, or impurities studies.
- The project is aiming to design more robust electrolyzers to produce hydrogen using "dirty" water. The project approach is to control ion flow using AEM and BPM electrolyzers. It is hard to capture what the unique approaches this team is trying to make are. AEM and BPM electrolyzer architectures are well known in the community. The only difference in this project is using unpurified seawater instead of the pure water used in other studies. In the approach section, there is no clear message on how impurities in the unpurified water are going to be dealt with. There are no specific pathways to deal with local pH change by seawater, which could be the key consideration to reach the proposed targets. No AEM material justification was provided. The choice of Sustainion or node-supporting materials for the project is unclear.
- There is a clear need to see the limits of alkaline water splitting electrocatalysis considering the larger overvoltage window in alkaline conditions. The approach is, however, missing key details such as thermodynamic limits based on the pH of "dirty water." "Dirty water" has not been clearly specified except that it contains NaCl; it is unknown if other components make sense in this regard. So, the approach would need more clarity, especially as to the basis for choosing 0.5 M NaCl and whether that choice has any relevance to "dirty water."

Question 2: Accomplishments and progress

This project was rated **2.8** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals, as well as the HydroGEN consortium mission.

- Good progress has been demonstrated toward meeting the project goals, including establishing testing capabilities and publishing project results. Work on the project so far has built a good foundation for future work, especially baseline development work and development of methods for the project, such as test stand construction, integrated reference electrodes, and analytical methods. The project appears on track to meet its milestones.
- The project demonstrated good AEM water electrolysis (AEMWE) performance using pure water. The project is unable to show long-term durability of the AEMWE. The catalyst $PbRuO_x$ might be highly unstable in alkaline media. The study on dirty water is not thorough, as there are many ions or impurities in saline water or wastewater.
- The project has been in place one year and had a possible delay due to COVID-19. Some progress was made using commercially available materials. The team accomplished measurement of the baseline electrolyzer performance, a water transport study, measurement of CO_2 effect, and measurement of ion crossover in NaCl solution. The PI talked much regarding the degradation via polymer oxidation, but this is a known fact from a previous study (see *Energy Environmental Science*, DOI: 10.1039/D0EE04086J [2021]). Likewise, water transport and CO_2 effects (NaOH vs. $NaHCO_3$) have been discussed in the community. It is okay to discuss those effects, but the PI should justify why these types of studies are related to the scope of the project. The project needs to define how to calculate the voltage degradation rate on slide 15. For the IrO_2 anode in pure water, the cell voltage increased from 1.87 V to 2.26 V for the first 100 hours. So, the voltage increase rate is 3.9 mV/h (0.39/100*1000), but it was reported to be 0.67 mV h^{-1}. Also, for the IrO_2 anode in 0.2 g $NaHCO_3$ case, the cell voltage increased from 1.9 V to 1.98 V during the first 110 hours, so the voltage increase rate is 0.72 mV/h, but it was reported to be 0.17 mV h^{-1}. The voltage increase rate during the first 20 hours shown on slide 12 seemed to be correct. The project needs to define

durability better. Electrolyzer durability should not be measured by voltage increase rate only. If the cell is stable only 10 hours before voltage goes to zero, then this cell is not durable. So, the project needs to add a minimum operation time (e.g., 1,000 hours) (please see what other HydroGEN projects are doing). The preliminary BPM electrolyzer performance shown in slide 18 is relatively poor, although the performance is approaching that of the AEM electrolyzer in pure water. The team should do a simple technoeconomic analysis that shows how much performance improvement is required to generate hydrogen at $2/kg. Any voltage over 3 V is not practical, but even the best-performing BPM electrolyzer has cell voltage >3 V at 2 A cm^{-2}. Adding dirty water and PGM-free catalysts will likely increase operating voltage. A few missing experiments the team probably should have accomplished in the first year of the project are the following:
  - Baseline electrolyzer performance using "dirty water"
  - Performance of PGM-free catalysts in rotating disk electrodes or electrolyzers
  - Planning for material design standpoint, as it is not clear what types of materials the team is trying to use for the proposed systems
  - Detailed planning for the modeling, ink formulations, and membrane electrode assembly fabrication.
- Accomplishments have clearly shown the risks, especially in terms of the choice of membranes and catalysts. Degradation rates are not acceptable. It is unclear what is causing this rapid decline in activity.

Question 3: Collaboration effectiveness

This project was rated **2.5** for its collaboration and coordination with HydroGEN and other research entities.

- The collaborations are yet to begin, and, hopefully, they will manifest themselves next year. It has been a challenge to foster collaborations in these circumstances.
- This project listed many node supports. The node supports seem to cover various areas of the project. However, no detailed tasks or accomplishments from the node collaboration are described. The node tasks listed in the presentation are general, except one from Weber. There are no plans or works with the benchmarking/protocols teams other than mentioning "ongoing collaboration with Nel Hydrogen."
- The project's integration with collaborators appears to be more in planned future work, without major collaboration demonstrated so far. However, the planned collaborative work does appear to make good use of HydroGEN node capabilities directed toward the project goals. The presentation made no mention of the project's use of the HydroGEN data hub or data sharing.
- Collaboration with HydroGEN is clearly presented. The project uses W7 membranes, but the role of SNL (Cy Fujimoto) is not clear.

Question 4: Relevance/potential impact

This project was rated **3.3** for supporting and advancing progress toward DOE Hydrogen Program goals and the HydroGEN consortium mission.

- The impact of this project is high, if it succeeds. It can eliminate expensive and scarce Ir catalysts. It can also help develop strategies to mitigate impurities in water. The AEMWE can also enable other inexpensive non-active components. Long-term durability needs to be thoroughly studied for a higher impact.
- The relevance of this effort is very high, though there will always be a tradeoff between external water purifiers and inherent tolerance to salts and other species.
- The potential impact of AEM electrolyzers using seawater or dirty water is high. Also, the plan for using PGM-free catalysts for AEM electrolyzers is relevant and well aligned with DOE's research, development, and demonstration goal. One concern is that the actual content and current accomplishment is not well aligned with the project target. The PI indicated that ion exchange in membranes, deposition of impurities, and Cl oxidation are the major barriers, but the project's remaining challenges listed are ionomer degradation, comprehensive modeling, and model studies. It seems that the chance of high-performing AEM electrolyzers using seawater and PGM-free catalysts at the end of the project is very low. The work should be more focused and resolve some specific challenges related to the project objective.

- The project's focus on electrolysis of truly "dirty" water provides unclear value to the community, and this is not obviously a promising path to meeting broader DOE Hydrogen Program goals for low-cost hydrogen production. However, much of this project's work is more broadly impactful (for example, understanding degradation and transport in AEM systems generally and understanding and mitigating contaminants from steel or non-PGM cell components). It is good for the project to keep its focus on activities that provide broader value to the community and improve understanding of AEM and BPM systems in use cases other than dirty water electrolysis as well. The work focused on baseline performance, stability and degradation, and the impacts of supporting electrolytes (including after attempts to flush the cell) is very useful to the community and fits well within the project scope of understanding ion impacts and degradation mechanisms.

Question 5: Proposed future work

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

- The planned work continuing to understand baseline performance and degradation in pure-water-fed cells is good, as this area is clearly still needing more clarification. The planned validation work supported by HydroGEN is a good inclusion to ensure reproducibility of results. Planned support from HydroGEN on modeling and understanding ionomers and membranes is also reasonable for achieving the project goals. It is not clear if the planned work will include non-PGM catalysts as described in the project vision.
- The future work is clearly described. The dirty water study is interesting but challenging. Work has to be conducted to differentiate the impact on catalyst and on membrane. Advanced characterizations can help to understand durability issues caused by the interface.
- They are well delineated in slide 20.
- The future work section is filled with rosy targets. No clear pathways and specific approaches were discussed. For example, the PI indicated that oxidation of ionomer is the big issue. Then it is unclear what the appropriate experiments or designs to mitigate the oxidation of ionomer are. It is unclear what the specific plan to decouple the degradations by membrane, catalyst, ionomer, and impurities is. It is unclear what the pros or cons for PGM-free catalysts for AEM electrolyzers' performance and durability are. It is unclear what the strategy to prevent impurity crossover to the cathode is. Also, collaborative works with HydroGEN nodes are not well described.

Project strengths:

- The project is achieving good scientific work to understand performance, transport, and degradation in AEM electrolysis that is of broad value to the community. The project includes interesting innovative work in alternative cell design approaches, such as bipolar membranes and anode water feed. Strong baseline development work has been completed so far, as well as method and capability development, and the project is actively publishing its results. The project appears to have a good approach to understanding and overcoming key challenges for AEM electrolysis.
- The concept of the project is a strength. The conventional electrolyzer requires pure water or alkaline solution for high performance. The project is aiming to use "dirty" water for generating hydrogen. The team looks to be capable to analyze the data and has some background knowledge on AEM and BPM electrolyzers. Also, it was reasonable to test the electrolyzer in a practical current density (500 mA/cm^2) (slide 12). The team also has access to some commercial and non-commercial materials for the project. They are looking at various aspects to realize highly performing and durable water electrolyzers.
- This project and hopefully others will provide the general limits to how pure the water needs to be under alkaline pH conditions. This is a clear pathway toward the technoeconomics of running this unit with either seawater or seawater after partial desalination.
- Using PGM-free catalysts and dirty water are great ideas. Good progress has been made in a one-year time frame. The reference electrode design is very helpful.

Project weaknesses:

- The major project weakness is the broadness of the scope of work. Some of the works are largely overlapped with previous and other current projects. The project task should be refined and more selective to accomplish the project targets. Current data show some important information regarding performance and durability limits but lack strategy to resolve the problems. DOE Hydrogen and Fuel Cell Technologies Office projects are milestone-driven. Understanding degradation modes is useful to set strategies but does not necessarily bring the project to meeting the targets. It is not clear how controlling ion flow in AEM and BPM electrolyzers can improve the performance at the target level. The go/no-go milestone is not clearly defined. It stated "pure water/gray water/salt-water" feed, but it needs to provide specific information or definition of gray water and salt water. Also, it shows 1 A cm^{-2} at <2 V and 1 mV/h. Reaching the target at the end of the project (<2 V at 2 A cm^{-2}) and the durability target (<4 mV/100 h = 0.04 mV h^{-1}) seem to be very challenging. Also, there is no information regarding operating temperature, non-PGM catalyst, catalyst loading, etc.
- The catalyst $PbRuO_x$ might be highly unstable in alkaline media. This can be seen from a Pourbaix diagram. There is no plan to differentiate the impact of dirty water on the catalyst and on the membrane. The poor durability of AEMWE has not been understood.
- At the moment, durability is of concern.
- Electrolysis of "dirty" water is not clearly a promising path to meeting broader DOE Hydrogen Program goals for low-cost hydrogen production. Many of the project efforts are focused on more useful topics to improve general understanding of AEM and BPM cell degradation and performance and on contaminants from other sources. This is good in a way, but having this disconnect between the project vision and ongoing activities is not ideal. Significant collaborative work with HydroGEN is planned, but it is not clear how active this part of the project is at this point. Efforts should be made to integrate the collaborators into the project sooner rather than later.

Recommendations for additions/deletions to project scope:

- Work on contaminant mitigation through feeding water to the anode side should include clear comparisons to feeding water to the cathode (and possibly also both electrodes). Including investigations of supporting electrolytes with the investigation of contaminants would be useful as well.
- Dirty water study is time-consuming. In fact, dirty water has to be processed before entering the electrolyzer. This work can be less significant in this project. Focus should be given to PGM-free electrode design to improve the performance and durability.
- CO_2-related work is irrelevant, so that work should be deleted unless there is strong justification. The project should reduce all work with pure water feed. Those studies were largely overlapped with other studies. The project should expand PGM-free catalyst work under seawater-fed conditions. The project should do more AEM electrolyzer tests under dirty-water-fed conditions to identify performance and durability limiting factors.
- A clear strategy to understand the origins of poor durability is recommended.

Project #P-188: Advanced Coatings to Enhance the Durability of Solid Oxide Electrolysis Cell Stacks

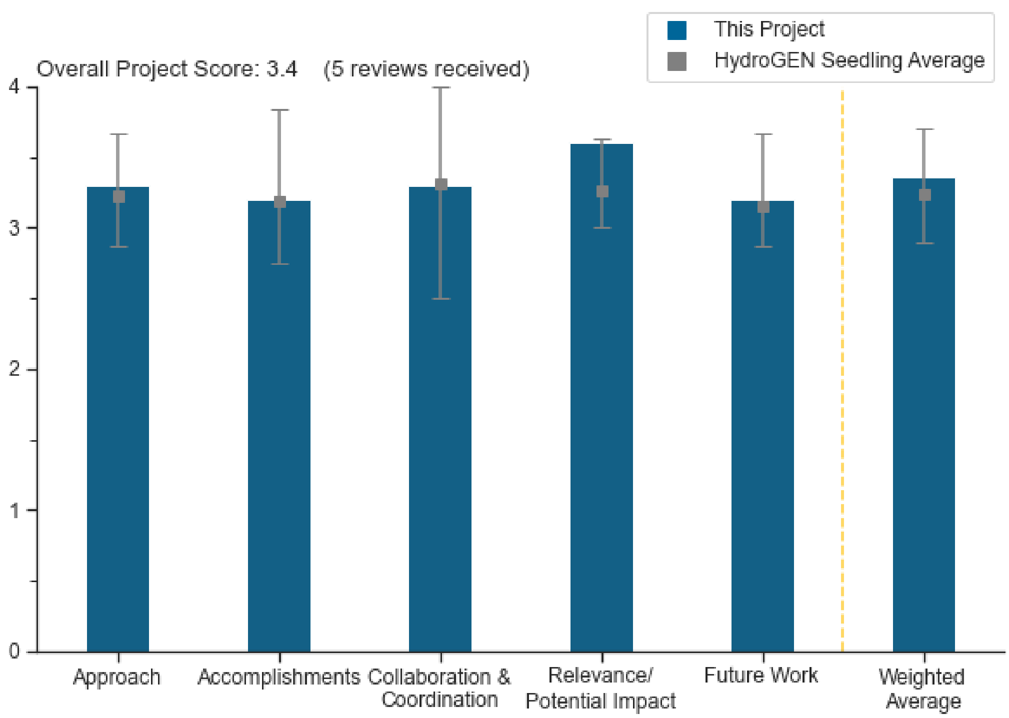
Neil Kidner, Nexceris, LLC

DOE Contract #	DE-EE0008834
Start and End Dates	4/1/2020 to 3/31/2023
Partners/Collaborators	University of Connecticut, Lawrence Berkeley National Laboratory, Idaho National Laboratory
Barriers Addressed	• Deconvolution of degradation mechanisms • Demonstration of coating technology at production-relevant scale

Project Goal and Brief Summary

This project will advance the technical and commercial readiness of solid oxide electrolysis cells (SOECs) by developing protective coatings and Cr getters to enhance system life. An integrated degradation mitigation strategy consisting of SOEC optimized interconnect (IC) coating, Cr getters, and a balance-of-plant (BOP) component coating will address the critical degradation mechanisms of metal corrosion and chromium evolution. These degradation mechanisms can be substantial, and mitigation strategies are essential to improving SOEC durability and achieving the U.S. Department of Energy performance and lifetime targets for SOEC systems. The efficacy of the coating strategy to reduce degradation will be demonstrated by testing on SOEC single cells and stacks, with a goal of achieving an equivalent (or better) reduction in stack degradation rate compared to single-cell testing.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by HydroGEN Seedling projects.

Question 1: Approach to performing the work

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

- The objective of this project is to develop an effective coating strategy to minimize the degradation caused by metal corrosion and chromium evolution from metallic components in SOEC stacks. If successful, the project will significantly enhance durability and accelerate commercialization of SOEC systems. The technical approaches are scientifically solid and are appropriate for achieving the project objective. Several critical barriers are identified and are being effectively addressed. The experiments are well-designed and are integrated with other relevant efforts.
- This is a good approach for an industry project. Taking existing coatings Nexceris offers for yttria-stabilized zirconia (YSZ) solid oxide fuel cells (SOFCs) and running a gap analysis for SOEC operational conditions is an efficient way to begin the project from the basis of Nexceris's extensive prior work (and extensive work in literature on manganese cobalt oxide [MCO], on which the ChromLok coating technology is based). Also, the scalability of the coating approach is already validated, so if it works for SOECs, it is commercially viable. Some constructive criticism is offered: The presenter admitted to not realizing that MCO will degrade in reducing atmospheres (slide 9) (an unexpected admission). That is well-documented in literature, but there may be differences in ChromLok, so characterizing the coatings' degradation was useful. Also, much of the project's novelty lies in the data-gathering to understand the operational parameter space; the coatings and future getters seem to be selected from reasonably well-researched materials. Having access to these insights will benefit the community, but it is also plausible that some information will be withheld; Nexceris is encouraged to find a balance that allows for broad data-sharing, when possible. A reviewer mentioned the Data Hub, which would be a good avenue.
- This project tries to address an important issue in the SOEC, i.e., Cr poisoning. Half of the project focuses on evaluating existing coatings for interconnect, and the other half focuses on loading Cr getters in BOP. These two approaches are technically unrelated, although they contribute to the common goal.
- This project effectively builds on prior technology to meet the project objectives. Collaboration with HydroGEN is effective.
- It is indicated that the barriers of the approach are deconvolution of degradation mechanisms and demonstration of coating technology at production-relevant scale. However, no discussion or work plan on these barriers was given.

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, as well as the HydroGEN consortium mission.

- The team has successfully completed the first go/no-go milestone, suggesting that the proposed coating is acceptable for further testing of SOEC stacks under typical operating conditions. Overall, good progress toward the outlined project objectives has been made against clearly defined and measurable performance indicators, such as required area-specific resistance and degradation rate. The accomplishments to date suggest that the researchers have made reasonable progress in addressing critical barriers to achieving DOE goals. It would be useful if the distribution of Cr and the Cr-containing phases in the porous electrodes could be characterized under different operating conditions, which could then be correlated with the performance of the test cells with and without the protective coatings.
- The positive is that the researchers met their Year 1 go/no-go targets. Some constructive criticism is offered: The existing coating has been determined to be adequate already, with minimal modification, for SOEC operation, and future milestones may necessitate much more significant modification to the coatings. Therefore, those should be evaluated early in Year 2 since they can take a good deal of time.
- The project exceeds the go/no-go criteria. It is not clear that the Year 2 go/no-go milestone will result in significant progress toward the <4 mV/kh degradation goal. Improvement toward the degradation goal relies instead on progress outside of the project scope (Generation 3 cells).

- From an interconnect Cr evaporation point of view, SOEC and SOFC operating conditions are similar to each other, so it is no surprise that the coating that worked under SOFC conditions also works under SOEC operation. It will be interesting to see more Cr getter results.
- Go/no-go decision point 1 was completed.

Question 3: Collaboration effectiveness

This project was rated **3.3** for its collaboration and coordination with HydroGEN and other research entities.

- Nexceris is adding to the company's baseline expertise with key partners at the University of Connecticut, Idaho National Laboratory (INL), and Lawrence Berkeley National Laboratory (LBNL). Prabhakar Singh's group is well-poised to contribute to the Cr getter work, as well as other Cr impact studies, based on the group's extensive experience. Some constructive criticism is offered: The INL and LBNL nodes are mentioned, but there was not much discussion of the laboratories' actual contributions to project goals. The presented work should communicate the national laboratories' roles more clearly, or descriptions of the work the laboratories contributed should be added. It would be interesting to know whether Nexceris participated in the 2B Benchmarking project activities.
- Valuable technoeconomic analysis is provided through collaboration. Collaborations appear to be effective.
- The team shows effective collaboration with Energy Materials Network nodes.
- The collaboration and coordination with the three partners, including the University of Connecticut, LBNL, and INL, appear to be reasonable. It would be helpful if the developed coatings could be independently tested by other HydroGEN team members.
- A more detailed description of the collaboration with INL and LBNL should be given.

Question 4: Relevance/potential impact

This project was rated **3.6** for supporting and advancing progress toward DOE Hydrogen Program goals and the HydroGEN consortium mission.

- Efficient, durable, and low-cost SOEC systems will accelerate the transition to renewable, energy-efficient, and low-cost hydrogen. The project objective is to develop a critical coating that will enhance the durability of SOEC systems. The project aligns well with the Hydrogen Program and DOE research, development, and demonstration (RD&D) objectives and has the potential to advance progress toward DOE RD&D goals and objectives. The successful completion of this project will significantly advance the hydrogen production technology and the commercial viability. The project is also leveraging the resources and framework of the HydroGEN consortium. The project has good potential to advance the discovery and development of novel materials for efficient water-splitting systems, which will enable meeting the DOE ultimate hydrogen production goal of $2/kg H_2.
- The project is sharply focused on demonstration and development of SOEC corrosion coatings for commercial deployment. This sharp focus on a critical component will have a significant impact toward DOE goals.
- The Cr impact of steel components needs to be addressed in most SOEC systems in a way that is cost-effective, so the relevance for this project is very clear. The magnitude of the impact will be more apparent when the actual change in long-term degradation values is reported and acceptable stack lifetimes are calculated, but the project is off to a good start.
- This project addresses one of the most important issues in SOEC stability: Cr poisoning.
- The project includes evaluation of coating under electrolysis conditions. However, it is not clear how the project has leveraged progress and used relevant data under fuel cell (SOFC) conditions.

Question 5: Proposed future work

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

- The project is well-planned-out.

- The proposed future work in budget period 2 includes further detailed studies and improvements of the existing ChromLok IC coating technology, identification of effective Cr getter materials, and the combination of IC coating and Cr getter materials.
- It is not clear whether the project covers any of the cell development. The presenter mentioned that Generation 3 will help get current densities and voltage ranges in the desired range. More detail on the coating and getter analysis plan would be helpful. The kinds of improvements being considered were not specified.
- The project should work on scaling up the coating technology.
- The effort should focus on the Cr getter development.

Project strengths:

- The project brings together a good team for the development of coatings at a major SOFC manufacturer. The coatings will be needed for most high-temperature electrolysis applications since Cr-containing steels are required in just about every design.
- The approach seems to be practical and effective in improving the durability of cell components susceptible to Cr poisoning. The probability of developing some effective protective coatings is relatively high.
- The project focuses on an effective low-cost coating technology and evaluates coating effectiveness under SOEC conditions.
- Thanks to leveraging previous technology, the project is likely to meet the project goals on schedule and within budget.
- The team’s expertise and the approach to addressing BOP are strengths.

Project weaknesses:

- Weight gain and lifetime studies would benefit from repetition of the trials. These types of data are prone to large sample-to-sample variability, so repetition would increase confidence that the results have not been influenced by spurious factors. Also, although Cr issues are undeniably important in solid oxide cells, it would be helpful to more explicitly highlight how these coatings will influence hydrogen production costs to help reach $2/kg.
- It is implied that meeting DOE's degradation rate target of <4 mV/kh is a project goal, but the project is narrowly focused on corrosion coatings. The degradation rate target is unlikely to be met by the project work alone.
- The project lacks microanalysis of the porous electrode surfaces contaminated by Cr or other contaminants. The project also lacks mechanistic understanding of Cr poisoning.
- The barriers identified for the project approaches are not addressed. The project activities are not closely coordinated (e.g., how the chromium getter work at the University of Connecticut has been incorporated into the project).
- Not much progress was demonstrated on the Cr getter approach in budget period 1.

Recommendations for additions/deletions to project scope:

- Thermodynamic analysis is mentioned, but there was no discussion of the approach to this type of analysis. It would be useful if the presenter would explain what he meant. Kinetics and electrochemistry analysis are clearer. If the existing ChromLok MCO coatings are deemed to perform well with minimum modifications, the scope should expand to evaluate more aggressive conditions. The time saved by not needing to make significant developments to the coating process would be re-invested into data-gathering for where the approach fails. That could provide information for future Office of Energy Efficiency and Renewable Energy decisions about Hydrogen Program goals and directions.
- It would be very useful to perform some microanalysis of the chemistry, phase, and morphology of the electrode surfaces contaminated by Cr or other contaminants. To gain some mechanistic understanding of Cr poisoning, it would be necessary to characterize the distribution of Cr and Cr-containing phases in the porous electrodes under different operating conditions.

- The IC coating effort should be reduced.
- No change to project scope is recommended.
- There are no recommendations for additions or deletions.

Project #P-189: Scalable High-Hydrogen-Flux, Robust Thin Film Solid Oxide Electrolyzer

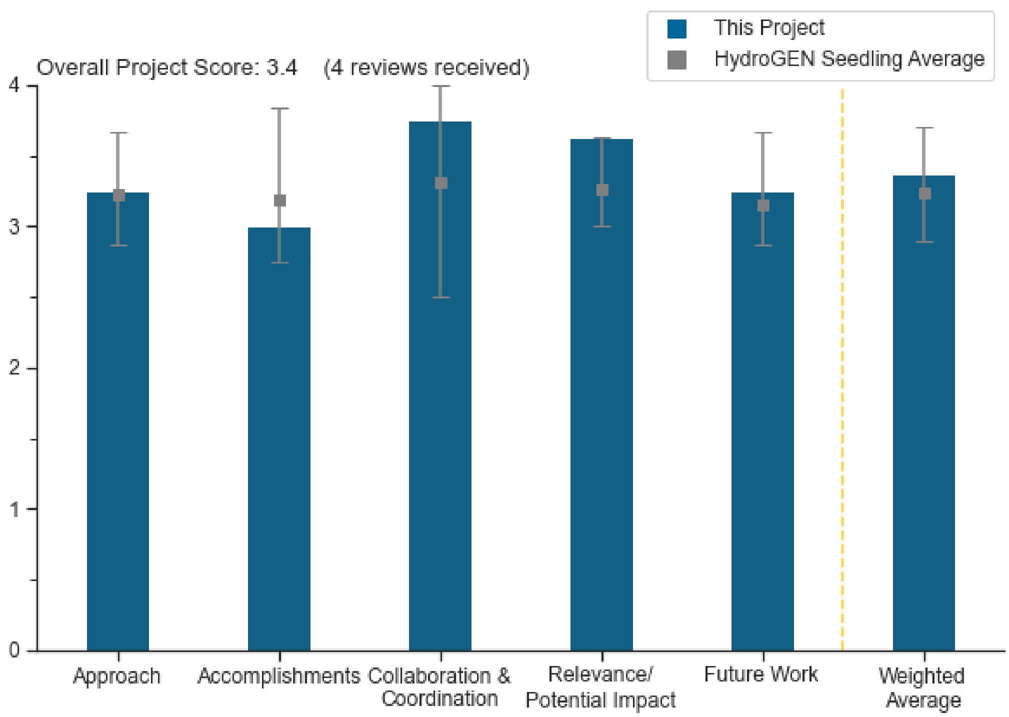
Colin Gore, Redox Power Systems, LLC

DOE Contract #	DE-EE0008835
Start and End Dates	5/7/2020 to 5/31/2023
Partners/Collaborators	Idaho National Laboratory, National Renewable Energy Laboratory
Barriers Addressed	• Optimizing stability, conductivity, or Faradaic efficiency in solid oxide electrolysis cell materials and devices typically requires tradeoffs in properties or processing

Project Goal and Brief Summary

The objective of this project is to demonstrate the technical and economic feasibility of solid oxide electrolysis cells (SOECs) based on a thin-film, multilayer, proton-conducting electrolyte. This project will develop a multilayer concept to block the electronic current with one layer, dramatically raising Faradaic efficiency (FE), and provide steam protection with the second layer, thereby mitigating long-term degradation and extending lifetime. The multilayers will be deposited by physical vapor deposition via methods scalable to high-volume manufacturing, overcoming high-temperature processing challenges that have hampered the development of conventionally processed high-performance proton conductors. The cost for H_2 production is expected to decrease significantly owing to a reduced stack size and decreased power consumption resulting from lower cell resistance of the thin film electrolyte layers and high FE.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by HydroGEN Seedling projects.

Question 1: Approach to performing the work

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

- The scope of work appears to be appropriate. Significant integration of advances from HydroGEN consortium members into Redox Power Systems, LLC's (Redox's) commercial cell fabrication expertise is a plus.
- The main objective of this project is to enhance the stability and FE of SOECs based on proton-conducting electrolytes using multilayer electrolytes and functional layers. It is hoped that the proposed SOEC systems have potential to meet the U.S. Department of Energy's cost and performance targets. The technical approaches seem to be reasonable for achieving the project objectives. However, some critical barriers may be difficult to overcome. For example, it could be difficult to fabricate coherent bi-layer electrolyte composed of different materials. Sputtered films often need high-temperature annealing to get the desired phases; critical challenges associated with high-temperature annealing may include chemical reaction, inter-diffusion, and delamination between the two layers of different materials. It would be helpful to provide some description on how to determine e-, h+, OH-, and O_2 leaks versus temperature by varying pO_2 and pH_2O using a mass balance system.
- The project proposes a bi-layer approach to addressing the electronic leaking issue related to BZCY (ceria- and yttria-doped barium zirconate) proton-conducting electrolyte.
- The project approach should place more emphasis on optimizing cell stability and efficiency (key barriers) via tradeoffs between cell component properties and processing parameters.

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, as well as the HydroGEN consortium mission.

- Some good progress has been made; preliminary results suggest that the protective coating material (the composition was not given) is relatively stable against high concentrations of steam at 500˚C for >200 hours. However, the claimed stability was based only on x-ray analysis of the samples, which may not be conclusive since x-ray diffraction is sensitive neither to surface change (the volume fraction of the new phases on the surface is too small) nor to non-crystalline phases (some of the hydroxides may be amorphous). It would be necessary to measure the conductivity of the samples after exposure to steam at 500˚C for >200 hours to confirm the stability against high-concentration steam. Overall, good progress toward the outlined project objectives has been made using measurable performance indicators. It would be useful to perform some careful microanalysis of the electrolyte surfaces (composition, phases, and morphology) before and after exposure to high concentrations of steam at 500˚C for >200 hours, which can then be correlated with the change in conductivity of the samples after the exposure to steam.
- Because of the coronavirus pandemic impacts, progress to date has been limited. Given the circumstances, the current progress is reasonable.
- It would be better if more bilayer data were presented.
- There is no significant progress on sputtering process development and scale-up.

Question 3: Collaboration effectiveness

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

- The team shows effective collaboration with Energy Materials Network nodes.
- Effective collaboration is a particular strength of this project.
- The collaboration and coordination with other partners of the project appear to be reasonable, including the HydroGEN consortium, with appropriate use of nodes.
- There is good collaboration with Idaho National Laboratory and the National Renewable Energy Laboratory but no collaboration or interaction with Lawrence Berkeley National Laboratory, Sandia National Laboratories, or Lawrence Livermore National Laboratory.

Question 4: Relevance/potential impact

This project was rated **3.6** for supporting and advancing progress toward DOE Hydrogen Program goals and the HydroGEN consortium mission.

- The SOEC systems based on proton-conducting electrolytes have demonstrated very high performance in terms of current density and efficiency. The successful completion of this project will significantly advance the hydrogen production technology and the commercial viability. The project supports and advances progress toward DOE Hydrogen Program goals and objectives and also supports the HydroGEN consortium mission. The project has good potential to advance the discovery and development of novel materials for efficient water-splitting systems, which will enable meeting the DOE ultimate hydrogen production goal of $2/kg hydrogen.
- This project is significantly advancing the development of novel materials. The project is not advanced enough to determine the likelihood of significant impact on meeting DOE targets.
- Progress has been made on material development. More emphasis should be placed on demonstrating stability, especially the stability of large-area cells.
- Electronic leakage is the most important issue for the FE and stability of proton-conducting electrolyte.

Question 5: Proposed future work

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

- The custom mass balance system developed by Redox is likely to be an effective tool to identify root causes of efficiency losses.
- The scope of approach for the proposed future work is good.
- Tasks proposed for budget period (BP) 2 (2021–2022) and BP 3 (2022–2023) appear to be reasonable. However, some details seem to be missing. For example, some specific description on how to enhance FE while reducing area-specific resistance would be very helpful. Also, it would be useful to explain how to determine the key performance degradation mechanisms and devise solutions to them.
- It is unclear why pulsed laser deposition (PLD) has been evaluated along with sputtering. If PLD is evaluated as an alternative to sputtering, then it is unclear when the down-selection is. It is suggested that the project focus on long-term stability and scale-up in the following budget periods, especially BP 3.

Project strengths:

- The proposed thin-film deposition methods seem to be practical and effective in exploration of the effect of protective coatings on cell performance, although a few key challenges are yet to be overcome to be successful in fabrication of coherent bilayer electrolytes. The probability of developing some effective protective coatings for durability and high FE is relatively high.
- Strong integration of the project with HydroGEN consortium members is a strength. Effective tools have been developed to significantly advance material development.
- The team members have the expertise to conduct the proposed work and leverage their strength in thin-film deposition in other solid oxide fuel cell systems.
- The project focuses on development of thin-film proton-conducting cells on hydrogen-electrode-supported substrates and evaluation of scalable and cost-effective sputtering processes.

Project weaknesses:

- There is a lack of detailed microanalysis of electrolyte surfaces exposed to high concentrations of steam at high temperatures and a lack of mechanistic understanding of steam–surface interaction or degradation mechanisms.
- More efforts (relative to the effort on performance improvements) are needed for evaluating material and cell durability and process scale-up.
- The early stage of the project makes it difficult to assess likelihood of success.

Recommendations for additions/deletions to project scope:

- To gain some mechanistic understanding, it would be necessary to perform some careful microanalysis of the electrolyte surfaces (composition, phases, and morphology) and to measure the conductivity of the samples before and after exposure to high concentrations of steam at 500˚C for >200 hours. The correlations between the changes in surface microscopic features and the changes in conductivity of the samples before and after the exposure to steam will be vital to gaining some critical insights into the mechanism of steam–surface interactions.
- It is suggested that the project team consider interdiffusion between the deposited layer and baseline electrolyte during SOEC operation.
- There are no recommendations for additions or deletions to project scope.

Project #P-190: A Multifunctional Isostructural Bilayer Oxygen Evolution Electrode for Durable Intermediate-Temperature Electrochemical Water Splitting

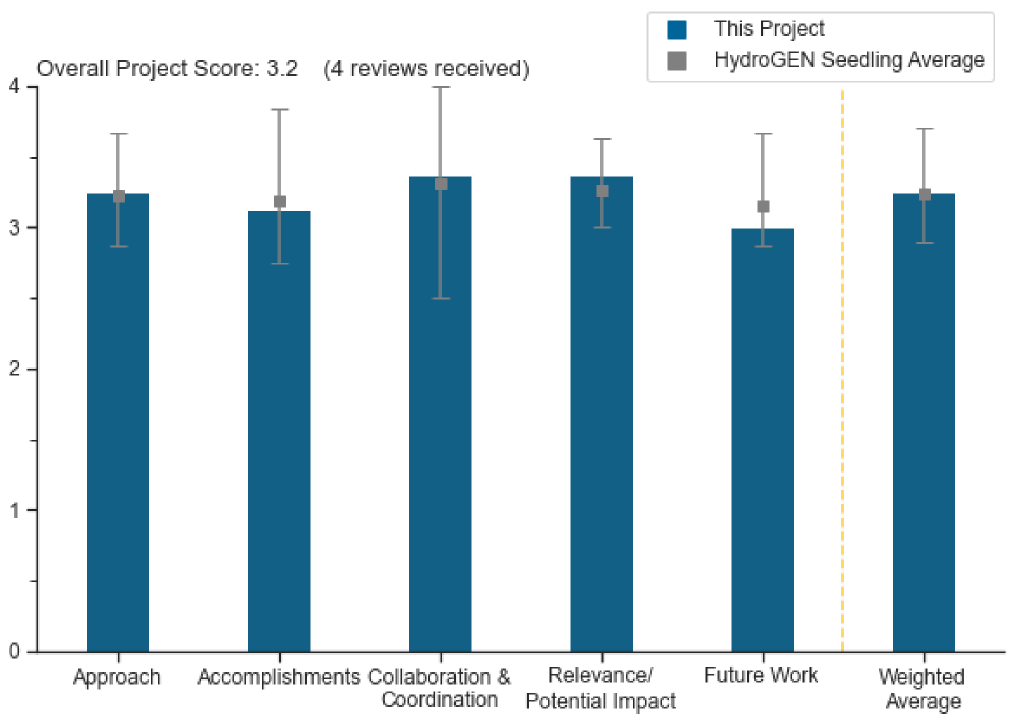
Kevin Huang, University of South Carolina

DOE Contract #	DE-EE0008842
Start and End Dates	4/1/2020 to 3/31/2023
Partners/Collaborators	University of South Carolina, University of Massachusetts at Lowell
Barriers Addressed	• Delamination and Cr-poisoning of OEs are the two leading causes for the performance degradation of SOECs. The new bilayer OE to be developed addresses these two critical issues at once.

Project Goal and Brief Summary

The two leading causes for solid oxide electrolysis cell (SOEC) performance degradation are delamination and chromium poisoning of oxygen electrodes. This project seeks to address these issues through materials innovation and theoretical modeling. The final product will be a highly active and chromium-resistant oxygen electrode for durable, high-efficiency, and high-rate hydrogen production via high-temperature SOECs. The University of South Carolina (USC) is collaborating with the University of Massachusetts (UMass) on this project.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by HydroGEN Seedling projects.

Question 1: Approach to performing the work

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

- The approach to this work seems very effective. USC is developing a unique oxygen evolution electrode, UMass is doing modeling work to explain the results, and Idaho National Laboratory (INL) is providing third-party testing to validate USC's results. The foundation of the experimental work seems solid; this material has already been developed by the USC team, so this project can focus on development more than materials research/discovery, which starts from a more mature position and reduces risk. It will be interesting to compare and contrast this work with that of Scott Barnett's HydroGEN project focused on overpotential and temperature impacts for different alternative oxygen evolution electrodes.
- The project aims at improving both performance and stability (against Cr poisoning) of SOECs via wet-chemical coating of the oxygen electrode.
- This is a good materials development project. It is unclear that building both planar and tubular cells is necessary to prove out the technology innovation.
- More emphasis needs to be placed on evaluating the interrelationship between electrode microstructure and electrode stability.

Question 2: Accomplishments and progress

This project was rated **3.1** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals, as well as the HydroGEN consortium mission.

- The oxygen electrode performance is excellent. However, it is unclear how the team will address the delamination issue that can lead to rapid degradation. This discussion should be expanded in the future.
- The team's 25% of $SrCo_{0.9}Ta_{0.1}O_{3-\delta}$ (SCT) coating can significantly improve the performance (polarization resistance [Rp] reduction) of the SOEC. Considering the high costs associated with both Ta and the process (25% loading takes multiple steps to achieve), the team may want to conduct a preliminary techno-economic analysis (TEA) to understand the costs and benefits of the proposed approach.
- Progress has been made in the electrode overpotential study and modeling, but limited work has been done on manufacturing (task 3).
- It is hard to conclude that 25% loading and 950°C processing is an optimized set of parameters when it's also the maximum loading tested and minimum temperature. While it is understandable that infiltration to higher wt.% is challenging and hits a plateau as pore volume decreases so that 25% may be the maximum feasible amount, it would be helpful to see repeat experiments of the degradation percent at 25% loading to add confidence to the result that the 950°C value does indeed decrease in trend after increasing from 5 wt.% to 15 wt.% and plateauing at 20 wt.%. Also, the team could add confidence to the results by testing one or two lower temperatures (perhaps just for the 25 wt.% loading case) to prove the properties worsen or by showing x-ray diffraction results to explain that 950°C is the minimum calcination temperature with no secondary phase formation. This was a fair amount of experimental work already, and these minor refinements are suggested to help get the best confidence out of all that work.

Question 3: Collaboration effectiveness

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

- The collaboration for this work seems very effective. USC is developing a unique oxygen evolution electrode, UMass is doing modeling work to explain the results, and INL is providing third-party testing to validate USC's results. INL testing adds extra confidence to the results; their facilities are world-class, and they have many highly experienced researchers. Kevin Huang's group is also highly regarded in the field. Mistakes in characterization are always possible, so the agreement of results between the two facilities is critical in trusting the data with great confidence for the impact of this SCT. It's not clear, though, what the split in modeling effort is between UMass and the National Renewable Energy Laboratory (NREL). It is unclear if NREL is doing the multiphysics modeling guided by Professor Jin. Her role in the project needs to be clarified so that her contributions are not lost. She is identified as an expert in the modeling, so it is expected that she's playing a notable role.

- Effective collaborations with Energy Materials Network nodes were clearly demonstrated.
- The team is working effectively with HydroGEN consortium members.
- The project shows good collaboration with INL and NREL on modeling.

Question 4: Relevance/potential impact

This project was rated **3.4** for supporting and advancing progress toward DOE Hydrogen Program goals and the HydroGEN consortium mission.

- Electrode material research is essential to improving the performance of SOEC systems. Results from this project are likely to inform the development of higher-performance stacks within the HydroGEN consortium.
- The main impact of this project is a better understanding of the oxygen electrode stability under high-temperature electrolysis conditions.
- Stability of the SOEC is a major issue for the life cycle cost of such systems. The project is trying to address such issues, especially the oxygen electrode, via coating.
- The coatings appear to help drastically reduce the overpotential at the oxygen evolution electrode. This is one of the largest challenges in the oxygen-conducting SOEC lifetime. The impact will be large if this is scalable and works on full cells. The impact on Cr tolerance will also be very important if it works at full scale since Cr poisoning is a major issue. If system components don't need to be coated, this could potentially decrease system costs. The conformality of the coating is very advantageous. It is unclear what the reason for it is and whether this is a benefit of low surface energy for the SCT. The usefulness of this infiltrated shell layer will depend on how it performs in full cells. It is understood that cell results are coming in budget period 2 (BP2), so those will be interesting to see. If new problems arise when not in symmetric cells and where current densities may be higher, the coating behavior may change. We'll stay tuned for more information next time. The scalability of infiltration processes is always a concern. Attaining 25 wt.% likely requires several repeat infiltrations and processing steps. It will be useful to know how labor-intensive these are at this stage so scaleup can be planned. Infiltration is possible at large scale, but it is still difficult. It was mentioned that SCT has very large coefficient thermal expansion, ~20, compared to that of lanthanum strontium cobalt ferrite–gadolinium-doped ceria, which is typically between 12 and 15. This could be problematic if there are larger thermal changes in large-scale cells during operation at high current densities or when changing from low load to high load, where rapid thermal changes are present.

Question 5: Proposed future work

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

- Proposed future work is reasonable.
- It would be good to also show the budget period 3 proposed work, even if in less detail than BP2. Perhaps the project should mention the partial pressure of oxygen ranges planned for the studies and the current density ranges.
- It is not clear how the future work that is proposed will address issues such as delamination.
- It is not clear when the work for task 2 (manufacturing of larger planar and tubular cells) and task 3 (performance demonstration at pilot scale) will be initiated and incorporated in future work.

Project strengths:

- The main strength of this project is to use the bilayer oxygen electrode approach to address the issues of electrode delamination and chromium poisoning.
- This oxygen evolution electrode coating impacts two of the largest issues in high-temperature electrolysis SOECs based on oxygen conductors (which is most SOECs at this time): the delamination at high currents due to high overpotential and the Cr susceptibility.
- The team has all the needed expertise to achieve the proposed work and goals.
- Innovative materials development is a strength.

Project weaknesses:

- The main weakness of this project is limited effort to evaluate the effect of electrode microstructure on electrode stability.
- It is unclear how delamination will be addressed to meet future project milestones.
- A primary concern is how scalable the infiltration-based process can be.

Recommendations for additions/deletions to project scope:

- The scope seems appropriate for this project.
- It is suggested to the team to conduct preliminary TEA to consider the cost of materials and process versus the proposed benefit.
- The project should down-select planar versus tubular cells for cell fabrication and testing.

Project #P-191: Perovskite–Perovskite Tandem Photoelectrodes for Low-Cost Unassisted Photoelectrochemical Water Splitting

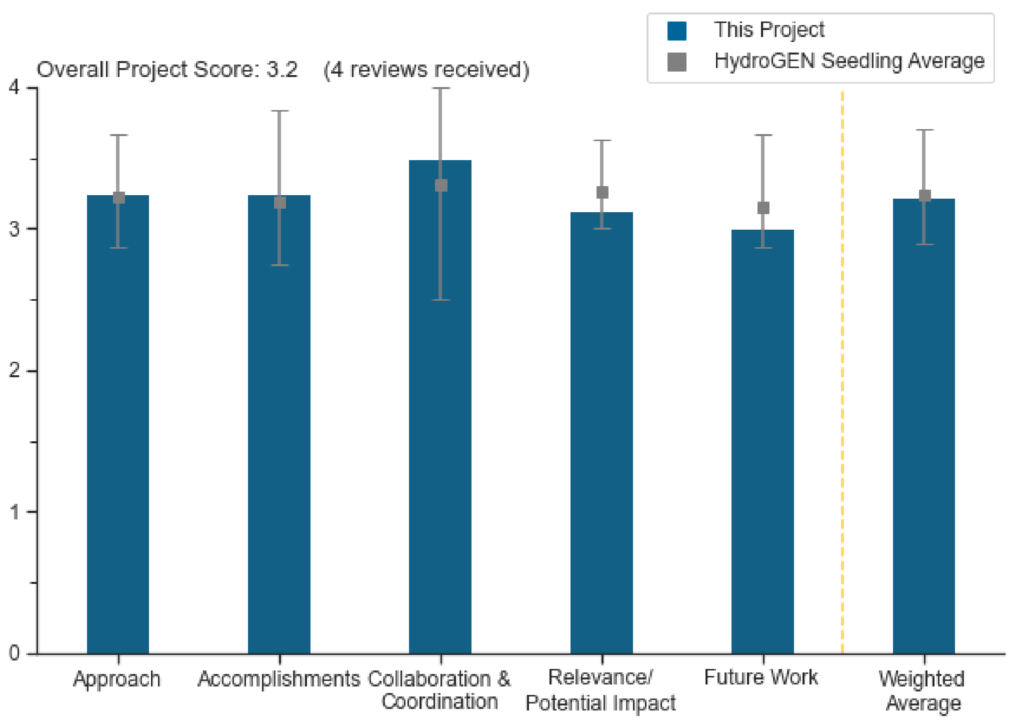
Yanfa Yan, The University of Toledo

DOE Contract #	DE-EE0008837
Start and End Dates	10/1/2019 to 9/30/2022
Partners/Collaborators	National Renewable Energy Laboratory, Lawrence Livermore National Laboratory
Barriers Addressed	• Materials Efficiency - Bulk and Interface: Identify absorber and interfacial materials for efficient hydrogen generation. • Materials Durability - Bulk and Interface: Investigate intrinsic stability; Develop durable protection layers Integrated Device • Configurations: Tandem cell and photoelectrode integration

Project Goal and Brief Summary

This project's goal is to enable cost-effective photoelectrochemical (PEC) water-splitting devices using monolithically integrated perovskite/perovskite tandem photoelectrodes, developed by the research team. If successful, the proposed PEC technology presents a significant technoeconomic advantage over the state-of-the-art spontaneous water-splitting devices. The team aims to demonstrate a high-efficiency and stable PEC system that shows potential to reduce PEC hydrogen generation costs to $2/kg. The University of Toledo is collaborating with the National Renewable Energy Laboratory (NREL) and Lawrence Livermore National Laboratory (LLNL) on this project as part of the HydroGEN consortium.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by HydroGEN Seedling projects.

Question 1: Approach to performing the work

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

- The work centers on the important materials class on perovskite materials and the design of tandem perovskite photoabsorbers. The authors have shown which perovskite materials show sufficient stability to be processed into monolithic devices.
- The project seems to be very well balanced in its approach. It is logically laid out with each major aspect identified and appropriate numerical metrics applied to each.
- The pragmatic shift in approach to wider-bandgap materials and Pb-based perovskite/perovskite tandem to improve stability is good.
- The need to revise milestones so early in the project suggests the approach was not well thought out. The principal investigator (PI) indicated stability was the critical metric, yet the plan is to demonstrate only 100 hours of “stable” operation, the criteria for which is not given.

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, as well as the HydroGEN consortium mission.

- The team showed that Sn/Pb-based photoabsorbers showed significant instability that was not able to be overcome. In addition, the team has identified the p-absorber water interface as the crucial next barrier to address; the reviewer agrees with this determination. Overall, the project progress has been very impressive, even in the presence of the global pandemic.
- The project has made substantial progress on a difficult and complex device structure. However, several of the numerical goals were revised downward. The reasons for the revisions were briefly mentioned but with insufficient details. Nonetheless, the revisions were generally slight and, if based on legitimate reasons, are acceptable. Identification of a suitable interconnecting layer is a big accomplishment. Demonstrating a working tandem device is also a significant achievement.
- The project is extending the community’s understanding of the challenges associated with perovskite devices for water splitting.

Question 3: Collaboration effectiveness

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

- Use of HydroGEN node members NREL and LLNL is exactly on-point for the HydroGEN consortium vision. It rates an “excellent” collaboration.
- The project seems well integrated with HydroGEN.
- The authors showed excellent progress on the project and outlook toward budget period 2. Budget period 2 shows increased engagement with the LLNL nodes; however, it would be beneficial to more explicitly show the type of engagements that have been initiated with NREL in the current period.

Question 4: Relevance/potential impact

This project was rated **3.1** for supporting and advancing progress toward DOE Hydrogen Program goals and the HydroGEN consortium mission.

- Perovskite materials have great potential for PEC hydrogen production. The construction and evaluation of effective monolithic devices are crucial to meeting the DOE targets.
- When the project goals are achieved, the project will have a meaningful impact on meeting the overall goals of low-cost PEC hydrogen.
- It appears the project had to scale back its ambitions around novel materials due to stability issues.

- The upside potential impact of this work seems low given the low stability of the perovskites. It's also tied to the potential impact of PECs in general. It is still a bit unclear how/why PECs will be advantageous in the future compared to photovoltaics plus electrolyzers from a cost, operations, and safety perspective.

Question 5: Proposed future work

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

- The schedule for future work is reasonable and logically described.
- The project identified key challenges that need to be addressed, including hysteresis and durability, but did not articulate a clear plan to address them.
- The projected work for the next budget period was outlined. It is unclear how the project is going to proceed and which tasks and milestones will be continued or changed. The team showed that in situ x-ray experiments are planned, but, beyond that, it was not discussed.

Project strengths:

- The methodical breakdown and investigation of each component of the tandem device is a project strength. The excellent collaboration and use of the consortium team members are strengths.
- Perovskite monolithic devices are being investigated as an integrated solution for solar-to-hydrogen production. The proposed in situ experiments are of particular interest for the development and in defining areas of research going forward.
- This is an exploratory project that will help define the potential for perovskite PEC. There is collaboration with national laboratories.
- This project has a well-rounded team and research approach. The quick changes in plan to mitigate stability issues are appreciated.

Project weaknesses:

- The research activities seem particularly reactive. A more "develop a clear hypothesis, test hypothesis" approach is recommended.
- There isn't verification that achievement of the project goals will lead to the target $200/m^2 device cost. The downward revision of the metrics is a worrying trend.
- The stability of the perovskite device is still a concern. The team needs to have a more concrete plan on the possible degradation pathways and mitigation strategies.
- From what is described in the material, there is no plan for investigating the effect of the studies undertaken on hydrogen costs.

Recommendations for additions/deletions to project scope:

- Besides the work described, a preliminary investigation of the effect of the developments on hydrogen cost is suggested.
- The PI should better explain the reasons for the downward revision of milestone targets.

Project #P-192: Development of Composite Photocatalyst Materials That Are Highly Selective for Solar Hydrogen Production and Their Evaluation in Z-Scheme Reactor Designs

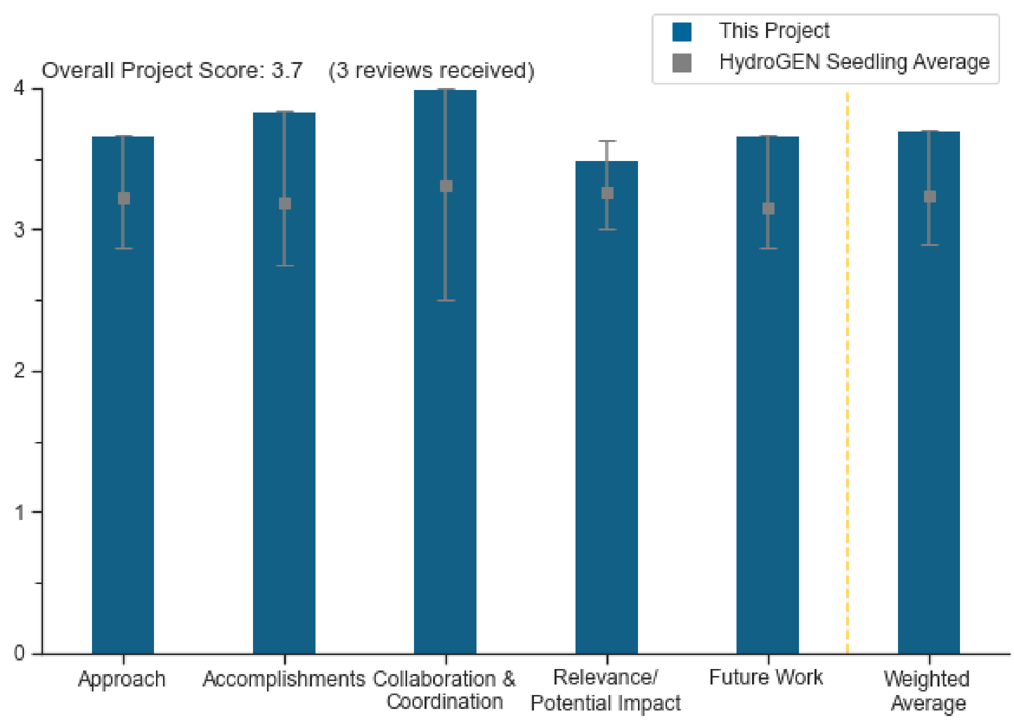
Shane Ardo, University of California, Irvine

DOE Contract #	DE-EE0008838
Start and End Dates	10/1/2019 to 3/31/2023
Partners/Collaborators	University of Michigan, Columbia University, National Renewable Energy Laboratory, Lawrence Livermore National Laboratory, Sandia National Laboratories, Tokyo University of Science, California Institute of Technology, Shinshu University
Barriers Addressed	• Few composite particles are known that selectively evolve H_2 and O_2 instead of performing undesired redox shuttle back reactions • Empirical and numerical results guide our design of ultrathin coatings for selective reactivity, and reactor dimensions for natural convective mixing

Project Goal and Brief Summary

This project aims to develop new photocatalyst particles and ultrathin oxide coatings for photocatalytic solar water splitting that can enable demonstration of the interim DOE target of 3% solar-to-hydrogen efficiency. The goal is to demonstrate a selective ultrathin oxide coating on particles that results in a ≥10 times larger hydrogen evolution quantum yield than for uncoated particles. Using an intrinsically safe tandem (Z-scheme) dual-bed particle suspension reactor design, the project also aims to validate high-efficiency and technoeconomically viable photocatalyst reactors for solar water splitting. The project team includes the University of California, Irvine; the University of Michigan; Columbia University; National Renewable Energy Laboratory (NREL); Lawrence Livermore National Laboratory (LLNL); Sandia National Laboratories (SNL); the Tokyo University of Science; the California Institute of Technology; and Shinshu University.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by HydroGEN Seedling projects.

Question 1: Approach to performing the work

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

- The integrated approach of combining theory, experiments, materials science, and techno-economics is excellent, as it brings the PEC story into a broader context.
- This project is checking all the boxes: experiments, modeling, characterization to develop novel materials, and process concepts.
- The team is clearly guided by techno-economic modeling to target relevant barriers. This work represents an important alternative approach from conventional photoelectrochemical (PEC) systems. Novel catalyst coatings are investigated as a cornerstone of this approach.

Question 2: Accomplishments and progress

This project was rated **3.8** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals, as well as the HydroGEN consortium mission.

- The project has shown excellent progress toward its goals. The team has investigated catalyst coatings that effectively avoid back-reaction of the Z-scheme redox shuttle. The team has also demonstrated the effectiveness of the approach using state-of-the-art photoabsorbers and has outlined how the next budget period will be leveraged to investigate new materials. Using techno-economic analysis (TEA), the team also shows that the project can achieve a significant impact on the cost of hydrogen. The project progress has been very impressive, even in the presence of the global pandemic.
- This project has hit all of the milestones, with deep scientific study behind each one.

Question 3: Collaboration effectiveness

This project was rated **4.0** for its collaboration and coordination with HydroGEN and other research entities.

- The team has been doing a pretty good job at collaborating across principal investigators (PIs) and the HydroGEN nodes.
- The team is clearly well connected with the HydroGEN nodes, as well as several other universities.
- The team has clearly shown how the various nodes have been engaged.

Question 4: Relevance/potential impact

This project was rated **3.5** for supporting and advancing progress toward DOE Hydrogen Program goals and the HydroGEN consortium mission.

- The team has done a great job in demonstrating the potential of this technology and in advancing it.
- While the current technology readiness level (TRL) of the proposed designs is less advanced than other projects, it underlines the importance of keeping all options open in order to achieve the goal of sustainable and cheap hydrogen.
- The overall project work is good; however, from an industrial scalability perspective, as well as the $2/kg target (now $1/kg), it is a bit unclear on how this will beat photovoltaic–electrolysis (PV–electrolysis) systems that are a lot more mature. The statement holds for PEC projects in general. Hence, this is something to be mindful of when funding future work in this space.
- The project is a good model for how to leverage DOE funding to advance PEC; however, this reviewer is skeptical that this approach to PEC can be scaled up and therefore will have an ultimate impact on the cost of hydrogen. It is worth some level of DOE funding, but the ultimate impact may be in applying some of the materials concepts to other water splitting approaches.

Question 5: Proposed future work

This project was rated **3.7** for effective and logical planning.

- The project has a well-thought-out plan and is on schedule despite the COVID-19 pandemic.

- The team has outlined how new, more efficient photoabsorbers will be investigated in the next project period.

Project strengths:

- The project team works well across multiple team members and shows great progress. Furthermore, it demonstrated how powerful an integrated approach of experimental development with TEA analysis can be in identifying roadblocks early and mitigating them. An example is the elimination of active circulation of the system that clearly was a problem that was solved elegantly by employing natural convection.
- This project's breadth and depth of scientific inquiry are strengths, as it is not Edisonian. Its collaborations are also a strength.
- This is a strong team with strong collaboration. The project has a good approach that combines modeling, materials science, experimentation, and technoeconomic modeling.

Project weaknesses:

- The main limitation of the project is the TRL level, which is still low. This makes this project even more crucial in order to ensure multiple technologies have a shot at solving the problem of sustainable hydrogen.
- Although the project team is doing some nice fundamental work around PEC systems, the scalability challenges related to the use of rare earth materials, such as Ir or Sr, need to be addressed. In particular, given the rare availability of such materials, it is not clear if it is even possible to build world-scale green hydrogen facilities (10–100 kilo tons/year). If not, it is questioned if there is an envisioned pathway to a more scalable set of materials.
- The project has no plan to demonstrate stability.

Recommendations for additions/deletions to project scope:

- The team should demonstrate stability. Ideally, the team should comment on how material or modeling innovations might translate to other HydroGEN targets.
- The team's suggestion of moving to more efficient photoabsorbers, which can be accessed due to the progress made so far, is of high importance to achieve a more efficient system. Furthermore, the construction of a functional device as a demonstration in the next phase is also very critical.
- The work on the TEA is interesting. The team should consider looking at the different reactor designs from an operability and safety perspective, especially when compared to PV–electrolysis. This reviewer worries that due to low overall yields per unit reactor volume for PEC, a hazard is being distributed (H_2 and O_2 in stoichiometric ratios) over fairly large areas versus a polymer electrolyte membrane electrolyzer, which has a smaller hazardous area.

Project #P-193: Highly Efficient Solar Water Splitting Using Three-Dimensional/Two-Dimensional Hydrophobic Perovskites with Corrosion-Resistant Barriers

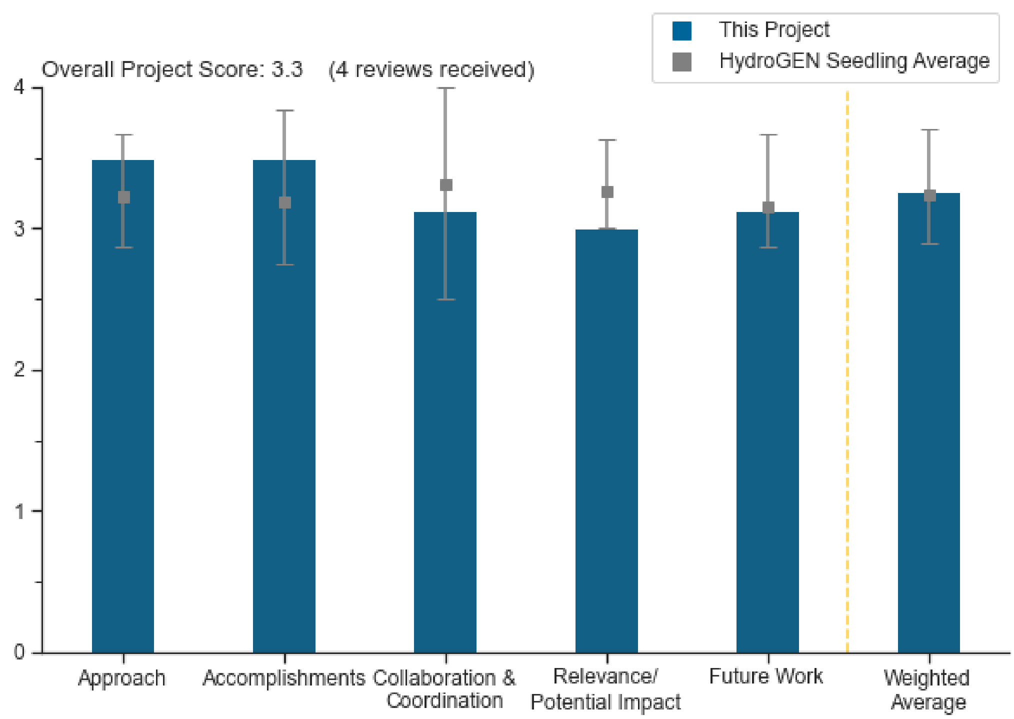
Aditya D. Mohite, William Marsh Rice University

DOE Contract #	DE-EE0008843
Start and End Dates	01/01/20 to 01/01/23
Partners/Collaborators	Lawrence Berkeley National Laboratory, National Renewable Energy Laboratory
Barriers Addressed	• Hydrophobic polymers, carbons, atomic layer deposition oxides

Project Goal and Brief Summary

Rice University aims to demonstrate an innovative concept with advanced materials for photoelectrochemical (PEC) cells based on direct water splitting to produce hydrogen fuel. The project team is combining high-efficiency, low-cost halide perovskite (HaP) solar cells with hydrogen evolution reaction (HER) and oxygen evolution reaction (OER) catalysts to demonstrate an integrated HaP-PEC cell with 20% solar-to-hydrogen efficiency and 500 hours of operational durability. If successful, this project, in collaboration with Lawrence Berkeley National Laboratory (LBNL) and the National Renewable Energy Laboratory (NREL) through the HydroGEN consortium, will demonstrate a water-splitting system that can produce hydrogen at scale using low-cost, abundant materials.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by HydroGEN Seedling projects.

Question 1: Approach to performing the work

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

- The approach is well-considered and encompasses the main elements of the complete device. The selection of HaP-PEC cells is an interesting approach.
- The team has identified a neat approach that combines the best possible performance of tandem perovskite cells by making the top and bottom cell in parallel. This will allow the best evaluation of the performance in the absence of the practical issues of layering absorbers and catalyst; however, this will mean a minimum 50% loss in theoretical efficiency. While this trade-off was clearly identified, the practical limitation on the cost of hydrogen was not clearly identified in the presented work.
- The project is focused on the most important challenge for perovskite PEC. However, it is not clear what the potential is for the project's materials innovation to be useful in other HydroGEN strategies.

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, as well as the HydroGEN consortium mission.

- Since the team chose a device design approach that utilized some of the state-of-the-art perovskite materials, the focus of the project and progress are on the stability of the system. The team showed significant progress toward a complete device operating under realistic conditions, as well as advances in the design of two- and three-dimensional perovskite materials that showed great promise. Owing to intellectual property considerations, the details of the protection layers strategy for the individual photoelectrodes could not be discussed, but the advertised properties were promising. During the stability measurements of the tandem device, the team showed a good initial performance, followed by a rapid decrease in performance. While the team could not share the experimental evidence showing that the stability issues were due to the OER catalyst, not the perovskite photoabsorber, the principal investigator provided detailed descriptions of the team's recent advances, which showed good progress on achieving higher stability. Most of the milestones set for this project period have been achieved or were described by the team as being under way; based on this, progress is on track to deliver on the targets identified. Overall, the project progress has been very impressive, even in the presence of the global pandemic.
- Demonstration of 12.4% efficiency with a perovskite PEC device is a very good Year 1 accomplishment. The evaluation of the four-barrier-layer concept and the down-select to one is a good achievement.
- The synthesis of the first-ever water-stable perovskite and identification of the patentable barrier are impressive.
- The team has been making good progress on the HER side of the reaction. However, challenges remain on the OER side. It will be interesting to see the innovative degradation mitigation strategies the team devises.

Question 3: Collaboration effectiveness

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

- Collaboration with NREL and LBNL is appropriate and worthwhile to the project.
- Rice University is collaborating with two national laboratories for benchmarking and characterization, but the impression was that these collaborations were only arms-length interactions.
- While the project showed great progress, it is unclear how the HydroGEN nodes have been leveraged and to what extent the team has an integrated approach.
- There are none in particular. The use of NREL as a collaborator, especially around benchmarking, is appreciated, but further use of NREL's capabilities is encouraged.

Question 4: Relevance/potential impact

This project was rated **3.0** for supporting and advancing progress toward DOE Hydrogen Program goals and the HydroGEN consortium mission.

- A perovskite-based PEC device should be low-cost, and if efficiency and durability targets are achieved, the overall project will be a large step toward achieving overall DOE PEC goals.
- The team showed that the proposed strategy can achieve high solar-to-hydrogen efficiencies of 10%. The impact of high efficiency on hydrogen cost was not demonstrated by technoeconomic analysis at the time of the Annual Merit Review but was identified as a later milestone. Still, a top-level description of such an approach would have been beneficial for evaluating its impact.
- It is an innovative idea to explore and develop the potential of low-cost perovskites as a way to enable cost-effective PEC.
- The potential impact of this work is tied to the potential impact of PEC cells in general. It is still a bit unclear how and why PEC cells will be more advantageous in the future compared to photovoltaics (PV) + electrolyzers from a cost, operations, and safety perspective, especially if the solar PV materials are similar to the PEC options and anion-exchange-membrane-based water electrolyzers become mature.

Question 5: Proposed future work

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

- Budget period 3 not only continues the progress gathered in periods 1 and 2 but also expands into technoeconomic evaluation to determine the impact of the discoveries. Of particular interest is the work on stabilizing coatings for both anodes and cathodes that show functionality in both OER and HER catalysis.
- There are logical and clearly stated future work plans.
- It is strongly recommended that the team prioritize/focus on improving the durability of the device with regard to the OER side.

Project strengths:

- The clearly defined device concept is a project strength. Achievement of >12% efficiency is a strength. The examination and test of multiple barrier concepts is a strength. Demonstration of a water-stable perovskite is a strength.
- The project uses an innovative approach and has an effective problem-solving team.
- The team has shown excellent progress in developing novel protective coatings for perovskite photoabsorbers, as well as producing a demonstration device.
- The progress on stable coatings for the halide perovskite, at least for the HER half reaction, is quite promising.

Project weaknesses:

- Lack of any cost targets or analysis is a weakness. The claim “demonstrated a near-ideal corrosion barrier” is not supported by the slides. It is not clear which barrier layer they are referring to. The rapid deterioration during the 2.5-hour solar-water-splitting test is described as OER catalyst degradation, but no explanation is given for its rapid and precipitous onset just after two hours.
- The project relies on a strategy that immediately reduces the theoretical efficiency by 50% compared to a stacked device. In addition, the cost of materials (owing to a doubling of the device size) is also going to increase the hydrogen price.
- Durability of the OER half reaction is a concern. The team should prioritize improving that or at least create a mitigation plan around it.
- There is a lack of modeling to complement the experimental approach.

Recommendations for additions/deletions to project scope:

- The technoeconomic analysis proposed as a milestone for the next phase of the project is important and should be accomplished. This will be key to determining the full impact of this work.
- A modeling component focused on elucidating the fundamental degradation mechanism should be added.
- More discussion of failure mechanisms in the project presentation is recommended.

Project #P-194: New High-Entropy Perovskite Oxides with Increased Reducibility and Stability for Thermochemical Hydrogen Generation

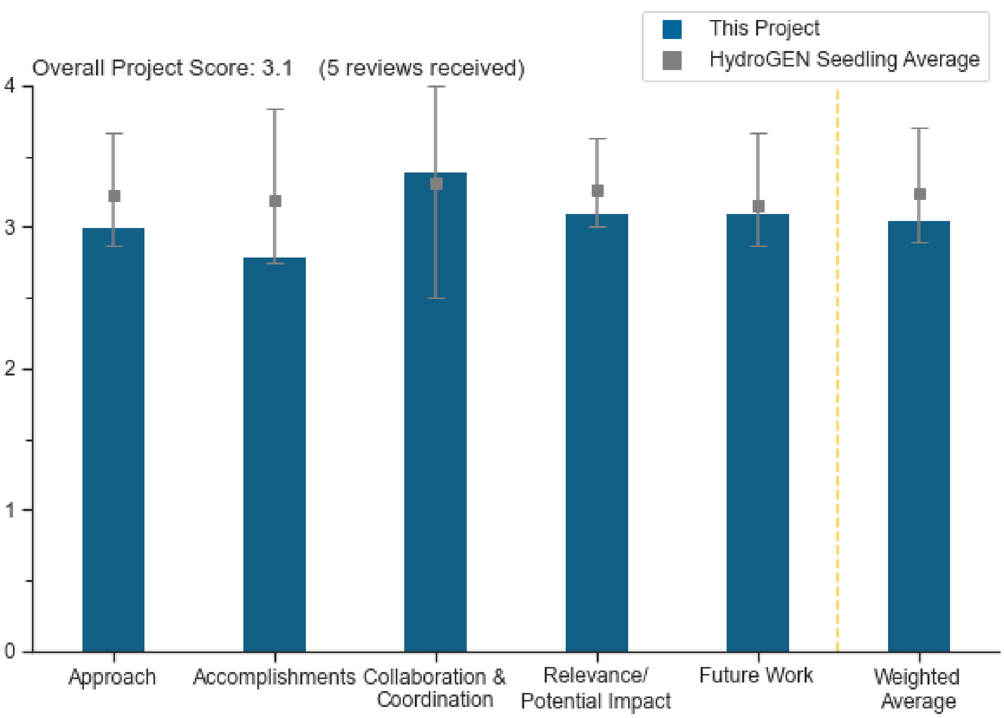
Jian Luo, University of California, San Diego

DOE Contract #	DE-EE008839
Start and End Dates	10/1/2019 to 1/31/2023
Partners/Collaborators	West Virginia University, Michigan State University, Brown University, Sandia National Laboratories, National Renewable Energy Laboratory
Barriers Addressed	• Extremely vast compositional space • Highly complex compositions and structures • Compositional controls in the synthesis of many-component oxides • Correlation of computation with experiment

Project Goal and Brief Summary

University of California, San Diego (UCSD), aims to design, synthesize, and test a transformative class of high-entropy perovskite oxides (HEPOs) as redox oxides to enable thermochemical hydrogen generation with improved stability, kinetics, and efficiency. If successful, this project will validate the usefulness of a new field of water-splitting materials and establish a new class of high-entropy redox oxides with a vast, unexplored compositional space. Along with project partners, UCSD will develop a HEPO that is able to deliver a H_2 yield of over 400 µmol per gram oxide and demonstrate high stability with less than 20% degradation after at least 50 cycles.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by HydroGEN Seedling projects.

Question 1: Approach to performing the work

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

- Overall, the approach is outstanding. The high-throughput synthesis technique appears robust and has allowed for the successful synthesis of some very challenging compositions. The computation effort is also on very solid footing, although it is concerning to move away from tolerance factors (TFs) and into vacancy formation energy calculations, as they may become exceedingly costly from the aspect of computational time. It does appear that the calculations are only informing the cation selection process rather than driving it, so the risk may be mitigated. It is not apparent if starting oxygen stoichiometry is being taken into account or if all compositions are assumed to be stoichiometric with enough transition metal available to compensate for charge imbalances. The experimental screening is the only part that could use some improvement. The cycling between N2 and air is nominally sufficient; however, more information can be captured by beginning the reduction from room temperature rather than cycling only from the oxidation temperature in air. It is possible that this information is already being captured but not presented; however, the onset temperature for reduction is an important metric and is worthy of inclusion. Finally, some caution should be heeded when evaluating the oxidation kinetics with air rather than steam, as it is unlikely to be representative. The performance of most perovskites evaluated by the community thus far have shown significant kinetic limitations during water splitting that were not apparent in air oxidation.
- The approach in this project is to design, synthesize, and test a "transformative class" of HEPOs as redox-active oxides with the objective of enabling thermochemical hydrogen generation with improved stability, kinetics, and efficiency. The goal is to develop new enabling design strategies and methods, including the synthesis of HEPOs, which have a vast compositional space and tunability. The project identifies barriers to realizing the approach but stops short of identifying what barriers need to be addressed to advance the technology. Hence, the barriers are not well-identified, and therefore, it is not possible to tell whether they will be addressed well through this project's innovation. That said, the approach to investigating a "new class" of materials that increases the possible composition space is interesting and promising enough that the approach is good. The project is well-designed to find a large number of redox-active materials with large potential oxygen off-stoichiometries that are also cyclable and have fast kinetics. However, whether it is well-designed to identify optimal water splitters is less obvious, as no water-splitting experiments were shown, and the criterion of high off-stoichiometry does not imply good water-splitting potential. It seems the project is well enough integrated with at least one HydroGEN consortium node at Sandia National Laboratories, but there is still not a single water-splitting experiment. To validate the project's technology innovation, the project must show a capability to split water and should not have a singular focus on materials with large $\Delta\delta$ quantification and relatively low energy vacancy formation energies. The attempts at developing a theory that can explain the material thermodynamics are commendable; this effort seems at an early stage, and more development along those lines is expected by next year's review, and that might help sharpen up the design criteria. The researchers have relaxed their original criterion related to the stability of the structure relative to cubic, which seems appropriate; however, the new criterion is now "the predicted $\Delta\delta$ (with oxygen vacancy interaction and distribution) meets the HydroGEN requirement." It is unclear what this criterion means, and therefore, it seems too non-specific to meet the objective of being a useful design criterion essential to the approach to provide guidance moving forward.
- The approach to solving the problem of evaluating HEPOs as redox mediators for water splitting is generally very good. The biggest opportunities for improvement are to leverage approaches and results developed and obtained in the earlier HydroGEN Seedling projects. This would allow the team to focus on answering questions that have not yet been answered, rather than rediscovering much of what has already been learned from other seedling projects.
- The project aims to discover materials for thermochemical hydrogen production. The hypothesis appears to be that such materials could exist among perovskite oxides, specifically those with high entropy. From a computational and materials standpoint, the approach is strong and seems likely to identify many materials with the target properties. To have relevance to water splitting, however, the overall approach needs an improvement in direction and a deeper understanding of actual thermochemical cycles. First, the team is targeting materials with a vacancy formation energy of 2–3 eV (slide 7 and slide 14). With the enthalpy of water splitting at 2.5 eV, materials below this value would be unable to split water and are therefore not relevant to the project. The upper part of the target range is at least thermodynamically feasible but is

highly unlikely. For reference, the vacancy formation energy for ceria is ~4.5 eV. Second, materials seem to be evaluated under conditions that are not relevant to water splitting. Specifically, the reversible oxygen capacity ($\Delta\delta$) is evaluated for reoxidation at 870□C in partial pressure of oxygen (pO2) of ~21,000 kPa. For comparison, the pO2 in steam (with no hydrogen) at the same temperature is ~0.15 Pa, or about 140,000 times lower. In the presence of hydrogen, this pO2 is lower still by many orders of magnitude. Therefore, reoxidation in air and the associated $\Delta\delta$ cannot identify promising materials for water splitting (slide 16), and it is unclear why such experiments were performed so extensively (perhaps they verify material stability). At best, the experiments can identify materials that are of no value (i.e., have a low $\Delta\delta$, even under these highly favorable conditions, or decompose). It would have been somewhat more relevant to use the same gas (i.e., 10 ppm O2) for both reduction and oxidation.

- The team is using the Goldschmidt TF in its modeling. There are newer factors that are better and that should be considered. The researchers used computation to identify potential B-site elements. They had two series of suggestions. To get the second series, they increased the TF range from 0.95–1.05 to 0.9–1.05. It would be helpful to understand why this adjustment was made.

Question 2: Accomplishments and progress

This project was rated **2.8** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals, as well as the HydroGEN consortium mission.

- The project has a number of notable accomplishments, especially in synthesizing a large number of compounds, doing a first-level screen on those compositions, identifying a number of interesting compounds, demonstrating redox activity and cyclability, and measuring some kinetics. In addition, there are efforts to produce thermodynamic models. All this is commendable, especially given that the compositional space is extremely large. The project has shown dual redox activity from cobalt and manganese in at least one combination, which is very interesting. Relative to the chosen milestones and go/no-go, the progress looks very good, and it is good to see a focus on the kinetics. What is not evident is whether the project is addressing barriers that will advance the technology. The researchers state that they have met the milestone BP 1 go/no-go target ($k \geq 7.5\times10^{-4}$ cm/s) and 90% of $\Delta\delta$ in an hour. What they do not state is why these are the right milestones to overcome specific barriers to advance the technology, and the team has not shown evidence that any of the materials that have been screened and used to meet milestones will split water.
- This group has a number of excellent accomplishments, from the large number of synthesized and screened materials to the modeling that produced oxygen vacancy formation energy calculations for quinary cation oxides. However, there are concerns that the project is pushing too hard on the goal of increased reduction (delta). With DOE's stated goal of solar thermochemical hydrogen materials that are viable at high conversion rates, it seems unlikely that a perovskite that reaches a delta >0.2 will be capable of splitting water under those conditions. The middle range of 0.1–0.2 seems more reasonable, and there may even be opportunities below 0.1. It would also have been helpful to see a list of any compositions the team could not synthesize, if any exist.
- It is not clear why the team increased the TF range for material identification. It seems the team increased it solely to give itself more materials to test (slide 13). High-throughput synthesis was thoughtfully done. On slides 18 and 19, phase stability is reported, but the conditions of the tests, including the number of cycles, were not given. Without knowing the experimental conditions, it is hard to determine whether the materials are indeed stable. The presenter did an admirable job of identifying where HydroGEN nodes were used. For the kinetics work, it would be helpful for the presenter to identify the kinetics goals—whether a 15-minute cycle is enough or it needs to be faster. The oxidation was done with oxygen gas. It is recommended that the project use steam to better match real operation. The team needs to show multiple cycles.
- The team is generally making good progress, but progress could be accelerated by not reinvestigating issues that have already been investigated by the seedling projects. Interaction with the seedling projects to learn the advances that have not yet been published, and using what they have already published, could greatly accelerate the project. For instance, using techniques developed within the HydroGEN consortium would have allowed the project to screen far more than the 100 compositions that it has currently screened. It was not clear whether progress had been made on screening for kinetics (or how this would be done).

- The project is making good progress toward goals but would benefit from aligning with DOE goals.

Question 3: Collaboration effectiveness

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

- There is clear evidence of collaboration with the HydroGEN consortium nodes at Sandia National Laboratories. The team has engaged with the benchmarking/protocols (2b) project by participating in the workshops. It is unclear whether the team has engaged with the HydroGEN Data Hub. It is hard to tell whether the partners are well-coordinated.
- The institution collaboration appears to be working well, with contributions coming from all three members. The groups also participated in the latest benchmarking and protocols workshop. The reviewer is unable to judge contributions to the Data Hub, however, as the public-facing count of zero datasets does not consider all submissions. The consortium nodes have provided meaningful contributions that directly support the project's successes.
- The project's internal collaboration is outstanding, while the use of the nodes is very good. Interactions with other teams within the consortium is limited but could greatly add to the success of the project.
- The team appears to be effective and making good use of HydroGEN research infrastructure.
- The presenter did a good job identifying what nodes were used and what was learned. The use of HydroGEN nodes is consistent. The team should have used the HydroGEN team to refine its experiments prior to doing the experiments.

Question 4: Relevance/potential impact

This project was rated **3.1** for supporting and advancing progress toward DOE Hydrogen Program goals and the HydroGEN consortium mission.

- By looking at HEPO materials and synthesizing these complicated systems, the project has already made contributions to the discovery of new materials. Going forward, the large number of materials and the data collected about them will help not only this project but others who want to mine the results to glean possible underlying principles.
- This project has the potential to make a significant impact toward the DOE HydroGEN consortium goals and objectives.
- The project supports the HydroGEN consortium mission of theory-guided materials discovery for redox-active metal oxides that can split water. The project has significant potential to advance the discovery and development of novel advanced water-splitting materials when the project establishes the appropriate screening criteria. An improvement over the state-of-the-art water-splitting material is necessary, although not sufficient to enable meeting the DOE ultimate hydrogen production goal of $2/kg H2. Project aspects align with some of the DOE Hydrogen Program and DOE research, development, and demonstration objectives, and the project is leveraging and contributing to the resources and framework of the HydroGEN consortium to some extent.
- The project would benefit from a realignment that would make otherwise excellent materials work relevant to DOE goals.
- It is too early to tell the potential impact of this project.

Question 5: Proposed future work

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

- The future plan should be effective and should contribute to overcoming most of the identified barriers to the success of the project. Additionally, the project appears to be on track to meet most, if not all, of the end-of-project goals and to advance the materials research mission of the HydroGEN consortium. The researchers have proposed some challenging new efforts, including trying to develop a new thermodynamic model for the relationship between partial pressure of oxygen, temperature, and, and most importantly, the team will be doing some water splitting, ideally in collaboration with the nodes. In addition, the intent to move beyond equimolar compositions will make the composition space much larger, thereby making it a challenging undertaking but potentially that much more interesting.

- Plans are in place to address one of the main project weaknesses, namely cycling conditions. This is a positive development and should be prioritized.
- The future work appears both ambitious and conservative. This is especially true for some of the new efforts. Expanding the compositional space to explore more non-equimolar compositions could be a very large rabbit hole to traverse if sufficient direction is not involved. There does not appear to be a plan for using the vast results from the earlier budget periods to guide this work. Some of the characterization work, on the other hand, seems interesting, but it is unclear what contributions it will provide, considering the overall thrust of the project. It would have been good to see a more concerted effort to produce water-splitting results on a large cross-section of the discovered and synthesized HEPO materials and attempt to make correlations to better guide both the theoretical modeling work and further synthesis.
- In addition to the proposed improvements to the model, the researchers should use a factor different from the Goldschmidt. For the foam samples, the team needs to include some tests on the mechanical strength. The project needs to clearly identify its performance goals.
- The sequence of looking at A-site mixing and then B-site mixing in the subsequent budget period does not seem effective. There is significant potential for coupling the effects caused by the separate alloying on the A-site and B-site sub-lattices. Learning this later in the project could lead to a much less successful materials discovery effort.

Project strengths:

- The approach of exploring the rich compositional space of high-entropy perovskite oxides is a strength. The high-throughput synthesis capability is a strength, as this team seems to have been able to screen a large number of compositions. Having demonstrated dual cation redox activity (cobalt and manganese) is a strength.
- The work division is a strong suit of this project. Each institution has a manageable scope of work and appears to be leveraging its individual strengths. The synthesis and initial characterization throughput is impressive.
- This is a strong, well-organized team with complementary abilities. The project has a generally good approach and methods, as well as good theoretical concepts to explore.
- This is a well-rounded team. The researchers are using good high-throughput production methods. The project is using the HydroGEN nodes well.
- The strength of this project is the strong computational materials basis and the effective team.

Project weaknesses:

- It seems that the project did not have the right equipment for the tests it was running. It seems it is building the test stands and will be able to use them in future tests. It would be helpful if the team included more description of the test conditions and what success looks like.
- The non-stoichiometry and kinetics experiments could use work. Too much importance is being conferred to the extent of reduction over other metrics, and the fitting of kinetic parameters for oxidation in air is of limited usefulness. It appears that not enough effort is going into leveraging the existing results to guide further development. The project is producing a treasure trove of results that should be utilized to a greater extent.
- There is a limited number of compositions evaluated to date. There is no screening for kinetics. The team is not using the lessons learned from previous HydroGEN efforts, and it was not clear how the project would determine which phase to use for predicting vacancy formation energies.
- The team has materials to characterize and yet has not demonstrated water splitting. The researchers have not given a rationale for their guiding criteria.
- The team does not appear to have a firm understanding of the nuances of the actual application, water splitting. Such understanding would significantly improve the project’s effectiveness and impact.

Recommendations for additions/deletions to project scope:

- The approach is sound and needs only a better focus to significantly increase the impact.
- Nothing formal needs to be changed, but some slight adjustments to the approach are likely sufficient.
- The scope is appropriate; one addition should be to determine design criteria with a clear, understandable rationale for the design criteria.
- The team did not have the right equipment for the tests the project was running. It seems it is building the test stands and will be able to use them in future tests. It would be helpful if the team included more description of the test conditions and what success looks like. It may be useful for this team to consult with HydroGEN and the Benchmarking/Testing Protocol project for the tests they run. Since the goal of the project is water splitting, it is recommended that the team test the material oxidation with steam and not oxygen.
- The project needs to be explicit about its plans for screening kinetics.

Project #P-195: A New Paradigm for Materials Discovery and Development for Lower-Temperature and Isothermal Thermochemical Hydrogen Production

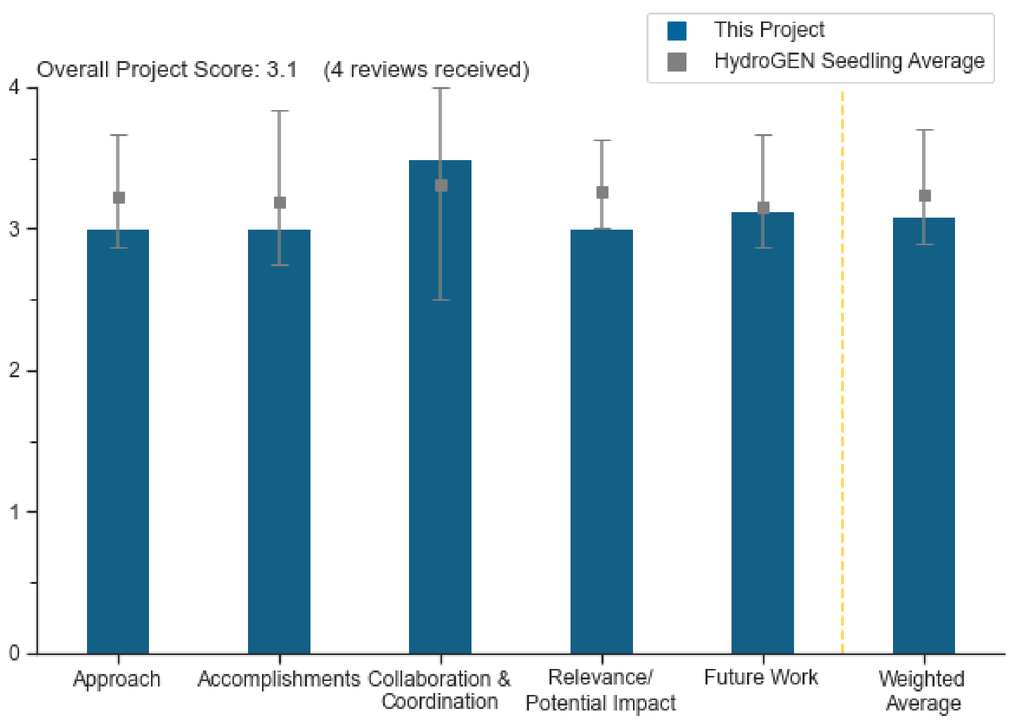
Jonathan Scheffe, University of Florida

DOE Contract #	DE-EE0008840
Start and End Dates	1/1/2020 to 1/1/2023
Partners/Collaborators	National Renewable Energy Laboratory, Sandia National Laboratories
Barriers Addressed	• Computational accuracy coupled with high throughput • Material stability and kinetics – phase stability and redox capability. Does it work? • Scale-up synthesis and stability – porous structure synthesis and characterization with simulator and laser heating

Project Goal and Brief Summary

University of Florida aims to combine computational and experimental efforts toward the development and demonstration of novel materials for efficient solar thermochemical hydrogen (STCH) production under isothermal operation. If successful, this project will provide a pathway to scalable and STCH hydrogen production with a solar to fuel efficiency > 26%, allowing STCH producers to reach the U.S. Department of Energy target of less than \$2/kg H_2.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by HydroGEN Seedling projects.

Question 1: Approach to performing the work

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

- The team is trying to lower the operating temperature and/or operate isothermally. Lowering the operation temperature for STCH is a worthy task. Isothermal operation may result in higher system efficiencies. The researchers are trying to develop foams instead of powders. This could result in some improvements. The foams the team is making should include mechanical strength testing, which is missing, or at least not reported, in this work.
- The main hypothesis of the project is that lower temperature and isothermal thermochemical hydrogen production will overcome the engineering difficulties of high-temperature, thermal swing thermochemical cycles. Although the hypothesis is highly questionable, not supported by thermodynamics, and contrary to approaches in highly successful fields (such as turbines), the team has undertaken a comprehensive and well-structured materials search under it. Assuming performance metrics omissions is addressed in future work, the project is highly likely to conclusively disprove the original hypothesis and help focus the field onto more promising directions.
- The approach is generally good, but there is not sufficient detail to know whether the team is using the lessons learned from previous HydroGEN materials discovery projects. From the information that was presented, it appears that there is significant opportunity to leverage advances made from the HydroGEN Seedling efforts.
- The approaches being taken by the three efforts within the project appear reasonable. However, they do not all seem to align with the broader goals of the project, nor the approach that is proffered on slide 4. Specifically, while the computational effort is looking for new manganates with higher oxygen vacancy formation energies, this does not address the stated goal of finding combinations of enthalpy and entropy that will make isothermal splitting viable. In fact, there is no mention of entropic effects being modeled at all. While the foam development is well designed, it seems better utilized on compositions that are already identified as viable, but the powder testing to identify those candidates is lacking in detail.

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, as well as the HydroGEN consortium mission.

- The project describes a number of accomplishments and has met most of its milestones. The foam development has produced excellent results, and it will be interesting to see how well it translates to newer compositions. With the help of the nodes, the project has also highlighted some of the challenges with working with strontium-containing perovskites, so the computational work that has investigated alternative A-site compositions could be very beneficial. The existing cycling work is also a good start on quantifying whether the surface issues seen in the lanthanum–strontium–manganite (LSM) family are a large concern. The project's accomplishments on milestones makes it clear that the team is poised to achieve the budget period 1 (BP 1) go/no-go target. Considering the early deadlines on the Annual Merit Review slides, perhaps the team has since tested the new compositions.
- The research team was able to synthesize and test foams. The results on slide 14 were obtained at a very high temperature of 1350°C. This is higher than other STCH materials and is in the opposite direction of the project's stated goal of lowering operation conditions. The researchers should be commended for showing experimental error in their data (for example, on slide 14). This is a best practice that should be encouraged. The data reported in slide 14 indicated that there was no difference in performance when the water hydrogen ratio was changed. It is unclear whether this was a product of the high test temperature. The team was able to cycle for 50 cycles, but only 10 cycles are shown. It would be helpful to show all 50, or at least the first 10 and the last 10. On slide 15, the cycle time appeared to be about 30 minutes. It is unclear whether this was the target. Slide 16 shows an x-ray photoelectron spectroscopy (XPS) graph. It would be helpful if the researchers stated what we are to learn from the graph. It would also be helpful if they would explain why the temperature used for this experiment was a different from those used for other experiments. Perhaps this was an equipment limitation.

- The team is making excellent progress toward meeting project goals, especially in terms of material identification, synthesis, and characterization. It must be noted, however, that the main technical performance metric (page 14) omits key elements and that, in its current form, it is difficult to evaluate the relevance of project outcomes to practical thermochemical hydrogen production. Recalling that no useful work can be performed by a cycle without a temperature difference (also the Second Law), the only useful work in the process, as proposed, must come from external work input. In this case, that work appears to be in the form of separation (of H2 from a H2/H2O(g) mixture and of O2 from an O2/N2 mixture). If such input is ultimately derived from heat, then conversion efficiencies should be accounted for. However, these key energy inputs (and associated parasitics, such as conversion of heat to separation and pumping work) are completely absent from the proposed metric (page 14). It is absolutely vital to include these inputs in the performance metric in future work.
- The project appears to be making good progress; however, it was difficult to evaluate how many materials have been screened, what lessons/principles were learned (specifically from the computational screening), and how those principles are guiding the continuing effort.

Question 3: Collaboration effectiveness

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

- The project has utilized several node resources and achieved meaningful results. The interactions with the nodes and with the broader advanced water-splitting community are excellent. Professor Scheffe is an active participant in the benchmarking efforts, both in developing protocols and in overall discussions during workshops.
- The team is collaborating with other team members and HydroGEN nodes effectively. There is an opportunity to increase the amount of interaction/collaboration for other projects funded by HydroGEN that would make both this project and other HydroGEN-funded projects more effective and successful.
- The team appears well-coordinated internally and in work with the HydroGEN infrastructure.
- The research team effectively used the HydroGEN nodes. The research team worked with the benchmarking team to develop protocols. The research team was composed solely of professors from the same university. The project may have benefited from some additional collaborators. For example, the team could have examined additional materials.

Question 4: Relevance/potential impact

This project was rated **3.0** for supporting and advancing progress toward DOE Hydrogen Program goals and the HydroGEN consortium mission.

- The relevance and impact of the project on DOE goals are twofold. First, as indicated earlier, the main premise of the approach is not supported by thermodynamics. It has, however, enjoyed periodic popularity, largely driven by difficulties in addressing truly hard engineering challenges. This project presents an excellent opportunity to conclusively disprove the premise and help focus the field. To do so, performance metrics must be revised to address key omissions. Second, the project is reasonably likely to create methods and capabilities that can be useful in the future.
- If successful, this project has the potential to discover new materials that can dramatically improve upon the hydrogen production capacity of existing materials. However, it is unclear from the report how quickly materials are being evaluated and whether the approach will be sufficiently directed and efficient to evaluate enough materials, and materials in the right chemical space, to discover superior materials.
- The larger impact has yet to be realized. It depends on the efficiency arguments, and those efficiencies lean heavily on hydrogen separation techniques, which are not included in any analysis. By the project's own results, steam-to-hydrogen ratios lower than 200 realize dramatic penalties in yield. This means active separation and recirculation will be critical, regardless of the hydrogen produced. Higher production will require commensurate increases in steam, which cannot just be condensed out of the product stream. The parasitic losses to the separation process could be difficult to keep below the heat exchange improvements afforded the isothermal process.
- It is too early in the material development to determine the potential impact.

Question 5: Proposed future work

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

- The proposed future work is both appropriate and relevant. One possible exception is the proposed fabrication of ceramic foams and their subsequent characterization.
- It is good that the researchers will be looking at additional materials. The work on the foams seems well-thought-out. The team needs to consider mechanical strength considerations.
- The proposed future work was not sufficiently detailed to enable understanding of what will be explored and how results and lessons learned are guiding adjustments to the work plan. If the project is successful, it should uncover principles that will motivate revising the future proposed work toward materials that are more likely to be successful, but the report does not provide information about what the lessons are and how they will be used and, in some cases, how what was discovered repeats lessons learned from previous work on STCH redox mediators.
- The proposed and planned future work is acceptable. It builds upon the finding from earlier budget periods and is in line with the initial roadmap. It is unclear whether the existing workflows will be capable of keeping pace with an increase in candidates to be synthesized and tested, however. The realistic solar testing seems premature/unnecessary, considering the effectiveness of the other testing proposed, but it could be valuable to know if there are large discrepancies between the in-laboratory testing and more real-world scenarios.

Project strengths:

- Perhaps the biggest strength is investigating isothermal cycles at all. It is still important work, and there are many unanswered questions that need to be either answered or marked as unknowable. The foam synthesis work has shown itself to be quite strong. The testing reactor is excellent and is providing beautiful data.
- The project has good collaboration and a well-thought-out materials development approach.
- The project is looking at a process different from others. The team is utilizing the HydroGEN nodes well. The work is a good combination of theory and experiments.
- The team is strong and experienced. The methods and approaches used are appropriate. Collaborations within the team and with the nodes are extensive.

Project weaknesses:

- By not addressing the hydrogen separation issue, the project loses impact. This is critical, considering there is also no investigation into reducing the higher steam-to-hydrogen ratios needed for isothermal splitting of perovskites found to date. While the DOE target of 10:1 or lower may be unrealistic for all materials but ceria, the efficient utilization of ratios 20–50 times higher seems equally so.
- It is recommended that the researchers consider additional material classes. Whenever foams are used, mechanical strength needs to be considered. A task characterizing the foam mechanical strength would be recommended.
- The reported results are vague, so it is difficult to access the progress and to provide suggestions. The project does not use many advances made from previous STCH efforts funded by HydroGEN, which is essential to accomplishing the ambitious goals of the project and exploring such a large composition space.
- There is a fundamentally flawed basic premise, with performance/outcome metrics constructed in a way that cannot reflect the actual potential (or lack thereof) of the approach.

Recommendations for additions/deletions to project scope:

- As nice as it would be to add an investigation into hydrogen separation and/or decreases in steam-to-hydrogen ratios, the scope increase is too great. Perhaps there are smaller analysis efforts that could be done to identify the impact of the problem and incorporate the results into a modified performance metric. The whole community would probably be overjoyed to hear that it is a non-issue.
- A stronger technoeconomic component would be beneficial, as it would help bring to light omissions in the efficiency metric and the likely real-world productivity.
- A task characterizing the foam mechanical strength would be recommended.

INFRASTRUCTURE

Project #H2-061: Innovating Hydrogen Stations: Heavy-Duty Fueling

Michael Peters, National Renewable Energy Laboratory

DOE Contract #	WBS 8.6.2.1
Start and End Dates	8/1/2019
Partners/Collaborators	Air Liquide, Honda, Shell, Toyota
Barriers Addressed	• Hydrogen safety, codes and standards: insufficient technical data to revise standards • Hydrogen delivery: other fueling site/terminal operations • Targets for Class 8 tractor–trailers: hydrogen fill rate

Project Goal and Brief Summary

This project aims to develop both digital and physical models of hydrogen fast-fill systems that can fill heavy-duty vehicle (HDV) hydrogen tanks at a rate of 10 kg/minute. This work will address a lack of data on fast hydrogen filling into representative medium- and heavy-duty storage systems.

Project Scoring

The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- This is the only project that is directly tackling the question of what kind of equipment is required to achieve the desired performance for HDV fueling. The objectives are timely, and if the project is successful, it will provide valuable insights for several stakeholders. The approach is well-constructed to collect the necessary details, providing significant and reliable demonstrations of equipment performance and capability as applied to HDVs.

- This project is very much in line with what is happening in the industry regarding fueling for HDVs. That, coupled with the publicly available tools and data, makes this an extremely valuable project.
- Having hardware to generate real data and rapidly validate both the one- and three-dimensional models is a great value to the industry.

Question 2: Accomplishments and progress

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

- The work is progressing very well and is exceptional, given the pandemic-related circumstances of the past year. The ability and opportunity to test some new equipment (the micro heat exchanger) are also advantageous. The project can work with and add to some good historical data (the 116 L and 36 L tank data). The team seems very tuned into the needs of those who are developing the protocols and who will be implementing this technology (HDV fueling) in the near future.
- Given COVID-19, the fact that the project has progressed to commissioning equipment installation—in less than two years—is remarkable.
- Several significant milestones have been crossed in the project, especially with respect to station equipment installation and testing equipment fabrication. Some of the final steps remain outstanding for the station equipment commissioning, though it does not appear that there should be any remaining roadblocks. The amount of time left in the project may be a little bit of a concern, especially given the desired scope of the outcomes. It appears that only six months are left in the project and that may be a strain on full-station testing and the ability to collect enough data for full characterization of station performance, while also collecting enough data to inform and validate computational fluid dynamics (CFD) and the one-dimensional model.

Question 3: Collaboration and coordination

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

- The project includes a wide range of collaborators, spanning industry members, academic institutions, regional and state government, and even an international partnership with a similar technical focus. In addition, this project leverages accomplishments from another DOE-funded project, the HDV simulator. The industry project partners are also appropriate, given the interests and expertise of those organizations.
- The project has a good deal of input from the industry and collaboration with other entities and projects, which enables receipt of comprehensive input. The monthly update meetings and the review of key details allow for a consistent exchange of information.
- The project has a good network of industry, academia, and standards organizations in the United States and internationally.

Question 4: Relevance/potential impact

This project was rated **4.0** for supporting and advancing progress toward the Hydrogen and Fuel Cell Technologies Office goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- This project's outcomes are absolutely necessary to ensure the viability of applying hydrogen fueling technology to HDVs. The work completed through this project appears to be a unique effort, or at least one without very many existing parallels. DOE has recently launched the Million Mile Fuel Cell Truck (M2FCT) initiative. As has been learned from the example of light-duty vehicles, success in vehicle design and development will not mean much without preceding success in fueling infrastructure. This project directly addresses some of the most pressing information needs in the critical, prerequisite scope of fueling infrastructure.

- The hydrogen industry has pivoted strongly toward HDV transportation, with large complex compound hydrogen storage systems (CCHSSs) as a second wave—after forklifts—of commercial fuel cell applications. Fueling those applications safely and quickly will be critical to enabling commercial success.
- This project is extremely relevant to the emerging, yet very fast-paced, HDV hydrogen fueling market.

Question 5: Proposed future work

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

- Future work is happening currently and is all excellent and timely.
- As presented, the project looks to be on track for completing and commissioning the test facility and for updating and releasing the revised Hydrogen Filling Simulation (H2FillS). It would be helpful to clearly explain how the industry and companies that are working on HDV fueling can access the tools to test and validate their protocols and systems, clarifying both the access to and support for H2FillS and the use of the National Renewable Energy Laboratory (NREL) site for validation.
- The proposed future work is well-aligned with the project objectives and follows logically from the accomplishments to date. One suggestion is to develop a more complete solution for how the CFD model could be shared with stakeholders outside of the national laboratories. At the least, a proposal could be developed for how organizations could collaborate with NREL to have the model available for further investigations in the future. Based on the latest state-of-the-art tools, it does appear that there are still several important design considerations for HDV tanks that need to be explored. The CFD model developed through this work appears to be quite powerful and accurate, so it should be leveraged significantly by future investigations, even outside of the national laboratories.

Project strengths:

- A major strength of this project is the combination of practical demonstrations—including some advancements being made in novel test equipment design—with detailed, multiple-scale modeling. This methodology and the project's wide scope of research seem effective at helping the project self-direct the necessary investigations to respond completely to the questions being asked by the project objectives.
- The upgrade of the testing site at NREL with "real" HDV hardware and testing capabilities is a huge strength of this project. This is a resource that does not really exist anywhere else, at least in a form that can be accessed widely by industry. This installation will provide safety and commercialization benefits and accelerate the development of infrastructure for fueling vehicles with large CCHSSs.
- Overall, this project is one of the most useful in terms of the "real time" aspect, which means working in parallel with industry as the technology comes along. The original H2FillS model was very useful, and expectations are that this version of it will be the same.

Project weaknesses:

- There are no apparent weaknesses with this project.
- The timeline is the only identifiable and significant concern; however, it appears that this project has simply been affected by the COVID-19 pandemic, like many other projects and the global economy in general. If the project does end up time-constrained, DOE should find some way to provide flexibility for the project's completion date.
- It would be good to see more information on how a developer of HDV fueling protocols and equipment can access the resources that the project is putting in place, especially with regard to support for modeling and validation.

Recommendations for additions/deletions to project scope:

- As mentioned in the presentation, HDV nozzle, hose, breakaway, and receptacle standards and hardware are under development and should be considered for integration into the project as soon as possible, as those elements will play a significant part in the safety and reliability aspects of HDV fueling infrastructure and there are many lessons to be learned by including them in both the hardware and modeling elements of the project.

- If any progress is made on protocol development within the timeframe of this project, the team should run some tests and incorporate them into the model. That being said, it might be a given with the level of coordination within the industry.

Project #IN-001a: Hydrogen Materials Compatibility Consortium (H-Mat) Overview: Metals

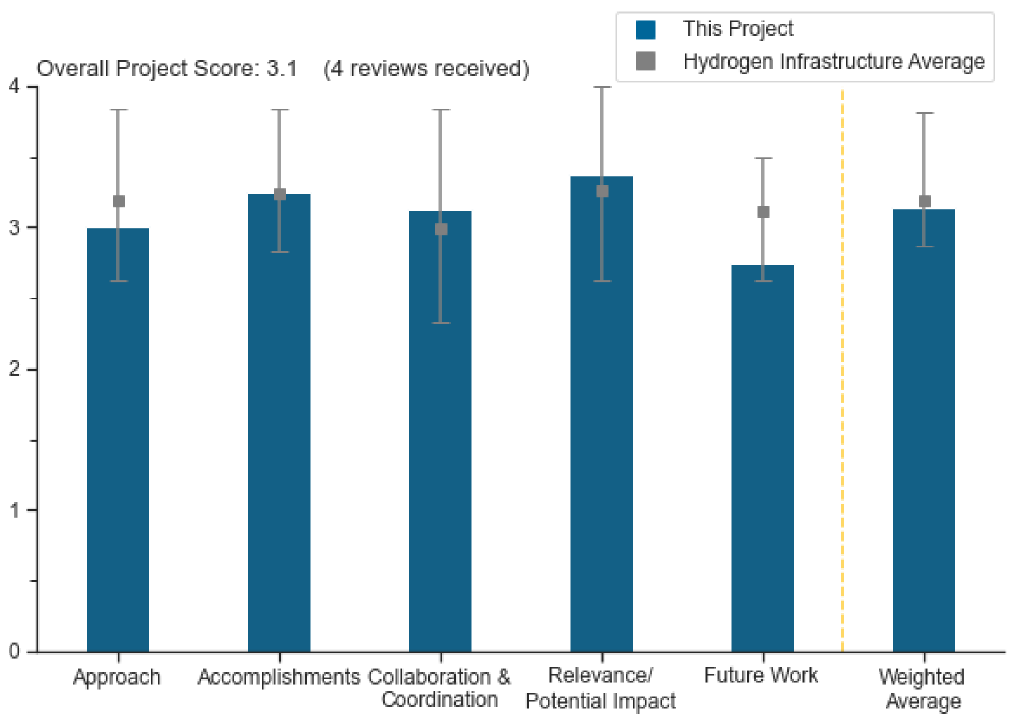
Chris San Marchi, Sandia National Laboratories

DOE Contract #	8.7.0.1
Start and End Dates	10/1/2018
Partners/Collaborators	Colorado School of Mines, University of California, Swagelok, HyPerformance Materials Testing, Massachusetts Institute of Technology, University of Alabama, UIUC
Barriers Addressed	• Reliability and costs of gaseous hydrogen compression • Gaseous hydrogen storage and tube trailer delivery costs • Other fueling site/terminal operation

Project Goal and Brief Summary

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.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- The work provides useful information in some areas of the project, and it should be focused on well-determined objectives to improve cost efficiency. The reviewer supports the following:
    - High-strength ferritic steel microstructures
    - High-strength aluminum alloys
    - Transferability of damage and crack nucleation
    - Microstructurally resistant, austenitic stainless steels
    - Materials for cryogenic hydrogen service.
- To approach this question, one must start with what is (somewhat) negative about the project. This is actually five projects being funded under a single contractual vehicle. One can see this on pages 2 and 4 of the presentation (under the Tasks heading), as well as in the size of the budget. It is odd to say that this is a bad thing, except that the presenter was provided the same amount of time allotted to single projects. In this case, each individual task (project) was provided two to three slides and not nearly enough presentation/ discussion time. As such, delivery of the important bits during the presentation and in the slides is sorely lacking. This is an unfortunate outcome, given the apparently excellent work being performed. It is strongly suggested that future U.S. Department of Energy Hydrogen Program Annual Merit Reviews view this grant as five projects and allot presentation time for each. With the above in mind, yes, the barriers were sufficiently defined, but the project design and feasibility were not communicated well. There was simply no time.
- This is a very broad program. It is challenging to have a rigorous critique of the approaches based on the abbreviated descriptions in the slides and the short talk. This is not the fault of the principal investigator but rather just a structural constraint of the review format. Generally, the approach is sound. In some areas, the goal/objective is very broad, and the link between the specific actions described is not self-evident. While in many instances the reported results seem useful, they would fall short of conclusively informing the very broad objective.
- The project would benefit from having more visible input from folks who are experts in specific material classes or test methods.

Question 2: Accomplishments and progress

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

- It appears that, to date, the project has performed a considerable literature review, a decent-to-considerable number of molecular dynamics simulations, a decent-to-considerable amount of high-strength steel fracture testing, in situ hydrogen slip experiments, and some cryo-temperature testing. While it is certain that the project has performed more work, the above is clear from the slides. This is quite a bit of work for a project that started in October 2018 and endured COVID-19. Having said that, more/supporting information is warranted for each task, given the claims that are stated in the slides.
- The testing and analysis could result in a breakthrough for achieving the identification of ferritic steel microstructures with tensile strengths up to 1,100 MPa and a 50% increase of fracture resistance in high-pressure hydrogen.
- Evaluation of the progress is difficult owing to the abbreviated material. That being said, all indications are that significant progress has been made; this is an achievement, given the COVID-19 environment of the past year and a half.
- It seems like much effort is being put toward modeling, and more should be placed in developing well-planned experiments.

Question 3: Collaboration and coordination

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

- It appears the project is collaborating relatively well within the hydrogen community. The project is strongly advised to look toward collaboration in the damage, damage parameter, and crack initiation community.
- There are extensive collaborations that are not just "on paper" but are actually being realized.
- The projects are all focused within DOE or at select universities. There could be much more progress in this area with more diverse participation.
- The project's collaborations are somewhat limited. It is likely that this limitation is a function of the project's being new, and collaboration is anticipated to grow. The team needs to establish a higher degree of collaboration for achieving goals common to the delivery and storage of high-pressure hydrogen.

Question 4: Relevance/potential impact

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

- One could not overstate the relevance or potential impact of any one of the five projects being studied in this grant.
- The posed questions/goals/objectives are highly relevant, and the work will make advances toward these goals. While the progress toward these goals will be tangible, it seems that there will be incremental progress rather than the more comprehensive solutions that would be inferred by the objective statements.
- The research and development relating to high-strength ferritic steel microstructures and high-strength aluminum may be a breakthrough for efficient pressure vessels that are used in delivery and storage.

Question 5: Proposed future work

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

- This project was marked fair because the detailed future work was not elucidated in the presentation or slides sufficiently such that someone could comment on the project's worthiness of future funding.
- The commentary on remaining challenges and the associated barriers is well-stated but a bit vague. However, some of the statements may be neglecting the significant work aimed at understanding and integrating nucleation behavior into linear elastic fracture mechanics predictions and environmentally assisted cracking in high-strength Al alloys.
- The project would benefit from having more visible input from experts in specific material classes or test methods, and more effort should be placed in developing well-planned experiments.
- The result of this ongoing project will determine the need for expanding it.

Project strengths:

- The current project and testing protocols should provide guidance with regard to applicability of high-strength ferritic steel microstructures or high-strength aluminum for suitable pressure vessels or pipelines that provide adequate strength and ductility for resisting hydrogen embrittlement.
- Project strengths include the collaborations, breadth of effort, the multiscale nature of the efforts, integration of testing and modeling, and identification of important knowledge gaps.
- The projects have good participation of DOE laboratories and some academic institutions. The folks working on the projects are experienced in effects of hydrogen on ferrous materials.
- This is a well-rounded approach that includes experimentation and modeling for all the projects in the grant.

Project weaknesses:

- There was not enough time for the presenter to provide enough information to elucidate any real strengths or weaknesses in the current and proposed work. This is unfortunate, as, in all review processes (e.g., peer-reviewed journal articles), reviewer feedback can help to open a line of thinking that may have been missed. While project teams may seem broad and diverse in their expertise and thorough processes, no team is sufficiently equipped with both depth and breadth. Once subsequent year funding is decided upon, this project would benefit from a more thorough peer review to support its path forward. The review should include people outside of those commonly involved with hydrogen.
- There are very broad objectives (that are likely not to be fully realized), intermittent disconnects between the broad objective and targeted modeling, and, in some instances, a lack of integration of work from other fields (could simply be due to the abbreviated format of the review).
- There is a lack of collaboration, mainly involving experts in specific materials classes who can propose new ideas with respect to the materials, microstructures, and mechanical behaviors of those specific materials. The project uses ferritic steels, austenitic steels, aluminum alloys, fatigue crack nucleation, and alloy development. It is unclear who the experts who are helping to guide the work are.
- There should be an extensive literature search to avoid repeating expensive testing. There were several tests of low and medium ultimate tensile strengths of ≤950 MPa for steel, so the results did not provide any new information.

Recommendations for additions/deletions to project scope:

- There was no chance to ask questions after the presentation, so the reviewer emailed the presenter afterwards. The recommendations here are somewhat based upon the email communication between the presenter and the reviewer. The images chosen for the "project goal" slide would lead one to believe that the project has interest in elucidating the effects of notches and/or cracks. The images also lead one to believe that the project is interested in the micromechanisms leading to the particular morphology of crack paths. The images are not congruent with an understanding of the project focus, as elucidated via email. The project would benefit from an explicit definition of what is sought from the "transferability of damage crack nucleation" project. Email communication made this clear, but the presentation did not. Along those lines, the term "critical damage accumulation leading to a crack of interest to fracture mechanics" (or something similar yet less convoluted) would be more apt, rather than the terms "nucleation" or "initiation." Those two terms have specific meaning to a sect of mechanicians. The project team is cautioned against the sole or primary use of monotonic deformation accumulation when going for a critical damage accumulation formulation. The components of interest for hydrogen use experience repeated loading, even if only hundreds of cycles. Materials of interest experience considerable kinematic and isotropic evolution, cyclic stress and strain redistribution, and cyclic load magnitude-to-cyclic damage accumulation rate dependence, all of which may occur in the first hundred to several hundred cycles of loading (even under R=0). To this end, load-controlled tests are unable to capture the material's history-dependent deformation response.
- It is recommended that this project continue with a new objective and goal to use the resource and strength toward achieving a new material that is better suited for hydrogen embrittlement.

Project #IN-001b: Hydrogen Materials Compatibility Consortium (H-Mat) Overview: Polymers

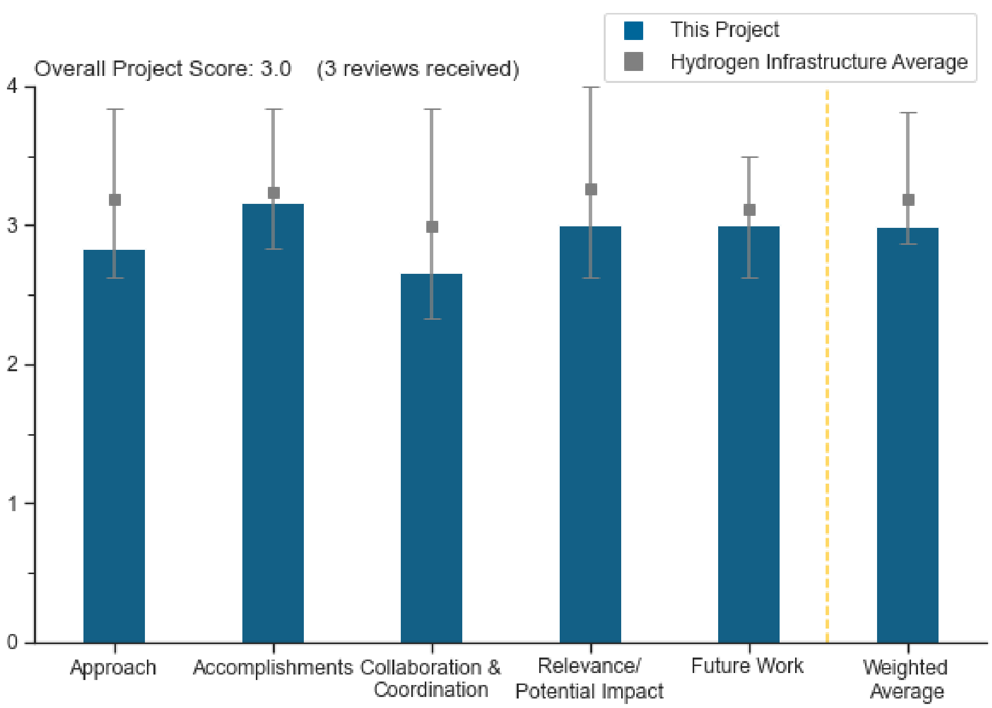
Kevin Simmons, Pacific Northwest National Laboratory

DOE Contract #	L062-1502
Start and End Dates	9/1/2018
Partners/Collaborators	Argonne National Laboratory, Sandia National Laboratories, Oak Ridge National Laboratory, Savannah River National Laboratory, Pacific Northwest National Laboratory, Swagelok, Takaishi Industries, Arlene, Zeon, Top Sector Energy, Chemours, Kyushu University
Barriers Addressed	• Limited access and availability of safety data and information • Insufficient technical data to revise standards • Limited participation of business in the code development process • No consistent codification plan and process for synchronization of code research and development • Reliability and costs of gaseous hydrogen compression • Gaseous hydrogen storage and tube trailer delivery costs • Other fueling site/terminal operations

Project Goal and Brief Summary

The project objective is to fill a critical knowledge gap in polymer performance in hydrogen environments. Investigators are gathering and assessing stakeholder input about the challenges, materials, and conditions of interest for hydrogen compatibility. Findings inform the project's development of standard test protocols for evaluating polymer compatibility with high-pressure hydrogen, characterizing polymers, and developing and implementing an approach for disseminating the information.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- The approach addresses key knowledge gaps with respect to failure mechanisms in polymer systems commonly used in hydrogen service and should provide useful feedback to make improvements. That said, the specific questions that are being investigated in the U.S. Department of Energy Hydrogen Program (the Program) are a small subset of the large range of materials and approaches that could have been assessed. It was not clear how these specific materials and improvement approaches were selected for evaluation, whether by the investigators themselves or with input from the broader research and commercial community.
- The project objective is clearly identified as demonstrating, by September 2022, an elastomer formulation with 50% less swelling compared to similar off-the-shelf materials. Regarding critical barriers, since this project purports to develop science-based strategies to design material (micro)structures and morphology with improved resistance to hydrogen degradation, it is expected that the project is identifying and addressing specific science-related barriers. However, such science-related barriers are not clearly established and are not linked to the technical accomplishments. Rather, there is more emphasis on particular modeling and experimental tools than on resolving specific knowledge gaps by applying the tools in a targeted fashion. To illustrate this impression, the titles of slides 7–11 all emphasize the tool applied, as opposed to the science question or knowledge gap that is motivating the application of the tool.
- The project objectives and critical barriers have been clearly identified. It is difficult to understand how all of the different pieces of work are tied together and pointing toward addressing the barriers and achieving the objectives. The relationship between the Hydrogen Materials Compatibility Consortium (H-Mat) website, Data Hub, and the technical work is not clear; they seem like many individual efforts.

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 results generated by the project team during the last year have provided useful insights into the behavior of compounded polymeric elastomers when exposed to high-pressure hydrogen. The accuracy of the molecular model as a predictive tool for a hydrogen pressure failure point is especially impressive, and its value in directing research toward new, more degradation-resistant formulations has already been demonstrated. Further progress could be made in having tools and conclusions more broadly available, especially with respect to the H-Mat and Data Hub.
- The project objective of demonstrating an elastomer formulation with 50% less swelling compared to similar off-the-shelf materials is clearly identified; however, it is not clear how the technical accomplishments represent progress toward that objective. Since the project does not identify specific science-related barriers and knowledge gaps that are linked to the goal, it is difficult to judge how the accomplishments represent progress toward achieving the goal. For example, in reference to slide 7, it is unclear whether it was postulated that swelling could be related to hydrogen accumulation at the silica–polymer interface, and thus these results would confirm the posited relationship and inform a pathway to mitigating swelling.
- The project has made significant progress, but it is difficult to see how the progress will lead to reaching project objectives.

Question 3: Collaboration and coordination

This project was rated **2.7** for its engagement with and coordination of project partners and interaction with other entities.

- The partners include DOE laboratories and a few companies, and among those partners, there appears to be sufficient collaboration. Participation by a broader group of organizations, including standards development organizations, pre-normative groups focused on materials, and the National Institute of Standards and Technology might help accelerate this work. This reviewer recalls the Hydrogen and Fuel Cell

Technologies Office director making a statement about all canoes rowing in the same direction; however, that approach is not seen here. It is not clear how the team knows that it has the right partners for success.

- The strong collaboration between national laboratories is a key component of this project and has been readily demonstrated through the technical results obtained by geographically and organizationally diverse teams. That said, while multiple commercial organizations are listed as team partners, their degree and form of involvement are unclear. Achieving DOE goals for the project will hinge on the near-term commercial impact of findings, so this linkage should be emphasized going forward.
- The project clearly identifies industry partners and research collaborators; however, it is not apparent how these relationships are enabling the accomplishment of the project goal (for example, how the collaboration with Kyushu University's contributes to accomplishing the project's goal).

Question 4: Relevance/potential impact

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

- The project's goal, "By September 2022, demonstrate an elastomer formulation with 50% less swelling compared to similar off-the-shelf materials," seems pretty conservative and achievable. The project objectives are aligned with DOE's goals and will help realize the goals if the project achieves those objectives.
- The project is intended to enable a more robust and reliable infrastructure, which is certainly in alignment with the Program goals.
- The broad goals of this project will have a high impact on the overall Program goals and objectives through the reduction of unanticipated maintenance events and hydrogen losses. However, it is unclear what percentage of the overall problem is addressed by the specific technical projects the investigators have selected to pursue. A broad survey of the materials in use and the impact of solving the specific issues as a percentage of the commercially deployed hydrogen seal market would assist an assessment of how impactful the project will be, assuming its technical success.

Question 5: Proposed future work

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

- The focus of investigators on broadening the applicability of technical results generated so far, and making them accessible to the community, is exactly the right direction for the project to go. Close collaboration with industrial partners will be necessary to ensure near-term impact of the work. Further thought may be necessary to determine the best approach to making H-Mat and Data Hub useful repositories for Program results, though this, too, is planned through beta testing by the team.
- There does not seem to be a plan to generalize the technical approach more broadly. It looks like the team is addressing a solitary problem. There does not appear to be a plan to engage stakeholders with the H-Mat site and Data Hub widely. A plan to ensure that the resources are what stakeholders need and to evaluate their use could help target the resources to address these needs.
- It is not clear how the proposed future work is informed by the reported accomplishments. Furthermore, it is not apparent how the proposed future work represents a pathway toward satisfying the project goal (by September 2022, demonstrate an elastomer formulation with 50% less swelling compared to similar off-the-shelf materials).

Project strengths:

- This project shows a high level of collaboration between multiple national laboratories, which has led to the generation of impressive technical results and predictive modeling tools. Providing these results to hydrogen seal manufacturers should yield near-term benefits in the design of more hydrogen-resistant materials and a reduction in seal-related unplanned maintenance events. By building in communication and dissemination tasks in the later portion of the project, sharing the findings from this work, as well as the subsequent positive impacts, is much more likely.

- The project's science-based approach can be fruitful toward the goal of improving the performance of materials when they interact with hydrogen gas.
- Establishing mechanisms, such as consortia, to enhance collaboration could be valuable in accelerating progress toward reaching DOE goals. The technical collaborators on the project are making good progress.

Project weaknesses:

- The breadth of the problem that this project hopes to address makes any reasonable workplan fall short with respect to addressing all the potential technical questions related to elastomeric materials for hydrogen service. The team has done excellent work on the technical problems selected for evaluation, but it is unclear how large a percentage of the underlying technical issues are being addressed. As the project moves into its final years, it will be challenging to broaden the findings to enough different materials to truly serve as a complete database.
- It is not clear how all of the pieces of the project are related or how they are driving toward the same goals. The engagement with key stakeholders for codes and standards development could be improved. A plan for how the team will address the stated barrier of "limited participation of business in the code development process" could be helpful.
- The team needs to explicitly identify science-based issues and knowledge gaps that must be resolved to meet the project goal (demonstrate an elastomer formulation with 50% less swelling compared to similar off-the-shelf materials). The suite of theoretical and experimental tools seems intended to give the impression that the project is science-based, but the purpose of each tool for answering a targeted science-based question is not clear.

Recommendations for additions/deletions to project scope:

- The original scope of this project is exceptionally broad and has not necessarily been narrowed down by the investigators. To maximize the relevance of the project results, it would be useful at this point in the project to resurvey commercial partners to ensure that their most pressing needs are being addressed by the selected technical activities. Further industry input on the specific list of elastomeric and thermoplastic materials that are investigated will maximize the project's near-term impact on DOE performance goals.
- It is recommended that the project consider whether all of the theoretical and experimental tools are necessary. The priority must be to pose the critical science-based questions that represent barriers toward the project goal, and then identify and implement the right tools to address these questions.
- No additions or deletions are recommended at this time.

Project #IN-004: Magnetocaloric Hydrogen Liquefaction

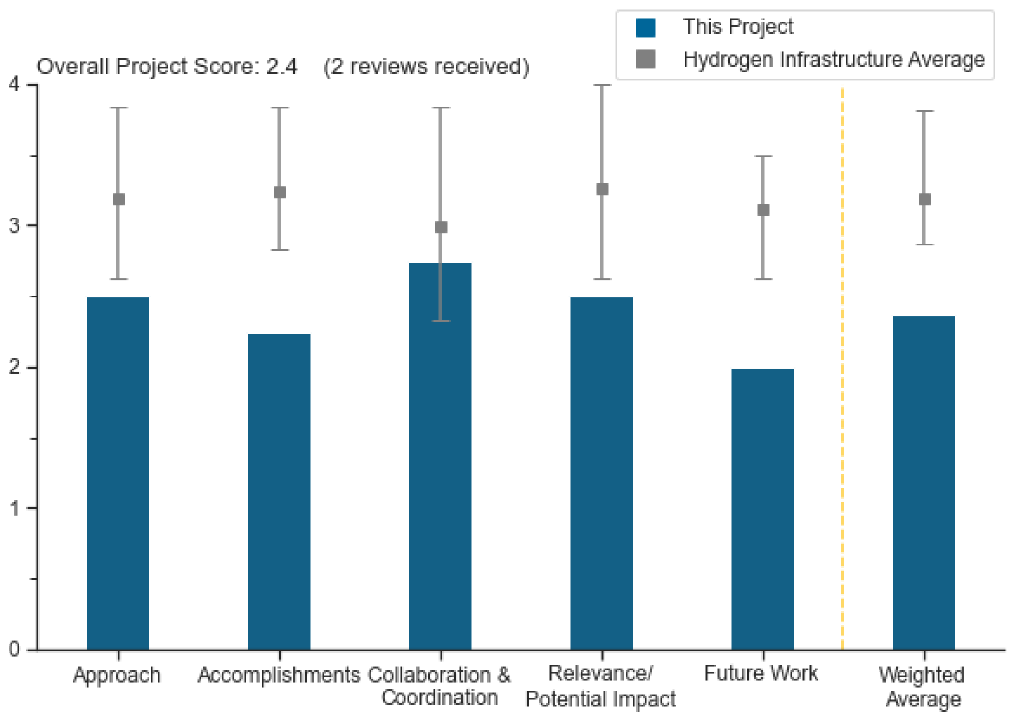
John Barclay, Pacific Northwest National Laboratory

DOE Contract #	3.1.0.2
Start and End Dates	10/1/2015
Partners/Collaborators	AMES Lab, Iowa State University
Barriers Addressed	• Low hydrogen liquefier efficiency • High liquefier capital costs

Project Goal and Brief Summary

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 of hydrogen. 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.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Because of late reviewer withdrawals and conflict of interest notifications, the minimum number of reviewers for a complete review panel (three reviewers) was not achieved for this project. The results are included here to inform future work and reviews, but the scores for this project are not included in the subprogram average.

Question 1: Approach to performing the work

This project was rated **2.5** for identifying and addressing objectives and barriers and for project design, feasibility, and integration with other relevant efforts.

- Critical barriers (FOM, capital expenditures, operations and maintenance, energy input) are clearly identified and addressed. They are also consistent throughout the years, which is good to see. The way the project is self-designed and whether it is commercially feasible are more difficult to quantify or qualify. There is obviously a good mix of modeling and experimental investigation, but it looks like a large amount of work has been devoted to characterizing and manufacturing the materials rather than looking at the system itself.
- This is the fourth year of reviewing this project. The fundamental premise is interesting; however, the lack of progress over the past two years suggests that it is time for the U.S. Department of Energy to provide other potential technologies the chance to prove their value. Much of the material in the presentation appeared to be very similar to what was presented in 2019. Furthermore, current commercial technologies in hydrogen liquefaction have reduced their energy input to levels approaching the project's goal of 6–7 kW/kg.

Question 2: Accomplishments and progress

This project was rated **2.3** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Since the last presentation in 2019, this project appears to have made very little progress in the area of materials development and testing, although the impact of the pandemic over the past year must be acknowledged.
- There were only three slides on accomplishments and progress (the fourth one is actually a response to reviewers). Granted, the third slide has many bullet points, but it is difficult to judge the level of effort (such as whether accomplishments listed in slide 8 have required as much effort as those listed in slide 10).

Question 3: Collaboration and coordination

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

- Many industries are listed. However, they are merely possible prospects, not active collaborators. For instance, it may be a long time before the technology readiness levels (TRLs) presented at the DOE Hydrogen Program Annual Merit Review (AMR) are acceptable enough to be of benefit to Raytheon Technologies and Nikola Corporation. There are no onboard liquid hydrogen (LH2) storage technologies available yet, and the hydrogen business model that Nikola Corporation must demonstrate is such that it is unlikely that the AMR is on the top of their list for another five years or so.
- It was good to see collaboration with Nikola Corporation, Woodside Energy Ltd., and Raytheon Technologies, but at the end of the day, none of these companies will develop liquefiers on their own.

Question 4: Relevance/potential impact

This project was rated **2.5** for supporting and advancing progress toward the Hydrogen and Fuel Cell Technologies Office goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- Relevance and impact are excellent. This is typically the type of work DOE should support; it is innovative, high-risk, high-reward, and low-TRL. There is one note, though: the claim that this project is a game-changer is a little far-fetched, as it does not address the cost of hydrogen production, which is central to the consideration of hydrogen production pathways and zero-emission transportation.
- Progress is slow, and convincing results are not available. Now that hydrogen consumption for mobility is ramping up, organizations that build hydrogen liquefaction plants are ramping up their investments to achieve energy-efficient liquefaction processes to compete in the market. Today, the three industrial gas organizations have commissioned, or are building, hydrogen liquefaction plants with capacities of over 30 tons per day. Chart Industries has invested in liquefaction technology and is likely working on

improvements. As demand grows, so will proven and reliable designs that adopt efficient technologies for a competitive edge.

Question 5: Proposed future work

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

- It is uncertain that the concept of proposed future work is relevant since the project is on a no-cost term extension, ending in September 2021. Work left to be performed until then seems challenging. The related slide shows very interesting mapping of technology options.
- It is suggested that DOE allow the project to end and look for other concepts to fund in the future.

Project strengths:

- This is a very innovative project, with a strong science basis and high potential of generating LH2.

Project weaknesses:

- There is a lack of clarity on cost, material selection, fabrication, and overall plant design. Producing well-shaped spheres seems to have been a challenge throughout the six years.
- Any innovative technology must face the reality of commercialization, which means productivity and reliability. This technology is interesting but has not demonstrated the potential to be commercialized. For that reason, when the funding is finished, there should not be a renewal.

Recommendations for additions/deletions to project scope:

- The project should be clearer on challenges and how difficult and unforeseeable they have been.
- No changes to the current project are recommended.

Project #IN-015: Optimizing the Heisenberg Vortex Tube for Hydrogen Cooling

Jacob Leachman, Washington State University

DOE Contract #	DE-EE0008429
Start and End Dates	1/23/2019 to 9/30/2021
Partners/Collaborators	Washington State University, Plug Power Inc.
Barriers Addressed	• Reliability and cost of liquid hydrogen pumping • High cost and low efficiency of liquefaction • Other fueling site/terminal operations

Project Goal and Brief Summary

This project aims to establish that Plug Power Inc.'s (Plug Power's) Heisenberg Vortex Tube (HVT) cooling system can effect the following improvements to cryogenic hydrogen systems: (1) a 20% increase in liquid hydrogen (LH2) pump volumetric efficiency through vapor separation and subcooling, (2) a 20% decrease in LH2 storage tank boil-off losses through thermal vapor shielding, and (3) an increase of supercritical hydrogen expansion from 31% to more than 40% through greater isentropic efficiency.

Project Scoring

The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- The reviewer has tracked this project for the past three years and is happy to see that the concept has evolved in a productive manner and found an application that may result in its productive use.

- Due diligence has been performed to identify the best application case. This looks like a great utilization for the HVT. Also, stratification is a key issue for stationary LH2 storage systems.
- The work to validate the science behind the concept of refrigeration provided by para-orthohydrogen conversion is sound. The project is about to enter the testing phase. This will test the efficacy of the project plan to meet its goals. There will be inherent challenges with this testing because of the number of variables involved. The three barriers listed on slide 3 do not seem directly applicable to the project for the following reasons: (1) the reliability and cost of LH2 pumps is not directly affected by the thermodynamic performance of the LH2 system (e.g., tank vent losses), (2) the high cost and low efficiency of LH2 liquefaction is not addressed, and (3) it is unknown what is meant by "other fueling/terminal operations."

Question 2: Accomplishments and progress

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

- The work on material selection and material shape is admirable. It was good to see the adoption of the catalyst in the HVT.
- The project has made progress in validating the technical concept of para-orthohydrogen conversion. There is no progress yet in terms of calculating or comparing the refrigeration effect to achieving the stated goals on slide 2. For example, it is not clear how the pump volumetric efficiency improvement of 20% and the reduction of vent losses will be achieved and measured reliably. It would be helpful if the project provided the baseline for both of these goals, to which the results can be compared later. It is also not clear why the same loss-reduction benefit could not be achieved by simply improving the operation of the LH2 pump and/or increasing the size of the boil-off compressor. The goals of this project are relatively narrow and apply only to the specific Plug Power systems. As such, it is not clear that the HVT will provide significant benefits to other systems or markets. For example, there are other cryogenic pumps on the market today that demonstrate performance that already exceeds the stated goals, even at the relatively small system size. As systems increase in size for the heavy-duty market, the benefit of a modest amount of in-tank cooling will diminish rapidly.
- There were few details on Task 2.1.2, which lasted 12 months. If this is proprietary to Plug Power, then public money should probably not pay for this, nor should there be any evaluation. Although Task 2.2.3 is interesting in principle (nice block diagram), it is difficult to estimate the progress there. It is unclear how many runs were completed or what the error bars on each result were. The team can perform uncertainties well. It is not clear why it was not done to compare experiments versus computational fluid dynamics. Also, it is not clear whether 1 K of cooling (53.7 K to 52.7 K) is enough for the process. Finally, it would be good to know what the accuracy of the sensor is. Objective 3 seems to have been well met.

Question 3: Collaboration and coordination

This project was rated **2.7** for its engagement with and coordination of project partners and interaction with other entities.

- It is a good choice to work with Plug Power, which utilizes installations that are ideally suited to adopting the HVT and utilizes the molecules vented from the process. Upon completion of the late 2021 testing, the results should be shared with the broader industry to gain greater visibility of the technology.
- Coordination between the two project members, Washington State University and Plug Power, seems to go well. However, there are no collaborations with outside members.
- Working with Plug Power and a tank vendor offers the ability to deploy and test the technology. The collaboration with Plug Power limits the ability for broader use, testing, and validation, though. As a result, the applicability might be relatively narrow and will help with only a subset of applications using specific equipment. The equipment of the entire system has not been described to help with understanding the HVT's effect on other systems and when used by other vendors.

Question 4: Relevance/potential impact

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

- The work is well-aligned with DOE objectives of reducing costs for LH2. Boil-off is a challenge for many applications. The implementation is realistic and backed up well by analysis.
- As the demand for LH2 for mobility and stationary applications grows, the potential value of the HVT will expand significantly.
- The underlying technology is sound, but the question is whether it will have enough of an impact to be commercially feasible for this market or for other markets. The nature of the work lends itself to a relatively narrow subset of applications where it would be helpful. For example, the project states that this technology is projected to be feasible for only 50% of Plug Power material handling sites, which is already a fairly narrow portion of the DOE Hydrogen Program. There are also other technologies and equipment that already meet the goals of improving pump efficiency and reducing vent loss on LH2 systems and that serve the same and comparable markets.

Question 5: Proposed future work

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

- The plan to proceed with a tank vendor to deploy this technology for further testing is good. Given the number of variables inherent in the design, use, and manufacturing of the equipment, it might be challenging to get useful results. The catalyst's degradation is critical to the HVT's overall long-term success since the catalyst is inaccessible after the manufacturing of the tank. It is good to see that this is a priority.
- The project team has developed a good plan for future work. With that in mind, the researchers will need to begin thinking about how to commercialize the technology for wide use.
- It is great to see a proof of concept in a real full-scale system.

Project strengths:

- The concept of using the para-orthohydrogen conversion process to recover refrigeration is one that has potential. Progress in this area is useful for future LH2 system optimization. There is a lack of data concerning the actual temperatures within an operating LH2 tank, particularly with regard to stratification of its contents. This project and its testing will provide additional insight in this area. While it will help the existing project, it also will benefit future research. The project has a good plan, and the plan is being executed.
- The project has adequately performed both the theoretical and practical testing to position the technology for a real-life demonstration.
- This is a down-to-earth application that has a great academic background and is partnering with a leading LH2 company.

Project weaknesses:

- The validation of the concept will be challenging because of the relatively small benefit when compared to external factors that may affect the overall heat leak and operation of the system. The modeling shown for the internal temperatures of an LH2 tank appears to be for relatively static conditions and does not reflect the dynamics inherent with the operation of the cryogenic pump (and its condition), the boil-off compressor, the tank autovent, and the pressure-build system. The relatively modest refrigeration, provided by the HVT, may get lost within these conditions. For example, slide 15 shows the relatively small temperature impact. The lack of ability to share key technical and economic information (because this information is proprietary) makes it difficult to truly validate the technology's performance and future viability. It is not clear how the goal of a 35% increase in boil-off compressor flow will be obtained from the HTV's use. The efficiency and output of the boil-off compressor are unlikely to be unaffected by modest refrigeration inside the tank. Additional detail should be provided on this point.

- Clearer communication is needed to explain what delta T (ΔT) is needed, what the rough energy balance is, and how the HVT improves it.
- There are no serious weaknesses.

Recommendations for additions/deletions to project scope:

- The team should provide a rough order of magnitude on what makes the technology relevant (e.g., ΔT, balance of energy).
- The project could add commercialization planning.
- There are no recommendations for additions or deletions to project scope.

Project #IN-016: Free-Piston Expander for Hydrogen Cooling

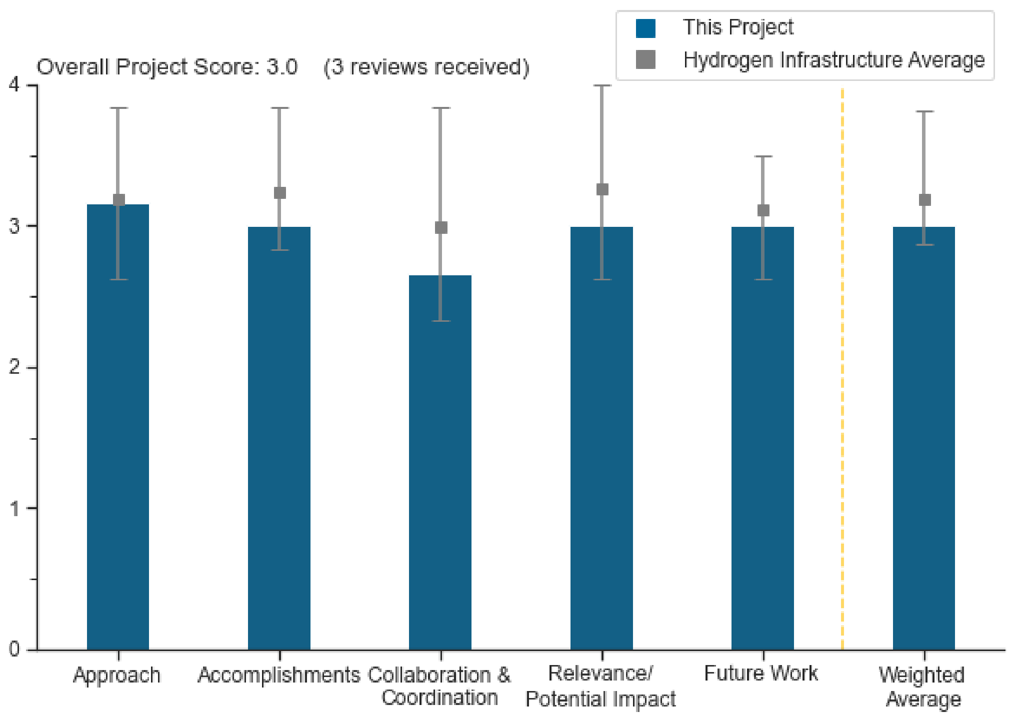
Devin Halliday, Gas Technology Institute

DOE Contract #	DE-EE0008431
Start and End Dates	1/2019 to 6/2022
Partners/Collaborators	Center for Electromechanics (University of Texas at Austin), Argonne National Laboratory, Quantum Fuel Solutions
Barriers Addressed	• Other fueling site/terminal operations

Project Goal and Brief Summary

The project team is developing a free-piston linear motor expander that can conduct hydrogen pre-cooling for light-duty hydrogen fueling, while producing energy that can be used to offset compression energy consumption. Pre-cooling units represent 10% of the capital cost of hydrogen fueling stations and impose significant operating costs as well. Replacing conventional pre-cooling units with expanders could reduce these costs, removing a major barrier to hydrogen fuel adoption.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- The concept is very interesting and, if successful, will be a major contributor to diminishing the cost of hydrogen fueling. The team appears to be analyzing both the theoretical and physical constraints of the concept.
- There is a linear approach from modeling to design to proof of concept.

- The project is attempting a new method of providing cooling for dispensing applications. No meaningful barriers are listed on slide 3, although several were apparent in the presentation and from observation of the project review. In particular, some significant barriers might exist regarding long-term operation and longevity (e.g., maintenance costs):
    - Long-term longevity of the expander seals. No information was provided regarding the expected life of these seals, despite their being a limiting aspect of many reciprocating designs. This applies both to internal seals (affecting efficiency and performance) and to external seals (affecting safety).
    - Valve life. Some information was provided, but there were no details to back up assertions as to cycle life of the valves, especially when modifications were made and they are in service that exceeds the manufacturer specifications.
    - Maintainability of the unit, especially a unit of this size. This will be critical in terms of the technology's ability to be used commercially.
    - Ability to stay within the closely defined SAE International fueling protocol targets, particularly over a wide range of flows and pressures. For example, it is unclear whether the equipment can meet the cooling requirements for partial fills with low differential pressure. There is no mention of being able to vary flow as needed to meet varying flow conditions.

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.

- There is good work on evaluating valve speed and considering improvements. More modeling of ground storage dynamics is required, and the fact that the researchers acknowledged that they had not considered cascade dispensing is appreciated.
- The proof of concept is under construction.
- Good progress is being made on the basic function of the machine. However, development of similar machinery has typically shown that units such as these will need extensive prototype testing after initial function is proven. The competitive cost targets used are relatively high compared to current designs. The system shown on slide 5 is dated and does not fully reflect the current technology available, and the long-term targets are above competing technologies. For example, the cooling costs are not consistent and are higher than those shown in other DOE Hydrogen Program Annual Merit Review presentations.

Question 3: Collaboration and coordination

This project was rated **2.7** for its engagement with and coordination of project partners and interaction with other entities.

- This is a great mix of public–private partnership.
- The project collaborators have limited station operating experience fueling H70 (70 MPa) vehicles. It would be helpful to get the support of and partnership with a sizable station operator that could provide meaningful feedback on operating and installation issues, in addition to a platform for the long-term testing needed to validate long-term viability.
- The collaboration is focused on academic evaluation. The team needs to expand its reach to organizations offering commercial solutions and maybe even look outside the hydrogen fueling environment to applications such as helium cooling or other products.

Question 4: Relevance/potential impact

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

- Pre-cooling is a key challenge for LH2 refueling, and this looks like a worthwhile approach.
- Developing a lower-cost, lower-power refrigeration option for fueling activities has an impact on DOE goals. The costs shown in the presentation appear dated (2015) and for station sizes that are significantly

smaller than those being deployed and proposed today. Costs and technologies for dispenser cooling have advanced beyond the state of the art in 2015. In particular, this expander is required for each dispensing point, as opposed to systems that might be leveraged across multiple units for cost-effectiveness, so this technology may not scale well to larger systems (capacity and multiple dispensers).

- It is too early to tell, but the concept may have relevance to numerous applications.

Question 5: Proposed future work

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

- There is demonstrated functional hardware in various applications that target real-life applications.
- The next steps are logical.
- For those barriers identified, the future work is appropriate. The challenge is that unidentified barriers run a high risk of interrupting progress. Additional consideration should be given to those barriers, particularly long-term operation and reliability. The future work does not specifically address needing to stay compliant with very prescriptive fueling protocols that are critical to its success. There is no capacitance in the device to level out performance and transients.

Project strengths:

- The project looks to be relevant and has potentially broad applications.
- There is a realistic approach and great application.
- A properly designed expander can operate to provide the cooling required for fueling vehicles. The project team has developed a prototype.

Project weaknesses:

- The concept requires that all gas be compressed to high pressure to reliably provide enough refrigeration energy. This might be problematic for direct filling and for multi-pressure cascade pressure stations, which account for the majority of station designs. Effectively, the system relies on additional compressor energy and equipment to provide the cooling energy to then provide power back to the compression, but there will be losses inherent to the process. The maintenance costs have not been evaluated as part of the economics. The physical size of the machine (12'), its orientation (linear), and its relatively small capacity (light-duty) make it challenging for deployment in current form. Matching power generated with compression power required will be challenging because of different usage profiles and timing.
- The project needs to account for real-life technology scenarios.
- There are not enough details (see next section).

Recommendations for additions/deletions to project scope:

- After the basic function has been demonstrated and once the technology has been assessed for commercial viability, a long-term test program would be needed to assess long-term performance, particularly regarding maintenance intervals and cost. If not already completed, a hazard and operability study should be performed to evaluate potential safety risks of mechanical operation close to a dispensing operation, as well as potential for unanticipated leakage either within or external to the machine.
- The project could use more details: examples of how the work could be really recovered, especially during regime fluctuations of the station; the size of the system; and major technical challenges.
- The team should keep working.

Project #IN-019: Ultra-Cryopump for High-Demand Transportation Fueling

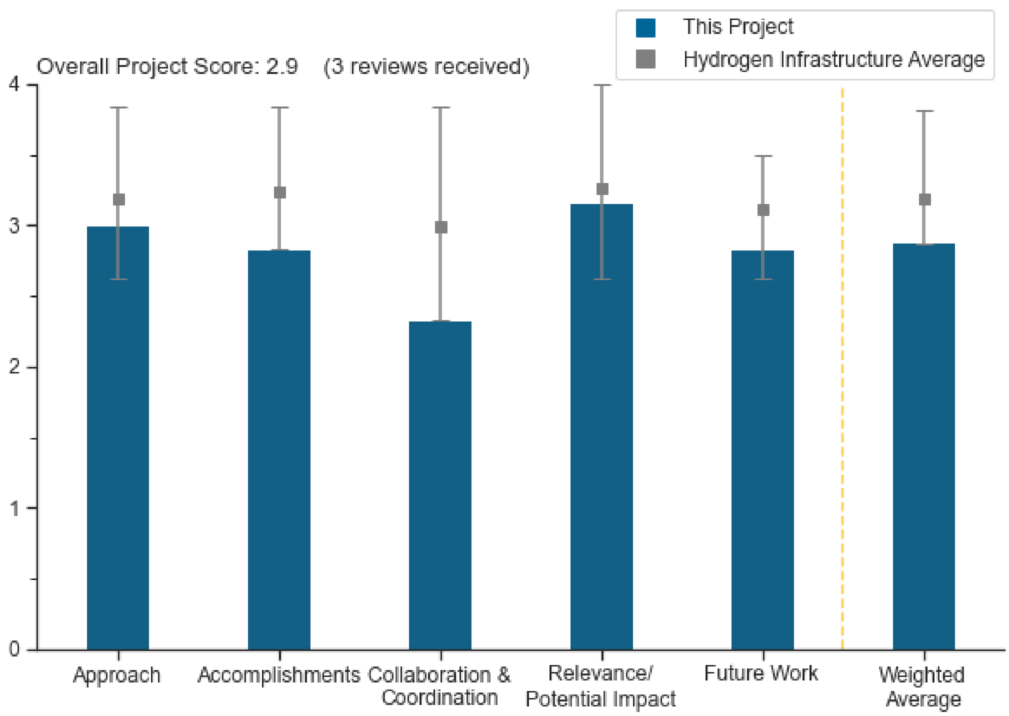
Kyle Gross, RotoFlow/Air Products

DOE Contract #	DE-EE0008819
Start and End Dates	2/1/2020 to 5/1/2023
Partners/Collaborators	N/A
Barriers Addressed	• Reliability and costs of liquid hydrogen pumping

Project Goal and Brief Summary

This project aims to help advance hydrogen refueling infrastructure for heavy-duty transportation by designing, building, and testing a liquid hydrogen pump with the flow and pressure necessary for bus and truck refueling. The work addresses challenges caused by refueling operating conditions (e.g., extreme pressure), in part by upscaling existing technologies by RotoFlow and making improvements to pump design, seal design, and motor–drive configuration. The intended final product is a cost-effective, reliable, high-flow, high-pressure reciprocating liquid hydrogen compressor system.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- The overall approach seems reasonable, and the tasks in the current budget period are showing good progress.
- This project's approach is methodical and measured.

- The project’s challenges to overcome were not really addressed adequately. The elephant in the room is seal life and maintainability. While these topics were mentioned, there was no proof provided that the issue of >350 bar cryogenic pump seal reliability has been addressed, nor was there at least an explanation of what is different about this approach compared to prior designs.

Question 2: Accomplishments and progress

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

- This project has made good progress with developing the necessary material to move toward actual deployment and demonstration that will happen later in the project.
- It is expected that component testing will be conducted to validate the design approach planned, such as accelerated life testing of seals on a bench rig. The mechanical design of a cryopump is not novel; the team should focus on the high-technical-risk elements and address that risk first.
- Direct articulation (a page or two) of the specific technical barrier(s) that must be overcome to meet the DOE technical targets for the cryopump should be provided, as comparable pumps that meet lesser targets already exist. This will help others in perhaps adjacent fields to offer potential solutions that could advance the technology but that would otherwise go undiscussed.

Question 3: Collaboration and coordination

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

- The collaboration within Air Products is well-coordinated and significantly benefits the project, but there are no external collaborations. Perhaps working with vendors is a possibility for the project. Also, it may be useful to engage a group (e.g., the National Renewable Energy Laboratory) that could help model the duty cycle of the pump as a function of station usage, particularly across different usage scenarios. That might help establish the best design.
- There is no collaboration and no voice of the customer.
- No external collaboration or coordination exists for this project.

Question 4: Relevance/potential impact

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

- Heavy-duty (HD) transportation is a clear target for hydrogen. The refueling infrastructure needs to be convenient and reliable. A cryopump that enables HD refueling is a key link in the chain.
- While this project focuses on the modification of existing commercial pump technology, if successful, the technology could have an immediate impact on the HD application market.
- High-pressure cryopumps that are reliable and have low operating expenses are a key limiting factor in high-capacity refueling and cost-effective distribution.

Question 5: Proposed future work

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

- This project’s proposed future work seems to logically follow the next steps for where the project's schedule and budget period currently is.
- The future proposed work seems to be on target.
- There is no component-level derisking or clear communication of the high-risk elements. If the investigators really have no concern, then it is unclear why this project is considered research and development (R&D).

Project strengths:

- Building off of internal company experiences and knowledge—particularly encouraged by engagement with Air Products experts for hydrogen refueling station designs—is a project strength.
- This project fits a gap in the current market.

Project weaknesses:

- The development plan does not credibly identify key risks or form an R&D strategy to focus on those risks before building a unit and testing it in a representative environment.
- Having no external interactions is a weakness.

Recommendations for additions/deletions to project scope:

- The mechanical design capability of the principle investigating entity is without question. It is recommended that the project focus on iterating seal designs quickly at a component level to prove they can last for durations that are commercially relevant, at full speed and full pressure.
- The pump capabilities should be evaluated against performance needs under different HD refueling station scenarios, with the help of a partner.

Project #IN-020: Self-Healable Copolymer Composites for Extended-Service Hydrogen Dispensing Hoses

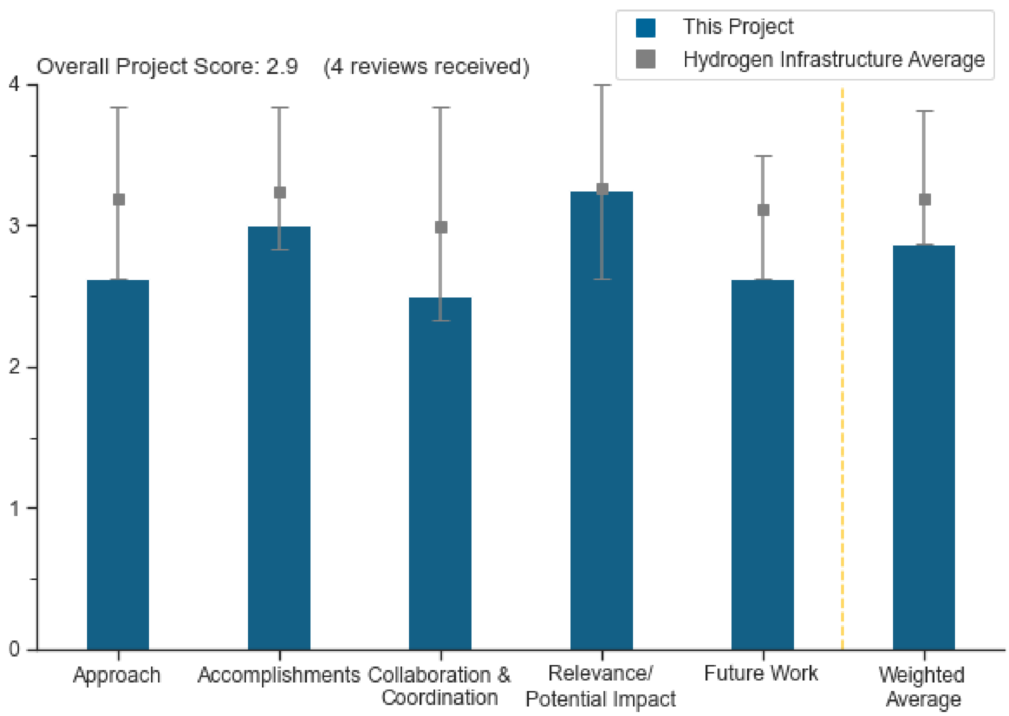
Marek Urban, Clemson University

DOE Contract #	DE-EE0008827
Start and End Dates	1/1/2020 to 2/28/2023
Partners/Collaborators	Savannah River National Laboratory, Sandia National Laboratories, Pacific Northwest National Laboratory
Barriers Addressed	• Fueling site/terminal operations • Reliability and cost of hydrogen fuel pumping

Project Goal and Brief Summary

This project aims to design, develop, and pre-commercialize a low-cost inner layer for hydrogen dispenser hoses that integrates a self-healable copolymer matrix with polypropylene fibers. Currently, hydrogen dispenser hoses develop microcracks after around 1,000 fueling cycles. This project could extend the service life of hydrogen hoses to over 25,000 cycles, making them far more cost-effective.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- The concept is extremely innovative and could be a real game changer for realizing reliable and cost-effective hydrogen fueling. It seems like some potentially useful blends might be screened out prior to environmental testing. Fundamentally understanding how the different environments affect self-healing could be an important aspect of the work. It is not clear whether the measurement of self-healing is reproducible or whether it is measuring the key properties. It is not clear whether scratches are the same as cracks. It would also have been helpful to understand how a damaged material would behave during a tensile test. Slide 14 shows undamaged vs. self-healed specimens, prompting questions of how a damaged specimen would behave.

- The principal investigator (PI) has taken a novel approach to solving an issue that is a key cost driver in hydrogen dispensing. A solid team of national laboratory experts is on board to perform materials evaluations, and the technical bar set—with respect to durability of a developed solution—is such that if the material can be deployed effectively, the self-healing attributes of the polymer should affect long-term hose durability. However, the omission of an overall scheme for how this polymer will be integrated into a full hose assembly makes it difficult to know whether the results can be translated into a deployable product.
- Ultimately, the project team would benefit by defining several key metrics that can be used to direct the project moving forward. These metrics should explain concepts such as the following: (1) what self-healing is, (2) whether self-healing includes restoration of strength and ductility or just hydrogen permeability/ability to seal hydrogen, (3) what the success level of self-healing is, (4) whether a scratch at 50% through thickness would be expected to self-heal more so than a completely severed membrane, and (5) how the properties and functionalities recovered through self-healing are defined (i.e., quantifying factors such as a membrane's ability to seal hydrogen, a specific measure of strength, or the elongation-to-failure measure). Each should be defined, and each metric should be quantified.
- The overall approach listed on slide 5 seems all right. From the subsequent slides in the presentation, it is not clear which of the work was done as part of U.S. Department of Energy (DOE)-funded Hydrogen Fuel Cell Technologies Office work versus what was already accomplished before this project started. It seems like many of the items listed in the approach were already known/published (e.g., design, synthesis, and characterization). Hence, it appears that the approach is repeating some of the prior work. Slide 3 describes a key barrier that this project is addressing as "reliability and cost of hydrogen fuel pumping." What is missing in the presentation is an explanation about the barrier associated with the "reliability" (of the hose, presumably) and whether it is attributable to the failure of the inner lining (that this project is trying to heal) or some other component of the hose (joints, crimps, cracking in outer layer, etc.).

Question 2: Accomplishments and progress

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

- The work has shown good progress to date. The results are promising. Once metrics are defined, it is believed that the DOE Hydrogen Program (the Program) can provide even more quantifiable support for the work that has already been performed.
- The project appears to be making good progress and is meeting its milestones.
- The PI has demonstrated multiple polymeric compositions that meet the cost, self-healing, and durability targets set at the beginning of the project. All of these successes are commendable; however, the order of tasks in the work plan, and the difficulty in translating the damage mechanism used to test the self-healing properties, make it difficult to determine whether these results can be translated to solve the issue of cracking of hydrogen dispensing hoses.
- The PI's work that was funded by the National Science Foundation (published in *Science* 2018, 362(6411) 220–225, 10.1126/science.aat2975) shows that the team had already developed self-healing chemistries prior to this project's start date in 2020. The stress–strain curves (slide 9) and molecular dynamic simulation (slide 10) are both taken from this *Science* article. Therefore, it seems that some aspects of the work described here have already been published, and new work is not clearly identifiable. The distinction between the prior work and that performed under current funding needs to be made. A self-healing response in the presence of moisture, temperature, and pressure (slide 27) was determined. These are useful experiments. Slide 10 states a high cohesive energy density (CED) is desirable for self-healing, and slide 27 shows that CED decreases with an increasing number of hydrogen molecules. It is unclear whether that means that the "healing action" will occur only when hydrogen is not being dispensed.

Question 3: Collaboration and coordination

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

- This project has a good team approach overall.
- The PI has assembled an excellent team of experts from national laboratories to perform accurate and detailed materials property testing. However, the team is very much in need of input from an industrial partner, ideally a hydrogen dispensing hose manufacturer, to assist in ensuring that the selected test metrics are the most relevant to the material properties necessary for the application. An industrial partner could also provide insight into the integrated design of a hose using these self-healing polymers and into additional materials testing that might be required to ensure the developed materials meet the required technical specifications of a polymer that could easily be utilized in such a design.
- Slide 16 lists three national laboratory partners. Gas permeability test data from one of the national labs was presented, but data from the contributions of the other two labs were not immediately obvious from the slides. Most of the work seems to be done at the PI's institution. The project could benefit from initially reaching out to national laboratories to learn about damage/failure mechanisms in hydrogen dispensing hoses in previous funded research, and then determining how the self-healing phenomenon could address those damages/failures.
- The collaboration and coordination taking place in this project were not explained well. From the review, it is not clear whether the appropriate collaboration is in place for success.

Question 4: Relevance/potential impact

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

- Developing inexpensive, self-healable copolymer fiber-reinforced composites to extend the hydrogen hose service life is critical to increasing the reliability and decreasing the cost of hydrogen fuel pumping. Resolving this issue would be a big win for the Program and would have a widespread impact.
- If successful, this project would provide great impact on hydrogen storage, transportation, and fitting applications. The work is very exciting, given its potential impacts.
- The project's high-level goal of developing a self-healing hydrogen dispensing hose would have a significant impact on the overall goals of the Program through cost reduction and elimination of unscheduled maintenance. However, the approach selected to try to meet this goal, and the order of tasks, waits until the end of the project to determine whether the developed materials will be able to perform in the intended environment. As such, the overall impact of the work will not be known without testing the self-healing ability of composites formed with these polymers.
- In general, the project has identified its main goal as improving hydrogen dispensing hoses so they can survive many cycles, which is aligned with the Program goals. The project goal is to use a "copolymer matrix with Innegra™ fibers," according to slide 2. However, all the experiments were done on damaged "neat" polymer, and it seems likely that the reinforcing fibers, Innegra, are unlikely to heal if cut or damaged. Therefore, an important question arises: even if the project were wildly successful, it is not obvious that a healed copolymer with damaged reinforcing fibers would still be a viable inner lining of a hose. Some information on the type of damage in hydrogen dispensing hoses currently used would have been useful to provide the context for this work and determine the value. The project assumes that it is the inner lining that is the limiting feature of a dispensing hose. It would be useful to know whether prior DOE-funded work on dispensing hose damage can validate this assumption.

Question 5: Proposed future work

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

- The proposed future work appears appropriate. The proposed future work can be made far more impactful if the team immediately determines the criteria for success (defining self-healing, defining metrics for recovered properties, defining successful recovery kinetics, etc.).

- A planned demonstration of self-healing for a reinforced composite mat, including polypropylene fibers, will help to resolve concerns that these materials will either not exhibit the mechanical robustness required for the application or will not provide the same self-healing durability within a strong composite. However, additional work is necessary to understand how such a mat could be integrated into a dispensing hose assembly and whether it could operate as intended. If the hydrogen permeability of the composite mat is too high, the external casing of the hose will experience cyclic exposure to high-pressure hydrogen, eliminating any benefit that the composite mat self-healing properties could provide.
- Several proposed tasks (slide 18) seem focused on tensile testing and increasing the number of damage–repair cycles, even though (1) the tensile strength does not seem to be a required property for the inner lining and (2) the relationship between a hose's duty cycle and the level of damage are not known. Slide 17 indicates effort to install a micro-scratcher to enable repetitive cuts. Since tensile failure is not the key failure mechanism, the project can consider some other means to impart damage repetitively (e.g., fatigue loading/unloading to a fixed number of cycles, followed by permeability testing). The Innegra fibers in the matrix are an overlooked part of the equation. The project can consider demonstrating the retention of required properties if the overall composite—and not just the copolymer matrix—is damaged.
- It is not clear how these polymers will perform in composite systems. This work seems to address the matrix material but assumes that there will be no loss due to fiber damage. It is unclear if this is the expected mode of failure. An understanding of the damage and healing mechanisms is essential for long-term manufacturability, reliability, and safety. Unfortunately, the project does not appear to have a plan to gain this understanding.

Project strengths:

- This project takes a novel approach to the long-standing, difficult problem of materials failure in hydrogen dispensing hoses. The PI has demonstrated the ability to synthesize multiple self-healing polymers that can be cast as films. These formulations exhibit the ability to restore their mechanical properties after damage due to a high enough number of cycles to be relevant in hydrogen service use. In addition, the materials have been developed with an eye toward cost, and their integration into a dispensing hose assembly would be financially viable.
- An excellent scientific basis, as well as chemical and modeling analyses, has been provided for a self-healing phenomenon.
- The project appears to be progressing very nicely with regard to materials processing. This will allow the team to build a solid foundation on which to move forward.
- This is innovative and potentially game-changing work to enable low-cost, safe, and reliable hose liners.

Project weaknesses:

- This project suffers from a lack of industrial input on how the materials would be integrated into a dispensing hose assembly. The composites developed need to be assessed for hydrogen permeability, as this is crucial in understanding whether they can be integrated into a hose in such a way that the self-healing properties will provide a benefit. The project also would have benefited from earlier testing of the self-healing performance of the composite mat, perhaps after the first polymer was synthesized, to see whether the self-healing properties of the pure polymer system will translate to the composite without further modification.
- (1) It is difficult to distinguish between pre-existing work and the new work being done. (2) Self-healing is expected to maintain the inner lining's permeability, but it is not clear that permeability of the inner lining is indeed the reason for failure in hydrogen dispensing hoses. (3) It is not clear whether any damage to the reinforcing fibers will render the lining (and thus, the hose) unusable even if the matrix is able to self-heal. (4) It is not clear who is providing the cost-share on slide 3. (5) The glass transition temperature seems to be close to room temperature or near 0°C on slides 6 and 7. It seems possible that such a transition temperature would make the copolymer brittle when filling in pre-cooled hydrogen and, therefore, more susceptible to widespread damage than the total number of healing cycles to which it is being tested.
- Understanding the damage mechanisms that are seen in liners today and matching the damage mode evaluated by the project to these mechanisms will be important to realizing the project's goals. There does

not seem to be a plan to accomplish this. Understanding the role of the fiber in the composite and how the self-healing matrix interacts with the fiber is largely ignored.

- The one weakness that is recognized in the project is that the project lacks detailed explanations for important concepts of self-healing and would benefit from defining and quantifying these components of the project.

Recommendations for additions/deletions to project scope:

- The project should clearly demarcate pre-existing work (experiments, modeling) from new work. Although self-healing in general is a very desirable material property for engineering applications, the project should focus on identifying the weak link in the hydrogen dispensing hoses and determine whether self-healing of the polymer matrix is the right property to be addressed.
- The project scope should be expanded to examine the hydrogen permeability of the composite polymer mat compositions. A commercial hose supplier should be added to the project team to assess whether additional materials property testing is required to meet the technical specifications necessary for dispensing hose integration and full commercialization.
- The team should consider focusing more on gaining a fundamental understanding of the damage and healing mechanisms. It will be difficult to gain the trust needed for code development and market penetration without a fundamental understanding of these mechanisms.
- The team would benefit from defining several key metrics that can help to direct the project moving forward.

Project #IN-021: Microstructural Engineering and Accelerated Test Method Development to Achieve Low-Cost, High-Performance Solutions for Hydrogen Storage and Delivery

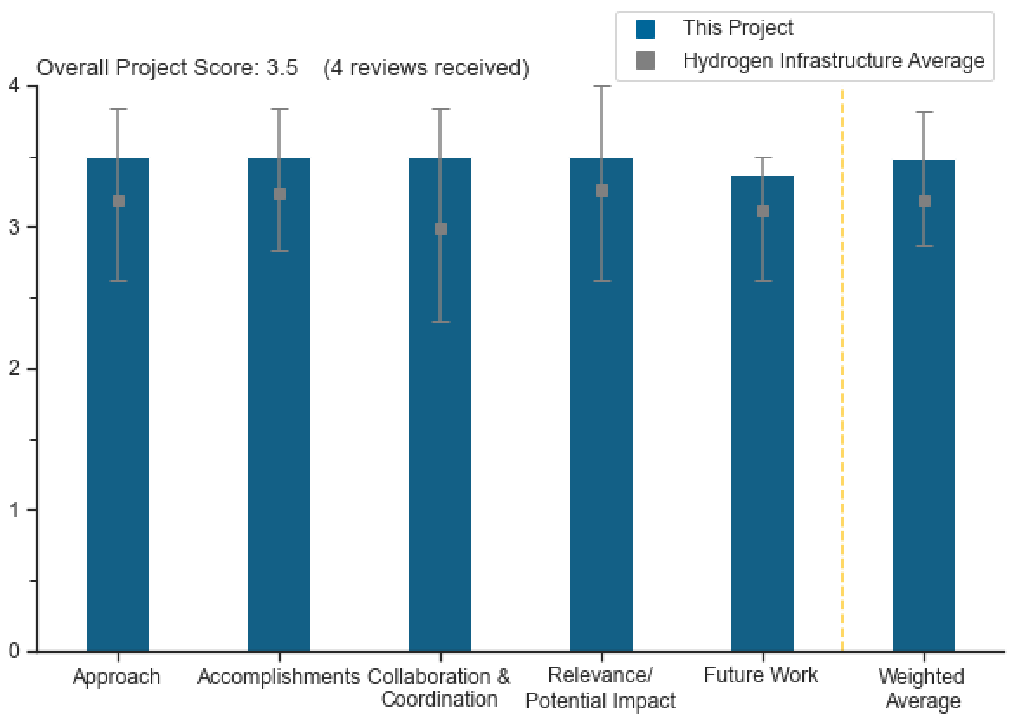
Kip Findley, Colorado School of Mines

DOE Contract #	DE-EE0008828
Start and End Dates	2/1/2020 to 2/28/2023
Partners/Collaborators	Los Alamos National Laboratory, National Renewable Energy Laboratory, WireTough Cylinders, LLC, United States Steel Corporation (U.S. Steel), General Motors Company, Hydrogen Materials (H-MAT) Consortium, Chevron Corporation, POSCO
Barriers Addressed	• Reliability and costs of gaseous hydrogen compression • High as-installed cost of pipelines • Gaseous hydrogen storage and tube trailer

Project Goal and Brief Summary

This project aims to use novel microstructural design techniques to develop lower-cost, high-performance steel alloys for use in hydrogen refueling infrastructure. The project will also develop and validate accelerated test methods for efficiently evaluating variations in alloy and microstructure design, enabling broader accessibility and lower-cost testing in hydrogen environments. The work could accelerate the implementation of hydrogen fueling infrastructure.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- The project clearly presents two objectives: (1) developing lower-cost steel alloys for use in hydrogen refueling infrastructure and (2) developing and validating accelerated methods to lower the cost of testing in hydrogen environments. These objectives focus on recognized barriers related to structural materials in hydrogen fuel infrastructure.
- The work provides very useful potential in development of infrastructure and acceleration of test methods to efficiently evaluate variations in alloy (austenitic steels [POSCO and commercial stainless steels]) and microstructure design to enable broader accessibility and lower-cost testing in hydrogen environments.
- There is a sound approach to the work. Comparison between electrochemical and gas in situ charging may be a little narrow, but it is a good first step toward being able to compare results from most labs that are able to do electrochemical and the few labs that have gas in situ testing capabilities. Going forward, it would be good to see a little more emphasis on the microstructure of the various steels—and specifically on enumerating the similarities and differences between them and how those may affect the steels' performance.
- The principal investigator (PI) nicely outlines the objectives and the approaches used to achieve these objectives. This is particularly true for the first two materials development tasks. The knowledge gaps and uniqueness of the third effort (the linking between electrochemistry and hydrogen charging) is less well-developed, and it is not clear that the approaches are going to provide clear insights. The fatigue crack growth testing methodology development is interesting but seems disjointed from the rest of 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 project formulates a clear and reasonable series of goals and milestones related to the two primary objectives of alloy development and materials test method development. The reported accomplishments represent progress toward these stated goals and milestones.
- The work on cost-effective austenitic steels (stainless steels) has great potential for design and construction of pressure vessels and pipelines that are suitable for hydrogen embrittlement performance of austenitic steels and lower-cost ferrite–austenite alloys that have intermediate hydrogen embrittlement performance, i.e., between austenitic stainless steels and lower-alloy ferritic steels.
- Progress is reasonable, given that it is early in the project and especially considering barriers due to the pandemic. Preparation work has been performed in designing and fabricating the alloys.
- Considering the effects of the pandemic on the last year, reported progress is excellent.

Question 3: Collaboration and coordination

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

- The industrial involvement is solid. To date, it does not seem like all of the benefits have been fully realized. This is simply due to the stage of the project; the feedback loop and early engagement with steel producers will ensure that candidate alloys are scalable and cost-effective.
- Great collaborations have been established with a number of highly qualified laboratories, industries, and institutions that will be essential in achieving the goal.
- The project is productively engaging with its partners to accomplish goals and milestones. One notable example is the relationship with U.S. Steel, which is producing experimental alloys that are designed to meet cost and performance targets.
- It looks like collaboration and coordination will increase as recovery from the pandemic continues, and it seems that the amount of collaboration, while good at the moment, will improve as the project moves forward.

Question 4: Relevance/potential impact

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

- The research and development relating to high-strength austenitic steels and commercial stainless steels could be a breakthrough for efficient and reasonable-cost pressure vessels that are used in delivery and storage of gaseous and liquefied hydrogen.
- The potential impact will depend largely on the ability to make a cost-effective alloy that meets the design criteria. (The PIs should be cognizant of how the data were collected with regard to the loading rate and that other important parameters will have strong impacts on the results.) Growth kinetics, other metrics of susceptibility to environmental degradation, and other property variations should be incorporated (and the PI has stated that the team plans to account for these). The impact and potential for success of the testing methods is questionable.
- This project aligns directly with the DOE Hydrogen Program's (the Program's) need for lower-cost materials that meet performance requirements in hydrogen service.
- The relevance to the Program goals is clear. The potential impact to relevant industries, if the project is successful, is clear. The relevance to the wider hydrogen embrittlement/testing community could be greater, though that would require expanding the scope of work. For example, while the current plan will allow comparison between the electrochemically charged and gas-charged in situ results with the specific sharp-notched sample geometry, it is not clear that it would be relevant for other conditions, which would be of great interest to the wider hydrogen community. However, extensive study beyond the stated scope of this project would be needed to answer that particular need.

Question 5: Proposed future work

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

- The proposed future work extends reported accomplishments and is consistent with the milestones and goals outlined for the project.
- The proposed future work shows a clear path forward.
- The results of Year 2 of this ongoing project will determine the potential for expanding the project to Year 3.
- The work is in its infancy, so future work will just execute the proposed work. Statements of the remaining challenges are broad, vague, and weak.

Project strengths:

- The current project for Year 1 and testing protocols provide potential for development of an accelerated testing for austenitic steel and commercial stainless steels that are suitable pressure vessels or pipelines that provide adequate strength and ductility for resisting hydrogen embrittlement. The highlight of this project is the development and implementation of an "electrochemical hydrogen charging setup utilized to evaluate fracture toughness of steels in the presence of hydrogen-containing environments."
- The alloy design strategy has a sound technical basis for its performance target, i.e., stacking-fault energy. The active roles of partners such as U.S. Steel, the National Renewable Energy Laboratory, and Sandia National Laboratories enhance the prospect that results from this project can have impacts on technology and concretely advance Program goals.
- Project strengths include collaboration with industry partners, logical material design strategy, and building on well-known literature to improve hydrogen embrittlement resistance by modifying stacking-fault energy.
- There is a strong technical metallurgical approach to solving a clear engineering problem.

Project weaknesses:

- This is somewhat of an informed Edisonian alloy design strategy with disjointed alloy design and testing efforts, and it is seemingly weak on determining knowledge gaps and a plan to address the electrochemical/hydrogen charging issues.
- The project needs to reduce the cost and speed the testing in Year 2, which will be a major factor in whether to continue the project.
- It is not clear that activities associated with partners Los Alamos National Laboratory (LANL) and WireTough Cylinders, LLC (WireTough) align with project goals. The need for permeation experiments at LANL to meet the two project objectives (alloy design and accelerated test method development) has not been demonstrated. In addition, the implication is that WireTough may consider the alloys developed in the project as alternatives to the incumbent liner material (A372 Grade J) in Type 2 pressure vessels. From a cost perspective, the idea that the highly alloyed steels proposed in this project could replace the A372 Grade J may not be realistic.
- More basic science aspects, such as neutron scattering analysis to determine mechanisms, feel tacked on to the project. If the project is successful, good insight could be obtained, but as presented, it has the feel of "Let's try it and see what we get."

Recommendations for additions/deletions to project scope:

- This project should continue with a new objective and goal to use the resources and strength toward the selection of new cost-effective austenitic steels (commercial stainless steels) that are suitable for delivery and storage of high-pressure gaseous and cryogenic hydrogen.
- Fatigue efforts seem tangential. Improvements to the electrochemical/hydrogen charging evaluation approaches are recommended.
- It is recommended that the project critically evaluate the roles for LANL and WireTough in the project.

Project #IN-022: Tailoring Carbide-Dispersed Steels: A Path to Increased Strength and Hydrogen Tolerance

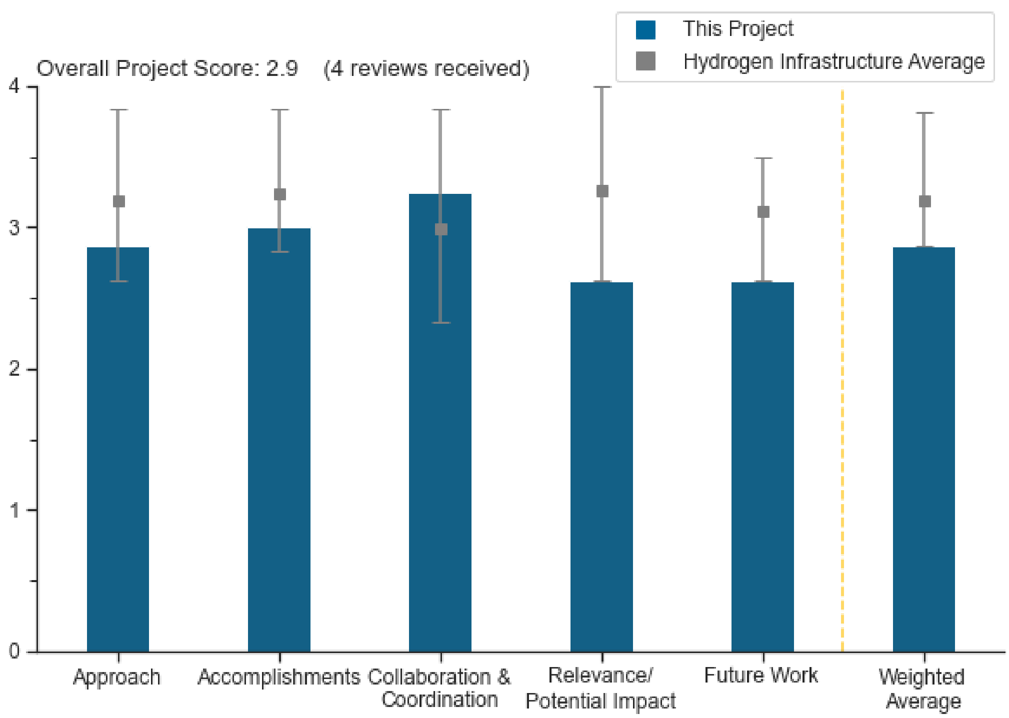
Gregory Thompson, The University of Alabama

DOE Contract #	DE-EE0008831
Start and End Dates	1/7/2020 to 1/31/2023
Partners/Collaborators	Colorado State University, Exothermics, Inc., Ames Laboratory, Army Research Laboratory, Hydrogen Materials (H-MAT) Consortium
Barriers Addressed	• Identification of the most sustainable transition metal carbides for a hydrogen trapping mechanism • Uniform dispersion of the trapping carbide particles • Forming metal-rich-carbide (hemicarbide particles)-dispersed steel alloy • Achieving the required compact steel alloy while retaining the target phase structure

Project Goal and Brief Summary

This project is developing a new carbide-dispersed austenitic/ferritic steel for hydrogen storage and dispensing. The alloy will have higher strength and hydrogen tolerance, which will increase the service life of hydrogen storage equipment, facilitating the expansion of hydrogen infrastructure while reducing its cost and environmental impact.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- This project addresses a critical need within the hydrogen technology community. The novel approach to avoid hydrogen embrittlement is well-thought-out and combines experimental and modeling approaches to the best use of both. There is a practical team in place to provide the input necessary to allow this technology, if successful, to transition to commercial use. The only potential gap in the approach is that long-duration hydrogen exposure experiments may be necessary to determine whether the hydrogen sequestration sites developed will become saturated and what effect this will have on the steel's mechanical strength (the presentation did not address the duration of proposed experiments).
- This project feels high-risk, high-reward. If successful at creating results and discovering how to transfer these results to conventional structural materials, this project could prove a fantastic advance in design. Otherwise, some fundamental science may be advanced, but the application will be minimal.
- The project proposes a combined experimental/first principles computation approach to disperse transition metal nano-carbides in ferritic/austenitic steels in order to improve hydrogen embrittlement and strength. The applications are directed toward hydrogen storage and hydrogen dispensing. The main thesis of the project is that the insertion of carbide nanoparticles in a steel microstructure increases the resistance of the alloy to hydrogen embrittlement. The idea is drawn from a similar approach to use oxide dispersoids to strengthen creep resistance of metallic alloys. Two critical barriers—the need for increased notched tensile strength and the need for high-yield strength—have been identified, but there is no specific quantitative objective. For instance, in the case of stationary gaseous hydrogen storage or dispensing, there is no reference to operating or refueling pressures at which these carbide-incorporating microstructures—achieved through powder metallurgy—are aimed. The statement that the proposed steel microstructures will have "comparable or better fatigue strength" is vague. There is not even a single reference or motivation in the proposed plan as to how dispersed carbides will bring about "better fatigue strength," specifically in the presence of hydrogen. The project uses density functional theory (DFT) to identify the most effective stoichiometric and non-stoichiometric carbides in trapping hydrogen, and the team will then use powder metallurgy to disperse the most promising carbides in a metal matrix. However, metrics of what constitutes promising carbides are not referenced. In the end, the project will use atom probe tomography to identify the location of hydrogen in the carbides. Incidentally, the project does not mention how it will quantify the overall trapping capability and distribution of the carbides because, in the end, it is this capability and density of the carbides that matter vis-à-vis resistance to degradation.
- It is not apparent why it is necessary to develop carbide-dispersed steels (CDSs) with optimized hydrogen trapping characteristics since the project does not identify the shortcomings of incumbent technologies or how the proposed solution represents an advancement. For example, it is unclear whether the incumbent SA-372 Grade J steels in the stationary hydrogen storage vessels are inadequate in their cost or performance or whether the CDS alloys can be demonstrated as the solution to such shortcomings. It is also not clear that the cost or performance of incumbent Grade 316 stainless steels in hydrogen distribution systems is a barrier to the deployment of refueling stations—or that the CDS alloys are a potential solution to this barrier.

Question 2: Accomplishments and progress

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

- The principal investigators (PIs) have demonstrated significant progress toward their stated goals, including developing multiple practical approaches to producing test samples. Future experiments will determine how effective the proposed solution is and how broadly applicable, but all tasks to date have been successful.
- The researchers made good decisions to maximize modeling and planning with collaborators during the limited-lab-access period due to COVID-19. The early stages have clearly been picking up speed as access has opened.
- DFT calculations were used to establish trap-binding energies for a number of stoichiometric and non-stochiometric carbides with calculated energies ranging from 10 to 100 kJ/mole. Interestingly, the

calculations indicate that as hydrogen is filling out the trap sites of a titanium carbide, its trapping capability reduces; in fact, it reduces drastically, as shown in the figure on slide 8. In addition, DFT calculations have been set to explore the activation energies for diffusion in the carbides. In summary, although trap-binding energies have been computationally determined, there has not been any reference to how such binding energy magnitudes underlie the project's goal, which is the mitigation of hydrogen embrittlement. On the experimental side, sufficient progress has been reported on dispersion and sintering of Fe with ZrC, ball milling of Cr into solution, and a route of rapid spark plasma sintering (SPS) dispersing of ZrC nanoparticles in Fe and 304L stainless steel. In general, within the project scope of developing carbide-strengthened microstructures, the project has made sufficient progress, in collaboration with the NASA Ames Laboratory SPS project.

- It is not clear how the performance indicators for the project were established. For example, the apparent metric for optimizing the hydrogen-trapping characteristics of the CDS is trap-binding energy exceeding 75 kJ/mol. An explanation is needed as to how this value was determined. In addition, trapping characteristics depend not only on binding energy but also on trap density, but it is not clear how trap density is being considered in the objective to optimize hydrogen-trapping characteristics. Other performance indicators are maintaining 95% of the notched tensile strength after hydrogen charging and yield strengths above 500 MPa. It is not clear how these performance indicators were established, particularly in the absence of any reference to incumbent technologies.

Question 3: Collaboration and coordination

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

- The PIs have assembled an excellent team with the right mix of expertise in fabrication, modeling, and property testing of materials. The team also includes multiple potential end users, including commercial end users, to ensure their input in the development process and to increase the chances of transitioning to becoming commercial if the approach is successful.
- The relationships with Ames Laboratory, Exothermics, Inc., NASA, and Sandia National Laboratories appear productive. Hydrogen technology stakeholders, such as Praxair, must be engaged to ensure the project is designed to address particular barriers and is guided by relevant performance indicators.
- For the experimental approach of carbide dispersing in Fe and austenitic matrices, it seems that the collaborations with Ames Laboratory, Exothermics, Inc. (processing through hot isostatic pressing), and NASA are effective and serve as a solid pathway for the project to synthesize the microstructures it promised. The computational aspects of the project seem to be uninformed of the existing understanding of hydrogen interactions with vacancies and carbides.
- It seems that the project's collaborations experienced limitations due to COVID-19, moving focus toward in-house activities. Future work appears to use collaborations heavily, which looks promising for producing progress.

Question 4: Relevance/potential impact

This project was rated **2.6** for supporting and advancing progress toward the Hydrogen and Fuel Cell Technologies Office goals and objectives delineated in the Multi-Year Research, Development, and Demonstration Plan.

- If the approach to reducing hydrogen embrittlement is proven successful experimentally, the impact could be significant. The approach, as detailed, should provide conclusive answers in the second year of the project. It remains to be seen how broadly applicable this technology can be; if the hydrogen sequestration sites quickly become saturated, hydrogen embrittlement may still occur. That said, there may still be short-term exposure applications (i.e., compression and dispensing) that could make use of the technology even if the protection from embrittlement is transient.
- Again, because this appears high-risk, high-reward, the potential impact is high if good results are achieved and processes to apply results to industrially relevant materials are found. The current status suggests less impact in the short term.

- Given the absence of references to incumbent technologies, it is not clear what impact the project will have on DOE Hydrogen Program goals and objectives. Specifically, it is not clear how the project will reduce costs or improve performance relative to incumbent technologies.
- The project addresses a classic and thoroughly investigated aspect of hydrogen embrittlement, namely, the trapping of hydrogen at microstructural defects. However, the trapping of hydrogen mainly affects the amounts and the spatial and temporal distribution of hydrogen in the material. Specifically, with the proposed carbide-dispersed-strengthened steel, the project's outcome may be that it can deliver carbide-strengthened steels in which hydrogen is all trapped at carbides and no hydrogen is available in the rest of the lattice to initiate embrittlement. However, this mitigation strategy needs to be analyzed relative to specific and targeted closed-system applications. It does not work for open systems in which hydrogen uptake is continuous.

Question 5: Proposed future work

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

- The team has outlined a solid plan to provide experimental evidence that the modeling work will translate to significant material property gains. The only additional work that may be required is long-term hydrogen-exposure testing to ensure the durability of the approach.
- Clear steps forward have been lined out to next steps, though not to long-term goals. "Discussions under way with industry partners," is a good statement but does not necessarily give concrete steps forward. The characterization techniques were not as clearly laid out in the presentation (although there were some answers in the question-and-answer period). Emphasis on the atom probe feels like the team is emphasizing the in-vogue technique, while ignoring other techniques that could provide the researchers with good characterization information (thermal desorption spectroscopy [TDS], diffraction analysis, and electron energy loss spectroscopy [EELS] in a transmission electron microscope are a few that come immediately to mind).
- The proposed work on phase stability of the carbide-rich microstructures and the tailoring of the carbide composition are reasonable next steps. However, what is missing is how and why these efforts relate to hydrogen embrittlement mitigation. Perhaps the project aims to determine whether martensite will precipitate under operating temperature or hydrogen-related conditions. If that is the case, it is not clear how such understanding affects the overall scope of the project. An interesting aspect of the project is the identification of hydrogen-trapping sites using atom probe tomography (APT), which is seen as the most important outcome of the project for its scientific value. Unfortunately, no reference to the DFT efforts is given, nor to the pathways that will be undertaken to relate atomistic insights with the APT results.
- No milestone list or project roadmap was presented, so it is not clear how the proposed future work represents a progression toward project goals.

Project strengths:

- This program has a very strong, well-rounded project team. The balance of modeling with experimental demonstration of theoretical results is excellent. The early success demonstrating the ability to formulate the desired compositions bodes well for the ability of this project to conclusively demonstrate the potential efficacy of the approach.
- The strength of the project is its experimental component. The development of carbide microstructures that are well-characterized relative to particle distribution, shape, size, grain boundary size, and structure may lead to potential ferritic or austenitic microstructures that are worth testing at Sandia National Laboratories and worth comparing with other existing microstructures.
- The coupling of modeling and experiments is a promising route toward more effective alloy design.
- This project has an innovative approach, and potentially, good fundamental science could result from this study.

Project weaknesses:

- The trapping of hydrogen at microstructural defects has been thoroughly investigated in the past 50 years, and its importance is recognized in its effect on the spatial and temporal hydrogen distribution in a component. The range of the carbide-trapping binding energies the project reported through DFT calculations is not at all different from those reported in the review article by Hirth (*Metallurgical Transactions A*, 1980, 11A, pp. 861). In addition, for an open system, trapping sites eventually saturate; in fact, they saturate very fast in ferritic systems, and eventually hydrogen becomes available at fracture initiation sites. Hence, the relevance of this project can be sought in the case of closed systems operating under known conditions of temperature and hydrogen content. Under such conditions, the project's outcome may be that it can deliver carbide-strengthened steels in which hydrogen is all trapped at carbides and no hydrogen is available in the rest of the lattice to initiate embrittlement. In fact, even this proposition needs to be carefully ascertained with regard to hydrogen effects in the carbide or the carbide–matrix interface. Definitely, hydrogen dispensing involves an open hydrogen system, and it seems unlikely that the proposed carbide mitigation strategy will not work. As for hydrogen storage, the proposed carbide strategy will depend on the hydrogen pressure and carbide distribution, but these aspects are not addressed as fundamental ingredients of the project. Another important aspect of the project is the nature of the carbide–matrix interface, which seems not to have been taken into account when the project was designed and proposed. If the carbides are incoherent, Tsuzaki (e.g., in "Effects of Hydrogen in Materials," *Proceedings of the 2008 International Hydrogen Conference*, pp. 448-455) has demonstrated in the case of TiC that these carbides do not trap hydrogen at room temperature, which may imply that the value proposition of the project is called into question since hydrogen embrittlement is of concern at room temperature. On the other hand, if the carbides are coherent or semi-coherent and trapping takes place at the interface at room temperature, it does not seem likely that interfacial trapping at room temperature will bring about any new mitigation strategy for hydrogen embrittlement. Furthermore, if the project is aiming at incoherent carbides (Mrovec et al., *International Journal of Hydrogen Energy*, 45, 2020, pp. 2382–2389) and trapping within the carbide vacancies, the project needs to address the activation barriers for vacancy trapping (Di Stefano et al., *Physical Review*, B 93, 184108, 2016) and how these barriers affect the overall scope of the project at room temperature. Lastly, it seems that the PIs have placed the emphasis on the binding energy. It is not clear how they plan to assess hydrogen embrittlement in relation to the density and distribution of the nano-carbides.
- The novelty of the approach to reducing hydrogen embrittlement under investigation leaves many potential unknowns yet to be proven out. Even if the sequestration approach is effective, with thermodynamic equilibrium dominating the hydrogen interactions, the sites able to sequester hydrogen may quickly become occupied and limit the overall impact. In addition, the industrial-scale cost of fabricating steel utilizing the techniques developed so far has not yet been evaluated but is likely to be a significant premium over the current processes.
- This project has the flavor of a solution seeking a problem. Without well-defined barriers related to the cost or performance of incumbent technologies, it is not apparent that there is a tangible problem whose solution resides in the objectives of this project.
- There is no clear path to the applications.

Recommendations for additions/deletions to project scope:

- The premise of this project is that hydrogen embrittlement resistance of steels can be improved through tailoring the trapping characteristics of transition metal carbides. In most cases, this concept applies only to closed systems in which materials subjected to stress contain a fixed hydrogen concentration. For fixed hydrogen concentration, benign traps with high binding energy and high density can deplete hydrogen from metallurgical sites that serve to activate hydrogen embrittlement. However, the materials in hydrogen containment components are effectively open systems, as the materials are subjected to stress and are exposed to an infinite hydrogen source. In this case, the hydrogen concentrations at all trap sites are in equilibrium with the hydrogen gas, so traps with high binding energies are not scavenging hydrogen from sites with low binding energies. For this reason, the target of maintaining 95% of the notched tensile strength after hydrogen charging will be misleading for the performance of the CDS in an open system. One property that may benefit from trap-site engineering in an open system is hydrogen-assisted fatigue crack growth rate, since this may scale with the effective hydrogen diffusion coefficient. It is recommended that the project not focus on notched tensile strength as a performance target but rather pursue the prospect that trap-site engineering can reduce effective hydrogen diffusivity and, therefore, affect hydrogen-assisted fatigue crack growth.

- It will be interesting for the project's APT effort to identify the magnitude of the hydrogen concentration in the lattice next to the carbides and that in the carbide or at the carbide–matrix interface, if there is such interfacial trapping (Takahashi et al., *Scripta Materialia*, 67, 2012, 213–216). Another important issue is the determination of the nature of the carbide–matrix interface because, as is discussed in the section on overall project weaknesses, it governs the trapping capabilities of the carbides. The computational component of the project needs to identify the activation barriers for hydrogen trapping in the carbides specifically at room temperature and by accounting for the nature of the interface. It is important that the results be validated experimentally. Calculated binding energies as shown on slide 7 are conventional and do not hold promise for mitigation, especially for open systems. Within the framework of the project, the density and distribution of the carbides also need to be determined, as they both affect the hydrogen populations and diffusion paths.
- If the approach proves successful in avoiding hydrogen embrittlement, the team should add long-hydrogen-exposure testing to the project to understand whether the effect is permanent or transient—and on what timescale.
- The characterization feels lacking. The project needs someone with strong credentials and access to good and varied capabilities to fully characterize the carbides and interfaces, not just cursory checks.

Project #IN-025: Hydrogen Delivery Technologies Analysis

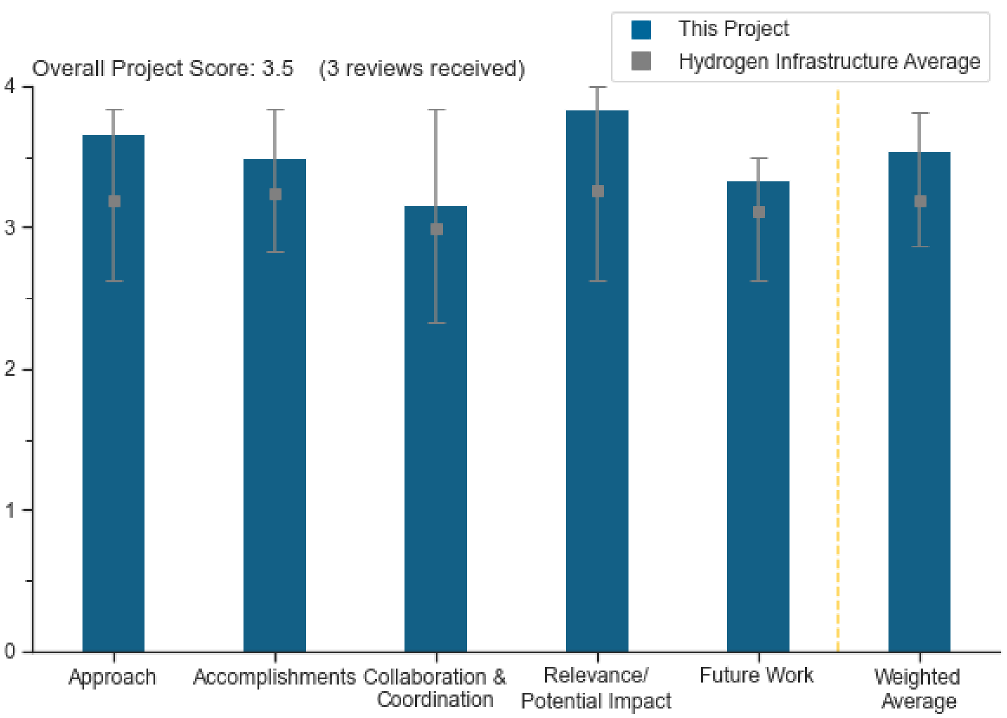
Amgad Elgowainy, Argonne National Laboratory

DOE Contract #	3.4.0.1
Start and End Dates	10/1/2005
Partners/Collaborators	Energy Technology Analysis, U.S. DRIVE Partnership
Barriers Addressed	• Inconsistent data, assumptions, and guidelines • Insufficient suite of models and tools • Stove-piped/siloed analytical capability for evaluating sustainability

Project Goal and Brief Summary

This project aims to evaluate the economic and environmental costs and benefits of hydrogen fuel and fueling infrastructure. Researchers will analyze various hydrogen technologies throughout their lifecycles and identify the technologies with the highest cost-effectiveness and lowest environmental impact. Argonne National Laboratory's (ANL's) Autonomie Team is collaborating with Energy Technology Analysis, the U.S. DRIVE Partnership, and other industry partners on this project.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- The approach this presenter took focused on how the team evaluated the impacts of various approaches for hydrogen delivery and the selection of hydrogen refueling station (HRS) components using models. The presenter explained the economic assumptions, after identifying the objectives of the project and the critical barriers, such as consistent data, assumptions, guidelines, tools, and capabilities for analyzing sustainability. When this presenter explained the team's evaluations of the overall fuel cell electric vehicle (FCEV) cost and energy storage for the fuel, he described output from the Hydrogen Delivery Scenario Analysis Model (HDSAM) and suite of models. The approach is strong and believable, given that ANL updates and harmonizes these models with other U.S. Department of Energy (DOE) models, i.e., Hydrogen Analysis (H2A) and Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET). To overcome insufficient data (a barrier) and prepare the models for use with emerging applications, such as refueling of medium-duty (MD) and heavy-duty (HD) FCEVs, ANL developed the Heavy-Duty Refueling Station Analysis Model (HDRSAM), which is used worldwide by thousands who seek to evaluate the cost of hydrogen refueling for various fueling station configurations and demand profiles. HDRSAM remains unique and is the only model of its kind that is available in the public domain. This way, DOE and the public have a consistent set of tools that are harmonized in their assumptions and thus overcome the barrier of stove-piped/siloed analytical capability for evaluating the sustainability of hydrogen delivery approaches and hydrogen dispensing based on the selection of components. The approach is valid in that it links technical, economic, and environmental performance to identify opportunities and challenges for different hydrogen supplies and refueling station components and various station costs (i.e., capital, operating, and energy). The project is very well-designed and also practical. The presenter explained that, for delivery options, the HDSAM scenario and delivery cost are evaluated over a wide set of options. Because of the wide set of scenarios, a user of the model can evaluate a variety of delivery options without investing in any one particular option. A user can evaluate regional power options, for example, without making an agreement with the power provider, and the user can examine greenhouse gas (GHG) emissions using HDSAM using different approaches to producing hydrogen, prior to any actual investment in the technologies for low-carbon pathways. The evaluation and assessment approaches, the presenter explained, are already proven, and using the model is feasible for numerous applications. The project approach also integrates a lifecycle analysis of GHG emissions for various transmission and distribution (T&D) options used for hydrogen. Since T&D is very expensive and often represents the bulk of a project's cost, this approach addresses a key part of the hydrogen value proposition.

- The scope of this project is timely and relevant to the deployment of HD vehicles such as Class 8 fuel cell electric trucks (FCETs). Furthermore, DOE's investment in this project highlights the importance of HD transportation in achieving the national goals to reduce emissions and decarbonize the transportation sector.
- The project meets its intended goal of providing technical, economic, and environmental analysis of hydrogen markets.

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 contributes to the DOE focus area of cost drivers. The project facilitates the ability of a wide audience of people who use HDSAM and HDRSAM to provide input to DOE so that the agency knows where to focus future DOE projects so that those projects can resolve challenges identified by the user community. For example, the models are "decision-making tools" for the user community to test out advanced low-/no-carbon pathways without investing in plants or informing competitors or suppliers. Additionally, the models cut research and development (R&D) costs for the private sector since those stakeholders can run the models, without investing in technologies and systems, to pre-determine the best opportunities for their firms. Those stakeholders can also minimize the risk of not knowing about the costs of particular hydrogen production and hydrogen refueling investments. The presenter explained that the models provide predictive tools that help the user community gain traction on their advanced applications for hydrogen and fuel cells, another stated goal of DOE. Those who use the models and provide results via an input form to DOE can assist the agency with its plan to focus on areas of research that are needed for the more advanced hydrogen technologies to gain traction. DOE has stated in this Annual Merit Review (AMR), as well as in previous AMRs, that the agency wants help and input on its projects, and HDSAM and HDRSAM provide ways for the user community to learn of the opportunities and pitfalls of hydrogen and fuel cell projects, which they can then report to DOE in a standardized approach. Those who use the models can also provide feedback to DOE on how to make the models better in terms of usability.

- The preliminary findings and costs associated with low-pressure liquid hydrogen (LH2) appear promising. These could drastically reduce the fueling infrastructure capital and operational costs, including those for high-flow fueling, which requires large quantities of precooled hydrogen for back-to-back fueling. However, understanding the state of the art, availability of LH2, and system reliability will be crucial to the feasibility of these approaches.
- The project provides analysis that is useful for companies aiming to understand the hydrogen pathway options, some of which are not intuitive. The barriers are clearly stated, particularly with regard to obtaining accurate data. Despite the attempted validation work with industry, it is still not clear that this information is accurate since companies are likely to hold certain information proprietary. The project would benefit from clear objectives that can flow down from the overall goal. It is not clear what sets the individual tasks in a given year. For such a long-term project, charting out objectives and pathways over multiple years would provide a better roadmap and a way to gauge progress.

Question 3: Collaboration and coordination

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

- When explaining how the team collaborates and coordinates with other institutions in the area of hydrogen and FCEV technology feasibility, the presenter mentioned that non-disclosure agreements are signed, as needed. This approach builds trust and confidence. When asked about the partners already participating in this project, the speaker mentioned the U.S. DRIVE Hydrogen Interface Taskforce (H2IT) and ANL's Autonomie Team, among others. When asked about hydrogen blending in natural gas pipelines and whether the models will be applied to this area, the speaker mentioned coordination with the related DOE consortia, which are focused on pipelines. The presenter convinced the audience that the team does not work in a vacuum but rather relies on input from others in the public and private sectors. The presenter described the benefits of the collaboration and coordination in highly practical terms: "We do this with others." This message instilled credibility in the 2021 AMR audience.
- A comment was made that there is industry input, but it would be helpful if more information could be shared regarding industry participants to better answer this question. Some of this information is better supplied from individual sources rather than from organizations where it might get watered down and become more general.
- The collaboration relies heavily on literature and published work but could benefit from direct end-user input. Unfortunately, much of the data available for high-capacity onboard storage are for buses with 350 bar systems, which are much less technically challenging and require less precooling than 700 bar, Type 4 tanks. A great portion of this work could inform the anticipated deployment of high-capacity HD HRS. More engagement and collaboration with key stakeholders could help direct this work toward current challenges with HD fueling infrastructure.

Question 4: Relevance/potential impact

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

- This project advances DOE goals to provide R&D that supports HD applications for fuel cells. The degree to which this project advances the DOE Hydrogen Program (the Program) is very high. This project provides comparisons of well-to-wheel GHG emissions of different hydrogen production technologies, including varying blends of hydrogen produced via steam methane reformation, and compares these to diesel used in HD fuel cell applications. Industry is currently attempting to conduct the same comparisons for investment decisions. When describing the outcome and evaluations of using the models, the presenter showed side-by-side comparisons that evaluated investments in the technologies for hydrogen production systems. The presenter demonstrated how one can remove the guesswork from investment decisions. As in the private sector—and most likely before the private sector—the DOE commitment to support the related R&D can focus on where the hydrogen technology/systems are predictably most beneficial. Both public- and private-sector focus and investment can result in an informed plan. This project advances DOE's stated Program goal, and those supported by its subprograms, of $1.5/kg of hydrogen in one decade by helping the overall agency decide where to focus its commitment on R&D in hydrogen and fuel cells. These likely represent the goals of industry, also, since DOE and industry use the same models and share their results.

- This project clearly aligns with the goals of the Program. The outcome from this project will directly inform industry on multiple HRS design options and further assess the impact and GHG reduction of various production paths.
- The project is aligned with DOE objectives to demonstrate the effectiveness of hydrogen to lower GHG emissions.

Question 5: Proposed future work

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

- The presenter explained the iterative process, which includes heavy stakeholder participation in determining the selection of future technologies on which to focus. The presenter explained the team's plan for future work in terms of the continual ("organic") process the project uses to address new technologies and to integrate those technologies into models. For example, HDRSAM for MD and HD FCEVs was developed only three to four years ago, and presently, the model is expanded to include new technologies such as LH2 onboard storage, including its impact on fueling costs. The MD and HD FCEVs were only starting to emerge three to four years ago, yet the model included them when they were still emerging. An argument could be made that DOE's future-proofing of this model could have contributed to the emergence of that industry. For a second example, many industry stakeholders are interested in hydrogen storage on board vehicles, and the presenter covered the influence of the hydrogen supply method (gaseous or liquid) on the cost of onboard storage. DOE may also need to expand the model to examine future applications such as rail, aviation, and marine, as was mentioned. Additionally, issues related to large-scale hydrogen export terminals will need to be added to the model, as this is another area of stakeholder interest. The presenter mentioned that the research team is in the process of adding hydrogen pathways in the GREET model for rail applications that use hydrogen fuel cells, with funding from the U.S. Department of Transportation. Additionally, the energy associated with hydrogen delivery and fueling is needed. The presenter mentioned that that can be sourced from expanding HDSAM/HDRSAM to incorporate those data and that information in GREET. The presenter explained that the team members either develop new models or expand existing models as the need arises to address new and emerging technologies and applications that are not in the current models. The previous scope of the models and the need to update for new and emerging issues was presented.
- The future work is appropriate, with the exception of further work on the economics of LH2. The analysis to date is sufficient. It would be good to see a longer-term horizon of future work and what constitutes the remaining 30% of the project. The costs listed on slide 13 are thematically correct, but it is surprising to see that gaseous hydrogen delivery does not get less costly than LH2 out to 500 km. This seems excessive, which makes me question the basis of the cost model.
- The future work could expand on more challenging fueling profiles. The 350 bar and Category D fueling scenarios are relatively limited. A more aggressive look at high-flow-fueling challenges can best inform industry on associated costs of high-flow fueling. In addition, a more detailed assessment of liquefaction costs at scale, impact on the environment, and available capacity and LH2 outlook could be of value, as HD vehicles are starting to roll out and are expected to displace diesel trucks in the next 15 years.

Project strengths:

- The overall project strengths include its contribution to the goals that Ned Stetson presented at the 2021 AMR, when he gave an overview of the cost drivers on which the projects focus, i.e., production, components, storage, and fueling stations. This presenter explained how the models can be used over a wide range of scenarios and that, when the scenarios become refined with experience, they can focus specifically in areas of importance. Evaluations can be run affordably prior to investment and commitment of time and money. This pre-planning through modeling saves resources, which drives down costs. Another strength of the models presented is their usefulness in evaluating the costs of HD FCEV fleets. Much of the cost of such fleets remains important in the investment decisions, such as which station capacity to use, which size of refueling components to use, and what really influences the overall cost of a fleet operation.

The answer to the latter could be the fueling cost, the station capital expenses, operating costs, energy costs, or cash flow.

- This project is working in an area of high importance for the successful rollout of hydrogen. Understanding the economics and demonstrating the best pathways will be helpful to those considering this technology. The future work proposed is all pertinent, especially the fueling protocol work for MD and HD vehicles. The material generated is useful for setting policy and goals elsewhere within DOE and at a high level.
- This project team is well-known in the field. This team has demonstrated knowledge and capabilities for this project to be successful and impactful; the assessment tools developed by ANL, the insight from U.S. Drive, and the direct input from industry are critical to the output of this project.

Project weaknesses:

- It would help if there were a clear pathway for this analysis to potentially affect codes and standards (C&S). It was mentioned that sometimes fueling protocols can be conservative, but the project should evaluate how it might influence the C&S in a more economical direction. The information and details will not likely be useful to industry at a detailed level to make decisions. The market will decide the successful pathways based on real-world economics. The project needs a feedback loop after several years to understand the accuracy of its analysis. It is time to chart a pathway to completion of the project over the next several years and then initiate a new project(s) as needed, with better-defined goals and objectives.
- From a broad perspective, the models appear to take a long time for users to learn how to use them adequately, although it appears possible to use the models without any background or training, and the investment in learning appears to be justified since the investments in hydrogen and fuel cells are typically high. Perhaps this is an overall project weakness. It would probably be useful to add comments to the presentation of the project about the learning curve for effectively running the models, recommended training for running the models, and online training/coursework that is available to start to learn how to use the models. Additionally, the models could possibly be enhanced with artificial intelligence to make them easier to run and even more “automatic.” If the models become easier to use, perhaps they could become more influential and drive the cost of hydrogen production and use down because questions about the value chain, GHG reduction, and the breadth of the opportunity to use hydrogen can be addressed.
- This project has strong partners and expertise. The project scope, however, could benefit by expanding into real-time challenges with HD fueling and high-flow infrastructure.

Recommendations for additions/deletions to project scope:

- It is recommended that information be provided on the potential to reduce (1) the cost due to the high level of expertise presumably needed to run the models and (2) the level of expertise required to run the models. In the agency in which this reviewer works, staff read the output from these models and use that output but do not typically invest the time to learn how to use the models, per se. It seems better to be self-reliant and learn how to use the models. Staff have suggested adding these reductions to the project scope. It is also recommended that time be put into the graphics to simplify them and to connect them directly to the areas of focus presented by Ned Stetson at the 2021 AMR. These recommendations do not take away from the excellent and outstanding ratings given here to the presenter and the work of the presenter. This work is invaluable and helps many people.
- The LH2 work is interesting, but it is not clear that there is much benefit to pursuing this further. Companies are building additional LH2 plants, including some with green pathways. This project will not directly influence those plans.
- The project scope could benefit by expanding into real-time challenges with HD fueling and high-flow fueling infrastructure.

Project #IN-026: Tailoring Composition and Deformation Modes at the Microstructural Level for Next-Generation Low-Cost, High-Strength Austenitic Stainless Steels

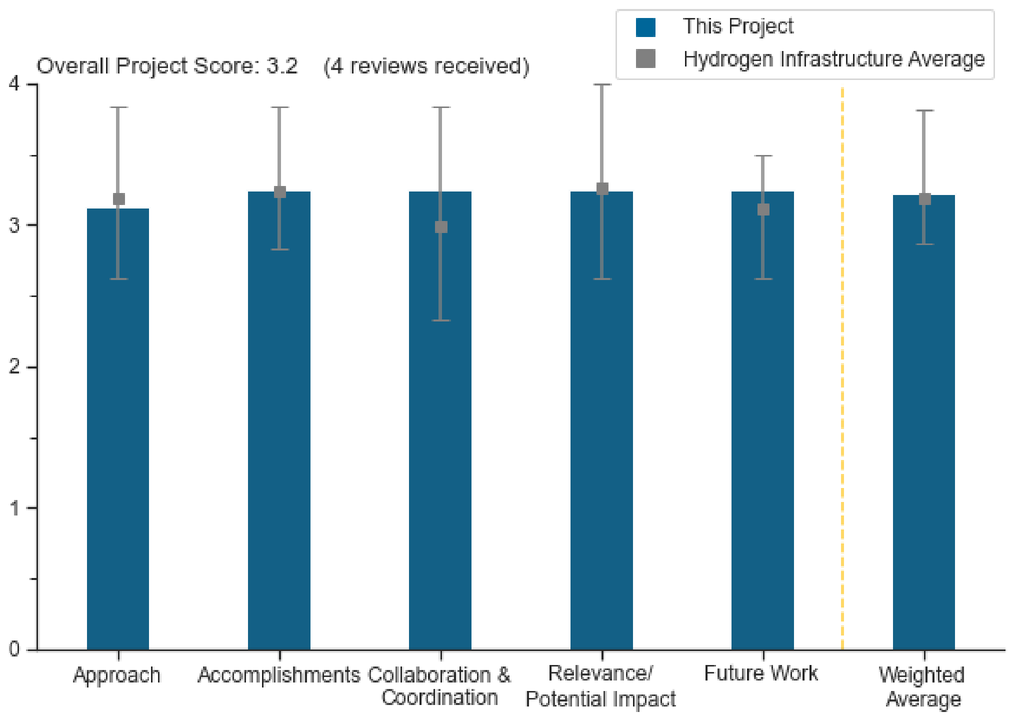
Petros Sofronis, University of Illinois Urbana–Champaign

DOE Contract #	DE-EE0008832
Start and End Dates	10/1/2019
Partners/Collaborators	Swagelok, Linde/Praxair, Arcelor-Mittal, Massachusetts Institute of Technology, Sandia National Laboratories, Lawrence Livermore National Laboratory, Oak Ridge National Laboratory, Argonne National Laboratory
Barriers Addressed	• Gaseous hydrogen storage and tube trailer delivery costs

Project Goal and Brief Summary

This project aims to establish detailed relationships between the chemical composition of alloys and localized plasticity caused by exposure to hydrogen. The results could enable the design of new, cost-effective alloys resistant to hydrogen embrittlement (HE). These materials could be used to construct and deploy economical hydrogen fuel infrastructure.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- Based upon the oral presentation and the slides, it is fair to say that the primary goal of the project, as worded by the project team, is establishing a mechanistic connection between local chemistry and local deformation behavior, in combination with the appropriate technoeconomic analysis that will enable design of cost-effective hydrogen-resistant alloys or, to put it a different way, to design more cost-effective austenitic alloys by tailoring microstructure at the local level. Neither the oral presentation nor the slides were explicitly clear in stating the project's primary outcomes and impacts. The above will be used as this reviewer's understanding relative to this review. If the understanding of the outcomes and impacts are incorrect, then this commentary may also be off-base. The approach to the work seems sound. The research team is first attempting to understand the local ordering of existing microstructure at a length scale below that which is typically probed. The research team is then performing atomistic and continuum-level models to predict the effects of chemical constituent ordering on dislocation dynamics. The research team then proposes to perform in situ experiments to elucidate these potential interactions. Finally, the research team intends to leverage the overarching team's metallurgical and metal processing expertise to create tailored microstructures that are more hydrogen-damage-resistant. Overall, the approach is sound. Based upon the recent literature, there is uncertainty about any in situ neutron or synchrotron experiments that will provide the measurement data that the team needs in order to be successful.

- The goal of the study is the development of a new HE-resistant material via compositional modifications. The work to date has identified highly novel clustering short-range order (SRO), and modeling suggests that this could affect the deformation in the absence of hydrogen and the segregation behavior of hydrogen. The use of the characterization and modeling approaches to address these concerns is world-class. Investigation of this novel phenomenon is important to understanding the grain-scale plastic deformation behavior, with possible implications on materials design. The only drawback is that the extent to which these SROs will actually affect the fracture and HE behavior remains uncertain. It is possible that they do not. This does not mean that the question should not be asked and investigated. However, the fact that it is possible that there is no clear link between these features and fracture/HE is a risk factor that should be acknowledged.
- The approach uses well-established methods and tools to identify candidate materials and explore chemical homogeneity and deformation modes. The knowledge gained from this approach addresses the cost and reliability of hydrogen transport and storage infrastructure if the work continues to reveal useful scientific understanding of the mechanisms governing hydrogen damage mechanisms. The work is very exploratory and does not appear to have multiple paths to overcoming scientific barriers. The inclusion of technoeconomic analysis is critical to the work's success.
- The cost reduction target was not presented. Perhaps it is based more on lower alloy cost with the addition of Mn, which is not likely to lead to significant gains, or on higher strength and reduced wall thicknesses, which is possibly more likely. The approach to the alloy development matrix is good, but elements/ranges may need to be broadened. The project should not put too much stock in the data comparing the behavior of the commercial-grade materials, Nimonic 40 versus 316, but should focus on modifications to 316 or 304 that may improve strength/increase HE resistance.

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 of the role of local compositional segregation as a potentially key contributor to the behavior of materials, when exposed to hydrogen, could be a critical advantage in designing lower-cost metal alloys with increased hydrogen damage resistance. The project is making excellent progress.
- The principal investigators (PIs) have made impressive progress, given COVID-19 restrictions. The emphasis on modeling/characterization is well done, as other paths for the project were hindered.
- Assuming the reviewer's understanding of the project goals and the work elucidated in the slides, the project has not been very fruitful, given the 16-month period of performance as of slide creation (February 21, 2021). The reviewer understands the effects of COVID-19, as the reviewer also had three federally funded grants occurring during fiscal year 2019. Even with COVID-19, the characterization of the existing metals' local compositional segregation, the atomistic modeling, and the continuum-level modeling fell short of what would have been possible.
- More progress could have been made on novel alloy development and less on characterizing commercial alloys.

Question 3: Collaboration and coordination

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

- The PIs list an excellent bevy of collaborators. In the talk, the collaboration with Kyushu University was highlighted. While opportunity is there, details of the collaboration with other partners were not provided. This will presumably be invigorated as the pandemic constraints are alleviated.
- The project team should be commended for going out and locating a new, unfunded partner to support project progress.
- Collaborators and their roles are clearly defined, and the project is making positive headway, but the actual role of collaborators was not evident in the review. It seems like the work is performed in-house at the University of Illinois, with limited interaction with collaborators. The transition from modeling and microstructure characterization to Milestones 1.1 and 1.3 would be a natural place for increased collaboration but was only touched on during the review.
- A project like this should have very close connections to a raw material provider and end user. If this is the case, it was not clearly presented during the review.

Question 4: Relevance/potential impact

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

- Establishing a mechanistic connection between local chemistry and local deformation behavior, in combination with the appropriate technoeconomic analysis, will enable the design of cost-effective hydrogen-resistant alloys that will have impacts on both the cost and reliability of hydrogen infrastructure. A key to realizing the potential impact will be how well the knowledge/technology can be transferred to stakeholders. This was not well-described during the review.
- Without any doubt, the successful completion of this project is relevant to progressing hydrogen's use as a clean energy carrier and, therefore, would have very high impact. As mentioned prior, the team's ability to perform measurements that sufficiently support what may come from the combined atomistic/continuum modeling is questionable. The measurement techniques to track dislocation mobility/accumulation in a bulk material, in either a hydrogen or laboratory atmosphere, in situ, are currently lacking.
- The topic area and new findings are certainly high-impact in the field of HE in stainless steel relevant to the hydrogen applications. The foundational knowledge gained will be very important. However, it remains uncertain whether the SRO is relevant to the deformation, fracture, and/or HE properties. As such, there is risk involved as to the direct relevance of detailed interrogation of these features to the design of HE-resistant alloys.
- This reviewer is not familiar with specifics of DOE Hydrogen Program goals, but it is unclear what the cost reduction target is and how much would it save if the "hydrogen economy" were to take off.

Question 5: Proposed future work

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

- The proposed work path is clear, logical, and state of the art to further investigating the impact of SROs on HE behavior. The linkage between the SRO work and the alloy design is not fully clear (as would be expected, owing to the novelty of the SRO observations). Linking the atomistic modeling and characterization to continuum approaches is clearly challenging, and there was not time during the talk for elaboration on how this future effort will be achieved.
- The future work should be focused on novel alloy additions that change the microstructure and lead to better properties or lower costs.
- Continued integration of scientific discovery with technoeconomic analysis will increase the likelihood of far-reaching impacts. The fact that the proposed beam line work is not likely to have the needed resolution may not be the best use of funding.

- The team's ability to perform measurements that sufficiently support what may come from the combined atomistic/continuum modeling is questionable. The measurement techniques to track dislocation mobility/accumulation in a bulk material, in either a hydrogen or laboratory atmosphere, in situ, are currently lacking.

Project strengths:

- The project has world-class characterization and modeling; identification of novel SRO clustering; a logically outlined, state-of-the-art framework to interrogate the impact of SROs on dislocation interaction and hydrogen distribution; clear plans to understand the impact of SRO on HE; and good collaboration with Kyushu University on alloy development.
- The project is making excellent progress in identifying new insights into the mechanisms that control hydrogen damage in materials. The insights may provide a new lever for resining low-cost reliable materials.
- The collection of people in the project is probably the project's primary strength at this point.
- The project topic is interesting and worth pursuing. It needs more industrial collaboration.

Project weaknesses:

- The unclear relationship between SROs and fracture/HE is a risk factor with regard to whether tailoring this microstructure feature will have an impact on the desired property. The in-depth characterization/modeling is current decoupled from the material design. (To be clear, the material design strategy is still solid but is currently proceeding decoupled from and in parallel with the main research effort.)
- The work does not appear to have multiple paths to overcoming scientific barriers. The extent of collaboration needed for success is not demonstrated in the review, and a careful consideration of knowledge/technology mechanisms is not evident.
- There is concern that, if the atomistic and continuum-level models cannot be validated by use of measurements, the work will not have the impact desired.
- The first year focused too much on the limited scope of work and should have focused more on alloy development.

Recommendations for additions/deletions to project scope:

- With the plan the PIs have put forth, increasing the collaborations will occur naturally. Right now, there is a focus on the dislocation interaction with the SRO and the impact on hydrogen segregation. It seems like there remains room to interrogate and link the failure behavior. This is likely outside the scope of the project; however, it would be the next step in the path to understanding the role of these SROs.
- Considerable time and effort should be expended, sooner rather than later, on determining an appropriate measurement technique to validate the modeling results.
- The project should look at the effect of cold work on the alloy and properties with different elemental additions and the effect of nitrogen on alloys and precipitates.

Project #IN-029: Reducing the Cost of Fatigue Crack Growth Testing for Storage Vessel Steels in Hydrogen Gas

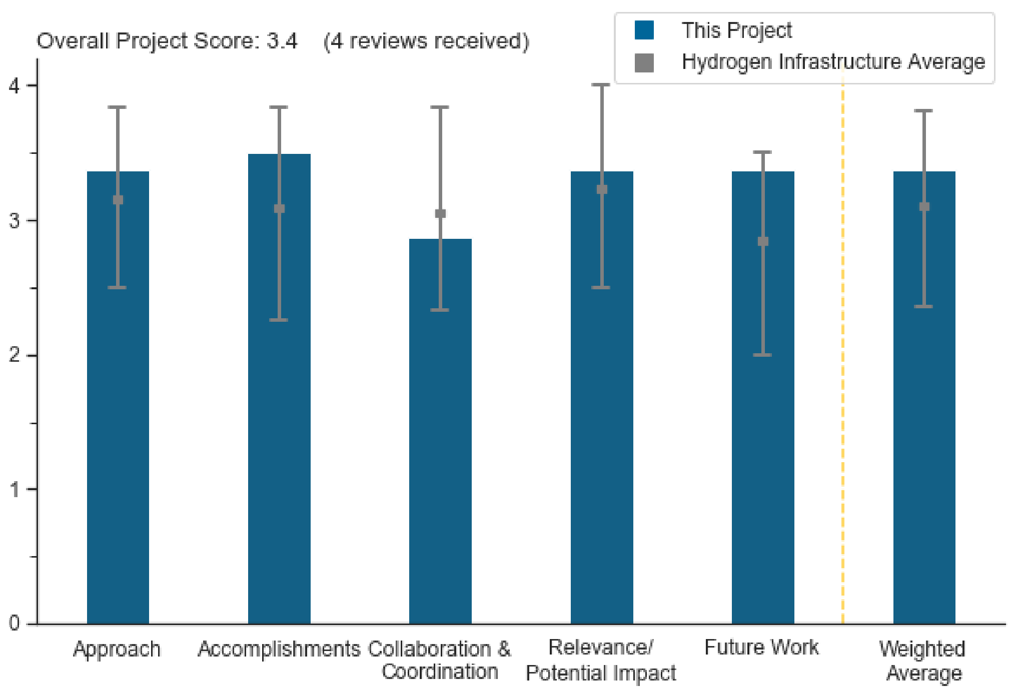
Kevin Nibur, Hy-Performance Materials Testing, LLC

DOE Contract #	DE-EE0008829
Start and End Dates	3/24/2020 to 2/28/2023
Partners/Collaborators	Someday Consulting, LLC, Sandia National Laboratories (via the Hydrogen Materials Consortium (H-Mat)
Barriers Addressed	• Permitting

Project Goal and Brief Summary

Hy-Performance Materials Testing, LLC, Someday Consulting, LLC, and the Hydrogen Materials Consortium (H-Mat) are designing efficient and affordable testing to measure fatigue crack growth rate (FCGR) in hydrogen gas storage vessels. The service life of hydrogen storage vessels at fueling stations is dictated by fatigue crack growth, and current FCGR testing methods are time-consuming and expensive. A more cost-effective approach to FCGR measurement would facilitate market adoption of hydrogen storage vessels at fueling stations.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- The approach to the project is systematic and laid out exceptionally well. This is supported by the results that have been produced to date.
- There is a strong systematic approach to addressing a specific technical problem.

- The approach seems logical, but a better understanding of how the proposed activity could affect the overall component development and acceptance process would be helpful. The approach does not seem to be novel, but tying it to acceptance and implementation is important.
- There does not seem to be as much value in reducing test time as increasing capacity and choosing the right tests to get the best answers to questions.

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.

- Project results to date indicate that a considerable reduction in cost to create hydrogen-accelerated fatigue crack growth (HA-FCG) data may be realized as a result of this work. It is very clear that DOE Hydrogen Program objectives are a primary focus of the project team. It is clear that the objectives met to date are far more valuable than the cost of the project to date.
- Progress appears right on schedule, based on the presented plan.
- This project seems to be well on track.
- Good progress has been made. It is not clear if there are any technical hurdles and challenges that will need to be overcome or how this improves the work that has already been done at Sandia National Laboratories.

Question 3: Collaboration and coordination

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

- The project team has a long history of working well together and is progressing well as a team.
- The team has good coordination and collaboration.
- It is not clear that collaborations are ongoing. There was a plan to coordinate with Sandia National Laboratories, but everything presented appeared to have been done in-house. However, the project does not appear to need collaborations at this point.
- The National Institute of Standards and Technology (NIST), Oak Ridge National Laboratory, and industry are not involved in the project. It is unclear what tests industry is pushing for where capacity is constrained.

Question 4: Relevance/potential impact

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

- Roughly 20 years ago, the relevant code bodies (American Society of Mechanical Engineers [ASME] B31.12 in particular) endeavored to modify the design codes specific to hydrogen. Specifically, they needed to make the codes more realistic relative to combined hydrogen mechanical loading damage mechanisms. Ultimately, it took Sandia National Laboratories and NIST on the order of 10 years to create a sufficient number of data to support a modification to the B31.12 code to allow for higher-strength materials and more realistic design requirements. The primary barrier to shipping and storing hydrogen in the United States in mass is twofold: reduction in the cost of hydrogen pipeline and storage infrastructure and the private sector's willingness to invest in the infrastructure relative to a risk posture that is supported by data. This project sets out to define a testing protocol that enables the creation of quality HA-FCG data at far faster rates than what can be performed currently. Given that data collection is the rate-limiting step to overcoming the two barriers mentioned above, this project will have exceptional impact.
- Updating testing standards to allow quicker and cheaper testing would reduce cost burdens to companies that need this type of testing and time burdens on the few facilities that can perform this type of testing.
- If an American Society for Testing and Materials standard is developed and ASME adopts it as well, it could open a possible lower-cost qualification pathway for hydrogen component developers.
- Added testing capacity from a commercial entity is a good step.

Question 5: Proposed future work

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

- Proposed future work for continuing the primary project looks appropriate and feasible. Proposed interactions with code committees look fine and presumably will be appropriately timed. Proposed future work that includes Sandia National Laboratories is rather vague.
- The proposed future work is aligned well with what the reviewer would expect.
- The project should examine crack closure more closely, especially for alloys such as aluminum that are very sensitive to oxide-induced closure.

Project strengths:

- This is exceptional work to date. The project has already shown results that will be greatly impactful to the hydrogen community at large. Strengths of the project are likely bolstered by the project team assembled.
- Having commercial test facilities for hydrogen materials testing is important for the hydrogen economy to evolve.
- There is a strong, systematic, technical approach to answer technical problems, and it appears to have a strong foundation for project success.

Project weaknesses:

- In terms of reaching overall goals, there is limited benefit to reducing the testing time.
- The project is light on scientific understanding, though that appears to be through design of the project scope.
- There are no noteworthy weaknesses.

Recommendations for additions/deletions to project scope:

- There are no recommendations at this time.

Project #IN-030: Micro-Mechanically Guided High-Throughput Alloy Design Exploration toward Metastability-Induced Hydrogen Embrittlement Resistance

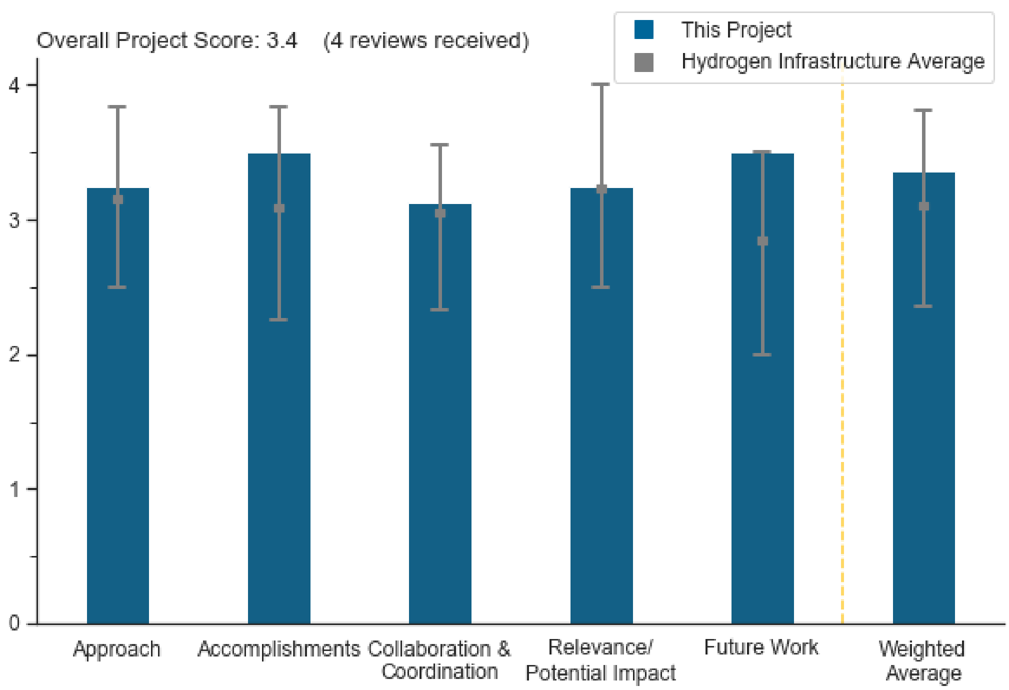
C. Cem Tasan, Massachusetts Institute of Technology

DOE Contract #	DE-EE0008830
Start and End Dates	4/1/2020 to 3/31/2023
Partners/Collaborators	Harvard University, Allegheny Technologies Incorporated
Barriers Addressed	• Gaseous hydrogen storage and tube trailer delivery costs • Materials of construction

Project Goal and Brief Summary

This project aims to develop a novel, high-throughput compositional and microstructural screening approach to developing new alloys with superior hydrogen embrittlement (HE) resistance. The research will focus on using metastability to enhance resistance. If successful, the project will provide novel testing methods that allow researchers to screen the hydrogen-related physical properties of multiple alloys simultaneously, thereby drastically reducing the research and development period for new alloy development.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Infrastructure projects.

Question 1: Approach to performing the work

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

- The approach leverages advances in high-throughput materials development methods to explore the materials space and identify candidate materials with increased hydrogen-degradation resistance. The team is clear about the advantages and limitations of different aspects of the methods, specifically using small-scale techniques to probe the response of bulk-scale materials. The plan to use small-scale experiments to identify areas to explore with modeling and bulk-scale approaches is likely to provide new insights at a much faster pace than conventional approaches. The approach is sharply focused on enabling safe, lower-cost containment technologies to address hydrogen storage and delivery barriers.

- Alloy development on thin films with gradient chemistries is a novel and interesting concept.
- The link between the identified knowledge gap and goal and the proposed high-entropy alloy (HEA) concept is reasonable. The use of metastable HEAs can be a solution in this space, but the scalability, production, cost, and other important factors are not adequately considered. The lack of investigation of other properties is concerning but will likely occur in future efforts. There are sophisticated methods and data science approaches to address the phase-stability issue, but the authors should comment on the trade-offs of this with other properties. The technical approach is reasonable and state of the art for design, fabrication, and quick screening. However, there are several potential pitfalls in extrapolating beyond the initial screening stage.
- The project objectives are clearly presented. However, the detailed hydrogen technology barriers motivating the objectives are not described. For example, it is unclear whether the technical approach is designed to address the high cost of incumbent structural metals in hydrogen infrastructure, such as 316 stainless steel. One of the stated project goals is to develop new alloys with superior HE resistance, but HE resistance is not a shortcoming of high-nickel 316 stainless steel. Rather, it is the high cost of 316 stainless steel that renders it impractical for widespread deployment in hydrogen infrastructure.

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 team has surpassed all of the Year 1 project milestones. The calculation of phase-diagram-based high-throughput alloy screening has led to the fabrication and testing of materials for γ phase metastability. The demonstration of combinatorial-style co-sputtering, followed by post-deposition heat treatments, to fabricate materials and control microstructure is impressive within the timeframe (especially considering the pandemic). The design and fabrication of an in situ hydrogen charging setup were completed and are in the process of being validated. Overall, the Year 1 accomplishments are excellent and promise to serve as the basis for future work.
- This project seems to be well on track. Much of the budget appears to have been spent in the early phases, presumably for equipment purchases.
- Solid progress has been made on this project with regard to modeling and design.
- The presentation is well-crafted, so it is evident that the technical accomplishments represent progress toward achieving the stated project goals. However, the project goals are not directly oriented toward resolving critical barriers in the deployment of hydrogen technology, e.g., the high cost of incumbent alloys such as 316 stainless steel.

Question 3: Collaboration and coordination

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

- The collaboration and coordination for the project are excellent. The project is engaged with appropriate expertise at various scales and between experimental and computational aspects of the project.
- The relationships with external partners Sandia National Laboratories and Allegheny Technologies Incorporated are described well, and the roles of these partners appear productive for the project. The goals of the project could be honed toward resolving technology barriers by including industry partners that are stakeholders in hydrogen infrastructure deployment.
- This project could consider an industrial partner, but the focus should be on developing the best compositional spaces for detailed commercial exploration. Therefore, it is not necessary to involve industry in this sort of project.

- The academic collaboration is solid; however, the applicability/relevance of the project will suffer without further engagement to understand the cost, scalability, and production considerations. Comments during the talk suggest that this is on the radar of the principal investigator (PI) but has yet to be realized.

Question 4: Relevance/potential impact

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

- This is a higher-risk project that could provide a significant new approach to materials design in general and specifically in support of DOE Hydrogen Program objectives. The length of time and cost to develop new alloys for hydrogen have been top barriers to realizing a hydrogen economy for years. This approach, if successful, could significantly advance progress to reach DOE objectives.
- The approach could be very beneficial for the Hydrogen Program, but it also has far-reaching potential in many other industries and applications.
- The foundational work in this study will inform the efficacy of the HEA approach to HE resistance. The atomistic modeling and other outputs will be of great importance to the academic community. However, the potential for this effort to result in actionable progress toward a material that can be applied is very low.
- With its currently stated goals, it is not clear that this project will have an impact on issues that need to be resolved to advance hydrogen technology. The project focuses on enhancing HE resistance of structural alloys, but this material characteristic by itself is not an impediment in the deployment of hydrogen technology. Rather, the HE resistance of cost-effective structural metals is an issue that must be addressed to enable the deployment of hydrogen infrastructure.

Question 5: Proposed future work

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

- The proposed work will achieve the scientific goals of the study with novel and state-of-the-art methods. The rapid screening approach is novel and well-conceived. The PI expressed plans for more detailed testing during the talk.
- The proposed future work represents a progression from the current accomplishments and is targeted toward achieving the stated goals of the project.
- The proposed work, looking at sputter-deposited materials, is appropriate and drives progress toward addressing HE barriers. It is concerning that the proposed work included no mention of how the work will extend to the bulk scale. This is a key component of the project and should be under consideration in the near future.
- The next phase of this project is more focused on alloy development, which should be more exciting than equipment/procedure development.

Project strengths:

- This project combines and leverages recent advances in materials design to address the challenge of hydrogen materials compatibility. The approach has the potential to have an impact on the materials used for hydrogen storage and transportation, but the approach could also extend to any other hydrogen application areas where metallic materials are employed. The team is appropriate, and its roles are clear. Year 1 progress was impressive and has set the stage for the rest of the project. The proposed future work addresses key challenges for the small-scale experimental and computational piece of the project.
- State-of-the-art modeling approaches, novel screening, and materials design and fabrication are high-throughput and unique. The scientific quality is excellent.
- The central concept of this project, i.e., integrated high-throughput alloy design, is sound and could lead to innovations in structural metals designed for hydrogen service.
- This project has a novel idea for alloy development and a good overall approach.

Project weaknesses:

- The primary weakness of this project is its unqualified goal of developing new alloys with superior HE resistance. The project must identify incumbent materials with shortcomings that are hindering the deployment of hydrogen infrastructure. For example, the primary shortcoming of the incumbent 316 stainless steel is its high cost. In this case, design goals for alternative alloys are twofold: (1) replicating the high HE resistance of 316 stainless steel and (2) lowering costs relative to 316 stainless steel.
- This is a multi-property assessment, with linkage to actionable engineering materials. Thus far, there is a need for more consideration of cost, scalability, and production.
- The microstructure effects of the different compositions should be considered. It is microstructure that affects the properties. Composition is used just to change the microstructure. The project should address scale-up in processing from depositing to bulk processing.
- The proposed future work, as stated in the review, does not include a clear pathway to bulk-scale experimental work.

Recommendations for additions/deletions to project scope:

- The project should include details of the next-level characterization of HE behavior and considerations of multi-properties.
- It is recommended that the project include industry stakeholders in hydrogen technology as project partners.

STORAGE

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

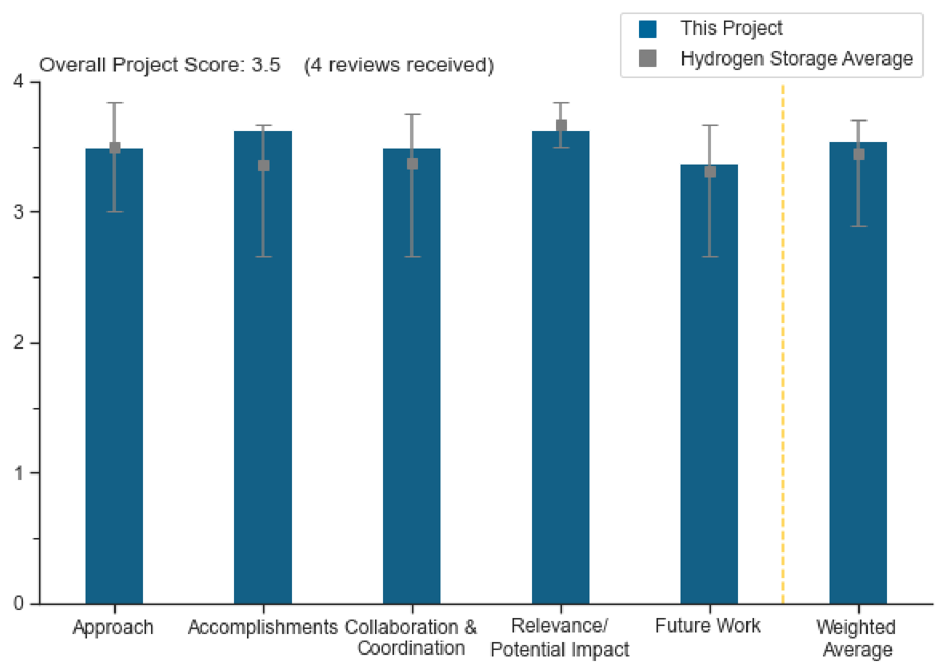
Rajesh Ahluwalia, Argonne National Laboratory

DOE Contract #	4.4.0.2
Start and End Dates	10/1/2009
Partners/Collaborators	Hydrogen Materials Advanced Research Consortium (HyMARC), Pacific Northwest National Laboratory, Lawrence Berkeley National Laboratory, Hydrogen Interface Taskforce, Argonne National Laboratory – Hydrogen Analysis (H2A) model, Argonne National Laboratory – Hydrogen Delivery Scenario Analysis Model, Hydrogen Materials Consortium, U.S. Army Tank Automotive Research, Development and Engineering Center, Lawrence Livermore National Laboratory, Ford Motor Company, Strategic Analysis, Inc.
Barriers Addressed	• System weight and volume • System cost • Efficiency • Charging/discharging rates • Thermal management • Lifecycle assessments

Project Goal and Brief Summary

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.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Storage projects.

Question 1: Approach to performing the work

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

- This is a continuing project, subject to annual direction and guidance from DOE. This important technical effort focuses on systems-level analysis of DOE hydrogen storage needs and options. The 2020–2021 work addresses three technology areas that have significant impact on the successful development of hydrogen-based options for future energy systems: (1) pathway analysis for (hydrogen) liquid carriers, (2) transmission costs for liquid hydrogen (LH2) transport, and (3) hydrogen storage for renewable energy systems. The analysis methodologies employed in all three areas have facilitated meaningful results and conclusions to be obtained on the project. The analyses address critical barriers and provide a compelling and useful way to assess and compare hydrogen transport alternatives, as well as requirements, costs, and scenarios for hydrogen storage in renewable energy applications.
- The principal investigator (PI) uses a thorough and consistent approach using the best available data within the DOE network of programs.
- The presentation was excellent, with plenty of information packed into 20 minutes. The scope of the project is large enough that getting into the details would take significantly more time.
- The project developed systematic approaches for the hydrogen supply chain pathway analysis by hydrogen liquid carriers with different transportation methods. Regarding this pathway analysis, if the liquid carrier is re-used after carrier decomposition, the transportation of the re-used carrier back to Texas should be considered.

Question 2: Accomplishments and progress

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

- The project provides a comprehensive analysis of hydrogen transport options, including technoeconomic comparisons of different hydrogen carrier systems for both stationary and transport applications. This work directly complements the ongoing materials work being conducted in the Hydrogen Materials Advanced Research Consortium (HyMARC) and other DOE projects, and it provides useful tools that help guide the materials development efforts. The Argonne National Laboratory (ANL) team has generated an impressive amount of detailed information that allows liquid carrier transport options (e.g., pipelines, rail, tanker ships) to be compared and correlated, and costs to be assessed. Likewise, useful new information is provided that quantifies the impact of different hydrogen storage approaches for off-load storage, backup power, and load leveling in renewable energy power generation applications. The results of the analysis in all of these areas are important for developing optimized storage and transport systems, in addition to providing a better understanding of how hydrogen systems compare with other (incumbent) storage technologies. On a side note, it is unclear why, in the two text boxes on slide 13, capital expenditures and levelized cost of electricity were expressed as $/kW instead of $/kWh.
- This project completed a good evaluation of the hydrogen transmission paths and costs. The scope of evaluation appears to be very thorough. It would be nice to see the cost for hydrogenation and dehydrogenation for each of the carriers in the charts. Also, cost comparison to energy storage methods other than hydrogen would be informative and interesting.
- The PI manages to efficiently communicate a tremendous amount of information in a short presentation. These presentations need to be reviewed afterward to dissect the tremendous amount of data and assumptions made. It is too much to cover in 20 minutes.
- The project demonstrated excellent accomplishments and progress, developed cost correlations for various transportation methods of different hydrogen carriers, and developed cost models for LH2 storage and shipping.

Question 3: Collaboration and coordination

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

- The valuable collaborations with the HyMARC core team and Strategic Analysis, Inc., are evident. The systems analyses being conducted in this project provide a useful complement and adjunct to the foundational studies in HyMARC. Continued close interactions with the HyMARC team will be essential to ensuring that DOE receives a meaningful "end-to-end" examination of hydrogen system status and challenges.
- This PI always pulls in the best available data from the DOE network of projects.
- The collaboration with other institutes looks to be very good. It would be nice if more input from the industry was available in the analyses.
- The project team should consider having more partners from industry to provide validation on the cost assumptions.

Question 4: Relevance/potential impact

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

- Hydrogen-based fuel cells are emerging as important elements in the DOE energy portfolio. The analyses being conducted in this ANL project are vital to understanding how this technology will be implemented on a large scale that is both efficient and economical. This ongoing project is closely aligned with the DOE research, development, and demonstration (RD&D) goals and objectives and continues to have positive impacts on the progress achieved in the DOE Hydrogen Program (the Program).
- Understanding the role of hydrogen and fuel cells in the grid energy system will be crucial for realizing their use and the adoption of renewable energy sources. It is important to conduct this kind of research.
- Understanding hydrogen supply chain and transmission cost is critical for the hydrogen economy as it moves into higher demand. Identifying the costs and opportunities early will help the industry capitalize on the correct technologies.
- The project is very relevant to DOE's current focus, as well as industrial perspectives, and has the potential to advance progress toward DOE RD&D goals and objectives.

Question 5: Proposed future work

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

- The proposed future work follows logically from the technical effort initiated in 2020–2021. The reviewer strongly supports the proposed effort to fully document the results of the hydrogen carriers and bulk storage work.
- The plans are clearly defined and well-planned. The team should consider adding one more study on supply chain development based on utilization of existing hydrogen storage infrastructure, such as hydrogen pipelines and hydrogen storage caverns.
- Heavy-duty fuel cell electric vehicle applications are attracting higher interest from industry. Understanding the cost of storage systems, as well as total cost of ownership (TCO) for vehicles, is important. It is important to consider fuel costs as part of the storage system, as changes in fuel cost become much more significant to TCO for heavy-duty vehicles.
- The next steps are appropriately chosen.

Project strengths:

- The project developed systematic approaches and scenarios for the hydrogen supply chain pathway analysis with various transportation methods. The project demonstrated excellent accomplishments and progress, developed cost correlations for various transportation methods of different hydrogen carriers, and developed cost models for LH2 storage and shipping. The project is very relevant to DOE's current focus and industrial perspectives and has the potential to advance progress toward DOE RD&D goals and objectives.

- This 2020–2021 project represents a new direction for the ANL analysis team. By focusing on hydrogen transport issues and storage for renewable energy applications, the researchers have expanded their impressive capabilities to address important new areas that underlie the successful development and implementation of hydrogen fuel cell technologies. The results provide an important complement to the ongoing materials and systems development work being conducted elsewhere in the Program.
- The project's strength is in its collaborations using the available data. The PI's experience in this area helps him to avoid making mistakes and choosing poor assumptions, parameters, etc.
- This project provides a good summary of various transmission methods and media and breaks the information down into understandable and actionable data.

Project weaknesses:

- This is a strong project with few deficiencies. The ANL team has demonstrated the ability to move quickly to effectively address new important issues that have impacts on the Program.
- The project can be further improved from the following points:
    - For the study on hydrogen storage for renewable wind and solar plants, in the scenario setting, toluene and methylcyclohexane (MCH) are considered as the hydrogen storage methods. Other hydrogen storage options should be considered, and more explanation should be provided on the cost assumptions of various transportation methods, such as pipelines, trains, and ships.
    - For Task 1, pathway analysis with transmission by trains, the project team might consider adding the LH2 as the reference, similar to LH2 ships. For the LH2 export study, the handling of boil-off gas during the trip is not clear; an explanation would be appreciated. Also, the team should consider the scenario using boil-off gas as the propulsion power supply, which may significantly influence the economics. The sensitivity analysis should be added on the hydrogen storage cost analysis for renewable wind and solar plants. For the study on hydrogen storage for renewable wind and solar plants, the study is targeted at 10 MW; a similar study should be carried out at a higher scale to show the effect of scale.
    - The project should include partners from industry to provide validation of the cost assumptions.
- This is not a project weakness, but there is simply too much information presented to be digested in 20 minutes. The PI should strive to provide high-level conclusions and trends up front to allow users to wrap their heads around the context of all that is being presented.
- The scope may be too large to get a good evaluation for each technology and comparison to current incumbent technologies. Significantly more work could be done on hydrogen storage for wind and solar.

Recommendations for additions/deletions to project scope:

- There are two recommendations for addition to the project scope. (1) The PI has stated that new pipelines will be required for hydrogen carrier transport (i.e., retrofitting of existing pipelines is apparently not workable). It will be important to identify infrastructure cost requirements (e.g., scope of the pipeline distribution networks and pipeline production and installation costs) to fully establish the efficacy of implementing the pipeline-based transport approach. (2) It is recommended that the hydrogen storage analysis for renewables include a comparison with existing incumbent storage technologies (grid-scale batteries, solar thermal, pumped hydro, etc.). Such a comparison is needed to fully benchmark the hydrogen storage technology against current storage approaches.
- The project should add sensitivity analysis to scalability of these processes and what the limiting steps are to scale up or down. It would help researchers to determine what storage material characteristics would be useful to allow scaling up or down. This is similar to the intent of the Hydrogen Storage Engineering Center of Excellence.
- Heavy-duty applications are expected to grow significantly over the next few years; identifying barriers to adoption and defining TCO in comparison to diesel will be critical for growth.

Project #ST-100: Hydrogen Storage Cost Analysis

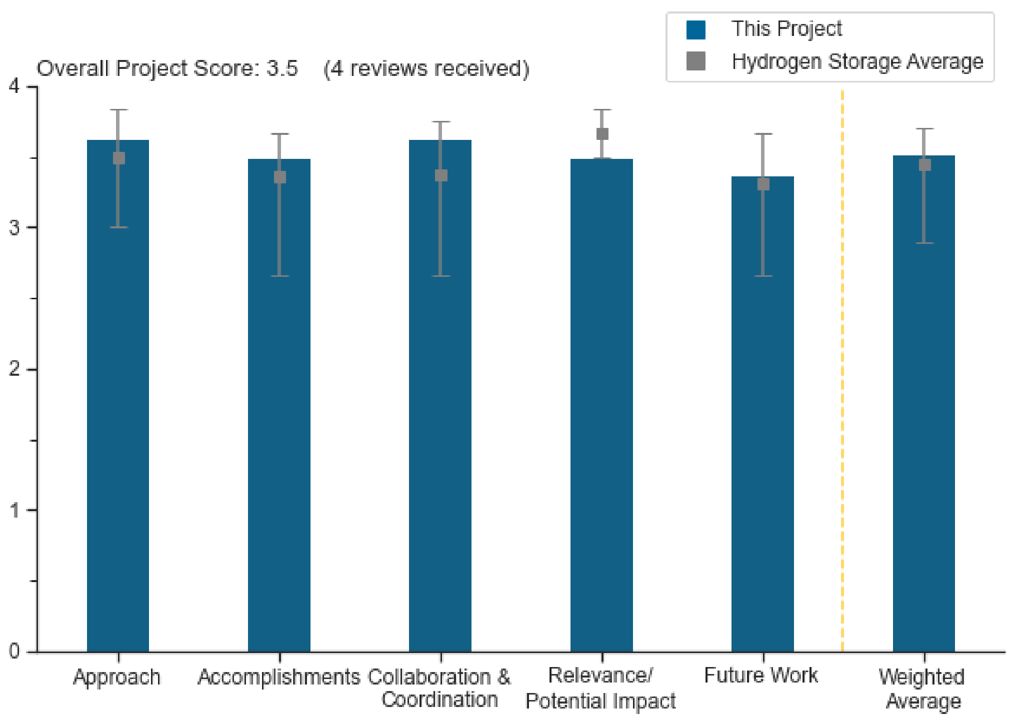
Cassidy Houchins, Strategic Analysis, Inc.

DOE Contract #	DE-EE0007601
Start and End Dates	9/30/2016 to 9/29/2021
Partners/Collaborators	Pacific Northwest National Laboratory, Argonne National Laboratory
Barriers Addressed	• System weight and volume • System cost • System lifecycle assessment

Project Goal and Brief Summary

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

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Storage projects.

Question 1: Approach to performing the work

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

- The DFMA methodology has proven to be a powerful analysis tool for predicting material and process costs and for identifying optimum design and manufacturing pathways. The approach was used effectively in the last year to evaluate and develop hydrogen storage system cost models for Class 8 long-haul systems. In addition, the approach has facilitated a detailed cost analysis for gaseous hydrogen (GH2) and liquid hydrogen (LH2) onsite stationary storage systems and analysis of low-volume 700 bar GH2 storage for light-duty vehicles (LDVs). This work complemented and extended prior work by the Strategic Analysis, Inc. (SA) team on analysis of LDV storage, fuel cell electric buses, and Type 4 natural gas storage systems. The approach adopted in this work also serves as a useful adjunct to the storage and transport system analyses being conducted by Argonne National Laboratory (ANL) and collaborating institutions.

- Compressed and liquid hydrogen are expected to be the primary on-board hydrogen storage systems for FCEV systems for the next several years, so understanding system costs and how these can be reduced is paramount to making FCEVs commercially viable. SA has done a good job of breaking down storage costs and identifying key contributors to costs at high volume.
- This is a very relevant topic and system selection, given the move to heavy-duty FCEVs.
- The project developed a DFMA methodology to track annual cost impact of technology advances. Cost models have been developed for hydrogen storage systems for Class 8 long haul FCEVs, light-duty FCEVs, and hydrogen refueling stations.

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 four areas. (1) The definition and preliminary analysis of a baseline Class 8 long-haul system based on a 700 bar, Type 4 hydrogen storage approach has been completed. A component-level cost breakdown was generated, and carbon fiber used in tank production was shown to dominate the system cost. (2) Analysis of LH2 storage and delivery for Class 8 long-haul vehicles has been completed. Collaboration with ANL showed that balance-of-plant (BOP) components and insulation dominate the system cost. (3) Companion cost and sensitivity analysis for LDVs provided a basis for estimating storage system costs in out-years (2025, 2030). (4) A bottoms-up cost analysis was performed for onsite compressed gas and LH2 storage systems at refueling stations. Cost breakdowns were provided for bulk liquid storage and industrial tube trailer systems used for compressed gas storage. Overall, these analyses and projections provide a useful foundation for more quantitatively assessing storage system costs and evaluating component and system design requirements.
- This principal investigator consistently delivers good progress every year. Presentations are clearly laid out and described.
- The project demonstrated excellent progress toward project objectives.
- For LDV hydrogen storage systems (HSSs), cost is a significant factor in vehicle adoption. Many of the DOE goals are based on passenger vehicles, and the progress toward these goals is good. However, on heavy-duty vehicles (HDVs), hydrogen storage system cost is much less significant, as fuel costs have become much more significant to total cost of ownership. This is outside of the scope of the project and is being addressed by other institutes, but it should be considered to provide a complete picture for HDVs.

Question 3: Collaboration and coordination

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

- Collaborations with national laboratories (ANL and Pacific Northwest National Laboratory) effectively support the technical effort in this project. The collaboration with ANL is especially noteworthy because it provides an overall systems analysis context for the cost and design analysis in this project. More extensive collaborations with industry have also been established during this reporting period. This inspires confidence that a "real world" perspective is being incorporated into the project. Closer attention should be paid to evaluating hybrid ideas involving compressed gas storage in tanks comprising hydrogen storage media. This will necessarily involve establishing more robust collaborations with organizations conducting the materials and system development investigations (e.g., the Hydrogen Materials Advanced Research Consortium [HyMARC] and Pacific Northwest National Laboratory).

- SA has a strong history of working with the full range of industry and DOE tools/collaborators to deliver the best available data on which to build modeling.
- It looks like an excellent collaboration with a good mixture of industry and institutes.
- Page 18 shows an excellent consortium for the work. It would be good to have more industrial partners for system validation.

Question 4: Relevance/potential impact

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

- As in the case of the ANL systems analysis work, the technoeconomic cost projections derived from this project support the DOE objectives, and these projections are useful in providing a more quantitative view of system efficacy and future market penetration. DOE is using this information to inform system and manufacturing development decisions and to project overall costs for stationary and onboard storage systems. Overall, the project is well-aligned with the objectives and goals of the Hydrogen and Fuel Cell Technologies Office.
- Storage systems are a significant cost of the system. A sensitivity analysis as to where costs can be reduced in future systems provides essential guidance to all parties.
- Compressed gaseous and liquid hydrogen storage is going to be the primary method of on-vehicle storage for the next several years, so this work is very important to early adoption of FCEVs.
- The project aligns well with the DOE Hydrogen Program (the Program) and objectives.

Question 5: Proposed future work

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

- HDVs have become of greater interest, as battery vehicles are becoming the better solution for short distances and light vehicles. Storage costs for HDVs and bulk ground storage need to be understood in conjunction with reducing hydrogen fuel cost to a cost level comparable to petroleum fuels.
- The proposed future work on Class 8 long-haul 700 bar hydrogen and LH2 storage systems and LH2 bulk storage is a logical and reasonable extension and follow-on to the 2020–2021 efforts. The proposed work should result in successful project completion in 2021. It would be helpful to include an analysis of hybrid ideas involving compressed gas storage in tanks comprising hydrogen storage media (e.g., adsorbents, nanoscale hydrides, etc.). Instituting a hybrid approach for tank and storage system design could result in relaxation of the demanding requirements currently imposed for tank design and construction.
- Future topics are on point with the Program and industry shift to heavy-duty applications.
- Though the project is close to the end, the team still provided a good plan for the future work.

Project strengths:

- The project does a good job of taking current dominant technologies and extrapolating to potential high-volume cost and comparing them to other possible storage solutions and cost estimates. Additional examination into bulk storage cost is of interest for understanding initial capital costs.
- An experienced and highly capable analysis team at SA and collaborating organizations is conducting important technoeconomic assessments and developing projections that are directly relevant to DOE hydrogen storage goals and system needs. The project is well-managed and -coordinated, and it is effectively aligned with other DOE storage systems analysis efforts.
- The project provides consistent results and modeling in line with work done at ANL and overall Program objectives. This is great work, as usual.
- The project demonstrated an effective approach by DFMA analysis to predict costs and provided insight into which components are critical to reducing the costs of onboard hydrogen storage and meeting DOE cost targets.

Project weaknesses:

- No notable weaknesses are apparent. The project is concluding in the third quarter of 2021. Detailed documentation and reporting on project results and recommendations are essential.
- This is not SA's fault, but the proprietary nature of data from industry limits the ability to further tune or improve the fidelity of the model. In general, SA does a good job of filling in the blanks and soliciting the maximum amount of information possible.
- As we move to larger systems that are expected to have lower fuel economy and higher mileage/usage, storage costs become less important in total cost of ownership unless fuel costs are equivalent. BOP costs are a small percentage of overall HSS costs, which currently has not been observed. This may be true as volumes increase but should be examined more closely to ensure assumptions for regulators and valves are valid for high-pressure hydrogen.
- (1) For the Class 8 long-haul system on page 7, it will be interesting to show which property has the potential for reducing system cost. (2) On page 8, how the safety factor is defined and how the relaxed safety factor at 2.0 will affect the safety should be explained. Also, the assumption for the DOE target of carbon fiber price reduction of 40% should be provided. (3) On page 9, the schematic of the LH2 system design for Class 8 long-haul (similar to page 7) should be shown, and comparisons with 700 bar GH2 systems should be made. (4) On page 10, for the figure on the left, the difference between PAN-MA and T700S should be explained. In addition, using the unit of $/kg hydrogen for the system cost would be consistent with previous system costs. The 2030 target and ultimate target should be added into the figure. (5) On page 17, whether the cost in the figure includes the compression cost should be explained, as compression is one major part of cost and one system has a pressure of 500 bar—much higher than the other two systems. (6) More industrial partners should be added for system validation.

Recommendations for additions/deletions to project scope:

- It will be useful to evaluate cost and performance trade-offs for hybrid system designs employing compressed gas in a tank containing a solid-state hydrogen storage medium. (Of course, this requires close collaboration and consultation with investigators conducting materials development for hybrid systems.) Since lower tank pressures would likely be required in the hybrid system, this could result in reduced cost and complexity of tank manufacturing and assembly.

Project #ST-127: Hydrogen Materials–Advanced Research Consortium (HyMARC) Overview

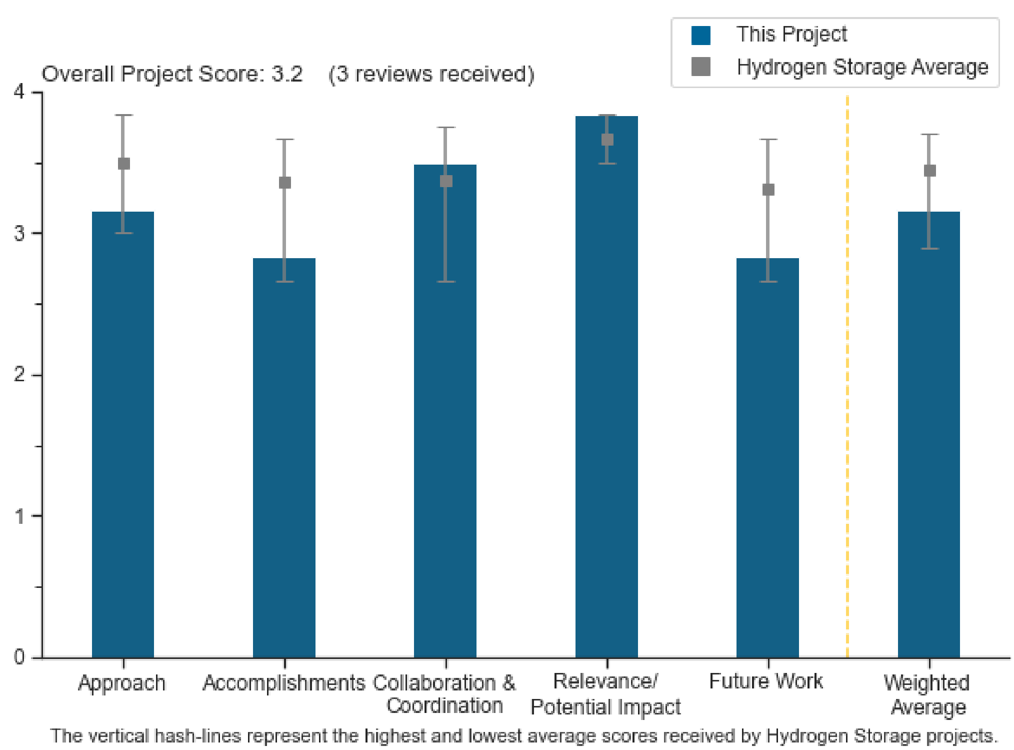
Mark Allendorf, Sandia National Laboratories

DOE Contract #	4.1.0.805 (SNL); 4.1.0.501 (NREL)
Start and End Dates	10/1/2015
Partners/Collaborators	National Institute of Standards and Technology, University of Nottingham, University of Uppsala, U.S. DRIVE Hydrogen Storage Tech Team, Colorado School of Mines, University of Hawaii, Université de Genève
Barriers Addressed	• Cost • Weight and volume • Efficiency • Hydrogen capacity and reversibility • Understanding of hydrogen physi-and chemisorption • Test protocols and evaluation facilities

Project Goal and Brief Summary

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 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.

Project Scoring



Question 1: Approach to performing the work

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

- The "co-design" approach adopted by the Hydrogen Materials Advanced Research Consortium (HyMARC) team is a rational and powerful way to address hydrogen storage material challenges. The incorporation of push projects serves to complement the core HyMARC effort and provides a means to readily expand the technical scope and depth of the consortium. The HyMARC team clearly understands the obstacles that must be overcome to implement improved hydrogen storage capabilities in both onboard and stationary systems. That said, it is not clear whether the goals of achieving "foundational understanding" are being effectively met by the consortium. At present, there are multiple, disparate efforts that address specific materials systems and approaches, but detailed information concerning reaction mechanisms, kinetic roadblocks, and reversibility issues seems to be limited.
- The approach is largely sound and clever. The consortium uses a multipronged approach that provides several paths to success. Theory and experiment are used to develop materials faster, with theory, at least in concept, providing an accelerated screening and experiment giving feedback to theory. The use of seedling projects with lower funding for high-risk areas, plus the use of push projects for high-possibility areas, is a very good way to spread funding. It is less clear if there is much pruning of dead ends. If this major allocation of funds is to be maximally successful and lead to jobs in a hydrogen economy, the leadership needs to make a call on projects that are not looking promising. This is not the National Science Foundation or even the Basic Energy Sciences office, and therefore, the allocation should flex to give maximal chance to the most successful avenues of inquiry—but not simply abandon whole areas of inquiry (e.g., do not defund all metal hydride work just because metal–organic framework [MOF] storage looks more promising, or vice versa, but definitely also do not award equal funding to all projects, regardless of progress).
- The objectives and barriers are clearly defined. The metrics for what success would look like are defined less well for the various pieces of the overall approach. The consortium has focused on push projects, which have brought some coherence to the consortium, and there is evidence that the seedlings are rather well-integrated into the overall consortium. The feasibility of the individual project areas was not well-defined. It is not clear how some of these very-low-capacity, somewhat reversible systems will find use.

Question 2: Accomplishments and progress

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

- Solid progress is being achieved on the core activities and push projects devoted to developing adsorbents containing multiple hydrogen adsorption sites. Likewise, as in past HyMARC work, the results from the theory and modeling efforts are providing useful guidance and support for the experimental efforts. The publication record in the last 12 months (more than 40 publications) is impressive. In addition to creating scientific impact, the publications serve as a useful way to document the consortium's progress. In the metal hydride arena, serious challenges concerning reversibility/cycling and sorption kinetics remain. For example, although nanostructured materials show great promise for improved hydrogen desorption rates, it is unclear whether the adsorption rates and reversibility/cycling can be improved to levels commensurate with practical systems applications; understanding those barriers is essential. High-capacity liquid-phase hydrides (borohydrides) are also intriguing, but again, finding a catalyst that can improve sorption kinetics seems to be elusive. Overall, development of an improved understanding of how additives and catalysts alter sorption reaction kinetics in complex hydride systems remains a serious challenge. The work on reversible storage in nanoconfined alane is new topic area; however, many questions remain, and it could be argued that the alane work is an unnecessary diversion that is tending to defocus the HyMARC effort. Even if confinement enables reversibility, it is not clear whether the gravimetric penalty imposed by the bipyridine-CTF (covalent triazine framework) host has significant impacts on the capacity. Likewise, it is unclear whether rehydrogenation can occur at useful temperatures and rates. The HyMARC team (mainly the Pacific Northwest National Laboratory group) is making good progress on hydrogen carriers, especially on understanding and improving catalyst deactivation in inorganic formates and developing a better understanding of kinetic bottlenecks and the full-cycle release of hydrogen in ethanol carriers. Questions remain about the extent to which the Advanced Light Source (ALS) diagnostics effort is providing meaningful impact. The new high-pressure x-ray absorption spectroscopy (XAS) system could yield important new insight, but progress to date has been slow.

- There has been good progress, given the conditions of the year. The consortium achieved adsorption of over two molecules H_2 per open K atom, a long-sought goal. The team increased both thermal conductivity and capacity of the bulk Li amide by using the surface energy of a nanoconfinement carbon structure. This system also achieved 50 cycles with no capacity loss. The team got 1.64 wt.% at 100 bar by first defining the correct energy range for 300 K operation, designing materials to get there, and then making the MOF, which is very encouraging. Also, there has been significant progress on Type 5 absorption curves. In a new area, the consortium performed an extensive theoretical search for destabilized metal hydride alloys and made a few, which perform as predicted. Verification of life and kinetics of these alloys would be a good addition.
- A roadmap and set of down-select criteria for each project area that define what must happen to enable "twice the energy density of compressed systems for hydrogen storage" were not described, and those down-selects are coming up at the end of fiscal year (FY) 2021. There were many unanswered but critical parameters that were not at least outlined, such as the minimum amount of scaffold that can still be used to obtain the energy density, the minimum amount of solvent, and the minimum amount of CTF to avoid the significant weight and volume penalties associated with these strategies. Also, it is unclear what use cases are being targeted for each of these projects and whether these weight and volume issues are obviated or exacerbated for these cases. There are a few areas that appear rather "sandbox-y"; the control of kinetics using coated MOFs is one of these areas. It was a nice trick, but with such a low capacity, it is unclear where this is heading. The carrier portion of the consortium's work seems solid; the team is addressing key problem areas effectively and using analysis of potential use cases to direct down-selects and focus areas for improvement. Some of that is apparent in the metal amide work, where the Tankinator model has been automated to help define where success lies. More of that sort of analysis-based approach should be applied more uniformly to each project area. The integration of theory and computation into the consortium has been, and continues to be, excellent. The machine learning exercise to find new metal hydrides is interesting but lacks a discussion of how this capability will be directed at other materials cases in the future.

Question 3: Collaboration and coordination

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

- The successful merger of the legacy HyMARC and Hydrogen Storage Characterization Optimization Research Effort (HySCORE) activities has created a more streamlined and manageable framework for consortium operations. The overall effort seems to be well-coordinated and -managed. Extensive industrial and international collaborations strongly support and complement the core HyMARC effort. Likewise, the push projects are a welcome addition, and they are serving to expand the scope and reach of the consortium into important new areas.
- This is an inherently massive collaborative; however, some projects (in their own reviews) seem to be more cooperative, while others are more lone experimenters. To get the most benefit of this large consortium, everyone needs to be researching as one team. This is hard to accomplish, but it would be a way to improve.
- There was good evidence supporting a high level of collaboration and coordination within HyMARC and with the seedlings. One problem area was illuminated in one of the seedling presentations: when one of the key high-pressure hydrogen sorption capabilities was taken offline because of an incident, the consortium did not quickly figure out how to cover that temporary loss of capability. It is unclear where the communication may have failed here, but perhaps it is an area on which to work. Overall, the seedling projects are well-coordinated and appear to be outproducing many of the core HyMARC projects.

Question 4: Relevance/potential impact

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

- HyMARC is a critical element of the DOE Hydrogen Program (the Program). DOE derives important benefits from the focus on developing an improved foundational understanding of processes and mechanisms operative during hydrogen storage reactions in solid-state and liquid materials. The project is well-aligned with Program research, development, and demonstration (RD&D) objectives.
- This consortium that is making materials and developing systems that enable hydrogen storage is perfectly aligned with the Program goals. The expansion of the goals to include stationary uses opens up many materials methods that were eliminated for vehicles. The consortium also helps other projects make progress toward their goals.
- The goals of HyMARC and the associated projects clearly support Program goals and objectives and are thus highly relevant to the potential advancement of the Program.

Question 5: Proposed future work

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

- Continuing the focused work on the push projects is essential, and the end-of-year go/no-go decisions should be made rationally, with close attention to progress on objective milestones. The consortium must adhere to its stated objective of providing foundational understanding and avoid the risk of investigating multiple unrelated systems that may show promise arising from speculative criteria. For example, careful consideration should be paid to continued work on diversions such as hydrogen sorption reactions in nanoconfined alane and photo-assisted hydrogen adsorption/desorption. Without a candid assessment of those and related ancillary activities, there is a serious risk of defocusing and fragmenting the core efforts. In addition, near-term decisions about the ALS diagnostics work must be made. ALS progress to date has been slow. A solid plan for accelerating the diagnostics work should be implemented.
- The future work is appropriate. Given the wider usage parameters in the new focus, all areas have potential to contribute. That said, focusing on those getting the most hydrogen out of the finished tank and materials systems and modest cost still makes sense, but more (not all) resources should be focused in the most promising areas.
- The proposed future work was not well-prioritized, as the top item listed was "international collaborations." With the push project down-select coming up in FY 2021, this would appear to be an all-hands-on-deck exercise to approach the down-select with an abundance of data. With the down-select rapidly approaching, a good discussion of the down-select criteria toward which the project teams are working is expected, along with discussion of the status in regard to reaching or not reaching a "go" decision.

Project strengths:

- The carrier projects do a nice job of integrating analysis, computation, experimental kinetics, and catalysis into their approach, which is very nice. Also, they have worked well with one of the key seedlings at the University of Southern California. The Lawrence Livermore National Laboratory theory and modeling capability continues to show high value to the projects, including the seedlings. The Lawrence Berkeley National Laboratory work on new materials for sorbents continues to do a beautiful job of pushing the envelope and helping to define what is truly possible in altering capacities, but also the heat of adsorption, via the really tough chemistry of designing and synthesizing open metal sites. The use of the automated Tankinator model to define the boundaries of practicality in the metal amide area is an important contribution. Defining barriers between success and failure is an approach that should be applied more generally to the other materials classes as well.
- HyMARC is a critical element of the DOE RD&D portfolio. Providing a foundational understanding of the thermodynamics and kinetics of hydrogen storage reactions and processes is vital to the development of storage materials that meet or exceed DOE goals. A strong and capable HyMARC core team has been assembled to conduct the work on this project. The addition of push projects is infusing new ideas and expertise into the consortium.

- The team is the project's greatest strength; it has excellent principal investigators. The addition of push and seedling projects to the main effort makes the consortium's structure a major strength. The breadth of approaches is a strength, as is the pairing of theory and experiment in a feedback loop.

Project weaknesses:

- The consortium must remain keenly focused on its stated objective to provide foundational understanding of hydrogen storage processes and mechanisms. The inclusion of projects and materials systems that are likely to be "non-starters" with little impact on overcoming obstacles is tending to defocus and fragment the overall effort. At this point in the life of the consortium, a thoughtful and detailed assessment of future directions must be made. In addition, even though the ALS diagnostics activity has significant potential, progress has been excruciatingly slow; a candid assessment should be made concerning the future of that effort.
- Size might be the consortium's biggest weakness, though of course, it comes with the breadth of effort requested of the consortium, which is a positive thing. It is very hard to properly manage so many projects and to move funds around in ways that might make the most progress. Also, it is hard to get communication across so many participants without wasting time in endless meetings.
- There continues to be a lack of a roadmap to success for many materials types. It is not clear what it will take, or what use cases are applicable, to get a low-capacity material across the finish line. This should be outlined for each materials project with the current status, the path forward, and the probability of achievable improvements.

Recommendations for additions/deletions to project scope:

- Four areas should be emphasized: (1) provide a greater focus on overcoming kinetics obstacles, especially in the amide and borohydride push projects; (2) conduct a careful evaluation of activities that have little or no bearing on foundational understanding and limited potential for meeting DOE goals; (3) conduct a candid evaluation of ALS diagnostics progress, and take remediation action as appropriate; and (4) provide a thoughtful and detailed view of where the consortium stands, what foundational information is missing, and what mid-course corrections should be made to address those issues. This information should be made available to Program management and the Hydrogen Storage Tech Team. On another note, for the benefit of the reviewers, it would be helpful if the team could provide what it considers to be the most noteworthy examples of foundational understanding that have emerged thus far from the HyMARC work.
- The work on MOFs and on carriers, especially formate, seems to be making the most aggressive progress, so the consortium should increase the resources (not necessarily monetary resources) provided to those projects to help them continue that progress.
- Any materials research and development that cannot be adequately defended as to what use case it will address, and how it will achieve success with respect to current status, should be considered for deletion from the work scope. From the few details given, it would appear that the MOF/PDMS (polydimethylsiloxane) and the alane/CTF projects are in this category.

Project #ST-209: Hydrogen Materials–Advanced Research Consortium (HyMARC) Seedling: Theory-Guided Design and Discovery of Materials for Reversible Methane and Hydrogen Storage

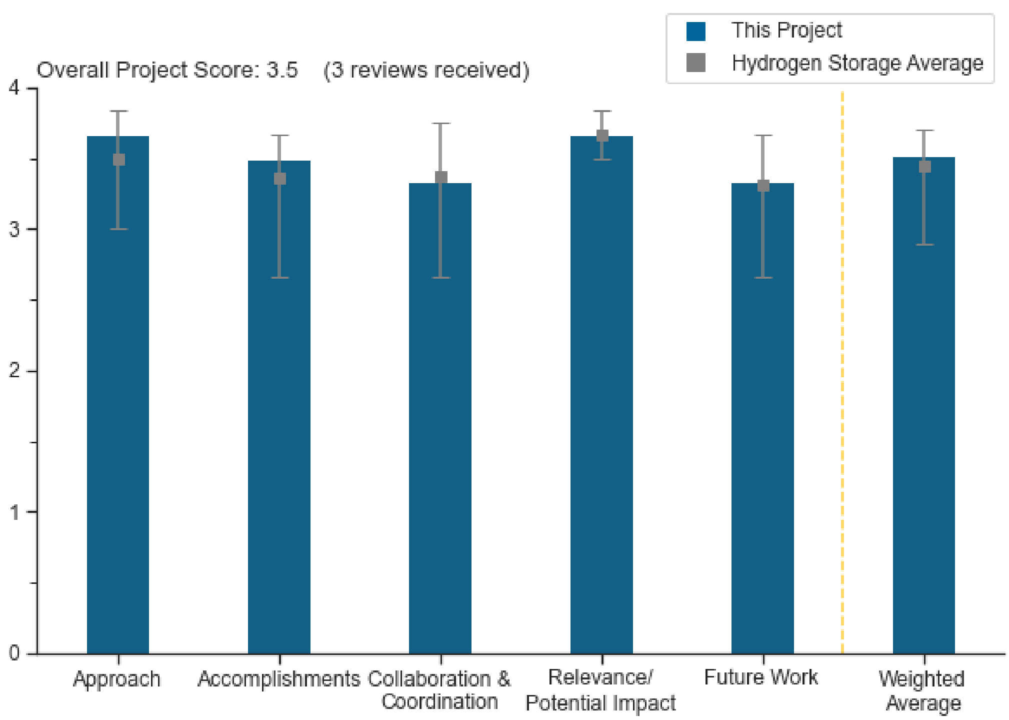
Omar Farha, Northwestern University

DOE Contract #	DE-EE0008816
Start and End Dates	1/1/2020 to 1/31/2023
Partners/Collaborators	National Institute of Standards and Technology, National Renewable Energy Laboratory
Barriers Addressed	• System weight and volume • System cost • Efficiency • Durability/operability • Materials of construction

Project Goal and Brief Summary

This project aims to exploit high-performance metal–organic framework (MOF) sorbents by combining synthesis with machine learning to find stable and scalable materials for hydrogen storage while maintaining a reasonable cost of production. The project researchers will use a machine learning algorithm to screen a database of materials for hydrogen uptake. Having identified the top candidate MOFs, researchers will synthesize and characterize them and study their behavior under pressure- and temperature-swing (PT swing) operation. The project team will also look at removing solvent molecules from MOFs to yield open metal sites for storing molecular hydrogen at near-room temperature. If successful, this project will advance economical hydrogen storage technology.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Storage projects.

Question 1: Approach to performing the work

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

- The project focuses on the use of computational screening and material synthesis to identify and demonstrate improved MOF adsorbents for high-capacity hydrogen storage. High-throughput computational discovery, coupled with machine learning, is used to identify materials with different structural topologies, allowing the team to predict which MOFs will have the highest capacities at practical operating temperatures and pressures. Subsequent synthesis and characterization are used for experimental validation. The approach is powerful and allows for rapid investigation of a broad parameter space. The project parallels very similar work on MOF identification and screening by Siegel, et al., at the University of Michigan, Savannah River National Laboratory, and Ford Motor Company. However, the principal difference is that this project focuses on exploring a broad range of different MOF topologies, leading to identification of promising candidates that are down-selected for subsequent synthesis and characterization. Overall, the approach is innovative and addresses critical obstacles to hydrogen storage in adsorbent systems in a novel way.
- Use of theory, experiment, and spectroscopy is a good plan for this MOF work. Machine learning to accelerate the search seems to be the method these days and makes sense as a means of isolating some good candidates. While setting up the algorithm used to generate MOFs to simulate, the project team used considerations of how easy or hard it would be to synthesize certain linker and corner groups; this is a good step.
- The modeling, simulation, and machine learning high-throughput approach, which is then validated through synthesis and characterization of promising structure types, may yield adsorbents with enhanced properties. This approach addresses critical barriers in developing improved sorbents for hydrogen storage.

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.

- An extensive parameter space has been explored, and several promising storage candidates have been identified. An energy histogram (volume fraction vs. energy) is used as a useful descriptor and discriminator. Initial experimental work has been performed to synthesize and characterize top candidates identified in the computation screening studies. Impressive performance, comparable to or exceeding the storage characteristics of the best incumbent adsorbents, has been demonstrated from several of the recently synthesized materials (e.g., MFU-4l, PCN-61). Although solid progress has been achieved, it is not clear whether a dominant chemical and/or structural property/topology has been identified as being most critical for enhanced storage. It will be important to focus on this issue to effectively guide future work. There is one minor point: creation of open metal sites requires removal of solvent molecules from the structure. Since incomplete solvent removal limits sorbent performance, it is worthwhile to consider whether removal of solvent molecules depends on the structure/topology.
- The project has demonstrated over 45 g/L and 13% specific capacity on a material basis from 100 bar using 83 K of temperature swing. Theoretical values were used to train the algorithm to seek the best MOFs. The researchers then made one of the best ones and again got over 45 g/L using PT swing. The researchers tuned it further and got 9.5% and over 50 g/L in PT swing. This material also could be reactivated after air exposure with little loss; that is an important point. Theory suggested that PCN-61 might be good, too. Results were a little shy of prediction, but 8% and 48 g/L were still delivered in PT swing.
- The researchers have rapidly approached the go/no-go milestone that they have set for themselves. They have made solid, steady progress in spite of COVID.

Question 3: Collaboration and coordination

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

- Useful collaborations have been established with National Institute of Standards and Technology (NIST) (primarily isotherm testing) and the National Renewable Energy Laboratory (NREL) (sorbent performance and experimental validation). The interactions among collaborating organizations appear to be well-coordinated and -managed. No other collaborations with the HyMARC core team are apparent. Also, it is surprising that, given the large overlap in project direction and scope, more active interactions with the University of Michigan group have not been established.
- There is very good coordination with the NIST in obtaining high-pressure sorption data, which have subsequently been validated by work at NREL.
- There is good cooperation within the team. There is value in getting structure and data from NIST and, of course, in NREL validation.

Question 4: Relevance/potential impact

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

- Providing experimentally verified rapid-throughput screening of a very large database of sorbent structure types can support the fundamental understanding of what barriers exist in breaking through the roughly 50 g/L hydrogen capacity "ceiling," which in principle could lead to materials that enable lower-pressure, lower-cost storage systems. This potential result is highly relevant and impactful to DOE's Hydrogen Program.
- This is a potentially impactful project that is well-aligned with DOE research, development, and demonstration objectives and goals for improved hydrogen storage in adsorbent systems. Although similar to the University of Michigan screening work, this project extends the computational parameter space to include a large number of different structural topologies. The project provides DOE with a new look at promising candidates for improved hydrogen adsorption, and the project has significant potential to advance progress in this important technology area.
- The project is well-aligned—it seeks to improve capacity and thus reduce mass and volume and cost of storage material through higher capacity and milder conditions.

Question 5: Proposed future work

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

- The future work is a direct and logical extension of the earlier technical effort. In addition, the investigation of MOFs functionalized with metal catecholates has been initiated, and experimental validation of simulation results is under way. This is an interesting new direction that has intriguing potential. Project milestones are unambiguous, and a quantitative go/no-go objective for Year 1 is in place. There is a minor point: there is no mention of future efforts to increase packing density, although it seems possible that limited packing density is an issue. If so, a description of a plan to increase the density would be helpful.
- This is a good team that combines theory and simulation with synthesis and characterization. The future plans take full advantage of these capabilities, and it is expected that the current level of productivity will continue.
- Extending the methods to 300 K (just pressure swing) materials that are predicted to exceed 2.8% and 18 g/L makes sense. One suggestion is that working to clean up the MOFs and/or look for damage while in transit to the test site would be good and a fast way to potentially improve the material results.

Project strengths:

- The project has an excellent team, combining strengths in characterization and synthesis with computation and simulation. The machine learning aspects and the development of a descriptor that contains the energy landscape of hydrogen molecules on or at the surfaces of sorbents seem to comprise a powerful approach. Apparently, hybrid descriptors are also in the plan and are expected to further advance the machine learning aspects of this project.

- This seedling project has potential to generate new, improved adsorbent materials for hydrogen storage. The research and development team and collaborators are highly capable and have expertise in all relevant areas. This is facilitating rapid and efficient progress.
- There is very active cooperation between the theory and experiment teams. Limiting the MOF theoretic analysis to those MOFs likely to be possible to make improves efficiency.

Project weaknesses:

- At this stage, no major weaknesses or deficiencies have been identified. To avoid duplication of effort, communication and collaboration with the University of Michigan high-throughput screening group might be helpful.
- The best results are based on a PT swing, but the researchers do not appear to have studied the impact on use.

Recommendations for additions/deletions to project scope:

- There are no suggestions for major revisions. There is one minor point: as was pointed out in the future work section, increasing the MOF packing density is an issue that could be explored further. Collaboration with the University of Michigan group would ensure that computation and screening efforts are not duplicated.
- Perhaps having some communication between this project and Jeff Long’s project vis-à-vis the catecholate approach could add value to both projects.
- All results are based on a significant temperature swing. In a real tank, this will have significant cost when it is time to refill, as the material and tank will need to be cooled. The project team should look at the overall scenario. Also, the MOFs should be checked for damage after testing to see if the validation values might be low because of changes that might have occurred in transport.

Project #ST-211: Hydrogen Materials–Advanced Research Consortium (HyMARC) Seedling: Optimal Adsorbents for Low-Cost Storage of Natural Gas and Hydrogen: Computational Identification, Experimental Demonstration, and System-Level Projection

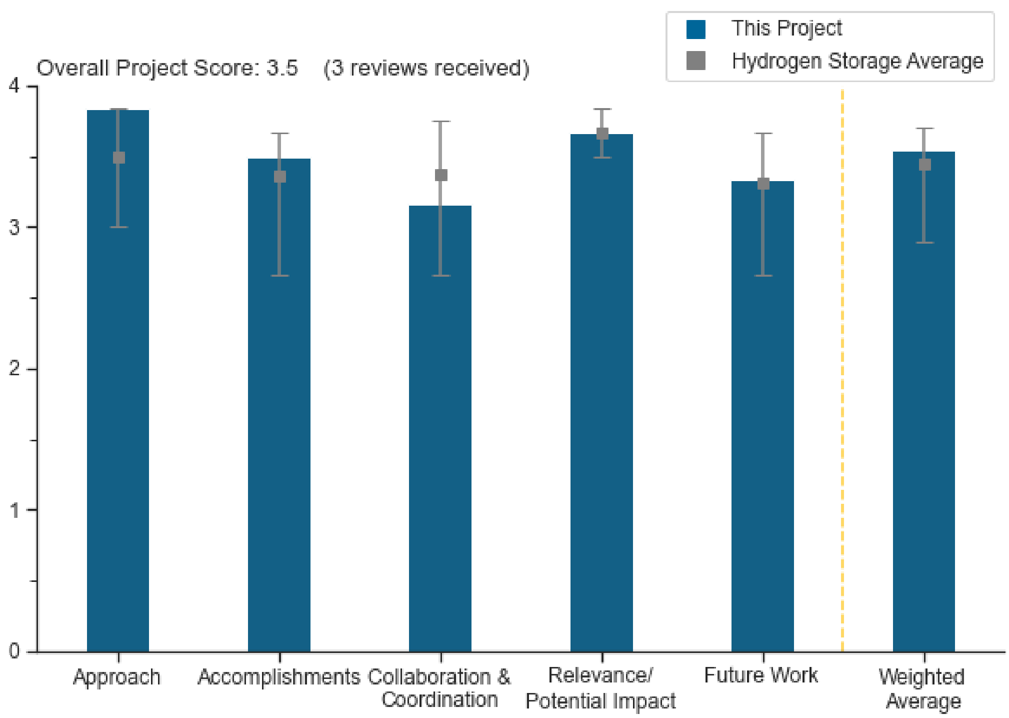
Don Siegel, University of Michigan

DOE Contract #	DE-EE0008814
Start and End Dates	1/1/2020 to 12/31/2022
Partners/Collaborators	Ford Motor Company, Hydrogen Storage Engineering Center of Excellence, Savannah River National Laboratory
Barriers Addressed	• Volumetric density • Gravimetric density

Project Goal and Brief Summary

This project aims to demonstrate adsorbents that, when incorporated into an adsorbed natural gas system, have the potential to surpass the capacity of compressed natural gas systems. If successful, this project will allow for smaller and lighter natural gas storage systems that operate at lower pressures, overcoming a significant barrier to the deployment of natural gas vehicles with long driving ranges. The project will use computational screening and machine learning techniques to identify target adsorbents with high capacities for synthetic exploration.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Storage projects.

Question 1: Approach to performing the work

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

- This project provides for the application of a significant amount of knowledge of hydrogen sorption and transfers it to an examination of materials for natural gas sorption. The focus is on searching for and experimentally validating the gravimetric and volumetric capacities of candidate materials that have been identified by modeling and simulation combined with machine learning algorithms. This approach appears to be very much state of the art and is directly addressing key barriers that are common in hydrogen and natural gas sorption for applications important to the U.S. Department of Energy, such as for onboard storage of natural gas or hydrogen for medium- and heavy-duty trucks.
- The project builds upon extensive prior work devoted to identifying optimum metal–organic framework (MOF) adsorbents for hydrogen storage. The high-throughput approach combines crystal structure analysis, grand canonical Monte Carlo simulation (GCMC), and machine learning to systematically identify MOFs for high-capacity methane storage. The approach is novel and powerful, allowing for the identification and evaluation of a huge number (>500,000) of real and hypothetical MOFs having high usable volumetric and gravimetric capacity for natural gas (NG). Usable capacities for candidates identified from application of the computational screening are compared with benchmark materials, and results from those characterizations provide the foundation for further materials synthesis and testing. The project is well-focused on overcoming limitations of conventional NG storage approaches (e.g., compressed natural gas [CNG] systems).
- The exhaustive computational process offers an excellent chance for progress, and the underlying theoretical fundamentals have been tested extensively, so this is a sound approach. Screening is based on machine learning, not direct calculation, for speed. The regression to complete simulation is quite good in general. Using a well-understood benchmark material and one that has excellent performance itself is a challenging but excellent strategy. The system-level component (while on pause at present) puts this effort above other similar projects that look only at materials.

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 was achieved on extending the prior work on hydrogen adsorbents to MOF adsorbents for NG storage. A high-throughput methodology similar to that used for the hydrogen work is employed, with an emphasis on evaluating different interatomic potentials suitable for open metal and non-metal adsorption sites. The comprehensive screening approach was successful in identifying several MOFs that potentially surpass the performance of HKUST-1, a leading incumbent. The computational results offer a straightforward pathway to subsequent synthesis and testing of MOFs optimized for NG storage capacity. Raman spectroscopy was demonstrated to be a useful method for near-real-time measurement of MOF sorption kinetics. The authors have used Raman spectroscopy to monitor the C-H stretch for bound methane at 2910 cm^{-1} in order to infer methane sorption kinetics. It could be possible to use the same method to monitor hydrogen adsorption in MOFs using the H-H stretch at ~4160 cm^{-1}.
- The researchers have made good progress, especially in this bizarre year. They passed their go/no-go milestone. The potentials they developed have been demonstrated as giving good agreement with data. Error is only a few percent of the value with a single set of generalized potentials. With experiments, the researchers showed that equilibration is on the order of seconds, which is important for control in fueling and use. The team has identified hundreds of materials that might give as much as 10% more volumetric capacity and double or triple the gravimetric capacity of useable CNG. This was determined using the longer GCMC modeling. The researchers proved out the machine learning model so that, in the future, they can test an order of magnitude more MOFs in a very short time. The project then made USTA-76, and it did outperform the benchmark in a pressure swing trial.
- The well-thought-out modeling and simulation approach coupled to a machine learning approach has led to a number of potential high-capacity candidates. Many of these have been experimentally validated through synthesis and characterization. The modeling work has been very successful in predicting the sorption

properties of MOFs. There is excellent progress toward the milestones. The unexpected departure of a team member that was providing storage system-level modeling has hampered that portion of the project; the remaining team members are actively working to mitigate this unforeseeable gap in the project.

Question 3: Collaboration and coordination

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

- Close and effective interactions among investigators at the University of Michigan (chemical and mechanical engineering departments), Savannah River National Laboratory, Ford Motor Company, and the Hydrogen Storage Engineering Center of Excellence have accelerated progress. The project is well-managed, and collaborations are well-coordinated. Future work may involve a more extensive collaboration with an industrial partner (e.g., Ford Motor Company) to facilitate potential product development. As pointed out by the principal investigator (PI), the departure of the co-PI, D. Tamburello, from the project has limited the system modeling work. Efforts are under way to find a person to lead this important aspect of the project.
- Up to the unexpected departure of the systems modeling expert on the project, the project exhibited good collaboration.
- There is not much collaboration outside the project, but there is good coordination inside it between three partners.

Question 4: Relevance/potential impact

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

- This novel and impactful project is well-aligned with DOE research, development, and demonstration objectives and goals for improved storage of natural gas. The project provides DOE with a comprehensive offering of potential candidates for improved CH_4 adsorption, and the project has significant potential to advance progress in an important technology area within the DOE energy portfolio.
- The project is addressing key issues and barriers for natural gas storage that have been leveraged by previous work on hydrogen sorption. These two areas are objectives of both the DOE Hydrogen Program and the Vehicle Technologies Office (VTO).
- While CNG might not seem relevant, the Hydrogen Program expanded and partnered with VTO, and this does fall squarely in the desired goal of increasing storage density of NG for use in transport. Additionally, there is some chance, if small, that this will help in our understanding of how to store hydrogen if there are fundamental and applicable rules of storage discovered.

Question 5: Proposed future work

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

- Future work follows logically and reasonably from the current technical effort. The future focus will undoubtedly be on synthesis and testing of MOFs identified in the computational screening work. An important issue that should be addressed is potential identification and mitigation of adsorption site poisoning by (non-methane) contaminants that may be present in the natural gas source. In addition, a detailed plan is needed to address the daunting challenge of efficiently down-selecting the most appropriate candidates for subsequent synthesis and characterization.
- Future work is on-task and seems sound.
- The team has future plans that will maintain the project’s focus on high-capacity sorbents and will seek out a path to mitigate the departure of the systems modeling collaborator.

Project strengths:

- This is a first-rate project that addresses an important DOE technology need. A highly capable team with expertise in all relevant areas is conducting the technical effort. The project is well-managed and -coordinated. (The PI continues to provide clearly stated, candid, and succinct presentation and review materials, which is appreciated.)

- There is a great team for theory and lab development of MOFs. The team seems to have the best monolayer system for rapidly and accurately screening MOFs. The project uses theory at material and system levels to work toward a viable product.
- There is a very cohesive, well-experienced team in the modeling and the experimental areas and excellent integration among the team members.

Project weaknesses:

- Only two (minor) weaknesses are evident:
    - The vast number of materials identified by computation cannot all be synthesized. Implementation of efficient method(s) for down-selection of materials for synthesis remains problematic.
    - As pointed out in the future work section, contaminants in the NG gas stream could create major problems. That issue should be addressed explicitly.
- The project lost the system partner and needs one to finish the plan.
- No weaknesses were detected.

Recommendations for additions/deletions to project scope:

- Apart from addressing the contaminant issue and identifying a partner to conduct system-level modeling (as discussed in the presentation), no additional revisions to project scope are recommended.
- It is suggested that the team also look at poisoning by S, P, and Si compounds found in landfill gas. Pacific Northwest National Laboratory or Argonne National Laboratory could be potentially good system partners, though someone in the gas industry would be even better.
- Depending on whether the team can identify a storage systems modeler, the project may or may not have to alter the project scope.

Project #ST-212: Hydrogen Materials–Advanced Research Consortium (HyMARC) Seedling: Methane and Hydrogen Storage with Porous Cage-Based Composite Materials

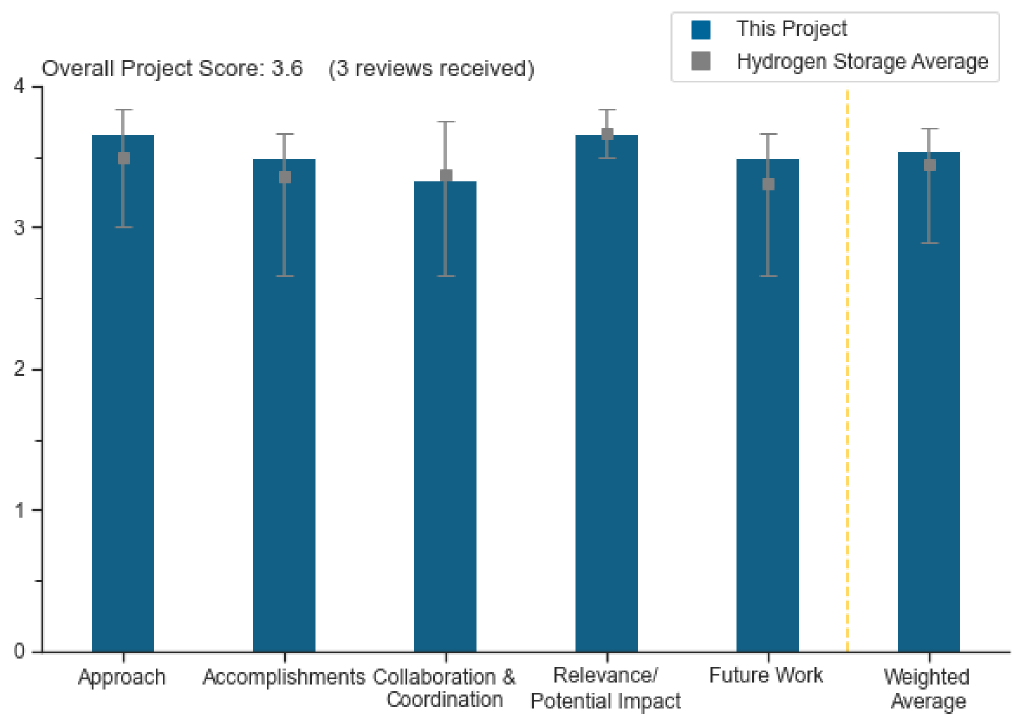
Eric Bloch, University of Delaware

DOE Contract #	DE-EE0008813
Start and End Dates	11/15/2019 to 11/30/2022
Partners/Collaborators	National Institute of Standards and Technology, National Renewable Energy Laboratory, Lawrence Berkeley National Laboratory, University of California, Berkeley
Barriers Addressed	• System weight and volume • System cost • Efficiency • Lack of understanding of physisorption and chemisorption

Project Goal and Brief Summary

Metal–organic frameworks (MOFs) have low bulk densities that present challenges to their use as methane and hydrogen storage materials. This project will attempt to address those shortcomings by preparing high-capacity soluble absorbents that can be placed in the space between MOF crystals, resulting in a porous cage–MOF composite with increased density and volumetric storage capacity.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Storage projects.

Question 1: Approach to performing the work

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

- The idea—to fill in between MOFs with "cages" that also hold hydrogen—is clever, and it seems to be playing out. Low-pressure screening is innovative.
- The approach is focused on maximizing the storage density of sorbent materials, which can have an impact on the barriers related to storage capacity, cost, and efficiency.
- Increasing volumetric performance of MOF powders or compacts (a primary goal of this project) is an important objective. It is unclear from the presentation how the densities of MOF powders are being measured (shaking for a given number of iterations, pressing, etc.) or whether these measurements are being conducted in a repeatable fashion.

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 approach of filling the voids between particles of a sorption material with a molecular sorbent is simple but elegant and appears to be novel. If successful, the team anticipates an improvement of 25% in the sorption capacity of these materials for hydrogen and methane/natural gas. This, in turn, has impacts on the goals of both the DOE Hydrogen Program and the Vehicle Technologies Office.
- The researchers made reasonable progress; they selected and made MOFs and cages and created a composite. The project met the go/no-go with 37% improvement over MOF alone. While results were based on total capacity, it appears that the usable capacity has also improved by enough to meet project goals. The researchers found that there is no real preferred cage for any particular MOF, so they can go for ones that are easy to make and to add to the MOFs—and, of course, favor ones that are cheap.
- Accounting for a slowdown due to Covid-19, this project has made excellent progress in its first year.

Question 3: Collaboration and coordination

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

- Given the circumstances of the last year, the project is doing well in collaborating with the National Institute of Standards and Technology and the National Renewable Energy Laboratory on materials characterization and validation and is having discussions with one of the key sorption experts within HyMARC: Jeff Long of Lawrence Berkeley National Laboratory and the University of California, Berkeley.
- There is good interaction for a smaller project, and the interactions have been useful to the team.
- The project team is collaborating adequately with others as needed.

Question 4: Relevance/potential impact

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

- The project addresses packaging problems with MOFs by placing a material between crystals, so it does address a relevant problem.
- Improving the volumetric and gravimetric capacities of MOF-type sorbents for hydrogen or methane supports the goals and objectives of the DOE Hydrogen Program, as well as those of the Vehicle Technologies Office.
- A successful outcome would have a positive impact on the practical energy densities of MOFs for storage of hydrogen or natural gas.

Question 5: Proposed future work

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

- Future work is directed at developing the necessary understanding of the interplay among the MOFs and the interstitial molecular adsorbent cages, including durability of these composite materials. The future work is well-focused on achieving enhanced capacities for gas storage, an important barrier to application.
- The team plans to look at compression and cycling durability and attempt to tune results to better capacity. These are all good ideas.
- The future plans are adequate.

Project strengths:

- This is a novel idea for easily improving capacity. It seems that there is no interaction between cage and MOF, so there will be no need to tune the cage to each MOF, so the team can look for an inexpensive and high-capacity material.
- This is a novel approach that is well-thought-out, well-explained, and well-executed.
- Many MOFs have been evaluated.

Project weaknesses:

- The methodology for densification needs to be standardized (or at least better explained) so that repeatability can be ensured.
- There are no serious ones. The team ought to focus on useable gas rather than total capacity.
- No weaknesses were detected.

Recommendations for additions/deletions to project scope:

- The researchers really need to show the usable capacity and would be better off doing their analysis and their predictions that way because that is what will matter to users. The project should look at the impacts of higher hydrocarbons and normal poisons in landfill gas (gases containing P, S, Si) on cage performance. It would be good to test kinetics relative to the same MOF with no cages.
- This project may benefit from collaboration with the team at the University of Michigan, which is performing similar experiments.
- The principal investigator seems to have the project well in hand.

Project #ST-214: Hydrogen Materials–Advanced Research Consortium (HyMARC) Seedling: Heteroatom-Modified and Compacted Zeolite-Templated Carbons for Gas Storage

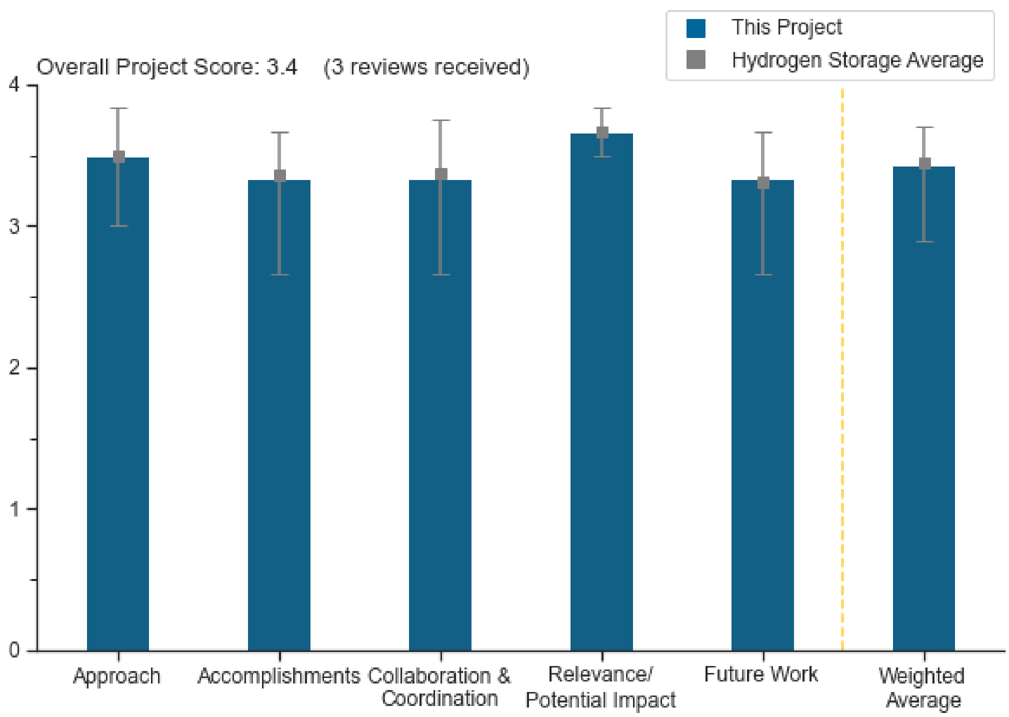
Nicholas Stadie, Montana State University

DOE Contract #	DE-EE0008815
Start and End Dates	10/1/2019 to 12/31/2022
Partners/Collaborators	California Institute of Technology, HyMARC, Tohoku University
Barriers Addressed	• Lack of understanding of (methane) physisorption • System weight and volume • System cost

Project Goal and Brief Summary

This project seeks to increase the volumetric energy density of zeolite-templated carbon (ZTC) as a methane storage medium, while reducing its cost. This research team will first determine the ultimate volumetric methane delivery limits in porous carbon framework materials, focusing on ZTCs as model absorbents. The team will quantify the effect of boron–nitrogen heteroatom dopants on methane adsorption thermodynamics and storage/delivery at near-room temperature. Researchers will then optimize conditions for the densification of graphene-like fragments in ordered pore networks suitable for methane adsorptive storage and delivery.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Storage projects.

Question 1: Approach to performing the work

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

- The use of doped ZTCs for methane storage is novel and innovative. The homogeneous, metal-free binding sites and controlled pore size in ZTCs provide a unique adsorption environment that can potentially overcome problems inherent in other (primarily heterogeneous) adsorbent systems. This approach is a departure from more conventional adsorption methods, and it provides an intriguing opportunity to facilitate high-capacity methane storage and improved delivery. B and N doping of the porous ZTCs may be useful for tailoring the CH_4 binding energy. Theoretical studies of dopant interactions are providing useful guidance for the experimental work. In addition, compaction of the adsorbent by densification into robust pellets is enabling achievement of higher volumetric densities.
- This is a good approach: using a metal-free framework to try to get a large amount of surface in the "perfect" binding energy region and then tuning that energy with heteroatom substitution. ZTCs are a scalable technology; they could be made at a potentially modest price in enormous amounts, if the approach is successful.
- Well-defined project objectives address barriers to enhanced methane/natural gas adsorption on porous carbons. The experimental and theoretical approach is well-thought-out and logical. As this is a "seedling" project, collaborations with the HyMARC team have been identified mainly in the area of spectroscopic characterization but also in an international collaboration on the compaction of high-density nitrogen-doped carbons.

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.

- Solid progress has been made on this project in 2020–2021. Increase in volumetric density has been achieved by hot-pressing the ZTC with a reduced graphene oxide (rGO) binder to form robust pellets. This densification approach provides a straightforward pathway to meeting DOE goals for CH_4 capacity. The initial studies of heteroatom doping are interesting and provocative. First principles quantum mechanical calculations suggest that nitrogen doping of the ZTC framework should yield enhanced methane binding, whereas boron doping actually suppresses binding. This difference is surprising and requires more detailed validation. The perceived disparity between dopants notwithstanding, the theoretical results clearly suggest that heteroatom dopants/additives can be effective in controlling the binding energy. However, retention of homogeneity with increased dopant/additive incorporation could be problematic. Overall, the dopant work is clearly important for further development of ZTC-based systems for methane storage, and it remains a critical topic for future work. Reversibility and efficient sorption cycling in the densified pellets are also important issues. Although the ZTC system apparently shows reasonable reversibility, swelling and breakage of the pellets occur upon cycling. The impact of pellet degradation during cycling on the incorporation of the densified ZTC material in a practical system remains an outstanding issue.
- The project has been productive so far, with modeling and simulations coupled with experiment well under way. Key measurements are being obtained, and early milestones are being achieved or approached. Higher binding enthalpies of methane have been observed at low to medium methane loading, which is a significant observation. One main goal is to synthesize homogeneously substituted nitrogen-doped carbon derived from the zeolite template approach. Computational simulations of a highly idealized model of nitrogen sites in a graphene-like environment have predicted enhanced binding of methane on such a site. So far, experimental verification of the homogeneity of sites, as indicated by a constant heat of adsorption versus loading, has been elusive. This may be related to the lack of site homogeneity in the realistic material as synthesized, where there are likely a variety of carbon chemical environments and, hence, a variety of different types of nitrogen-containing doped sites (pyrrolic, pyridinic, etc.) that likely have a variety of binding enthalpies with methane. The plan allows for more characterization using HyMARC capabilities to explore the local electronic and chemical structure at the nitrogen and carbon sites that may shed additional light on this matter. Approaches to densification of the zeolite-templated, nitrogen-doped carbons have been quite successful so far. However, the principal investigator (PI) indicates that, with

several adsorption–desorption cycles, the mechanical durability may be compromised. More work is planned.

- There is good progress on pellet formation with minimal function loss. The project is close to crystalline values of surface area versus density, especially in a specific surface area basis—basically, 200 v/V delivery at room temperature and max 100 bar pressure. The researchers used theory to suggest a poor chance of progress using boron, so they were able to focus on a nitrogen heteroatom addition. They were able to add nitrogen to the matrix and found increased binding but a high slope of the binding energy curve.

Question 3: Collaboration and coordination

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

- Effective collaborations with the California Institute of Technology (Caltech), Tohuku University, and HyMARC have been established, and these interactions are enhancing progress on the project. The research and development (R&D) team is highly capable and has expertise in all relevant project areas. The project is well-managed, and all external collaborations are well-coordinated. A more active collaboration with the nuclear magnetic resonance (NMR) group at Pacific Northwest National Laboratory (PNNL) is recommended.
- There is good collaboration with colleagues at Caltech and at Tohoku University, with good plans for expanded collaborations with HyMARC capability leaders in the future.
- The collaboration is about right for a project of this size. There is cooperation between partners but also use of DOE resources to get high-end work done.

Question 4: Relevance/potential impact

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

- This innovative project is a welcome addition to the DOE R&D portfolio. The project is a useful and potentially impactful complement to other gaseous adsorption projects. DOE has identified methane storage as an important emerging technology area, and this project has potential to overcome many of the critical obstacles to achieving high-capacity storage and delivery. The work is well-aligned with DOE research, development, and demonstration objectives and goals.
- Methane adsorption is now part of the DOE Hydrogen Program goal structure and also supports DOE's Vehicle Technologies Office. Plus, the general learnings may be clues to how to do the same thing with hydrogen. The project is working on reducing the system's cost, volume, and mass, all of which are spot on for relevance.
- Striving to enhance adsorption of methane on high-surface-area adsorbents via novel approaches may advance the state of the art and is relevant to the goals of both the DOE Hydrogen Program and the Vehicle Technologies Office.

Question 5: Proposed future work

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

- Future work focuses on improving N doping and densification while maintaining structural homogeneity. These are challenging tasks, but a reasonable approach for meeting the goals is in place. Cycling and reversibility issues are not inconsequential, and greater emphasis on finding ways to maintain homogeneity and capacity during sorption cycling is recommended. Likewise, questions remain about the accuracy of the first principles calculations with regard to heteroatom doping type and efficiency. Based on the calculations, nitrogen has been down-selected. However, if time and resources permit, synthesis and densification of a boron-doped sample would be useful in order to validate the theoretical predictions and to probe a different doping environment. Although PNNL is a collaborating institution, future work on solid-state NMR at PNNL is not mentioned. That powerful diagnostic capability could be employed effectively to probe structural details and provide important mechanistic understanding; the project team is urged to utilize that capability more extensively in future work.

- The researchers are focused on regaining homogeneity through densification and heteroatom addition, which is exactly what they need to do. N-bearing precursors have to be smaller than benzene to make ZTC; multiple precursors might be needed. The researchers should try getting as much N in as possible. They may look at properly packed powders to get dense powders.
- The project team is very good, highly capable, and will likely overcome many of the experimental hurdles and move the project forward. Whether the project's ultimate goal of achieving homogeneously doped carbons to enable high heats of adsorption independent of loading can be achieved is still to be determined.

Project strengths:

- This is a unique and potentially impactful project being conducted by a highly capable R&D team. The approach is innovative, and it serves as a new and important complement to related methods for methane storage and delivery in adsorbent systems.
- This is a fairly flexible method. The project uses the center resources effectively. The PI is open to suggestions for improvement, more so than most PIs.
- There is a strong team combining simulation and modeling with experiment.

Project weaknesses:

- Homogeneity of the material is lost in both densification and heteroatom addition. This may be overcome yet, so it is not an inherent weakness.
- This is a minor weakness: the project may want to avoid getting too closely wed to the highly idealized homogeneous model of nitrogen-/boron-doped sites.
- Maintaining structural homogeneity at high dopant/additive concentrations and pellet densification remains a serious challenge to efficient reversibility and cycling. Although overcoming this obstacle is being addressed, the effectiveness of the proposed approaches remains an open question.

Recommendations for additions/deletions to project scope:

- As pointed out in the future work section, three topics for additional work are recommended (as time and funding permit): (1) synthesis and characterization of a boron-doped ZTC sample and subsequent comparison with a nitrogen-doped sample to validate the first principles predictions, (2) a more detailed characterization of sample integrity and reversible capacity upon sorption cycling, and (3) use of solid-state NMR (PNNL) to probe chemical environments and sorption mechanisms in the doped ZTC system.
- The project should use polydisperse crystals to get higher packing with no need to compress the sample. Maybe the team could make melamine inside the zeolite, if the precursor used is small enough. The project might benefit from a look at precursor methods to make melamine that would be compatible with zeolites.

Project #ST-215: Hydrogen Materials–Advanced Research Consortium (HyMARC) Seedling: Developing A New Natural Gas Super-Absorbent Polymer

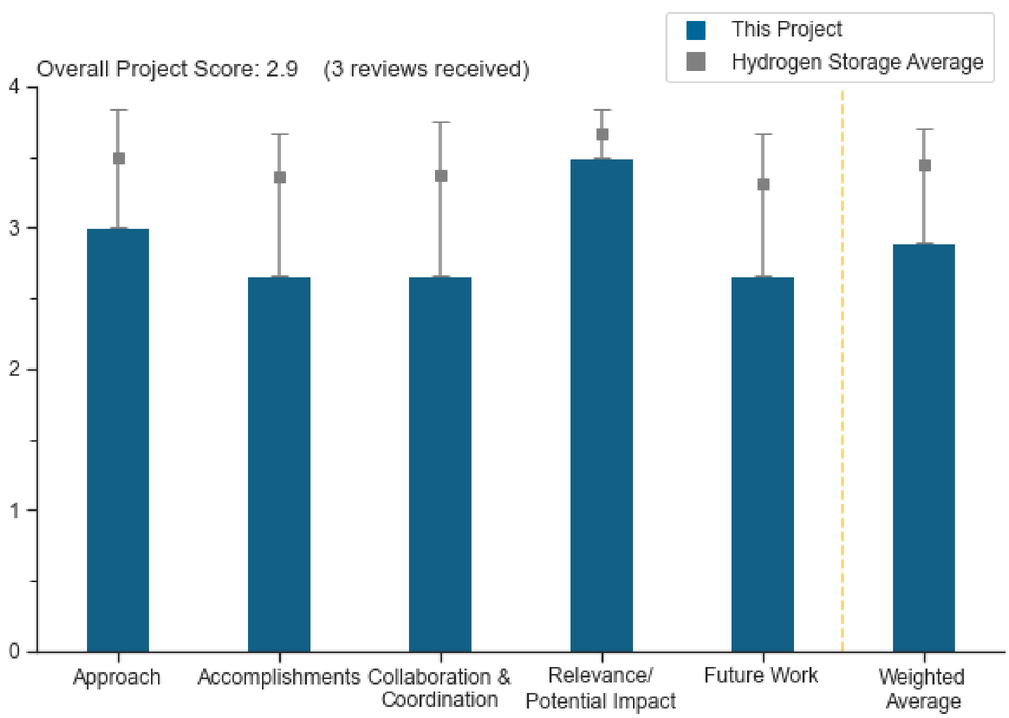
Mike Chung, The Pennsylvania State University

DOE Contract #	DE-EE0008811
Start and End Dates	10/1/2019 to 1/31/2023
Partners/Collaborators	HyMARC, National Renewable Energy Laboratory
Barriers Addressed	• Natural gas storage density, temperature, pressure • Polymer absorbent synthesis • High polymer surface area • Suitable natural gas binding energy • Charging/discharging rates

Project Goal and Brief Summary

Reduced-pressure natural gas storage in a materials-based system provides significant cost advantages over conventional liquefied or compressed natural gas storage. Such materials-based systems have the potential to reduce, or possibly eliminate, the need for expensive carbon fiber in natural gas fueling infrastructure. This project aims to synthesize new hydrocarbon polymers for use in materials-based storage systems for natural gas.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Storage projects.

Question 1: Approach to performing the work

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

- The project uses inexpensive materials with known affinity for higher hydrocarbons. The researchers adjust backbone and substituent and cross-link content to achieve the best uptake. They are considering the cost of materials as an important factor, so they have a good chance of making a commercial product if they get good performance. The project is attempting to make the expansion internal, which should help with functionality in real systems.
- The use of polycyclic hydrocarbon polymers is a novel approach for uptake and delivery of methane. Synthesis of both doped and undoped hydrocarbon polymers, characterization of methane binding energies, and sorption capacity are the primary elements of the project. The project provides a pathway to overcoming at least some of the barriers (especially limits to volumetric capacity). However, thus far, insufficient focus is placed on the important issues of methane sorption kinetics and reversibility/cycling in the polymer materials.
- The project goals are adequately defined, and important barriers are recognized. The experimental design appears to be somewhat chaotic; design rules for development of polymers that might lead to enhanced methane adsorption have not yet been communicated.

Question 2: Accomplishments and progress

This project was rated **2.7** for its accomplishments and progress toward overall project and U.S. Department of Energy (DOE) goals.

- Solid progress has been made in the synthesis and characterization of polycyclic aromatic networks having a variety of pore sizes and volumes and methane binding energies. A promising polycyclic boron-doped aromatic network candidate system (PAH – "B-pitch") has emerged from the initial studies. The methane binding energy (~18 kJ/mol) and volumetric energy density (~264 cm^3/cm^3) at 100 bar and room temperature are commensurate with a practical polymer-based methane storage system. However, there are many important issues concerning the suitability of the material for storage and delivery applications that must be addressed. These include, for example, what happens to the petrogel structure when the methane is liberated—i.e., whether the swelling is reversed. It is also unclear whether data are available concerning the sorption kinetics and cycling efficiency (i.e., although it is suggested that mesoporous channels in the petrogel matrix can facilitate fast kinetics and good cycling efficiency, no supporting data are provided). Also, since the polycyclic hydrocarbon swells dramatically during methane adsorption, packing density could be a serious issue. Thus far, the dominant emphasis has been on synthesis of hydrocarbon polymers. Future work must focus keenly on kinetics, reversibility, and the impacts of structural changes and packing loss issues on methane sorption efficiency.
- The researchers are focusing on making the swelling internal so that they get excellent kinetics and packaging. They are choosing side groups and cross-linking to accomplish this. The team will work with Pacific Northwest National Laboratory and do high-pressure methane nuclear magnetic resonance experiments to evaluate swelling. The project is getting 19–20 kJ/mol binding energy, which is a good range for 300 K operation, with 700 m^2/g area. Pitch was used for lower cost and better area up to 1800 m^2/g. The project does get 264 v/v at 295 K, but only about half is useable at the material level. The researchers still get roughly twice the capacity of pressure alone. It is somewhat hard to evaluate progress numerically based on the data presented. It is unclear whether they know how well (or not well) they are doing toward reaching goals.
- The progress has been satisfactory. It is unclear at this point that a logical progression in polymer structure and properties is being developed that can lead to logical design of polymers with improved methane sorption at high loading.

Question 3: Collaboration and coordination

This project was rated **2.7** for its engagement with and coordination of project partners and interaction with other entities.

- Useful collaborations with other investigators at The Pennsylvania State University and National Renewable Energy Laboratory (NREL) on design, synthesis, and characterization of new hydrocarbon materials are enhancing progress on the project. The principal investigator (PI) and his team are experts in hydrocarbon synthesis and characterization. The core HyMARC team could provide additional support and expertise for the essential work on kinetics and reversibility. The project seems to be well-managed and -coordinated, and detailed milestones and go/no-go decision points have been formulated.
- The collaboration is adequate for this work; it is mostly internal, with verification at NREL.
- While it is early days in this project, little detail was given as to what the plan is for collaborating with NREL to validate methane adsorption in polymeric systems, particularly where there may be issues of swelling.

Question 4: Relevance/potential impact

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

- High-capacity storage and efficient delivery of methane at suitable temperatures and pressures are important DOE research, development, and demonstration objectives. Although this innovative project provides a path to achieving the goals, many questions remain, and they must be addressed in future work. Overall, the project has potential to advance progress on methane storage and to successfully address limitations of incumbent approaches.
- Natural gas storage at lower pressure is part of this Hydrogen and Fuel Cell Technologies Office/Vehicle Technologies Office area. The project is aligned.
- The goals of the project are well-aligned with DOE Hydrogen Program and Vehicle Technologies Office goals.

Question 5: Proposed future work

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

- The future work is a reasonable extension of the prior studies. However, the future work statement lacks sufficient detail to assess what will actually be done to achieve the stated objectives given in the summary. The most critical concern is the limited information regarding plans for evaluating and improving kinetics, reversibility, and cycling and packing density. These are vital project needs that require a thoughtful and detailed research plan. Also, a critical evaluation of major technical risks and mitigation strategies is needed.
- Listed areas are all valuable but seem to be sort of a scatter, with no real plan.
- A list of areas to investigate was presented without sufficient rationale or prioritization. It is difficult to tell what the specifics of the future experimental plan are going to be.

Project strengths:

- The PI and his team have considerable expertise and background in synthesis and testing of the novel methane sorption materials being developed in the project. The approach is innovative and has the potential to meet many of the DOE objectives for methane storage.
- The approach in using organic polymers for methane sorption is leveraged off of prior work in hydrocarbon sorption. The approach complements other Hydrogen Program projects that are focused on metal–organic frameworks and other porous materials approaches.
- This is based on previously proven technology and uses low-cost materials.

Project weaknesses:

- The project focus thus far has been almost entirely on materials synthesis and characterization. As stated above, the critical issues of sorption kinetics, reversibility, cycling efficiency, and impact of large structural changes on packing density and capacity have not been adequately addressed. Moreover, detailed plans to address these issues have not been provided.
- It is hard to numerically evaluate progress. Results are mostly qualitative with regard to gas uptake and release. The plan underlying the work is either loose or not well-communicated.
- The experimental design is not well-described and appeared rather chaotic.

Recommendations for additions/deletions to project scope:

- The project might be able to use neutron scattering to evaluate change in shape. The project needs to understand cycling durability and the impact of higher hydrocarbons and S and P and Si impurities, and the team needs to increase bulk density of the material and increase, or at least not lose, capacity in crystallites.
- As pointed out in earlier sections of this review, a keener focus on methane sorption kinetics, cycling efficiency, and the impact of large structural changes during sorption is strongly recommended.
- The project needs to accelerate the validation of sorption versus loading at NREL.

Project #ST-216: Hydrogen Materials–Advanced Research Consortium (HyMARC) Seedling: Hydrogen Release from Concentrated Media with Reusable Catalysts

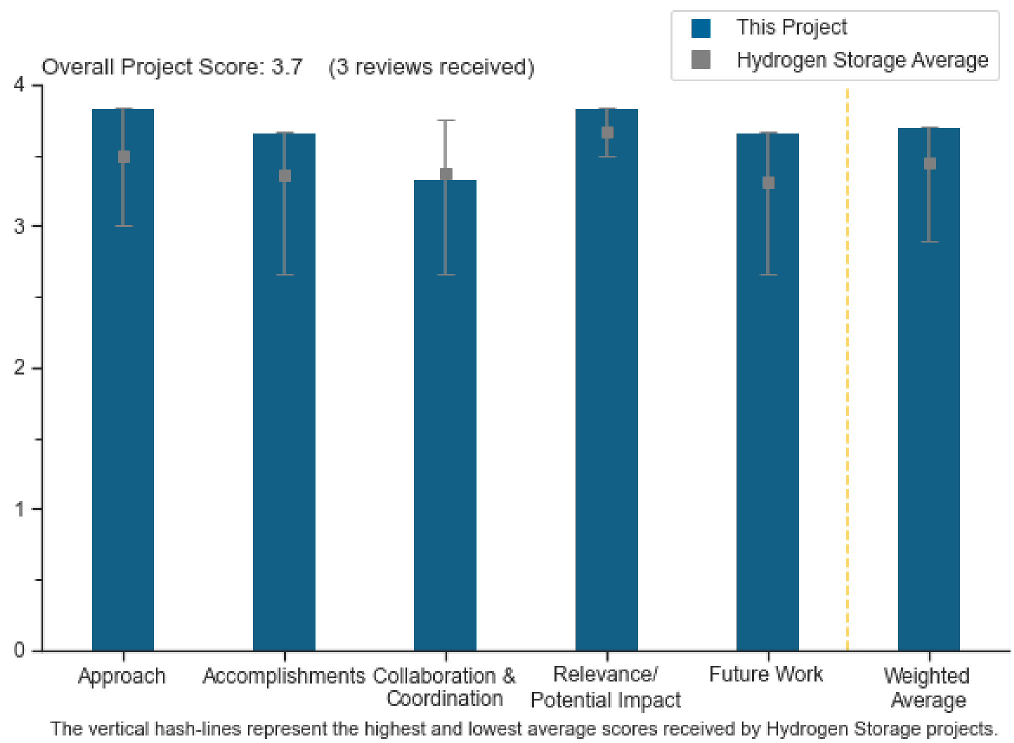
Travis Williams, University of Southern California

DOE Contract #	DE-EE0008825
Start and End Dates	10/1/2019 to 3/31/2023
Partners/Collaborators	Los Alamos National Laboratory
Barriers Addressed	• Generate hydrogen from formic acid: increase reaction scale, demonstrate hydrogen throughput, and remove CO_2 from output stream • Apply technology to blended fuels • Understand the molecular mechanism

Project Goal and Brief Summary

Hydrogen carriers such as formic acid have the potential to improve hydrogen delivery and storage pathways over existing compressed or liquid methods. This project aims to demonstrate on-demand hydrogen evolution from formic acid and formic acid fuel blends using a demonstration-scale flow reactor. Researchers will conduct molecular mechanistic studies to optimize the catalyst and fuel blend. A successful project outcome will further the ability to make hydrogen fuel available in distributed locations, which is vital for transportation applications.

Project Scoring



Question 1: Approach to performing the work

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

- This is a good approach; using a catalyst is logical, and if it is regenerable, the use of Ir is only an upfront cost. Developing operating conditions is critical and is a good plan. The project is also looking for better catalysts and is building a pilot reactor to look for scale-up issues.
- The principal investigator has a very thorough understanding of what the barriers are and how to approach the landscape of experiments to make solid progress. The project is well-designed, well-thought-out, and well-executed.
- The project addresses relevant barriers and objectives.

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.

- Substantial progress on the very basic science aspects of the combined theory and mechanistic work has led to significant progress on the experimental front. The project is rapidly moving from small-scale batch reactors to moderate-scale continuous flow reactors to enable the exploration of the more applied aspects of catalyst durability, recovery, and hydrogen stream purification, among others. The high-level mechanistic work has led to the realization that the initial catalyst was modified at higher-temperature operation conditions and accessed a wholly new reaction manifold that could dehydrogenate mixed methanol/formic acid to generate hydrogen. The recent progress with regard to the project milestones not only has been met but also has far exceeded the researchers' expectations. Certain aspects of this progress have been validated at Pacific Northwest National Laboratory (PNNL). Good progress is being made in constructing a flow reactor to explore this chemistry at higher throughputs in a continuous mode more closely allied with future applications. This is daunting, as the safety requirements are strict for this sort of activity and the levels of hydrogen production anticipated. The project leadership is also highly cognizant and capable of forming the technology transfer opportunities this project presents.
- The project achieved over 3.9 L/hour at low conversion, which was well over the go/no-go criterion. PNNL demonstrated a rate of over 100 L/hour at pressure. Los Alamos National Laboratory (LANL) almost has the continuous reactor ready; this will be a key test. There is progress toward methanol tolerance. No data are presented, but the project may be able to use a carbene version of the catalyst at pressure (but not at low pressure). This ought to lower synthesis cost.
- No plots of hydrogen release versus temperature (T) and versus pressure (P) were presented. It is unclear whether these have been conducted. The presentation also did not include measurements or estimates of the efficiency for regeneration of the liquid carriers.

Question 3: Collaboration and coordination

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

- The collaboration with computational chemists at LANL has nicely augmented the University of Southern California mechanistic work, which has helped to move the project rapidly forward. The collaboration with the chemical engineering capability at LANL to assist in the design of the continuous flow reactor has also accelerated progress. This collaboration may also enhance the CO_2 separation tasks that lie ahead. Collaboration with scientists at PNNL to validate and confirm the reactivity and throughput at a somewhat higher scale also helped to advance the project in its early stages.
- There is good coordination with LANL. The project is also tied into PNNL for testing.
- LANL is a formal partner. PNNL is a collaborator.

Question 4: Relevance/potential impact

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

- Formic acid is a compelling hydrogen carrier by mass and has excellent thermodynamics from an energy consumption point of view, and the cost is modest. It could be implemented in bulk fairly readily if the release portion of the cycle could be figured out; thus, this is clearly aligned.
- The project is closely aligned with the objectives of the DOE Hydrogen Program and supports the work in developing hydrogen carriers for a variety of use cases. This work can lead to reductions in costs of delivered hydrogen by enabling the transport and eventual conversion to hydrogen at higher pressure, avoiding some compression costs for certain use cases.
- A viable liquid carrier for hydrogen would be a major breakthrough.

Question 5: Proposed future work

This project was rated **3.7** for effective and logical planning.

- This plan is sound; developing the flow reactor is critical and a high priority in the researchers' minds. It is important to show durability at peak rate concentration in a pressurized flow reactor. If they can run continuously at a high degree of conversion (and so high catalyst density), they will greatly increase their rate. The second critical issue is CO_2 separation from hydrogen. This is also a good issue to start solving.
- The future work flows logically from the progress to date and addresses key issues such as the development of a continuous process for the generation of hydrogen at pressure, the use of mixed methanol/formic acid fuels, catalyst durability under continuous processes, and hydrogen purification from CO_2 in a continuous process.
- No mention of regeneration efficiency is mentioned.

Project strengths:

- The catalyst has high rate at moderate temperature and generates high-pressure hydrogen. The catalyst seems to have good durability. The researchers are working with a commercial product in mind, so they are taking on the right issues. They are making a pilot-scale reactor to look at something closer to the intended system in commercial use.
- The project has a highly motivated and capable team and collaborators.
- This is a relevant project focused on an important goal.

Project weaknesses:

- Co-generation of CO_2 and hydrogen requires separation, which may be expensive.
- The apparent lack of data on hydrogen release versus T and P is a weakness; regeneration efficiency is unclear, and plans to measure efficiency are not communicated.

Recommendations for additions/deletions to project scope:

- The project should measure hydrogen release versus T and P and regeneration efficiency.

Project #ST-217: Hydrogen Materials–Advanced Research Consortium (HyMARC) Seedling: A Reversible Liquid Hydrogen Carrier System Based on Ammonium Formate and Captured Carbon Dioxide

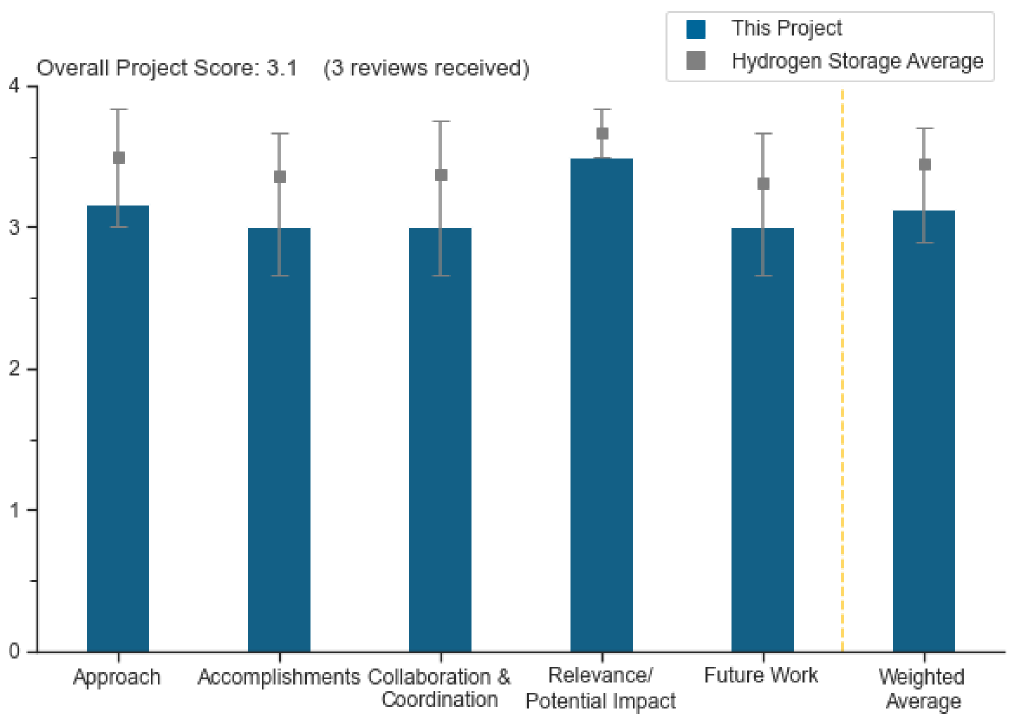
Hongfei Lin, Washington State University

DOE Contract #	DE-EE0008826
Start and End Dates	10/1/2019 to 1/31/2023
Partners/Collaborators	8 Rivers
Barriers Addressed	• Catalyst cost • Energy efficiency • Durability

Project Goal and Brief Summary

This project aims to build a prototype ammonium formate-based hydrogen uptake and release system and evaluate its technoeconomic potential for commercialization. If successful, this project will develop and demonstrate a new generation of hydrogenation/dehydrogenation catalysts superior to commercially available catalysts. Washington State University is collaborating with 8 Rivers and members of the Hydrogen Materials Advanced Research Consortium (HyMARC) on this project.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Storage projects.

Question 1: Approach to performing the work

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

- This project's approach includes a technoeconomic analysis (TEA) of hydrogen production from a liquid carrier, ammonium formate, and will assess a baseline cost for the hydrogen generated. As such, this project supports the objectives of the U.S. Department of Energy Hydrogen Program to drive toward reducing the cost of hydrogen and providing for a pathway for transportation of hydrogen in the form of a hydrogen carrier.
- This project has a good approach using parallel development of the catalyst system, creating both the support and the active metal/alloy.
- The project addresses important barriers.

Question 2: Accomplishments and progress

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

- The project's progress is good. The project developed a support that uses half the active material but evolves more hydrogen, developed other supports and dispersions and selected one with 88% yield and stability over at least five runs, and developed a bimetal catalyst using metal that is 100 times cheaper than palladium with good stability in six runs and good hydrogen yield. The researchers think they are near that best alloy now. Their studies suggest deactivation could be due to leaching, but they see no clear evidence.
- The TEA has been delayed by COVID-19; the focus of the project to date has been on catalyst optimization. Without the guidance of a baseline TEA, it would appear to be difficult to know where the research and development (R&D) emphasis needs to be focused. It could be that catalyst development, optimization, and characterization is premature.
- The data presented did not appear to include measures of hydrogen release versus temperature and pressure. Measurements or estimations of reversibility and the efficiency of regeneration also appeared to be absent from the presentation. It is difficult to assess the promise of this approach without these data.

Question 3: Collaboration and coordination

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

- There is decent cooperation inside the team, and it is getting help from Pacific Northwest National Laboratory (PNNL).
- Monthly conference calls with PNNL/HyMARC are occurring; it is unclear what the impact of these meetings have been from the materials presented.
- Some collaborations with Lawrence Livermore National Laboratory were noted. It is unclear what 8 Rivers is contributing to the project; no TEA was presented.

Question 4: Relevance/potential impact

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

- The potential impact of the proposed R&D could be high if successfully and logically executed. Developing cost-effective hydrogen carriers for certain use cases is relevant to the Hydrogen Program's objectives. In practice, the impact of this project will depend upon how well the TEA is performed, what the baseline cost for delivered hydrogen via this carrier system is determined to be, and to what extent additional identified R&D can impact that cost.

- Formate carriers could help deliver hydrogen over long distances. This aligns well.
- A viable liquid hydrogen carrier would be an impactful development.

Question 5: Proposed future work

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

- The plan hits the main needs: try to lower or remove palladium by better understanding the mechanism and using that understanding to seek alternate pathways; optimize ammonium formate concentration to get the best performance; TEA to look at where cost needs to come out; determine rate-limiting step and what the catalyst needs to do to help that. Looking at the flow reactor data will help the researchers understand the true nature of reaction and durability.
- The incorporation of the reactor work should provide data that supports the TEA in providing kinetics data, information on the concentration of impurities and carbon dioxide separation, and information on catalyst deactivation/regeneration, all of which can affect overall costs but also focus future work.
- The project needs measurements of hydrogen release versus temperature and pressure, regeneration efficiency measurements, and TEAs.

Project strengths:

- Lowering platinum group metal content and simultaneously developing support and active material to get a well-tailored catalytic system are strengths. The team is well situated to do its respective tasks, catalyst development, and TEA.
- This is an interesting system to explore as a hydrogen carrier that is potentially of high capacity and can deliver hydrogen at high rates at some pressure that is to be determined.
- The project is focusing on a promising material.

Project weaknesses:

- The focus on catalyst refinements appears premature in the absence of guidance from a baseline TEA.
- Many key measurements and analyses have not yet been performed. There is no mention of these in the future work statements.
- There is no ability to look at technical scale-up issues. There is no meaningful data on the durability of the catalyst, which may lead the team down an avenue with modest up-front cost but no chance of good durability, resulting in a system that is thus economically untenable overall.

Recommendations for additions/deletions to project scope:

- Start generating data on conversion and kinetics (rate) versus cycles or hydrogen produced since the start of the test. That data can tell the project if the catalyst under test has suitable durability. A high-level TEA should be able to give a target that the project must beat.
- The project should accelerate the baseline TEA and identify the major cost drivers based on the currently available catalyst and product stream characteristics/impurities. The project should pay attention to impurities other than CO_2 in the evolved gas stream. Separations costs can be a major contributor to the overall cost of a process, and any issues surrounding this can have a significant impact on cost if no mitigation strategies are put in place.

Project #ST-218: Hydrogen Materials–Advanced Research Consortium (HyMARC) Seedling: High-Capacity, Step-Shaped Hydrogen Adsorption in Robust, Pore-Gating Zeolitic Imidazolate Frameworks

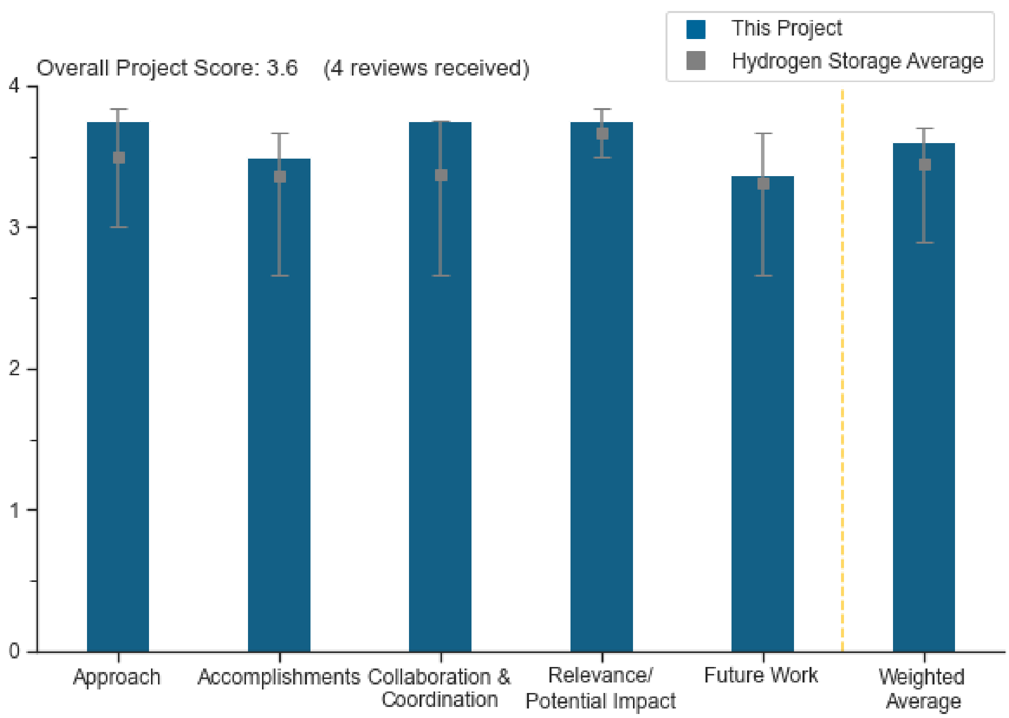
Michael McGuirk, Colorado School of Mines

DOE Contract #	DE-EE0008823
Start and End Dates	2/27/2020 to 2/28/2023
Partners/Collaborators	National Renewable Energy Laboratory, Lawrence Livermore National Laboratory, National Institute of Standards and Technology, SLAC National Accelerator Laboratory
Barriers Addressed	• The cost of producing and delivering hydrogen from zero- or near-zero-carbon sources must be reduced • Compact, lightweight, and low-cost hydrogen storage systems must be developed

Project Goal and Brief Summary

Current approaches to hydrogen transport and delivery entail extreme pressures or cryogenic liquefaction—both energy-intensive processes that increase costs. An alternative is using porous adsorbents that can densify hydrogen under milder conditions by providing enhanced surface area for hydrogen molecular adsorption. However, most porous adsorbents adsorb hydrogen most strongly at low pressures and temperatures. This project is exploring stimulus-responsive porous adsorbents that, through step-shaped adsorption–desorption profiles, can deliver their entire adsorbed capacity with minimal energetic input. These materials could store large quantities of hydrogen under mild conditions, as well as transport and deliver hydrogen with only small swings in pressure and temperature.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Storage projects.

Question 1: Approach to performing the work

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

- The approach adopted for the development and implementation of "stimulus-responsive porous adsorbent" materials for high-capacity hydrogen transport and delivery is novel and innovative. This project exploits the unique characteristics of selected zeolitic imidazolate frameworks (ZIFs) that undergo reversible porous/non-porous structural transitions to generate step-shaped sorption profiles. The work in progress includes sorption measurements in baseline systems and modification of organic linkers to facilitate step-shaped sorption in a model system (CdIF-13 [cadmium imidazolate framework 13]). This work should provide a foundation for future efforts devoted to optimizing adsorption and desorption properties in relevant pressure regimes. Overall, this is an exciting approach that can potentially overcome many of the barriers faced by other (rigid) porous-adsorbent approaches for high-capacity hydrogen storage and delivery.
- This project has an excellent approach. The use of flexible metal–organic frameworks (MOFs) offers a possibly unique opportunity to greatly increase usable capacity (by all but eliminating the five bar capacity, so total capacity and usable capacity almost match). There is a suitable plan to work from known flexible MOFs toward ones with higher capacity. Also, it is a good plan to tune opening pressure and capacity via new linkers and adjusting the shape of the adsorption curve with metal center substitution. The use of a family of Cd-based ZIFs to learn more about how metal impacts these materials' functional properties is probably a good plan; certainly there is every reason to think that valuable knowledge will come from this approach. The key is to gain sufficient knowledge that can be used (with a high probability of success) to design a high-functioning material with low safety risk.
- This is a high-risk, high-reward opportunity that leverages what has been learned from methane adsorption in pore-gating MOFs and attempts to port that to the adsorption of hydrogen. The project is focused on the critical barriers of employing pore-gating MOFs to enhancing hydrogen sorption above and beyond conventional MOFs, potentially reducing the cost of storing and delivering hydrogen.
- The objectives and critical barriers are clearly defined and are being addressed.

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.

- Significant progress has been made in 2020 and 2021. Notably, the first direct proof of step-shaped hydrogen adsorption validated the principal rationale/hypothesis for the project and provided a solid basis for future work. Neutron and x-ray diffraction diagnostics provided insight into the structural changes that accompany step-shaped hydrogen sorption processes. In situ, variable temperature neutron diffraction proved to be a powerful tool to identify primary adsorption sites. In addition, computational modeling was used effectively to identify possible intermediate structures present during the phase change in CdIF-13 and ZIF-7 model systems. The progress to date is impressive, and it inspires confidence that this approach will ultimately result in superior material systems for hydrogen storage and delivery. On another note, packing density is often an issue for adsorbent systems capable of meeting goals for hydrogen capacity. It would help to know whether the ZIF systems studied in this work require increased packing density to achieve acceptable storage and delivery capacity and performance.
- This project has made nice progress. It showed that it could get a stepped adsorption curve with ZIF-7. The best performance occurred at 100 K and 110 K. The project made 13 members of the CdIF-13 ZIF family ahead of schedule. It is anticipated that these members will have a flatter pre-step adsorption (relative to ZIF-7) based on a structure that more tightly closes the "door" of the ZIF in the non-activated state. The researchers were able to show with propane that they could tune the threshold pressure by changing the ratio of linkers. They were also able to show that the ratio of fluorinated linkers can greatly shift the pressure of the step. The team hopes that the Jeff Long laboratory can validate the results. Using x-ray and neutron diffraction, the team also measured the exact structure in open and closed formations in situ. It identified adsorption sites before and after opening the structure. It showed that the material functions as hypothesized, and it validated the concept that the electron-rich substituents on linkers increase the heat of

adsorption. The project modeled the energy of intermediate states, which cannot be captured experimentally.

- This project has moved forward in a logical, well-thought-out manner. Delays due to COVID-19 have largely been mitigated, which is a remarkable accomplishment on its own. The effort in the syntheses of key materials has been very productive, and the project has demonstrated step-like adsorption of hydrogen—a major milestone. There are some issues regarding access to the Hydrogen Materials–Advanced Research Consortium (HyMARC) high-pressure hydrogen adsorption capabilities that are temporarily offline; the principal investigator (PI) is actively looking at workarounds. The team has solved single crystal x-ray and powder neutron diffraction analyses of key materials, including some in various states of adsorption, in collaboration with the National Institute of Standards and Technology (NIST). These measurements provide some valuable details as to the structural/energetic landscape along the trajectories of hydrogen adsorption in these materials that provide guidance to future experimentation. The collaboration with the HyMARC simulation and modeling capability at Lawrence Livermore National Laboratory (LLNL) has provided insight into the energetics of adsorption at various gas loadings that will also help direct materials modification efforts.
- This project was significantly slowed down by COVID-19. Some good progress on synthesis has been made. The delays in staffing the project and in measurements of hydrogen uptake were noted. The first go/no-go milestone has not yet been assessed due to these delays. The team should not be penalized for these delays, as the delays are beyond the team's control.

Question 3: Collaboration and coordination

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

- Extensive and valuable collaborations with HyMARC investigators at the National Renewable Energy Laboratory (NREL), NIST, LLNL, and Stanford Linear Accelerator Center (SLAC) are evident. The numerous contributions from those collaborating institutions are significantly enhancing progress on both the experimental and computational modeling aspects of the project. The collaborations are well coordinated, and the overall project is well managed. The core research and development team and collaborating researchers are highly capable, having expertise in all relevant areas of this seedling project. Collaboration with Pacific Northwest National Laboratory (PNNL) on the characterization of transition intermediates using in situ nuclear magnetic resonance (NMR) is recommended. A HyMARC project on MOFs with step-shaped adsorption isotherms has been initiated (see slide 12 in Annual Merit Review 2021 presentation ST-127). Discussions with the HyMARC investigators on how that project might relate to the present work could be useful.
- This project has collaboration in the best sense with some of the best in the business, such as Brandon Wood for theoretical guidance and Craig Brown for powder neutron diffraction of structure.
- This is a well-integrated team of collaborators utilizing key HyMARC capabilities to accelerate the progress of this seedling project.
- Many collaborations exist in this project and it is on the right track.

Question 4: Relevance/potential impact

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

- The project is highly aligned; it is developing an innovative way to get a lot more hydrogen in a MOF at a higher temperature, which would be a direct enabler for solid material hydrogen storage to compete with compressed gas on a mass cost and volume basis.
- This is an exciting seedling project that has direct relevance to DOE research, development, and demonstration objectives, and it could significantly advance progress toward meeting DOE hydrogen storage and delivery goals. The project complements other hydrogen adsorbent work supported by DOE; however, the novel approach adopted here could offer significant advantages over approaches implemented in related efforts.

- If step-like adsorption of hydrogen in these pore-gated materials can be realized at significant capacities, this will provide a pathway for greater overall useable hydrogen capacity. This can impact the cost of stored and delivered hydrogen, which are factors that are key objectives to the DOE Hydrogen Program (the Program).
- This is a relevant project with good potential for impact.

Question 5: Proposed future work

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

- The proposed experimental and computational efforts are a logical and well-formulated extension of the current work. The future work on tuning the step-shaped behavior is especially important because it provides a solid basis for understanding the hydrogen-induced stimulus response and tailoring the systems having improved performance. The project milestones and an appropriate go/no-go decision point are clearly stated. The project has the potential to overcome many of the notable barriers facing high-capacity storage and delivery in adsorbent systems. A recommendation for future work is to include experiments designed to probe the reaction and structural intermediates during the step-shaped transition in the future work plan. That work could provide valuable insight concerning the transition mechanism. The additional experiments might include, for example, in situ solid-state NMR (maybe in collaboration with PNNL) or some other structural or optical diagnostic capable of providing either time-resolved or "stopped-flow" information during the transition.
- This project has good plans for future work. The key plan is making new MOFs guided by the theory that they should operate at higher temperature and with a better useful capacity and lower pressure peak capacity. The work at SLAC will help the team understand the opening and closing of the new materials. The team feels that if it can get the computations going based on the data in hand, it can accelerate the progress. Also, the project needs the NREL facility to reopen or to get another source of high-pressure hydrogen testing.
- The work on understanding the phase-change transitions is anticipated to help guide the synthesis effort to explore opportunities to increase hydrogen sorption capacities to an even greater extent. The combination of theory-guided experiment involving an LLNL/HyMARC collaboration and in situ diffraction at SLAC is a well-thought-out plan. Replacing Cd with a more benign cation is perhaps desirable, but perhaps lesser priority. It is also a higher-risk activity in that it is unknown at this point whether these same structure types are accessible via other, more electropositive cations as proposed.
- The project needs some extra time to catch up after the COVID-19 slow-down.

Project strengths:

- This is a first-rate project that is innovative and well formulated. The PI and his team have expertise in all relevant project areas, and solid progress is being made on demonstrating and optimizing the novel step-transition behavior in ZIFs for improved hydrogen storage and delivery.
- The project's strength is the use of these rare but special structures to eliminate residual hydrogen in the 'empty' state. It has a really superior supporting team in theory and spectroscopy.
- This is a very well-thought-out, planned, and executed project that is highly relevant to the Program objectives. It has very effective use of HyMARC and external collaborations at LLNL and NIST, and in the future at SLAC.
- The project has good collaborations and a sound scientific approach.

Project weaknesses:

- In this reviewer's opinion, there are no notable weaknesses or deficiencies.
- The project is focused on a group of materials that will teach the team about what makes a better material, but those materials under test are clearly not the ones the team seeks. Thus, the team is taking the bet that it will learn enough so that at the end it can pull a high-quality material out of the hat; that may not be the case, though.

Recommendations for additions/deletions to project scope:

- Two questions/issues could form the basis for additions to the project scope: (1) if increased packing density is required in order to achieve adequate hydrogen capacity, an experimental plan should be developed and described; (2) the ability to probe intermediate states during the step transition could provide powerful and useful information concerning the step-transition mechanism. A possible collaboration with PNNL (in situ NMR) or another organization capable of performing time-resolved or "stop-flow" diagnostics might be considered.
- The work needs to focus on getting the data the theory team needs to validate models and generate high-probability guidance toward high-capacity metals and linkers.
- A minor recommendation is to think about the priority of the cation-replacement tasks; it could be that the effort is better expended on the framework/ligand modification tasks and characterization.

Project #ST-223: Cost Assessment and Evaluation of Liquid Hydrogen Storage for Medium- and Heavy-Duty Transportation Applications

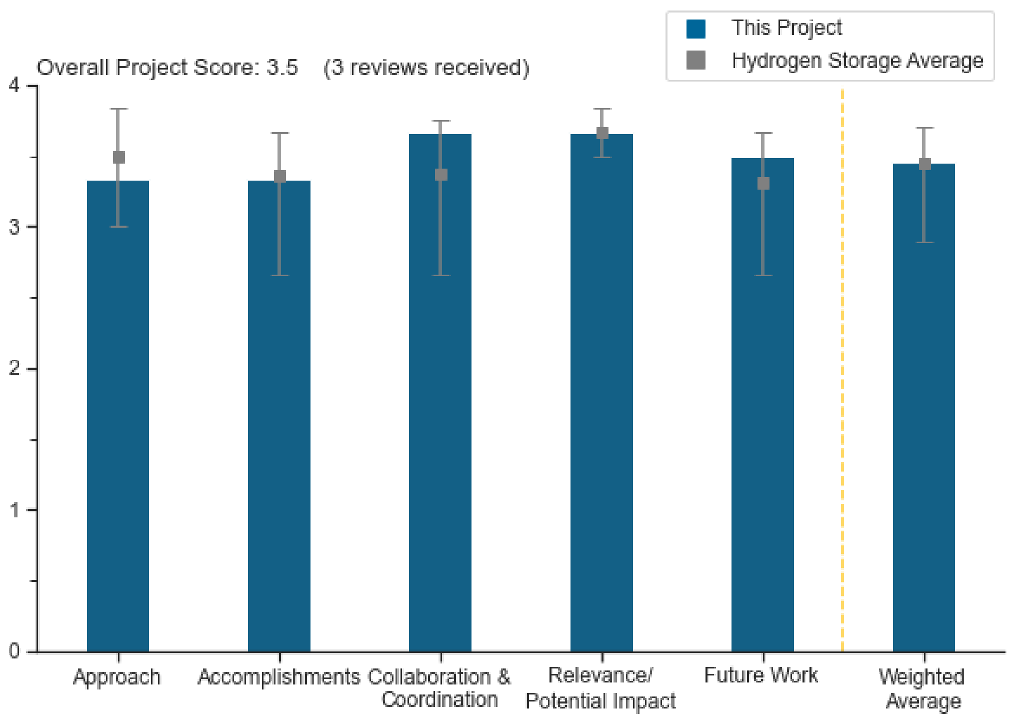
Rajesh Ahluwalia, Argonne National Laboratory

DOE Contract #	4.4.0.2
Start and End Dates	10/1/2020
Partners/Collaborators	Lawrence Livermore National Laboratory, Sandia National Laboratories, Strategic Analysis
Barriers Addressed	• System weight and volume • System cost • Efficiency • Charging/discharging rates • Thermal management • Lifecycle assessments

Project Goal and Brief Summary

This project will analyze the cost and performance of onboard liquid hydrogen (LH2) storage concepts for heavy-duty trucks. The analyses will look at capacity, insulation and dormancy, refueling rate, and hydrogen venting loss. The project will explore the design parameters best suited to at least three different heavy-duty truck vocations to inform the design of LH2 storage systems optimized for the needs of medium- and heavy-duty trucks. Argonne National Laboratory is collaborating with Lawrence Livermore National Laboratory, Sandia National Laboratories, Strategic Analysis, Inc., Air Liquide, Cummins, General Electric, and Navistar on this project.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Storage projects.

Question 1: Approach to performing the work

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

- Fuel cell electric vehicle (FCEV) use is moving toward heavy-duty (HD) applications; understanding the capability and limits of various storage methods is essential to enabling this sector. Evaluation of LH2 in this area is of practical interest.
- As fuel cells become of more interest to heavy-duty vehicles (HDVs), larger storage capacity is also becoming increasingly critical. LH2 is a clear possible solution to meet HDV range requirements. Understanding the cost of large on-vehicle LH2 storage is needed to help determine the correct selection of vehicle storage system.
- The project developed a systematic approach for system analysis of Type 1 vacuum-insulated cryogenic vessels for LH2 storage systems for medium-duty and HD trucks. The project also developed an ABAQUS finite-element analysis of liner and shell failure modes, liner/tank materials of construction, sloshing behavior, and tank weight. The project should provide necessary explanations of basic assumptions and some terms ($\delta_{\text{Liner mass}}$, $\sigma_{\text{allowable}}$, etc.) used for the analysis in the appendix for the reviewers.

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.

- As usual, Rajesh never disappoints in delivering consistent and strong results; there is often too much data to absorb in 15 minutes.
- The project demonstrated excellent progress toward the objectives with several achievements.
- The project only started in October 2020, so the results of the work that could be used by industry for selection of systems were not available for the DOE Hydrogen Program Annual Merit Review. However, evaluation of various system design requirements such as with or without a pump, vessel design, balance of plant, etc., should be on the path to finish work within the remaining time in the project.

Question 3: Collaboration and coordination

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

- There is a good selection of industry and institute collaboration to identify critical considerations in design. There is good coordination in and between laboratories to divide tasks and provide feedback to the group.
- Rajesh always does a great job of coordinating and aggregating data from all the available data inputs from DOE and industry.
- The team showed strong collaboration with a well-organized task assignment as shown on page 18.

Question 4: Relevance/potential impact

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

- Gaseous hydrogen (GH2) and LH2 are still the predominant onboard storage solutions for vehicles in the near term. Understanding LH2 cost benefits and challenges is critical for development of HDVs in the fuel cell applications.
- The project aligns very well with the DOE objectives. One suggestion is to show the number or percentages of different types of medium-duty vehicles (MDVs) and HDVs in the current market to ensure the analysis is focusing on the major types.
- Focus on HD applications is where the FCEV market is heading. This project work is timely.

Question 5: Proposed future work

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

- Understanding boil-off given the different usage scenarios is the next practical step. DOE has significant history in evaluating these criteria. Rajesh will use the best available data to deliver on his next presentation.
- The scope of future work is appropriate and important for the HD truck industry to make decisions for hydrogen storage systems until better technology than GH2 or LH2 is developed.
- It is suggested to include a hazard identification analysis for the safety assessment.

Project strengths:

- Doing ground-up analysis for systems larger than those that have been used for passenger vehicles is very beneficial to the HDV industry as they move into fuel cells. The project does a good job at considering various requirements and challenges of LH2 in the HD environment.
- The project developed a systematic approach and advanced analysis methodologies, showed strong teamwork, and achieved several significant accomplishments.
- There is strong modeling by a very competent and experienced team to deliver useful models in a relevant subject.

Project weaknesses:

- As a summary of the previous comments, the project can be further improved from the following points. (1) It is suggested to provide necessary explanations of basic assumptions and some terms ($\delta_{\text{Liner mass}}$, $\sigma_{\text{allowable}}$, etc.) used for the analysis in the appendix for the reviewers. (2) For the stress analysis and effects of sloshing studies, as the speed of vehicles may significantly increase the stress and sloshing effects, it is suggested to carry out the analysis and studies under different vehicle speeds. (3) On page 6, different LH2 storage systems (size, mounting methods) are provided. It is suggested to carry out the stress analysis and effects of sloshing studies under different mounting methods and tank volumes. (4) It is suggested to use a table to compare the features and parameters of the two systems with and without pump on pages 7 and 8. (5) The percentage of LH2 in the tank during the dormancy for the analysis should be explained on page 9. (6) On page 5, the engine-off period in the third bullet point for semi-trailer truck is different from the number in the red summary box; this should be corrected. (7) On page 10, the definition of usable hydrogen in this study should be provided. (8) It is suggested to provide the number or percentages of different types of MDV and HDV in the current market to ensure the analysis is focusing on the major types. (9) It is suggested to include a hazard identification analysis for the safety assessment.
- Only looking at the storage system and not considering refueling station issues/cost and fuel cost is a weakness. This might give a skewed picture of total cost of ownership for HDVs.
- There is too much information to be presented in 15 minutes. Rajesh needs to focus a bit more at the beginning and end doing the boil-down of the results and provide context to the results in terms of how they relate to the targets and what industry needs to do to get to targets based on his model outcomes.

Recommendations for additions/deletions to project scope:

- The two tanks shown in the analysis don’t seem to be the maximum amount of fuel that can be stored on the vehicle. It would be interesting to establish the bookmark of what the maximum fuel storage amount could be, what range that gets, and if that’s useful to the industry given the lack of infrastructure availability, or if it would be excessive given the cost or length of routes run. Surely the deciding factors for the sizing have already been discussed at length and were perhaps presented in other projects, but it would be nice to provide some quick context up front as to why that particular sizing was selected.
- It would be beneficial to see a comparison to GH2 systems with total cost of ownership as a reference.

Project #ST-227: Integrated Onsite Waste-Heat-Driven Hydrogen Carrier System for Steel and Renewables

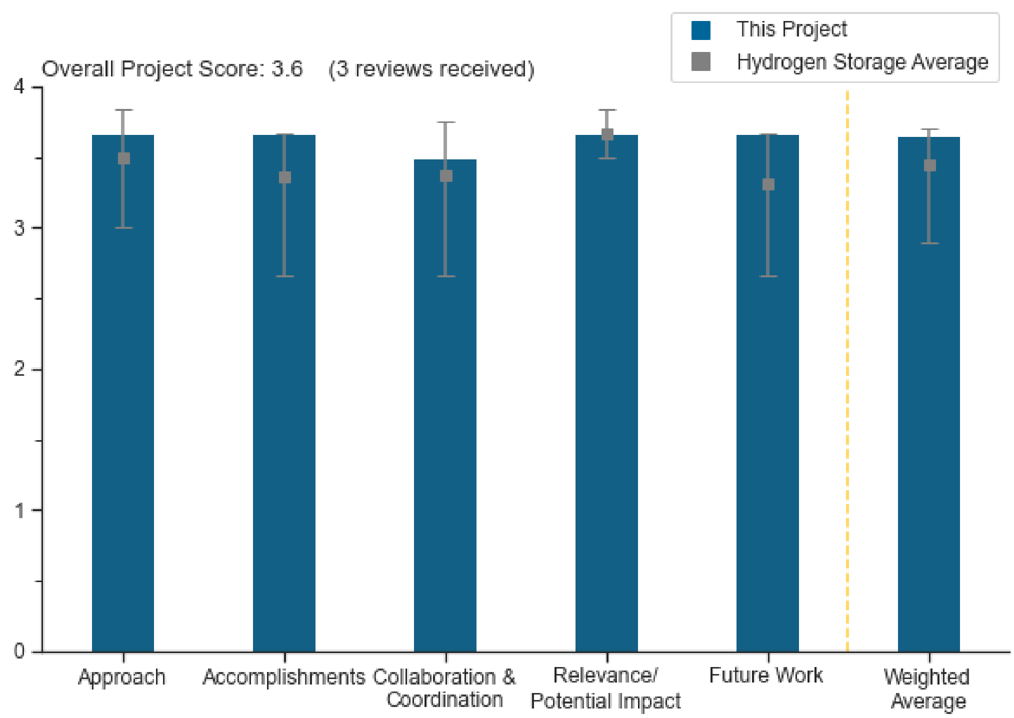
Hanna Breunig, Lawrence Berkeley National Laboratory

DOE Contract #	4.4.0.204
Start and End Dates	10/1/2020
Partners/Collaborators	Lawrence Berkeley National Laboratory, Argonne National Laboratory, Pacific Northwest National Laboratory
Barriers Addressed	• System cost • Efficiency • Codes and standards • Thermal management • System lifecycle assessments

Project Goal and Brief Summary

Traditional iron ore reduction creates significant amounts of carbon dioxide. When hydrogen replaces carbon monoxide in iron ore reduction, the only byproduct is water vapor. However, iron reduction with hydrogen has been demonstrated only at pilot scale. This project aims to develop and use models to analyze the performance and cost of a methylcyclohexane (MCH)-based hydrogen storage system for delivering hydrogen to iron and steel processes. If successful, this project will verify the feasibility of renewable hydrogen in iron and steel processes, enabling a more resilient, efficient, and low-carbon industry. Lawrence Berkeley National Laboratory (LBNL) is collaborating with Argonne National Laboratory (ANL) and Pacific Northwest National Laboratory (PNNL) on this project.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Storage projects.

Question 1: Approach to performing the work

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

- Systems analysis is important to evaluate hydrogen carriers for specific applications, as seen here for steel. The study comprises market analysis and industrial outreach, as well as comprehensive scenarios of the usage of MCH-based hydrogen storage and delivery systems for the specific application of the iron and steel processes. A transparent system design and process model is developed, systems analysis conducted to gauge the performance of the hydrogen carrier system, results provided to materials developers, and industrial outreach conducted. The results are to be compared to the use of compressed hydrogen gas (CHG) and liquid hydrogen (LH2) storage and delivery in the frame of the targeted application. This approach is very persuasive and it is difficult to improve.
- This project is well poised to take a critical look at a baseline techno-economic analysis (TEA) of hydrogen and hydrogen carrier-driven steel manufactured from renewable energy sources to assess where the current technology lands in terms of costs and particularly delivered hydrogen costs. These costs are critical barriers in meeting the objectives of the U.S. Department of Energy's (DOE's) H2@Scale activities. The project has well-defined, rational milestones. The output of this project will provide guidance to future research and development (R&D) endeavors.
- The project developed a process model for the use of hydrogen carrier storage systems integrated with green hydrogen production and application to iron and steel processes. One comment on the approach is that for Task 1 on page 8, the analysis set 200 metric tons per day (MTPD) H_2 production as the fixed target to develop the scenarios and do the analysis. It is suggested to extend the approach to study the impact of hydrogen production capacity.

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.

- A very transparent system design and process model has been developed. The required hydrogen quantities and purities have been determined. ProSim process simulation was performed to design and model different scenarios for hydrogen use in direct iron reduction units. Material and energy balances from process simulations determined and provided for exergy and efficiency analysis. The design for a dual-reactor system, including system size, capital and operating cost, and efficiency, has been finished. Geographics of mills were compared considering the renewable energy profiles. Electrolyzer and storage operation has been modeled, as well as the co-located toluene hydrogenation and dehydrogenation process.
- The project is well posed, logical, and is being executed by a talented team of researchers and collaborators. A logical spectrum of renewable energy scenarios is being analyzed against conventional technology and reasonable hybrid cases. The project takes advantage of the leverage provided by prior work on the MCH carrier at ANL and the analysis of geographic profiles of renewables potential in Texas and Illinois performed by PNNL. Process models of the MCH hydrogenation/dehydrogenation, coupled with the geographic renewables profile, have allowed for preliminary estimates of renewables inputs to costs, and thus, the project is making excellent progress toward meeting the objectives of the project's overall TEA, which will be very relevant to the DOE Hydrogen Program's (the Program's) interests.

Question 3: Collaboration and coordination

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

- It is readily observed that there is excellent communication, collaboration, and coordination among the national laboratories engaged in this project. LBNL has led the stakeholder engagement activity with a key industry participant.
- This is a very well-balanced and interconnected project of the core partners: LBNL, ANL, and PNNL. Monthly project meetings were held. The team has reached out to key stakeholders (iron and steelmakers in

North America). It is not clear, however, how much this work has been performed in collaboration and coordination with external groups and especially international groups.

- The project demonstrated close collaboration and excellent coordination among national laboratories.

Question 4: Relevance/potential impact

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

- This project, regardless of the outcome, is highly relevant to the objectives of the Program, as the output of the TEA, as a function of several renewables scenarios, will help to focus future R&D in carrier development and demonstration and establish benchmark costs for hydrogen generation from renewables, all the way through to transport/storage by carriers, and on to dispensing to a major industrial activity.
- In this project, the performance and cost of MCH-based hydrogen storage is analyzed for the specific application of iron and steel processes. Such a carefully performed analysis is required not only for iron and steelmaking applications, but others as well. It is really needed to generate the required knowledge to design our future clean energy-based society and to identify the best solutions for places, technologies, and energy transport.
- The project provided a good example of integrating green hydrogen production with industrial application. However, as green hydrogen production will be significantly impacted by geographic location, which may not be well connected for the iron and steel application, it would be interesting to show actual geographic connections in the United States and demonstrate the potential impact of the study in real situations.

Question 5: Proposed future work

This project was rated **3.7** for effective and logical planning.

- The evaluation of safety, codes and standards (SCS) and siting is an important part that is required to know which hurdles have still to be taken and what has to be done to prepare for the future use of these technologies. While many technologies are already quite mature, there will definitely be research gaps, which have not yet been dealt with. Finishing the TEA and benchmarking the performance of the system with at least two incumbent technologies is very important, as well as the case studies with different H_3 production and demand scenarios for a range of deployment scales. Also, the look at other new carriers or carriers in the development is extremely important. Furthermore, it should be analyzed if and which of the results could be transferred to other refining industries.
- With this being a very short-duration project, the team must remain very focused, and its future plans need to take this into account. The proposed future work focuses on the key remaining questions. Particularly important are the issues surrounding siting of these large-scale hydrogen activities with large-scale industrial processes regarding SCS.

Project strengths:

- The project provided an interesting opportunity to apply green hydrogen production with an iron and steel manufacturing application. This project provides a comprehensive analysis of the potential need and advantage of having hydrogen storage as a component of this low-carbon transition.
- This project has an experienced team with a good plan and excellent execution. The project has a high value and is impactful to the Program's goals and objectives relating to their H2@Scale activities.
- A very good consortium is doing a very profound and comprehensive analysis of the use of liquid organic hydrogen carriers for hydrogen transport and storage in steel and iron industries.

Project weaknesses:

- The only weakness this reviewer can assess is that it is not clear how and if the results of other international research groups are taken into account. International collaboration is not mentioned.

Recommendations for additions/deletions to project scope:

- The project can be improved from the following points. (1) For Task 1 on page 8, the analysis set 200 MTPD H_2 production as the fixed target to develop the scenarios and do the analysis. It is suggested to extend the approach to study the impact of hydrogen production capacity. (2) On page 8, in cases 1b and 1c, the balance hydrogen came from MCH dehydrogenation. Please explain the source of hydrogen used for toluene hydrogenation to MCH. In addition, it is suggested to add another scenario to produce additional hydrogen from electrolysis using grid power to meet the target of 200 MTPD. (3) It is suggested to consider other storage options such as CHG and LH2 for this study. (4) On page 12, please provide an explanation of why wind requires larger toluene and MCH storage capacity. Similarly, please explain why solar requires a larger hydrogenation plant versus wind. (5) In the green NH_3 production process, the hydrogen storage cost could be reduced by increasing the flexibility of the Haber-Bosch process for NH_3 synthesis. Similarly, it is suggested to study the flexibility of the iron reduction process with reduced hydrogen input for reducing the hydrogen storage cost. (6) The title of waste-heat-driven hydrogen carrier system seems not much reflected in the studies. There is not much analysis in the slide showing the waste-heat recovery and energy balance. (7) As green hydrogen production will be significantly impacted by geographic location, which may not be well connected for the iron and steel application, it would be interesting to show actual geographic connections in the United States and demonstrate the potential impact of the study in real situations.
- Case studies for different hydrogen production and demand scenarios and different scales might change the results. Furthermore, this work should be extended to the use of other carriers as well.
- This project is well conceived, so there are no changes recommended.

Project #ST-228: Determining the Value Proposition of Materials-Based Hydrogen Storage for Stationary Bulk Storage of Hydrogen

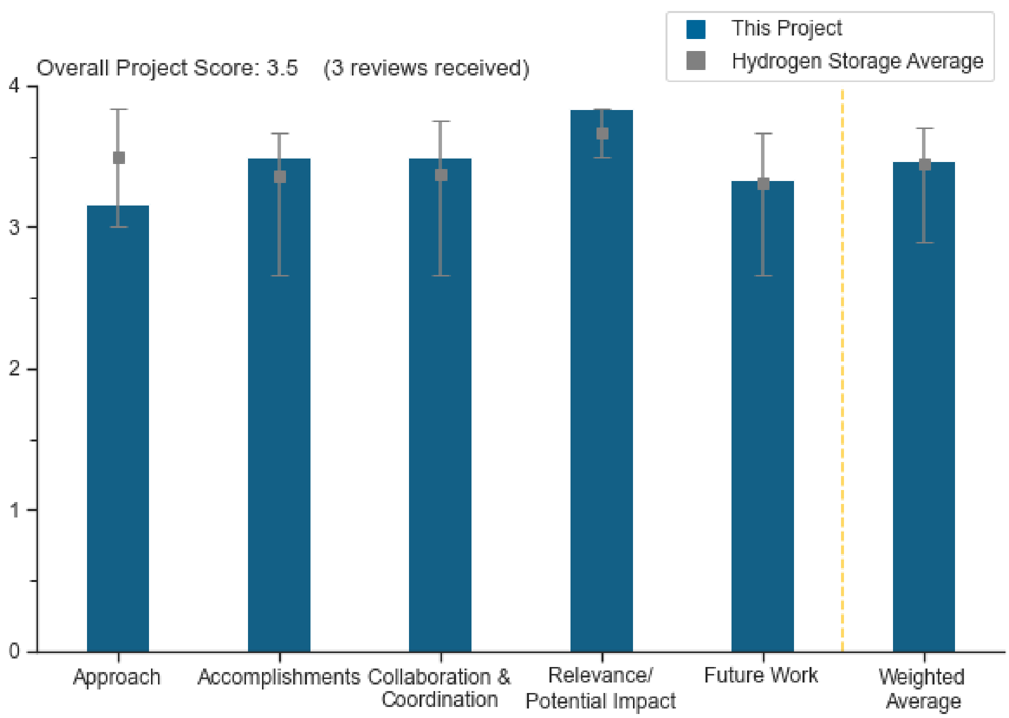
Bruce Hardy, Savannah River National Laboratory

DOE Contract #	WBS 4.4.0.905
Start and End Dates	10/1/2020
Partners/Collaborators	National Renewable Energy Laboratory, Savannah River National Laboratory
Barriers Addressed	• Technoeconomic analysis for cost challenges • Heat source availability and system size • Ability of system to supply H_2 flowrate to power a 20 MW data center for 72 hours at the required fuel cell pressure • Identify transient heat required rate for hydrogen discharge

Project Goal and Brief Summary

This project aims to evaluate the capability and design of materials-based stationary bulk hydrogen storage for backup power applications, starting with fuel-cell-powered data centers. The research team will leverage technoeconomic models developed by the National Renewable Energy Laboratory (NREL) to understand the value proposition of hydrogen and fuel cells for data centers. Researchers will determine a priority list of reversible materials, develop a detailed model to validate the suitability of a metal hydride-based storage system, and identify parameters and designs that yield the most significant performance improvements.

Project Scoring



The vertical hash-lines represent the highest and lowest average scores received by Hydrogen Storage projects.

Question 1: Approach to performing the work

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

- The project is primarily an economic assessment effort to assess current baseline technology and the ability to achieve system-level performance for backup power for a data center. As such, the project will develop relative costs versus other competing incumbent technologies. This approach is feasible and supports U.S. Department of Energy objectives in the H2@Scale activity.
- The project has developed a systematic approach for the analysis, including technoeconomic analysis (TEA), performance/integration, and space considerations. The project is suggested to use a figure to clearly illustrate the relationships among "information technology (IT) load," "data center total load," and "fuel cell (FC) system/data center thermal output" on page 6.
- The approach to evaluate the usage of fuel cells and metal-hydride-based hydrogen stores as backup systems for data centers is wisely chosen. Also, to perform the TEA, the performance/integration analysis as well as space considerations and comparisons of the different storage options (gaseous, liquid and metal hydride [MH]) are very important tasks. However, since there is quite a huge number of possible hydrides to be used for such an application, either a variety of different hydrides has to be taken into account or the specific hydride has to be chosen very wisely to allow for a fair comparison with the different storage options. The chosen hydride is by far not the most suitable. For such an application where several thousand tons of hydride are required, the chosen hydride, $(Ti_{0.97}Zr_{0.03})_{1.1}Cr_{1.6}Mn_{0.4}$, is much too expensive. There are much cheaper room-temperature interstitial metal hydride options available. Only 6 bar hydrogen pressure is required by the fuel cell. An equilibrium pressure of 73 bar, therefore, is not required. Also, for such an application, the $NaAlH_4$ is a poor choice since it requires high operation temperatures. The consortium should have taken more care for selecting the more appropriate low-cost room-temperature hydrides.

Question 2: Accomplishments and progress

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

- The project has made excellent progress in analysis of a baseline case for an MH-based storage system to provide hydrogen to fuel cells to provide extended backup power to a data center. The MH case has been baselined against potential competitive technologies. Preliminary results and accomplishments indicate key advantages and disadvantages of this MH–fuel cell approach and so can drive future research and development (R&D) to mitigate the disadvantages and maximize any advantages of this approach. In this regard, the project supports the objectives of the DOE Hydrogen Program.
- Given the poor choice of the project team's initial selection of hydride candidates, the consortium did a very good job characterizing and assessing the different systems.
- The project can be further improved from the following points. (1) A 5 megawatt (MW) data center is selected, as explained on page 5. However, as on pages 3 and 20, a 20 MW data center is the initial target, so the project is suggested to carry out a similar study for the 20 MW data center as well and show the effect of the scale on the TEA analysis for different scenarios. (2) On page 7, the project is suggested to add another scenario into consideration: hydrogen delivery in metal hydride plus metal-hydride-based hydrogen (MH2) stationary. (3) It would be interesting to show how the reduced footprint could influence the capital expenditures (CAPEX), etc. (4) It is unclear what the volume of storage tank is for gaseous hydrogen (GH2), liquid hydrogen (LH2), and MH2 in the TEA analysis on page 9. (5) As two different types of metal hydride were used for pressure swing and temperature swing models, it is suggested to provide detailed comparisons between these two models, including tank volume, CAPEX, operating expense (OPEX), etc.

Question 3: Collaboration and coordination

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

- There is good coordination and collaboration among the principals. A logical division of tasks has been developed, which is being executed.
- The partners seem to collaborate very well with each other. Considering the huge know-how of U.S. and international institutions and researchers in the field of different hydrides, however, it would have been important to collaborate much more with such materials researchers to make the most suitable preselection of hydrides.
- The project demonstrated good collaborations between partners. It is suggested to add more industrial partners to validate some assumptions for the analysis.

Question 4: Relevance/potential impact

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

- This type of project is highly relevant to the DOE H2@Scale activity. By performing TEA of incumbent and emerging technologies, this project can uncover advantages and disadvantages of various approaches, which will help to focus future R&D to remove the barriers identified.
- Energy security is of major importance, not only in the frame of the rise of renewable energy usage. Backup power, therefore, is utterly needed. Also, the importance of the digitalization is rising exponentially. Therefore, to analyze the usage of hydrogen and fuel cells as backup power for data centers is extremely wise and important.
- The project aligns well with the Hydrogen Program and DOE objectives.

Question 5: Proposed future work

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

- The proposed future work is excellent. Nevertheless, to be able to compare the costs of those different technologies, of course the prize of the metal hydrides chosen is of utter importance. Therefore, cheaper room-temperature hydrides should be assessed.
- As LH2 storage shows significant cost advantage, the project is suggested to do more studies on LH2 storage options. Based on the project goals, it seems that a list of reversible metal hydride materials will be evaluated for this application; however, only two hydride materials are considered in the analysis. The reason should be explained.
- The current preliminary costs for the MH system chosen are high; much of the cost appears to be associated with the MH costs. Future work is appropriately directed at exploring the influence of a variety of MH materials with a variety of properties and material costs.

Project strengths:

- The project demonstrated an interesting study to evaluate the capability and design of materials-based bulk storage system options for data center application and developed a detailed model to identify and validate the suitability of a metal-hydride-based storage system and identify parameters and designs that yield the most significant improvements in performance.
- This project aims at backup power solutions for data centers. This topic is of utter importance and should be given a larger budget to investigate and assess the cheapest choices of hydrides to be used in such an application.
- There is a good team and a well-posed approach to the problem. The project is identifying key parameters for the MH system to integrate with the fuel cell backup power system.

Project weaknesses:

- There is a minor weakness. It would be nice, but probably beyond the scope of the project, to automate the MH search space to accelerate the search for an optimum MH, if there is one.
- As a summary, the project can be improved from the following points. (1) The project is suggested to use a figure to clearly illustrate the relationships among "IT load," "data center total load," and "FC system/data center thermal output" on page 6. (2) A 5 MW data center is selected, as explained on page 5. However, as on pages 3 and 20, a 20 MW data center is the initial target. The project is suggested to carry out a similar study for the 20 MW data center as well and show the effect of the scale on the TEA analysis for different scenarios. (3) On page 7, the project is suggested to add another scenario into consideration: hydrogen delivery in metal hydride plus MH2 stationary. (4) It would be interesting to show how the reduced footprint could influence the CAPEX, etc. (5) It is unclear what the volume of storage tank is for GH2, LH2, and MH2 in the TEA analysis on page 9. (6) As two different types of metal hydride were used for pressure swing and temperature swing models, the project is suggested to provide detailed comparisons between these two models, including tank volume, CAPEX, OPEX, etc. (7) The project is suggested to add more industrial partners to validate some assumptions for the analysis.
- Unfortunately, only two hydrides are considered in this study. One of those hydrides is much too expensive. The other one requires high temperatures for operation.

Recommendations for additions/deletions to project scope:

- This is a well-conceived project and it is being well executed. No changes are suggested.
- As LH2 storage shows significant cost advantages, the project is suggested to do more studies on LH2 storage options. Based on the project goals, it seems that a list of reversible metal hydride materials will be evaluated for this application; however, only two hydride materials are considered in the analysis. The reason should be explained.
- The focus of the analysis must lay on cheap room-temperature hydrides. A much cheaper alternative would be, for example, FeTi-based alloys.