Fuel Cell R&D Subprogram Overview

INTRODUCTION

The Fuel Cell R&D subprogram supports applied, early-stage research and development (R&D) of fuel cell technologies for transportation applications, as well as stationary and cross-cutting applications, with a primary focus on reducing cost and improving durability. Early-stage research areas include catalysts, membranes, and fuel cell performance and durability. The subprogram seeks a balanced, comprehensive approach to fuel cells for near-, mid-, and longer-term applications. The development of fuel cells for transportation applications is a primary focus due to the nation's goal of significantly reducing its energy and petroleum needs and the benefits inherent in fuel cell electric vehicles (FCEVs) (e.g., high efficiency, long driving range, zero emissions). Transportation applications also include medium- and heavy-duty trucks, rail, and marine fuel cell propulsion. Stationary applications include the development of fuel cells for distributed power generation, including combined heat and power (CHP) for residential and commercial applications. Existing early markets and near-term markets generating market traction for adoption of FCEVs include primary/backup power for critical infrastructure such as data centers, auxiliary power units, and specialty applications such as material handling equipment. The subprogram's R&D portfolio is primarily focused on polymer electrolyte membrane (PEM) fuel cells, but it also includes longer-term technologies, such as alkaline membrane fuel cells, reversible fuel cells, and higher-temperature fuel cells like molten carbonate fuel cells for stationary applications.

Durability and cost are the primary challenges to fuel cell commercialization. Improvements in multiple components are required to concurrently meet these challenges. The subprogram's fuel cell tasks are delineated in the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan, with R&D focused in the key areas of fuel cell components and materials, as well as fuel cell performance and durability.

GOAL

The subprogram's goal is to advance fuel cell technologies for transportation, stationary, and cross-cutting applications.

OBJECTIVES

The subprogram's key objectives include:

- Developing a 65% peak-efficient, direct hydrogen fuel cell power system for transportation that can achieve 5,000-hour durability (ultimate 8,000 hours) and be mass produced at a cost of $40/kW by 2025 (ultimate $30/kW).
- Developing distributed generation and micro-CHP fuel cell systems (5 kW) operating on natural gas that achieve 45% electrical efficiency and 60,000-hour durability at an equipment cost of $1,500/kW by 2025.
- Developing medium-scale CHP systems (100 kW–3 MW) by 2025 that achieve 50% electrical efficiency, 90% CHP efficiency and 80,000-hour durability at a cost of $1,500/kW for operation on natural gas and $2,100/kW when configured for operation on biogas.

FISCAL YEAR (FY) 2018 TECHNOLOGY STATUS AND ACCOMPLISHMENTS

Reducing cost and improving durability while maintaining performance continues to be the key challenge facing fuel cell technology. For platinum group metal (PGM)-based catalysts, both a reduction in PGM loading and an increase in membrane electrode assembly (MEA) areal power density are required to reduce material costs. Current state-of-the-art MEAs with very low cathode PGM loadings experience a higher than expected reduction in performance when operating at high power (i.e., close to rated power), but FY 2018 saw continued progress toward addressing this performance loss. Commercial fuel cells are expected to use PGM-based catalysts in the near term; however, reaching cost competitiveness with conventional automobiles in the long

term favors a transition from PGM-based catalysts to PGM-free catalysts. The subprogram's consortia—Fuel Cell Performance and Durability (FC-PAD) and the Electrocatalysis Consortium (ElectroCat)—made great strides in materials innovation and phenomenological understanding of fuel cell catalysts and electrode components through increased cooperation between the national laboratories and industry and academic partners.

One of the most important metrics used to guide the subprogram's R&D efforts is the projected high-volume manufacturing cost for automotive fuel cells, which is tracked on an annual basis. The subprogram is targeting a cost reduction to \$40/kW by 2025. Long-term competitiveness with alternative powertrains is expected to require further cost reduction to \$30/kW, which represents the subprogram's ultimate cost target. This year, the preliminary cost projection for an 80-kW_{net} automotive PEM fuel cell system based on next-generation laboratory technology and operating on direct hydrogen is \$50/kW_{net} (2016 U.S. dollars) when manufactured at 100,000 units/year, and \$45/kW_{net} when manufactured at a volume of 500,000 units/year.[1] For comparison, the estimated cost of automotive PEM fuel cell systems that are based on currently deployed commercial technology in 2018 is approximately \$210/kW_{net} when manufactured at a volume of 1,000 units/year, which is a closer reflection of current manufacturing rates. The expected cost for an analogous system based on state-of-the-art materials is \$181/kW_{net}.

Power system cost projections are based on beginning-of-life stack performance using MEAs made with de-alloyed PtCo on a high-surface-area carbon (PtCo/HSC) cathode catalyst developed by General Motors (GM), Pt/C anode catalyst, and a 14-micron reinforced perfluorosulfonic acid membrane. The total PGM loading is 0.125 mg_{PGM}/cm^2. Experiments and modeling of the PtCo/HSC cathode catalyst were consistent with an increase of power density at rated power from 1,095 to 1,183 mW/cm^2. The estimated system cost is shown in Figure 1.

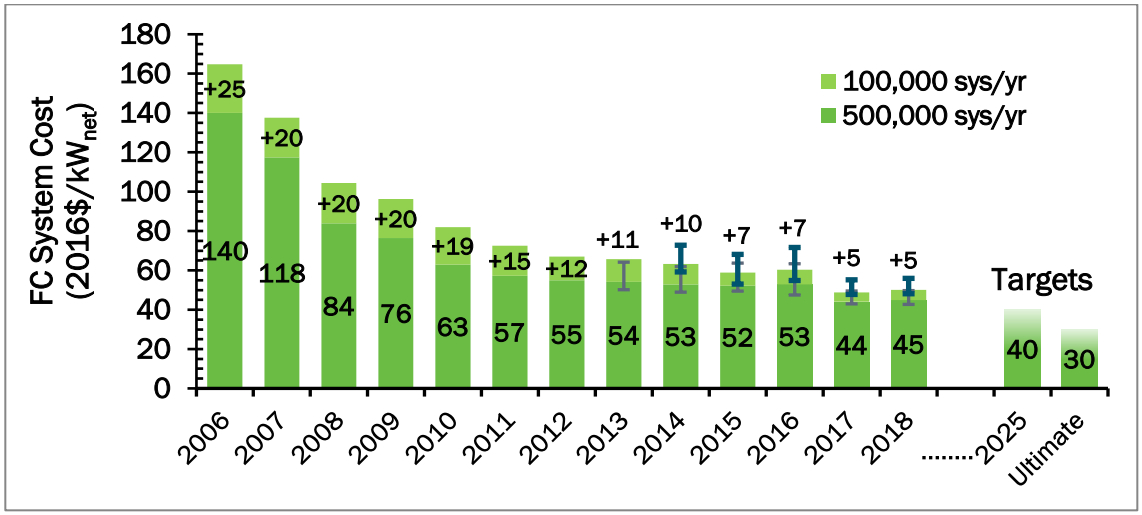

Figure 1. Modeled cost of an 80-kW_{net} PEM fuel cell system based on projection to high-volume manufacturing (2016 U.S. dollars)

The system cost analysis also includes the system description and state-of-the-art component materials and manufacturing cost for a 160-kW_{net} fuel cell system suitable for medium-duty vehicles. This system power output is equivalent to fuel cell systems for buses, and the analysis shares many common assumptions with previous analyses conducted for fuel cell buses, most recently in 2016. The system assumes a higher Pt

[1] Based on 2018 draft Fuel Cell System Cost record, to be published at https://www.hydrogen.energy.gov/program_records.html.

loading, 0.35 mg_{PGM}/cm^2, for increased durability. The current high-volume manufacturing system cost is estimated to be \$80/kW_{net} at 100,000 systems/yr (\$87/kW_{net} at 50,000 systems/yr).

To enable vehicle commercialization, fuel cell systems must also meet the subprogram's durability targets. These targets vary by application; for automotive systems, DOE has set a 2025 target of 8,000 hours, which is meant to represent the durability requirement in terms of miles driven (150,000 miles) for a larger range of drivers.

The durability of fuel cell electric buses has been evaluated since 2000 in transit agency demonstrations and has continued to increase after having surpassed the 2016 interim target of 18,000 hours in 2015. There are 12 buses that have now been on the road for more than 18,000 hours. The current bus maximum lifetime is more than 29,000 hours and was set by a bus that continues to operate, surpassing the ultimate target of 25,000 hours.[2] Furthermore, four other fuel cell systems have surpassed the 25,000-hour interim target. These are encouraging demonstrations of durability by several buses.

Consortia

ElectroCat (Electrocatalysis Consortium)

The subprogram established ElectroCat in FY 2016 under the umbrella of DOE's Energy Materials Network to address the materials problem of developing high-performance, low-cost, PGM-free catalysts for automotive fuel cells. ElectroCat aims to accelerate PGM-free catalyst and electrode development by coordinating relevant expertise and tools at the national labs to facilitate access to external researchers. In FY 2018, the core group of national laboratories continued to make progress in areas of catalyst and electrode development and optimization, understanding the structure-function relationship of catalytic active sites through advanced characterization, electrochemical and simulation techniques, and high-throughput modeling and synthesis of PGM-free catalysts.

- The core consortium team began working with the four awardees of the 2017 ElectroCat funding opportunity announcement (FOA): Carnegie-Mellon University, Giner, Inc., GreenWay Energy, LLC, and Pacific Northwest National Laboratory.
- **Improved catalyst synthesis and electrode optimization:** Cyanamide and polyaniline, (CM+PANI)-Fe-C, was improved to 27 mA/cm^2 at 0.9 $V_{IR\text{-}free}$ in H_2/O_2 on its second polarization, exceeding the 2018 milestone of 25 mA/cm^2 (Figure 2). This was achieved by modifying the catalyst break-in procedure and optimizing the ionomer to carbon ratio and catalyst loading in MEA testing.
- **Catalyst performance from FOA projects:** Carnegie Mellon University demonstrated 113 mA/cm^2 at 0.8 V and 268 mW/cm^2 at 0.7 V in H_2/air using a University of Buffalo-developed Fe-N-C catalyst. Giner demonstrated 16.5 mA/cm^2 at 0.9 $V_{IR\text{-}free}$ using a University of Buffalo-developed Mn-N-C catalyst, which is exceptional performance for a Mn-based PGM-free catalyst (Fe-free).
- **High-throughput, combinatorial methods for PGM-free catalyst R&D:** Continued work to optimize (AD)-Fe-N-C catalyst synthesis using a high-throughput robotic system, and tested samples via high-throughput oxygen reduction reaction (ORR) activity-testing using a 25-electrode combinatorial fuel cell hardware. The local atomic structure, phase composition, and near-surface composition were also characterized using X-ray diffraction, X-ray absorption spectroscopy, electron energy loss spectroscopy, and X-ray photoelectron spectroscopy.
- **Spectroscopic and electrochemical experiments with probe molecules to count active sites:** (AD)Fe-N-C catalysts exposed to nitrite solution (as a source of the nitric oxide probe molecule) showed that the

[2] Eudy, L. and Post, M., "Fuel Cell Buses in U.S. Transit Fleets: Current Status 2018," https://www.nrel.gov/docs/fy19osti/72208.pdf.

active site density is *ca.* 3 10^{12} sites cm^{-2} (*ca.* 0.5/Fe atom) and that the turnover frequency for ORR on these sites at 0.80 V is 1.7 e^- $site^{-1}$ s^{-1}. Complementary density functional theory study has identified that probe molecules bind to Fe and to the graphene only in the presence of defects or epoxides local to the FeN_x sites, as evidenced by calculated binding energies of potential probes/poisons to various active site structures.

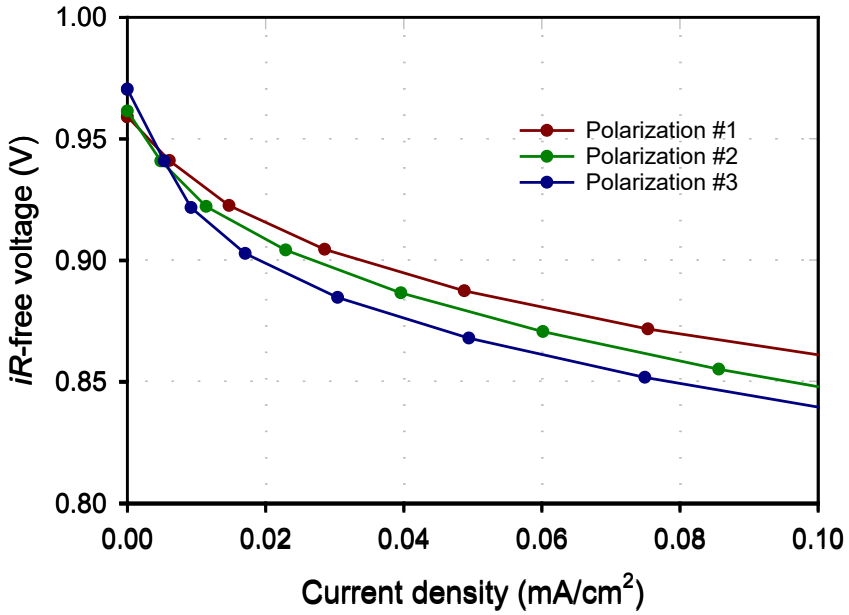

Figure 2. First three polarization curves of (AD)Fe-N-C catalyst in H_2/O_2, 1.5 bar pressure

Late in FY 2018, a second group of FOA projects were announced, and their goals address the key challenges in PGM-free catalyst development mentioned above. Northeastern University is leading a project developing M_x-N-C catalysts with a high density of multiple-metal-center sites. Indiana University-Purdue University Indianapolis is leading a project focusing on mesoporous PGM-free catalysts based on hierarchically porous carbon sphere and ionomer/catalyst interface controls within the cathode layer. Vanderbilt University is leading a project focusing on incorporation of PGM-free catalyst powders into sub-micron-diameter electrospun particle/polymer fiber catalyst layers. Pajarito Powder, LLC is leading an industry partner-led project with EWII Fuel Cells, LLC focusing on improved scalable catalyst synthesis and MEA integration. United Technologies Research Center is leading a project focusing on transition-metal oxide PGM-free catalysts based on high-throughput computational methods to identify corrosion-resistant oxides and a guided-design approach to optimization of oxygen reduction activity.

FC-PAD (Fuel Cell Performance and Durability)

The FC-PAD consortium continues to advance performance and durability of PEM fuel cells (PEMFCs) working in three main areas: (1) improving high-current-density performance at low PGM loadings, (2) developing the knowledge base for high-performance, highly durable PEMFC components, and (3) developing new diagnostics, characterization tools, and models. FC-PAD works to develop MEAs meeting the 2025 technical targets in cost, PGM loading (≤0.125 mg_{PGM}/cm^2), performance, and durability concurrently. National laboratory members also work to support four DOE-funded FOA projects led by 3M, GM, United Technologies Research Center, and Vanderbilt University, facilitating the access of industry and academic partners to national laboratory expertise and capabilities.

- **Novel array electrodes:** FC-PAD results have indicated that state-of-the-art electrode structures are hindered by severe mass-transport limitations during high-power operation, in part due to transport resistance induced by the ionomer, particularly as the Pt loading decreases. FC-PAD is pioneering free-standing arrays of vertically oriented ionomer channels to serve as non-tortuous pathways for proton conduction with catalyst filling between the pillars, thereby improving performance with decreased ionomer needed in catalyst ink formulation.

- **Catalyst conditioning protocols:** Several commercial Pt and PtCo catalysts supported on high- and low-surface-area carbons at loadings of 0.05–0.15 mg_{Pt}/cm^2 were subjected to an FC-PAD-developed conditioning protocol, and the mass activity was measured to determine optimal conditioning. The number of conditioning cycles needed was unique to each catalyst. The Pt particle size distribution did not change substantially for Pt/C, whereas the average particle size in PtCo/C catalysts increased significantly. With proper conditioning it was possible to increase the mass activity by as much as a factor of 3, indicating the importance of catalyst conditioning in MEA testing.

- **On-road PEMFC technology benchmarking:** To guide future R&D needs, the consortium analyzed two PEMFC components from the Toyota Mirai (as approved by Toyota): one MEA from a Mirai stack operated for 300 hours and one MEA from a Mirai stack operated for 3,000 hours. An initial analysis of the two MEAs showed no readily evident differences between them, demonstrating excellent durability. When the individual MEA components were subjected to the DOE-US DRIVE Fuel Cell Tech Team's accelerated stress test, the materials failed tests for catalyst and support lifetime, and there was significant growth in Pt particle size and preferential Co leaching from the PtCo/C cathode catalyst. The stability of the MEA materials in a real-world drive cycle suggests that the mitigation strategies implemented in the complete Mirai fuel cell system successfully prevent MEA degradation to a great extent, albeit it at higher catalyst loadings than the DOE target.

Low-PGM Catalysts

A project led by GM has continued to make progress on the results of their accessible porous carbon-supported PtCo catalysts from 2017, which exceeded the activity target of 8 kW/g_{PGM}. The benefit of the accessible porous carbon, particularly at high current densities, is evidenced by the polarization curves in Figure 3. Remarkably, the PtCo/HSC-f catalyst matches or surpasses the performance of a catalyst used in commercial FCEVs despite having less than one-fifth the platinum loading. The PtCo/HSC-e catalyst is the same catalyst on which the state-of-the-art system cost estimate is based. Although these catalysts demonstrate exceptional performance, both need further improvement to meet performance and durability targets concurrently.

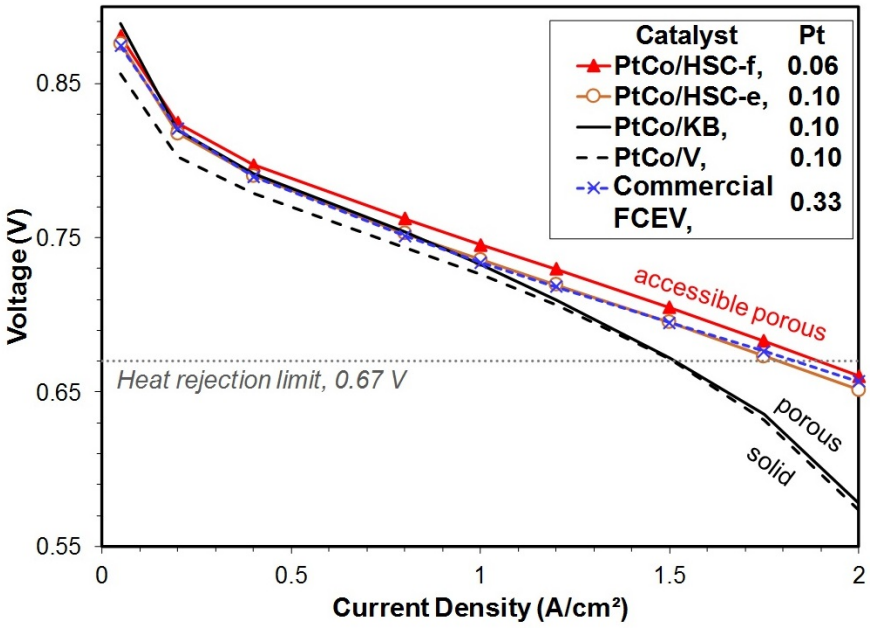

Figure 3. Fuel cell polarization curves of low-PGM-content PtCo/HSC catalysts with improved high-current-density performance compared to PtCo alloy catalysts supported on high- and low-surface-area carbons and on-the-road fuel cell technology

A project led by 3M demonstrated further progress on ultrathin-film catalysts, building on the understanding that a thin underlayer of Ir or Ta can stabilize Pt and the increased activity and stability resulting from surface additives such as Ru, Ir, Ta, and Cr. The electrocatalysts are based on a stabilized, layered catalyst structure consisting of a surface Pt layer and an underlayer of Ir between the Pt surface layer and the nanostructured thin film (NSTF) perylene red 149 whisker support. Both MEAs contain <0.1 mg_{PGM} and exceed the PGM utilization target of 8 kW/g_{PGM}: the 31Pt/26Ir/NSTF catalyst achieved 9.1 kW/g_{PGM} and the 50Pt/11Ir/NSTF catalyst achieved 9.4 kW/g_{PGM}.

Innovative Fuel Cell Concepts

Intermediate-Temperature Membranes

Intermediate-temperature fuel cells have the potential to reap the benefits of favorable kinetics and decreased sensitivity to fuel impurities (e.g., CO), both of which enable reduced PGM catalyst usage as well as higher efficiency due to the production of useful waste heat and/or the elimination of balance-of-plant components. Los Alamos National Laboratory is leading a project to enable fuel cells operating over a temperature range of 80°–220°C by implementing a highly conductive, non-leachable solid phosphonated ionomer into the electrodes with a phosphoric acid-doped, ion-pair-coordinated quaternary ammonium polymer membrane. Performance testing of these MEAs in H_2/O_2 results in peak power densities of 1,130 mW/cm^2 and 1,480 mW/cm^2 at 160°C and 200°C, and significant improvement over current state-of-the-art polybenzimidazole (PBI)-based membranes across all temperature ranges (Figure 4). The improved performance of the membrane coupled with the fluorinated electrode ionomer was attributed to a combination of lower phosphoric acid poisoning, exceptional water tolerance, and higher anhydrous proton conductivity at elevated temperatures.

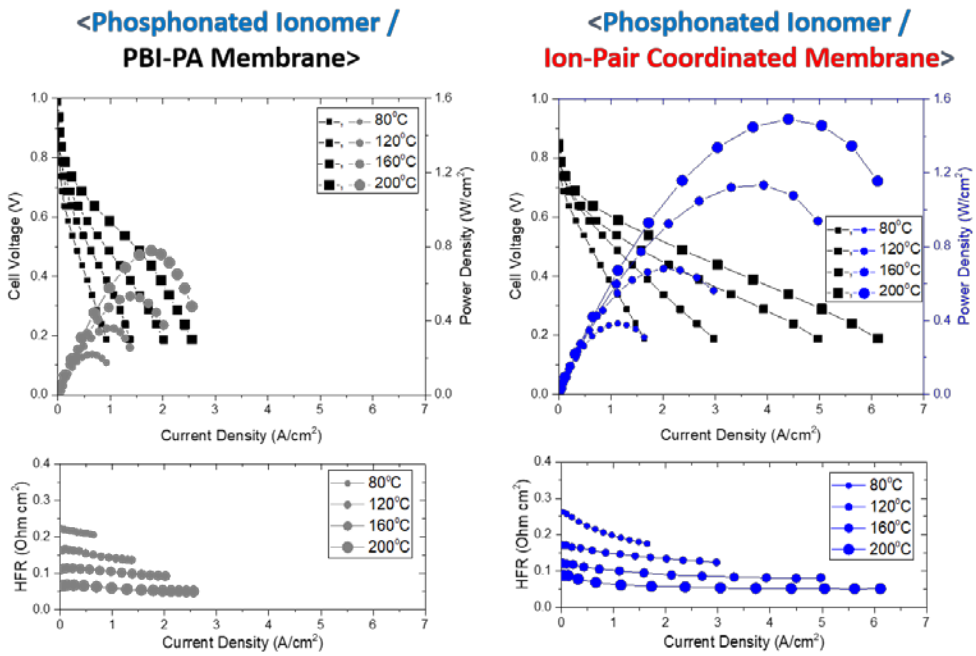

Figure 4. Polarization and power density curves (top) comparing the performance of conventional PBI-PA membranes and Los Alamos National Laboratory-developed ion-pair-coordinated membranes across operating temperatures 80°–200°C and corresponding high-frequency resistance measurements (bottom)

Reversible Fuel Cells

Reversible fuel cells (RFCs) are able to provide easily dispatchable power and are sufficiently flexible to address grid and microgrid reliability, resiliency, and other needs. RFCs contain both fuel cell and electrolyzer functions in a single device, and they are of interest for several stationary applications, including their ability to address intermittency issues with solar and wind power with a minimal footprint.

The viability and cost-competitiveness of RFC technologies requires continued foundational R&D in materials compatibility, enhanced durability, and optimized bidirectionality to improve roundtrip efficiency and meet long-term targets of less than $1,250/kW capital cost and a 5,000-cycle lifetime. Maintaining performance during repeated cycling between fuel cell and electrolysis modes as well as maximizing performance and efficiency in both modes are key challenges across the full range of potential RFC types.

Giner, Inc. led a project ending in FY 2018 that focused on an improved PGM-free bifunctional oxygen electrode capable of high activity for the ORR and the oxygen evolution reaction (OER) for use in reversible fuel cells. Giner and SUNY Buffalo have developed transition-metal-based oxide nanocomposites and heteroatom-doped graphene tube (carbon nanotube) catalysts with high ORR/OER activities and limited durability. These catalysts were integrated with selected anion-exchange ionomers and membranes. The MEAs were tested under both fuel cell and electrolyzer operating modes. Supporting electrolyte (KOH) decreased overpotential in electrolyzer mode and enabled demonstration of 600 h durability. The low oxidative stability of the membrane and ionomer was cited as a key barrier to cycling of the cell between electrolyzer and fuel cell modes and operation without KOH solution.

In 2018, FCTO awarded four new RFC-based projects covering all the technologies:

1. Lawrence Berkeley National Laboratory: Novel Bifunctional Electrocatalysts, Supports and Membranes for High-Performing and Durable Unitized Regenerative Fuel Cells, focusing on low-temperature reversible PEMFCs
2. Giner, Inc: High-Efficiency Reversible Alkaline Membrane Fuel Cells
3. Northwestern University: Efficient Reversible Operation and Stability of Novel Solid Oxide Fuel Cells
4. Georgia Tech: Durable, High-Performance Unitized Reversible Fuel Cells Based on Proton Conductors, focusing on ceramic-based high-temperature proton conducting electrolyte fuel cells.

BUDGET

The FY 2018 appropriation was $32.0 million for the Fuel Cell R&D subprogram. In FY 2018 the subprogram funded early-stage R&D efforts in key areas focusing on fuel cell stack components to increase performance and durability while reducing cost, broken down into the five areas represented in Figure 5. Funding was primarily directed toward improving catalysts and electrodes to increase performance and reduce catalyst cost by developing ultra-low-PGM or PGM-free catalysts for oxygen reduction. The ElectroCat consortium, four existing FOA projects, and five new FOA projects are included in the Catalysts and Electrodes funding area. Nearly an eighth of the total funding was dedicated to the FC-PAD consortium, including the core national lab membership and its four associated industry-/university-led projects. The remaining funding was provided to award and forward-fund 16 projects from the Fuel Cell Technologies Office FY 2018 FOA covering testing, lower-cost membranes, PGM-free catalysts, and RFCs.

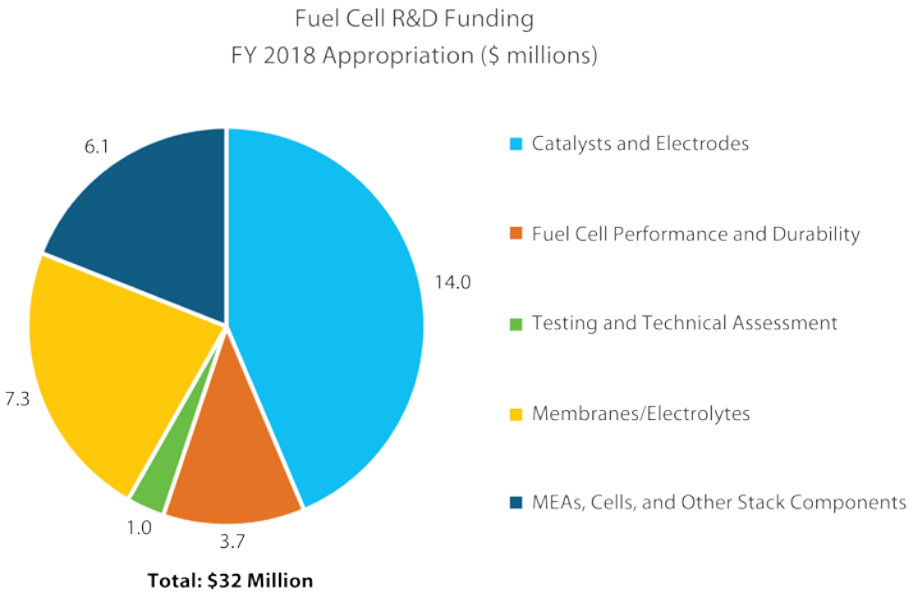

Figure 5. Fuel Cell R&D subprogram FY 2018 appropriation

FY 2019 PLANS

In FY 2019, the Fuel Cell R&D subprogram will continue early-stage applied fuel cell R&D for diverse applications that employ a variety of technologies (including PEM and alkaline membrane fuel cells). R&D will focus in the key areas of fuel cell components and materials, such as catalysts, alkaline and non-water-

dependent membranes, electrodes, and component innovations (such as components for reversible fuel cells), with an emphasis on cost reduction and durability improvement.

The subprogram's consortia will continue fostering national lab capabilities and collaborations with stakeholders and the research community. The subprogram will place particular emphasis on expediting the development of PGM-free catalysts and electrodes through ElectroCat. The Fuel Cell R&D subprogram will also continue efforts to advance fuel cell performance and durability through FC-PAD. Both consortia are now working with FOA projects, which will further increase the cooperativity and effectiveness of the respective communities. Ongoing support of modeling will guide component R&D, enabling exploration of alternate system components and configurations. Efforts to advance fuel cell performance and durability will increase emphasis on the challenging efficiency and durability requirements of medium- and heavy-duty applications, including trucks, rail, and marine.

The subprogram will further pursue early-stage R&D on components such as membranes, catalysts, and electrodes to optimize them for unitized RFCs that store energy and generate power as required in support of DOE's Beyond Batteries initiative. The subprogram will also explore the potential of stationary fuel cells to supply primary/backup power to data centers.

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