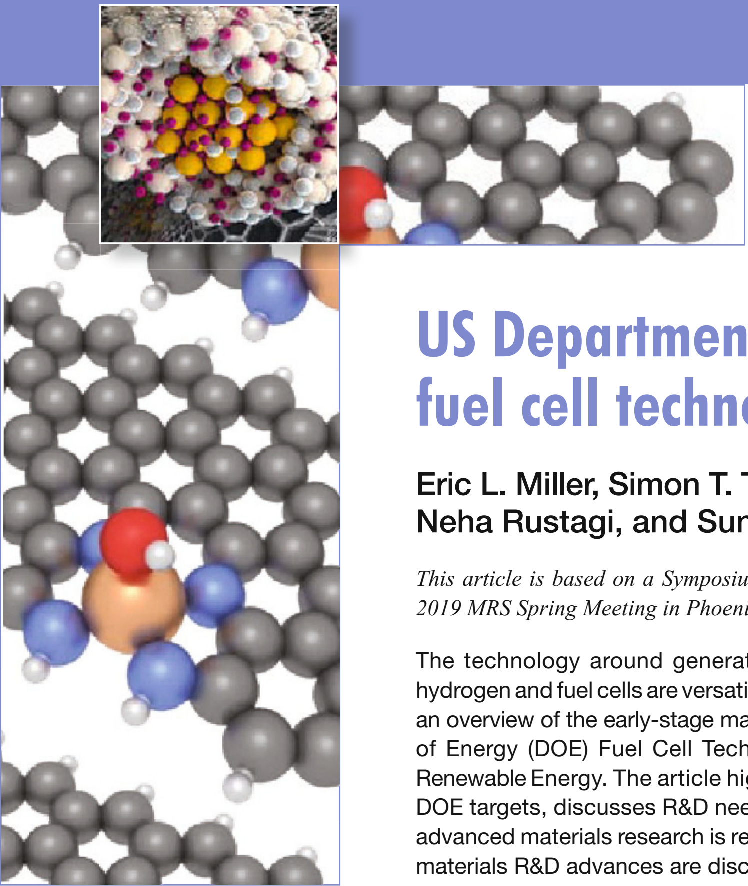

US Department of Energy hydrogen and fuel cell technologies perspectives

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The technology around generating efficient and sustainable energy is rapidly evolving; hydrogen and fuel cells are versatile examples within a portfolio of options. This article provides an overview of the early-stage materials R&D in hydrogen and fuel cells at the US Department of Energy (DOE) Fuel Cell Technologies Office within the Office of Energy Efficiency & Renewable Energy. The article highlights technology status and progress toward achieving DOE targets, discusses R&D needs and challenges, and provides specific examples where advanced materials research is relevant to addressing those challenges. For broader context, materials R&D advances are discussed in the context of DOE's H2@Scale initiative, which is enabling innovations to generate cost-competitive hydrogen as an energy carrier, enabling renewables, as well as nuclear, fossil fuels, and the grid, to enhance the economics of both baseload power plants and intermittent solar and wind, enhancing resiliency and avoiding curtailment.

Introduction

Hydrogen and fuel cell technologies, as important parts of a comprehensive energy strategy, offer an efficient and versatile means of producing, distributing, and utilizing clean energy across multiple sectors, including power-generation, transportation, chemicals production, and industrial manufacturing. Fostering the R&D necessary to enable widespread adoption of these technologies is a key objective of the US Department of Energy's (DOE) H2@Scale initiative,[1] in support of the DOE's broader aim to enhance US energy security, resiliency, and economic prosperity.

Foundational materials- and systems-level R&D has laid the groundwork for continued advancement of the hydrogen and fuel cell industries integral to H2@Scale. The fuel cell market, for example, has seen consistent growth in the last few years, with nearly 70,000 fuel cell systems and 800 MW in fuel cell power shipped worldwide in 2018, and with approximately USD $2.3 billion in fuel cell revenue.[2] Globally, there are more than 300,000 stationary fuel cells in operation, 14,000 hydrogen-powered fuel cell cars on the road, and approximately 300 hydrogen refueling stations. In the United States, we currently have more than 500 MW of stationary fuel cells, more than 7800 fuel cell cars, and more than 28,000 hydrogen fuel cell forklifts operating at major companies.[3]

Ongoing hydrogen and fuel cell commercialization efforts in the United States have been supported by government agencies, leveraging research with partners from industry, universities, and the national laboratories. Early pioneers, in addition to the DOE, have included federal agencies such as NASA and the US Department of Defense. Currently, public/private R&D activities under H2@Scale are being supported by DOE's Fuel Cell Technologies Office (FCTO) in DOE's Office of Energy Efficiency & Renewable Energy (EERE). More than 960 US patents have been issued over the years as a result of FCTO funding,[4] enabling roughly 30 commercialized technologies and an additional 65 prospective commercial technologies. These can range from specific components such as catalysts or membranes to complete systems such as electrolyzers and storage tanks.

Today, the FCTO mission focuses on early-stage materials- and system-level R&D that enables industry to develop and demonstrate hydrogen and fuel cell technologies that are

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cost-competitive with incumbent and advanced alternative technologies. This article briefly describes the H2@Scale initiative, including specific techno-economic goals and targets. It also discusses specific examples of the supporting foundational materials R&D being led by FCTO in areas of fuel cells, as well as hydrogen production, storage, and infrastructure.

H2@Scale

The H2@Scale initiative[5] aims to develop and enable transformational technologies that can sustainably produce and efficiently utilize large quantities of affordable hydrogen across sectors.[6] Hydrogen is a unique and versatile energy carrier due to the diversity of domestic options for large-scale hydrogen production (including utilization of natural gas, coal, water, biomass, nuclear power, electricity, and direct sunlight), as well as the broad spectrum of industrial end uses, as shown in the H2@Scale vision illustrated in **Figure 1**. The versatility of hydrogen across sectors is illustrated in the following examples:

- Hydrogen is essential to petroleum refining, biomass upgrading, and ammonia production.
- Combining hydrogen produced from renewable resources with captured carbon dioxide enables synthesis of renewable hydrocarbon fuels and chemicals.
- Fuel cells and turbines can efficiently convert renewable hydrogen into clean electricity or combined heat and power, both at large scales and at smaller distributed scales.
- Hydrogen fuel cells and electrolyzers coupled to the electricity grid can support grid stability under high penetrations of renewable generation.
- Renewable hydrogen can provide the fuel for advanced, highly efficient zero-emission fuel cell technologies for light-, medium-, and heavy-duty transportation.

Industry has also taken steps to foster expansion of hydrogen supply and demand opportunities. The Hydrogen Council, for example, is a global industry-led partnership established in 2017 whose members have committed to significant investments to enable commercialization of relevant technologies. Their roadmap, titled "Hydrogen, Scaling Up,"[7] is a comprehensive assessment of the potential impact of hydrogen, concluding that by 2050, hydrogen could meet 18% of world energy demand, create a $2.5 trillion market, support 30 million jobs, and reduce carbon dioxide emissions by 40–60% in transportation, industry, and residential sectors.

In addition to industry partnerships such as the Hydrogen Council, international government partnerships have been formed recognizing the important economic potential of hydrogen in global economies. Notably, the International Partnership for Hydrogen and Fuel Cells in the Economy, formed in 2003 to facilitate and accelerate the transition to clean and efficient energy and mobility systems using hydrogen and fuel cell technologies across applications and sectors, now includes 19 countries and the European Commission.[8] Other notable international efforts include the Hydrogen Energy Ministerial launched in 2018 by Japan's Ministry of Economy, Trade and Industry (METI) with the New Energy and Industrial Technology Development Organization (NEDO);[9] the Mission Innovation Challenge on Renewable and Clean Hydrogen, launched in 2018;[10] and the Hydrogen Initiative launched by the Clean Energy Ministerial at the CEM10 Meeting in Vancouver, Canada, in 2019.[11]

Through such partnerships, US and international stakeholder communities continue efforts to enable H2@Scale (also often referred to in other countries as "sector coupling"), recognizing that successful widespread adoption will depend ultimately on cost reductions being realized, in part, by concerted materials research and development. For example, over the past decade, expensive components of fuel cells, such as platinum group metal catalysts, have been minimized significantly through materials R&D, resulting in a 60% reduction in fuel cell capital costs.[12] Similarly, in the last decade, research has enabled an 80% reduction in capital costs of water electrolyzers for large-scale hydrogen production.[13] Since the trajectory of H2@Scale is closely tied to cost-competitiveness, technology-specific cost reduction targets have been developed to help guide the ongoing research.

Cost targets and status

The DOE FCTO conducts peer-reviewed techno-economic analyses and comprehensive systems studies to (1) establish targets for cost competitiveness for hydrogen and fuel cell

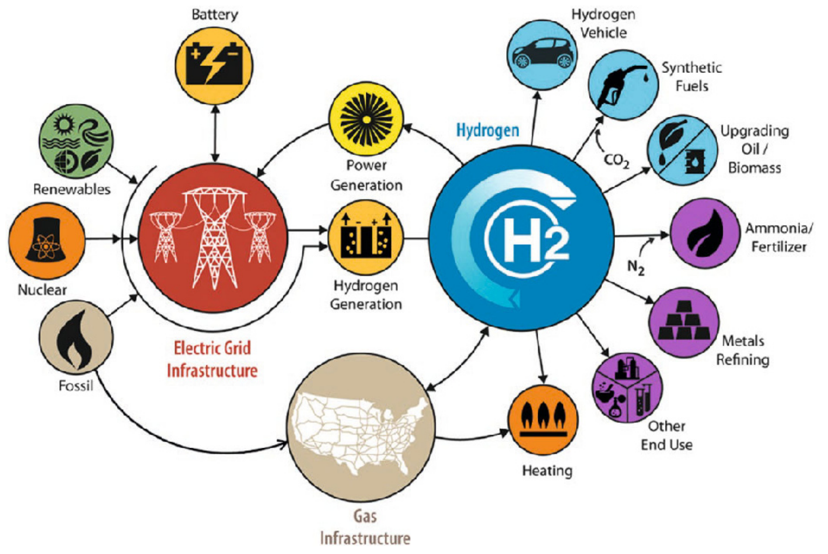

Figure 1. The H2@Scale Vision-Hydrogen can be produced from diverse domestic resources and is a central input to many important end uses in the industrial, chemical, and transportation sectors. Reprinted with permission from the US Department of Energy Fuel Cell Technologies Office.

technologies in current and emerging applications; (2) assess cost status versus the targets; and (3) help guide R&D priorities to enable needed cost reductions. Examples include targets established for fuel cells as well as hydrogen production, delivery, and storage for transportation applications, described next.

Fuel cells for transportation

Transportation fuel cell R&D at FCTO focuses on early-stage innovations, including medium-/heavy-duty transportation applications, including trucks, rail, marine, and other applications, while continuing to work on light-duty vehicle requirements. An ultimate price target of $30/kW by 2050 for an 80-kW$_{net}$ polymer electrolyte membrane (PEM) fuel cell system has been established to enable long-term competitiveness of light-duty hydrogen fuel cell cars. The system encompasses the fuel cell stack and the balance of plant needed to support its operation (e.g., air loop, fuel loop, cooling loops, humidifier, and electronics controllers and sensors). Note that the battery and electric motor are not included in the cost. This value was set by the DOE in collaboration with the US DRIVE partnership (a nonbinding and voluntary government–industry partnership focused on advanced automotive and related energy infrastructure technology R&D), the United States Council for Automotive Research LLC (USCAR), energy companies, and utility companies.[14] Although current high-volume cost projections for such fuel cell systems are as low as $50/kW (roughly $75/kW, including durability requirements), nearer-term estimates based on current low-volume manufacturing are as high as $210/kW.[15] Reaching the ultimate target of $30/kW will depend on achieving economies of scale in manufacturing coupled with continued R&D to reduce costs, including fundamental research to develop new materials for lower-cost catalysts, membranes, ionomers, and balance-of-system materials for fuel cells.

Hydrogen production and distribution

In the transportation sector, the combined cost of hydrogen and its use in a fuel cell vehicle must compete with conventional and other alternative technologies on a cost-per-mile basis to the consumer. For hydrogen-fueled cars to be competitive in the long term, FCTO has established an ultimate cost target of $4/kg for dispensed hydrogen,[16] with $2/kg apportioned for production and $2/kg apportioned for delivery and dispensing costs.[17] Additionally, an interim cost target of $7/kg has been established for fuel cell vehicles to be competitive in early markets.[18] Since current cost estimates for dispensed hydrogen range up to $16/kg (comparable to today's fueling station prices), R&D of production and delivery technologies is still needed to reduce costs. Specific materials research priorities in these areas include catalysts, membranes, energy-conversion materials (such as semiconductors and redox materials), and hydrogen-compatible materials for hydrogen infrastructure, among others.

Hydrogen storage

Cost targets for onboard hydrogen storage are based on the quantity of hydrogen stored in a fuel cell car required for a conventional driving range (e.g., greater than 300 miles) within the constraints of weight, volume, and durability. The DOE in coordination with the US DRIVE Partnership has established an ultimate storage cost target of $8/kWh to enable significant market penetration of hydrogen-fueled vehicles.[19] In current automotive use, compressed hydrogen is stored at high pressure (up to 700 bar) in expensive fiber-reinforced tanks, with current cost projections as high as $24/kWh.[20] Materials research to reduce cost of the carbon fibers used in these tanks offers one pathway toward meeting the targeted cost. Longer-term options include materials-based hydrogen storage (e.g., in metal hydrides, chemical adsorbents, metal–organic frameworks, and others), currently at the fundamental materials research stage.

Figure 2 summarizes the DOE targets and status for the three categories previously described. The low-volume estimates shown in the figure represent the current range of commercially available technology options in each category.[21] The different high-volume estimates incorporate projected technology advances as well as assumed benefits of economies of scale. For example, the $45/kW high-volume projection for fuel cells envisions near-term improvements in performance and cost of fuel cells being manufactured at a rate of 500 k/yr. In contrast, the $75/kW projection is based on 100 k/yr, with adjustments based on fuel cell technologies that can meet strict 150,000-mile durability targets. In all categories, ongoing R&D is needed for next-generation technologies that can achieve the ultimate cost goals, as detailed in FCTO's Multi-Year Research, Development and Demonstration Plan.[22]

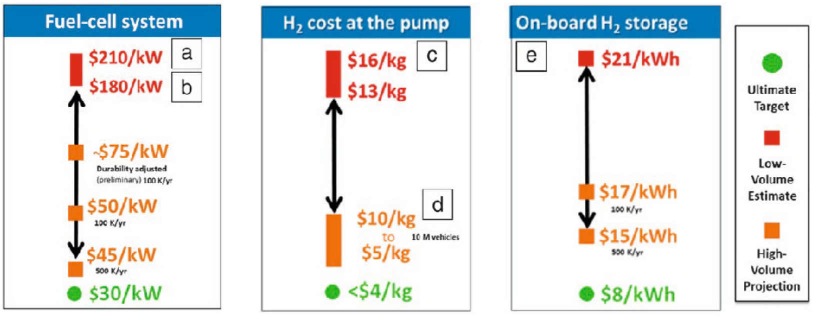

Figure 2. Status versus market-driven targets in fuel cells, hydrogen production and distribution, and on-board hydrogen storage displayed as low-volume estimates and high-volume projections. The current status costs are revised periodically based on real-world data. (a) Based on commercially available fuel cell electric vehicles; (b) based on state-of-the-art technology; (c) range based on H_2 produced from natural gas (NG), delivered dispensed at today's stations (~180 kg/d); (d) range based on H_2 produced from electrolysis at $5/kg and NG at $2/kg; and delivery and dispensing using advanced tube trailer or pipeline technologies ranging from $3–5/kg, and current liquid tanker technologies at scale at $5/kg. High-volume assumptions include electrolyzers manufactured at 700 MW/year; and >10,000 stations at 1000 kg/day capacity, to serve 10 million vehicles; (e) storage costs based on preliminary 2019 storage cost record. Reprinted with permission from the US Department of Energy Fuel Cell Technologies Office.

Foundational materials R&D

The FCTO supports early-stage and applied R&D for improving performance and reducing costs in hydrogen production, delivery, storage, and fuel cell technologies. Accelerated materials discovery and development through dedicated multilaboratory research consortia are central to the research approach. Such consortia, funded and managed by DOE, comprise core national laboratories offering state-of-the-art capabilities and expertise that university and industry partners can access to accelerate materials breakthroughs and innovations. As described next, the DOE has spearheaded the establishment of several successful research consortia within its Energy Materials Network, specifically to address a number of high-priority materials challenges in hydrogen and fuel-cell technologies.

The Energy Materials Network

The Energy Materials Network (EMN) is an innovative DOE approach to reduce the traditional timeline of 15–20 years-to-market for advanced materials by addressing vital challenges to expedite more widespread adoption of high-impact clean technologies.[23] The national laboratory consortia of the EMN conduct research on specific energy-related materials challenges by employing the resources and expertise at national laboratories and fostering industry and academic collaboration through DOE- and industry-sponsored projects. To accelerate progress through a multidisciplinary team approach, state-of-the-art resources in materials theory, synthesis and characterization at the DOE national laboratories are being leveraged, including innovative combinatorial and high-throughput techniques as well as advanced data management and informatics. The consortia are based on common foundational principles to create the collaborative research environment for rapidly building on R&D successes.

FCTO's EMN Consortia include (1) the ElectroCat Consortium on Platinum-Group-Metal-Free Electrocatalysts for Fuel Cells; (2) the HydroGEN Consortium on Advanced Water Splitting Materials; (3) the HyMARC Consortium on Materials-Based Hydrogen Storage; and (4) the H-Mat Consortium on Hydrogen Compatible Materials.

ElectroCat

The FCTO, leveraging the ElectroCat EMN Consortium,[24] is addressing the technical challenges to the development of low-cost, platinum-group-metal-free (PGM-free) and low-PGM electrocatalysts and electrodes, specifically for fuel cells.[25] One priority is increasing the performance and durability of PGM-free oxygen reduction reaction catalysts for fuel cell cathodes.[26] Success in this area is considered critical to bringing down the cost of an automotive fuel cell system to the $30/kW cost target, where the need for durable Pt-based catalysts in current fuel cell electric vehicles remains a key cost driver. The laboratory capabilities being harnessed by the consortium include modeling of catalysts, electrodes, and fuel cells; catalyst and model system synthesis; fabrication of electrodes and assemblies; advanced materials characterization; electrode and cell characterization/diagnostics; and data management.[27]

ElectroCat has had several successes to date in advancing the fundamental understanding of active sites in PGM-free electrocatalysts. In one example, identification of active sites was enabled by the relatively homogeneous structure of atomically dispersed Fe-N-C catalyst synthesized by ElectroCat partners at Los Alamos National Laboratory.[28] Scanning transmission electron microscopy (STEM) of this structure demonstrates the atomic dispersion of active sites, seen as bright spots in the dark-field image shown in **Figure 3**a. Figure 3b shows the electron energy-loss spectra corresponding to the indicated areas in Figure 3a, indicating co-location of Fe and N atoms at active sites (plots 1 and 2), and providing strong

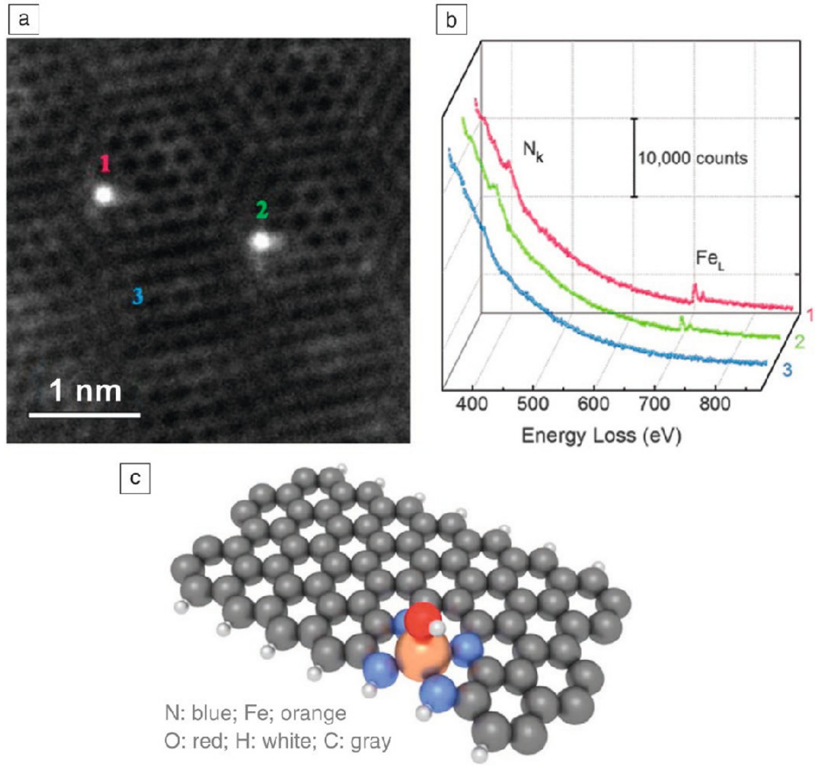

Figure 3. Experimental characterization and theoretical modeling of the atomically dispersed Fe-N-C as a promising platinum-group-metal (PGM)-free catalyst for the fuel-cell oxygen reduction reaction, including (a) scanning transmission electron microscope image highlighting active sites; (b) corresponding electron energy-loss spectroscopy spectra indicating co-location of Fe and N at the active sites; and (c) model of an active site on the edge of a graphene sheet established for computational investigations. Adapted with permission from References 28 and 29. © 2017 AAAS and American Chemical Society, respectively.

evidence of the Fe-Nx macrocyclic nature of these sites. To computationally explore the activity and stability of Fe-N$_4$ sites within and on the zigzag edge of a graphene sheet, models based on density functional theory (DFT) were developed. Model calculations indicate that edge sites that consist of bonds that can be broken more easily (particularly the N atoms) are most active.[29] Identifying and understanding active sites in the Fe-N-C catalysts are providing the key insights needed to prepare catalysts with increased active site density and stability, addressing two of the most significant challenges to PGM-free fuel cell catalysts.

HydroGEN

The primary focus of the HydroGEN EMN Consortium[30] is to accelerate materials research of advanced water-splitting technologies to enable widespread use of clean, sustainable hydrogen and related fuel cell technologies, consistent with the H2@Scale vision.[31] The consortium's R&D covers low- and high-temperature electrolysis as well as direct photoelectrochemical (PEC) and solar thermochemical (STCH) water splitting. While low-temperature electrolysis has been commercialized and high-temperature electrolysis is nearing commercialization, PEC and STCH are longer-term options for direct solar water-splitting with potential for low-cost, large-scale hydrogen production in the future. HydroGEN recognizes that each of the technologies poses disparate but interrelated materials R&D challenges, for example, in energy conversion, catalysis and separations; its core labs host more than 80 unique research "nodes," each of which includes state-of-the-art scientific tools or techniques coupled with the specialized expertise to employ them for these water-splitting approaches.[32] Early achievements include world records in direct PEC solar water-splitting efficiencies[33] and the development of standard protocols for systematically benchmarking progress from groups within and outside the consortium.[34]

As a good example of HydroGEN's cutting-edge approach to materials research, its collaborators at the University of Colorado Boulder are utilizing machine-learned models coupled with *ab initio* thermodynamic and kinetic screening calculations to accelerate the research, development, and demonstration of new STCH materials.[35] The team has focused in part on the perovskite family of materials, which has shown potential for STCH applications. The team has effectively utilized machine learning methods that screened more than 10^{10} potential descriptors to discover a new single descriptor, τ, based solely on chemical composition (including oxidation states and atomic radii) that accurately predicts perovskite stability. In the basic structure of a ABX$_3$ perovskite, shown in **Figure 4**a, there is an expansive number of possible compounds based on different combinations of elements; however, not all will form the stable structure.

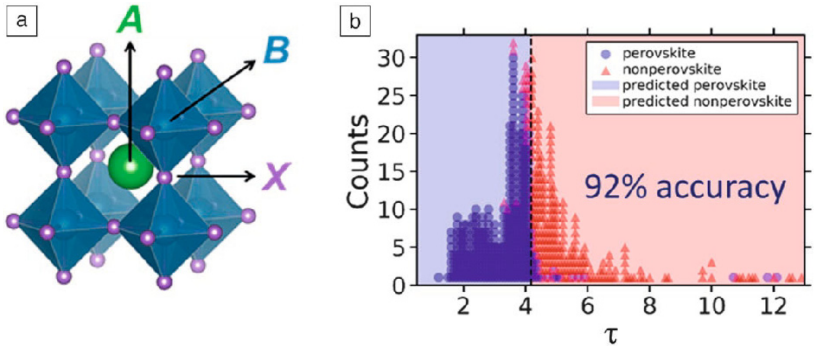

Figure 4. (a) The basic structure of a stable ABX3 solid. (b) Accuracy in predicting stability of known ABX$_3$ perovskite materials using τ, the newly machine learned descriptor based on basic chemical parameters of constituent elements. Blue dots in the blue sector and red dots in the red sector represent correct predictions. Adapted with permission from Reference 35. © 2018 Nature Publishing Group.

As shown in Figure 4b, when applied to an experimental data set covering nearly 600 ABX$_3$ solids, the new descriptor, τ, successfully predicts perovskite stability with 92% accuracy. With this accuracy, the descriptor can be used as a first screening step in exploring new perovskite-based STCH materials, significantly reducing the potential material space worthy of more in-depth and time-consuming density functional theory calculations, accelerating the materials discovery process.

HyMARC

Hydrogen storage is one of the critical enabling technologies for creating hydrogen-fueled transportation systems, and the HyMARC EMN Consortium[36] addresses the specific materials challenge posed by the low volumetric density of gaseous hydrogen for current onboard storage in pressurized tanks. A promising alternative is the use of solid-state materials, such as novel adsorbents[37] and high-density hydrides,[38] which have the potential to deliver hydrogen at lower pressures and achieve higher onboard storage densities to meet DOE storage targets. HyMARC's goal is to expand the foundational understanding of the phenomena governing thermodynamics and kinetics of hydrogen release and uptake in all classes of hydrogen storage materials, including adsorbents, metal hydrides, and other hydrogen carriers. Toward this goal, it is developing computational modeling tools, as well as implementing state-of-the-art diagnostic and characterization tools to provide experimental data for model development and validation.[39] Insights gained from these studies are being applied toward the design and synthesis of next-generation hydrogen storage materials, with exciting early results.

One impressive HyMARC accomplishment has been its success in advancing fundamental understanding of cycling behavior in high-capacity metal hydrides for hydrogen storage. The practical use of hydrides such as LiNH$_2$ is often hindered by slow kinetics for hydrogenation and dehydrogenation reactions that proceed through complicated multistep pathways. HyMARC teams at Sandia National Laboratories and Lawrence Livermore National Laboratory have discovered that rates of both reactions increased by a factor of two relative to bulk Li_3N when Li_3N particles are confined within nanoporous carbon structures to suppress the Li_2NH intermediate phase formation.[40] As illustrated in **Figure 5**, experimental and computational investigations showed no evidence of the Li_2NH intermediate phase formation in the nano-confined materials. Based on molecular dynamics modeling of the thermodynamic factors responsible for the creation and growth of this phase, it was determined that keeping the particle size below a certain threshold value avoids its formation and allows reactions to proceed in one step and at a higher rate. The new and experimentally validated computational models establish the importance of controlling nano-interfaces in solid-state hydrogen storage reactions, and introduce a new paradigm for enhancing performance by controlling the nanostructural architecture.

H-Mat

The Hydrogen Materials (*H-Mat*) Consortium[41] focuses on materials compatibility with hydrogen for safe, durable, reliable, and affordable infrastructure necessary for H2@Scale. H-Mat addresses hydrogen-materials interaction challenges such as hydrogen embrittlement that can adversely affect mechanical properties of component and storage materials, including fatigue life, tensile strength, and toughness. An important goal is to achieve a more robust understanding of the mechanisms of hydrogen effects to enable lower-cost methods of developing components for hydrogen service. Research focus areas span from foundational mechanisms to engineering solutions in the context of in-service temperatures and pressures. H-Mat partners are studying hydrogen-materials interactions across length scales from the atom to the power grid, including high-pressure mechanical and component materials testing, hydrogen transport properties, hydrogen-sensitive surface science, and advanced computational materials science.

H-Mat's R&D activities build on related materials work at the national laboratories investigating hydrogen compatibility in both metals and polymers. The "master curve" derived by Sandia National Laboratories for metal alloys has been incorporated into The American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel Code Section VIII Division 3,[42] and can reduce or even eliminate much of the experimentation required of industry stakeholders when obtaining certification of new vessel designs. Additionally, their test methods for polymers (including characterization of tribology, density changes, and changes in microstructure), have been incorporated into the Canadian Standards Association CHMC-2 standard.[43] A noteworthy advance has been the novel use of unique visualization tools, such as helium-ion microscopy to identify microstructure changes in model polymers after exposure to hydrogen. Microscopic results from an investigation of hydrogen exposure effects in ethylene propylene diene monomer rubber are shown in **Figure 6**, indicating significant void formation that would need to be mitigated in any hydrogen services.[44]

H-Mat and all of FCTO's materials research consortia are advancing hydrogen and fuel cell technologies for H2@Scale. Foundational to the success has been a paradigm based on integration of materials synthesis, characterization, multiscale modeling, and experimental model validation in a comprehensive feedback framework.[45]

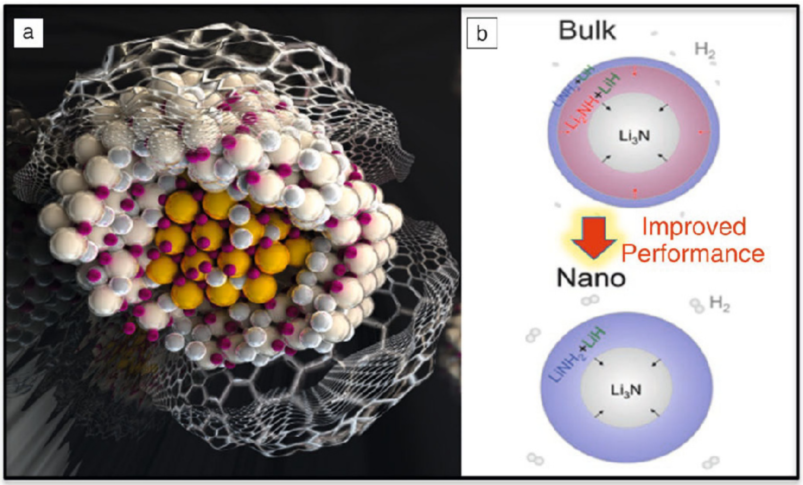

Figure 5. (a) Schematic showing one-step phase transition between Li_3N core and $LiNH_2$ + LiH outer shell during (de)hydrogenation (Li–purple, H–small white, N–yellow and large white spheres, nanoporous carbon–hexagonal net). (b) Schematic showing difference between two-step bulk and one-step nanoconfined (de)hydrogenation reactions. Adapted with permission from Reference 40. © 2018 American Chemical Society.

Summary

With projected growth in demand for hydrogen across multiple sectors, the affordability of hydrogen and fuel-cell technologies is becoming more important to national energy security and economic growth. The Fuel Cell Technologies Office at the US Department of Energy, as part of the H2@Scale Initiative, is focused on R&D to improve performance and reduce cost of key technologies relevant to hydrogen production, storage, distribution, and utilization. Addressing foundational materials challenges in these technologies is a strategic priority, as discovery and development of advanced materials for integration into commercially viable products are key to widespread adoption and deployment. To meet the challenges, FCTO is successfully leveraging the DOE Energy Materials Network consortia model, bringing industry and academia stakeholders together with national laboratory capabilities and expertise in collaborative research. Current focus areas include PGM-free and low-PGM catalysts for fuel cells, advanced water-splitting materials, breakthrough hydrogen storage materials,

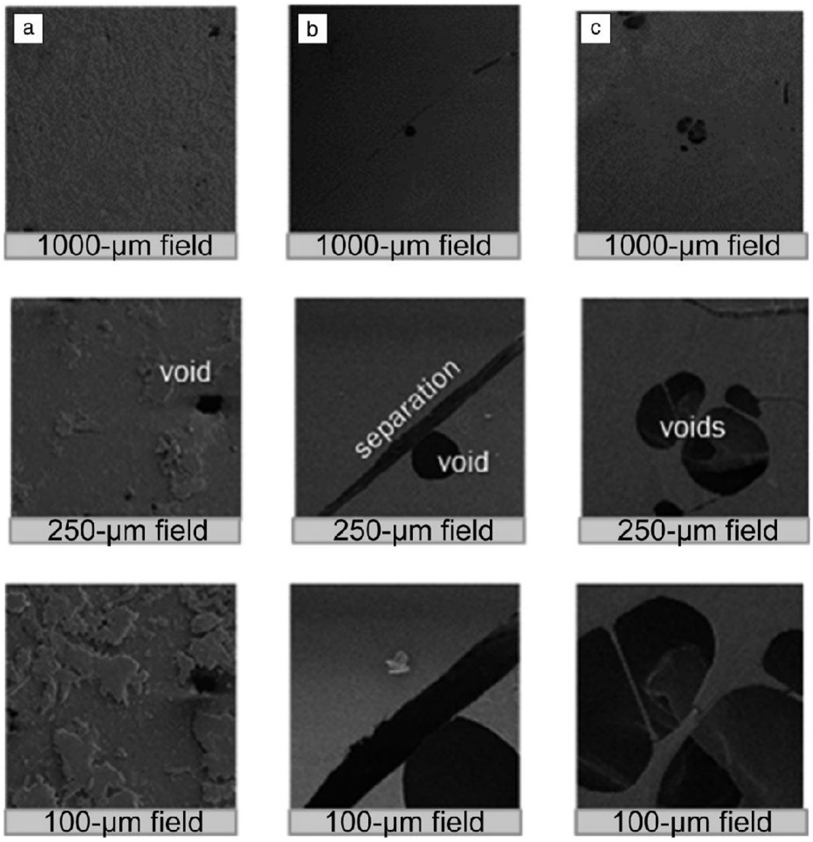

Figure 6. Helium-ion microscope images of commercial samples of ethylene propylene diene monomer rubber (a) as-received; (b) after cryo-fracture, subsequent to 16 h of exposure to 27.6 MPa helium; and (c) after cryo-fracture, subsequent to 16 h of exposure to 27.6 MPa hydrogen. Results indicate that saturation with helium results in phase separation of plasticizer, and saturation with hydrogen additionally results in formation of voids within the material.[44]

and hydrogen-compatible materials for distribution infrastructure. Progress has been impressive to date, but continued R&D is needed for meeting cost and performance targets that can pave the way to H2@Scale's vision for the future.

Disclaimer

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