V.0 Fuel Cells Sub-Program Overview

INTRODUCTION

The Fuel Cells sub-program supports applied, early-stage research and development (R&D) of fuel cell technologies for transportation applications, as well as stationary and early market applications, with a primary focus on reducing cost and improving durability. Efforts predominantly concentrate on R&D of fuel cell stack materials and components but also include system balance-of-plant components to achieve low-cost, high-performance fuel cell systems. The sub-program 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 due to the benefits inherent in fuel cell electric vehicles (FCEVs) (e.g., high efficiency, long driving range, zero emissions). One stationary application for fuel cells is distributed power generation, including combined heat and power (CHP) for residential and commercial applications. Existing early markets and near-term markets generating traction for adoption of FCEVs include backup power, auxiliary power units, and specialty applications such as material handling equipment. The sub-program's R&D portfolio is primarily focused on polymer electrolyte membrane (PEM) fuel cells but also includes longer-term technologies, such as alkaline membrane 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 sub-program's fuel cell tasks are delineated in the Fuel Cell Technologies Office (FCTO) *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 sub-program's goal is to advance fuel cell technologies for transportation, stationary, and early market applications.

OBJECTIVES[1]

The sub-program's key objectives include:

- Developing a 65% peak-efficient, direct hydrogen fuel cell power system for transportation that can achieve 5,000-h durability (ultimate 8,000 h) and be mass produced at a cost of $40/kW by 2020 (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-h durability at an equipment cost of $1,500/kW by 2020.
- Developing medium-scale CHP systems (100 kW–3 MW) by 2020 that achieve 50% electrical efficiency, 90% CHP efficiency, and 80,000-h 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) 2017 TECHNOLOGY STATUS AND ACCOMPLISHMENTS

Reducing cost and improving durability while maintaining performance continues to be the key challenge facing fuel cell technology R&D. For platinum group metal (PGM)-based catalysts, both a reduction in PGM loading and an increase in membrane electrode assembly (MEA) area 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 (e.g., near the rated power point), but FY 2017 saw continued progress towards 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 will require a transition from PGM-based catalysts to PGM-free catalysts. Two low-PGM catalyst projects produced catalysts exceeding the 2020 technical target for specific power output (8.0 kW/g_{PGM}). The sub-program's consortia (Fuel Cell Performance and Durability [FC-PAD] and the Electrocatalysis Consortium [ElectroCat]) made great strides in materials innovation

[1] Note: Targets and milestones were recently revised; therefore, individual project progress reports may reference prior targets.

and phenomenological understanding of fuel cell catalysts and electrode components through increased cooperative application of national laboratory capabilities.

One of the most important metrics used to guide the sub-program's R&D efforts is the projected high-volume manufacturing cost for automotive fuel cells, which is tracked on an annual basis. The sub-program is targeting a cost reduction to $40/kW by 2020. Long-term competitiveness with alternative powertrains is expected to require further cost reduction to $30/kW, which represents the sub-program'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}$ when manufactured at 100,000 units/yr and $45/$kW_{net}$ when manufactured at a volume of 500,000 units/yr. For comparison, the estimated cost of automotive PEM fuel cell systems that are based on currently deployed commercial technology in 2017 is approximately $230/$kW_{net}$ when manufactured at a volume of 1,000 units/yr. The expected cost for an analogous system based on state-of-the-art materials (lower platinum loading catalyst, coated stainless steel bipolar plates) is $180/$kW_{net}$.

The 2017 projected system cost is significantly less than that of the previous year (the cost decreased from $53/$kW_{net}$ to $45/$kW_{net}$ at 500,000 systems/yr or from $60/$kW_{net}$ to $50/$kW_{net}$ at 100,000 systems/yr). The main change that reduced the system cost compared to the 2016 analysis is an increase in power density (749 mW/cm^2 to 1,095 mW/cm^2) with a concomitant decrease in total platinum (Pt) loading on the cathode (0.134 mg/cm^2 to 0.125 mg/cm^2). This reflects a change in the catalyst basis from a de-alloyed PtNi/C catalyst in 2016 to a PtCo/HSC (high surface area carbon) catalyst developed by General Motors (GM). The proprietary HSC support led to increased performance, which surpassed that of the PtNi/C catalyst; hence, PtCo was used as the 2017 baseline. In 2016, Argonne National Laboratory (ANL) modeled the de-alloyed PtNi/C catalyst under optimized conditions, and although PtCo/C had not been modeled for optimized conditions, there was a consensus between ANL and GM that de-alloyed PtNi/C would have similar performance to PtCo/C if both catalysts were supported by GM's proprietary HSC support. The similarity in performance is due to the expectation of similar cathodic kinetic reaction rates whether using PtNi or PtCo. The catalyst synthesis cost was based on PtNi. The synthesis method of PtCo is proprietary to GM; however, GM has said that the synthesis costs of the two catalysts would be equivalent. Additional changes that reduced cost compared to 2016 included improved bipolar plate stamping process assumptions and the removal of the hydrogen sensors from the fuel cell system design (but not from within the vehicle). The decreased system cost is shown in Figure 1.

To enable vehicle commercialization, fuel cell systems must also meet the sub-program's durability targets. These targets vary by application; for automotive systems, DOE has set a 2020 target of 8,000 h, which is meant to represent the durability requirement in terms of miles driven (150,000 mi) for a larger range of drivers. An independent assessment of the current fuel cell durability test data from leading fuel cell developers found that the current average state-of-the-art laboratory durability status, defined as average hours to 10% voltage degradation, is approximately 3,700 h.

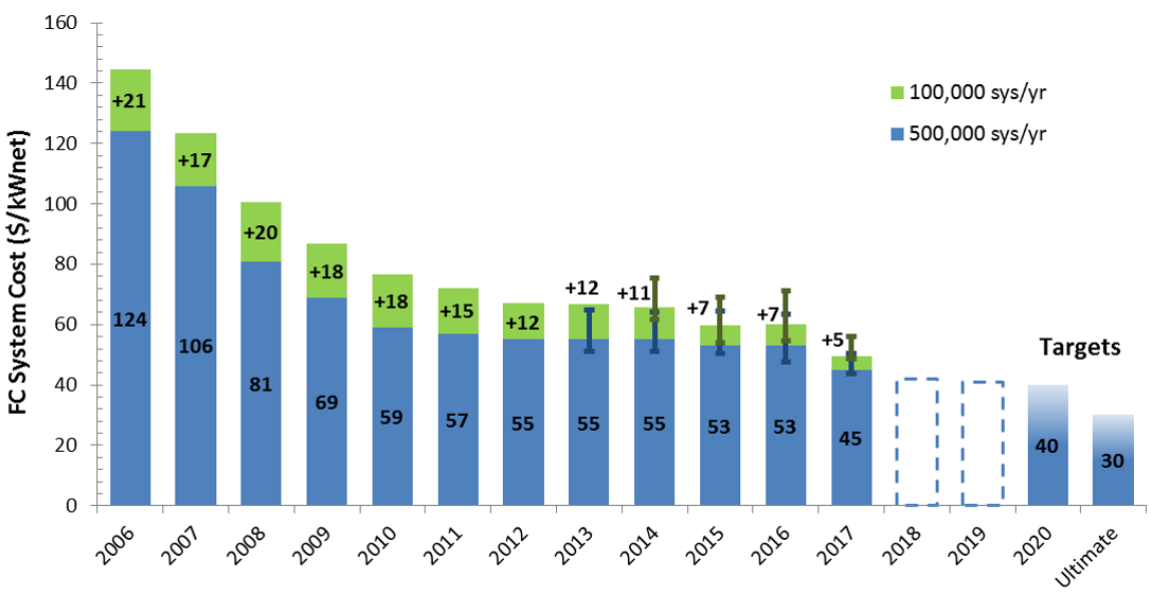


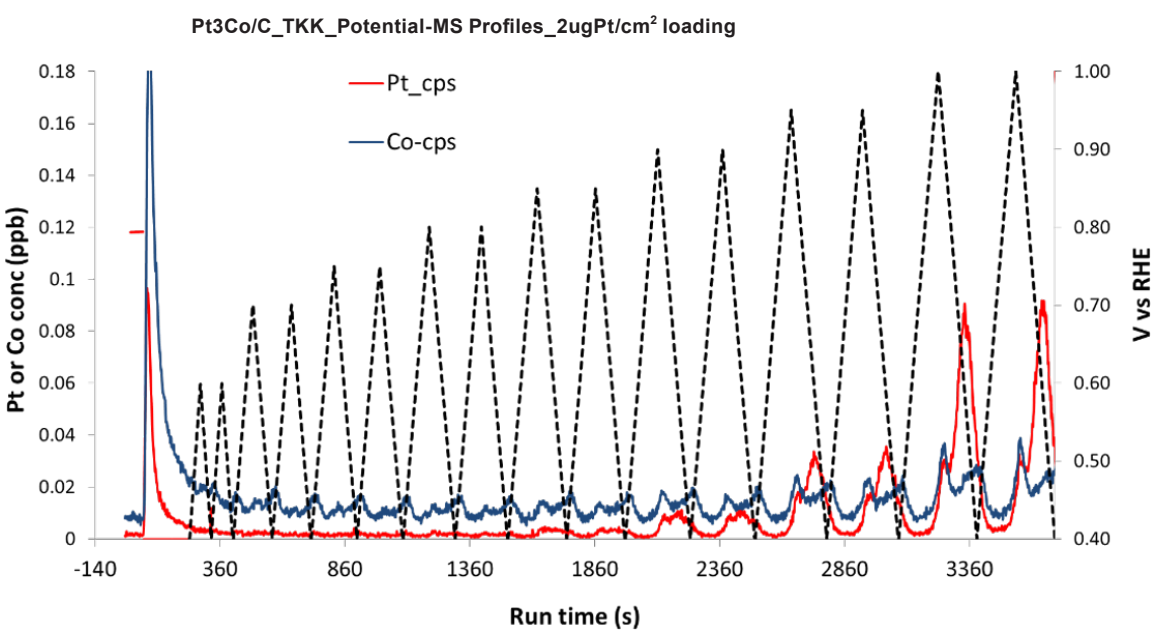
FIGURE 1. Modeled cost of an 80-kW_{net} PEM fuel cell (FC) system based on projection to high-volume manufacturing

Meanwhile, DOE independent validation of on-road FCEVs showed a more than four-fold increase in the maximum projected durability of fuel cell systems, increasing from 950 h in 2006 to over 4,100 h in 2017. Additionally, the maximum operating hours recorded for a single FCEV has remained at 5,600 h. The durability of fuel cell electric buses has also been evaluated since 2000 in transit agency demonstrations and has continued to increase after having surpassed the 2016 interim target of 18,000 h in 2015. The current bus maximum lifetime is over 25,300 h and was attained by a bus that continues to operate, surpassing the 2020 target of 25,000 h. High operational hours have been demonstrated by several other buses as well: nine have surpassed the 18,000-h interim target, with six demonstrating over 20,000 h in operation.

Consortia

Fuel Cell Performance and Durability (FC-PAD)

The FC-PAD consortium was formed to advance performance and durability of PEM fuel cells to further enable their commercialization. This R&D consortium is led by a team of national laboratories and began operations in FY 2016. Overall objectives of the consortium include building a knowledge base and optimizing transport and structures for highly performing, durable PEM fuel cell components; improving high current density performance at low PGM loadings; improving component stability and durability; and developing new diagnostics, characterization tools, and models. In FY 2017 work continued at the national laboratories in three main areas: cerium migration and cation interaction with membranes and ionomers; modeling and testing of ionomers, interfaces, and diffusion media; and characterization and testing of low-PGM alloy catalysts. The FC-PAD team evaluated a series of state-of-the-art PtCo catalysts from various sources (GM, Umicore, EWii) in terms of size, composition, and morphology using microscopy, inductively coupled plasma-mass spectrometry, and X-ray scattering methods, in beginning of life (BoL) and end of life (EoL) conditions (following several different accelerated stress test [AST] protocols). The Co leaching during AST was quantified (Figure 2), and it was determined that regardless of the starting structure and composition, all the alloy catalysts behaved similarly in terms of compositional stability, e.g., all catalysts lost greater than 50% of the Co, which resulted in (1) the electrocatalysts becoming more Pt-like and (2) Co migration into the membrane.



RHE – reference hydrogen electrode; TKK – Tanaka Kikinzoku Kogyo

FIGURE 2. Inductively coupled plasma-mass spectrometry quantification of Pt and Co leaching from a Pt_3Co/C TKK catalyst during potential cycling demonstrates the constant baseline leaching of Co with increased loss during cathodic sweep, and peaks due to Pt leaching during anodic sweeps at high potential (>0.95 V)

Significant progress also was made in other materials necessary for the function of electrodes, including ionomers, membranes, and electrode structures. The effect of cerium migration through ionomer and membrane polymers is important to increasing fuel cell durability by optimizing its incorporation into electrodes. Migration and diffusion coefficients were quantified at low (50%) and high (100%) relative humidity. Cerium zirconium oxide (CZO) was demonstrated to enhance membrane durability much better than the increased addition of cerium cations. Work also demonstrated advances in modeling of membrane structures and detailing water uptake with various cationic species. Combined modeling and experiments to understand interfaces demonstrated the importance of water droplets at the gas diffusion layer/flowfield boundary as well as the possible increase in performance through catalyst layer architecture optimization. Limiting-current diagnostics across a systematic study of different catalyst layers demonstrated the importance of accurate, effective active area measurements as well as the apparent dominance of diffusion-like transport at the local ionomer scale.

FC-PAD's approach couples national lab capabilities with funding opportunity announcements (FOAs) for an influx of innovative ideas and research. This year marked the beginning of the cooperation of the FC-PAD consortium with FOA projects led by 3M, GM, United Technologies Research Center, and Vanderbilt University.

ElectroCat (Electrocatalysis Consortium)

The sub-program also established a second consortium 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. The ElectroCat consortium aims to accelerate PGM-free catalyst and electrode development by coordinating relevant expertise and tools at the national labs to provide easy access to external researchers. ElectroCat's capabilities consist of high-throughput combinatorial methodologies, computational tools, and PGM-free catalyst expertise. In FY 2017, the core group of national laboratories established a list of capabilities and evaluated their value to the community by performing state-of-the-art experiments. Major improvements to fuel cell catalysts included an atomically dispersed (AD)Fe-N-C catalyst, developed to create single atomic Fe sites on the catalyst surface (no nanoparticles, minimal Fe clustering), as seen in high-angle annular dark field scanning transmission electron microscopy images (Figure 3a). The catalyst (AD)Fe-N-C comprises leaf-shaped structures with hierarchical pore structure, the result of Zn evaporation during pre-treatment. The oxygen reduction reaction (ORR) activity of this catalyst (half-wave potential, $E_{1/2}$) was increased by 20 mV (Figure 3b). This catalyst is the same one chosen for high-throughput synthesis at ANL and for which direct microscopic and spectroscopic evidence was found for a majority of Fe sites being on the surface. These Fe sites were shown to be more stable than those in another state-of-the-art catalyst that incorporates cyanamide and polyaniline, (CM+PANI)-Fe-C.

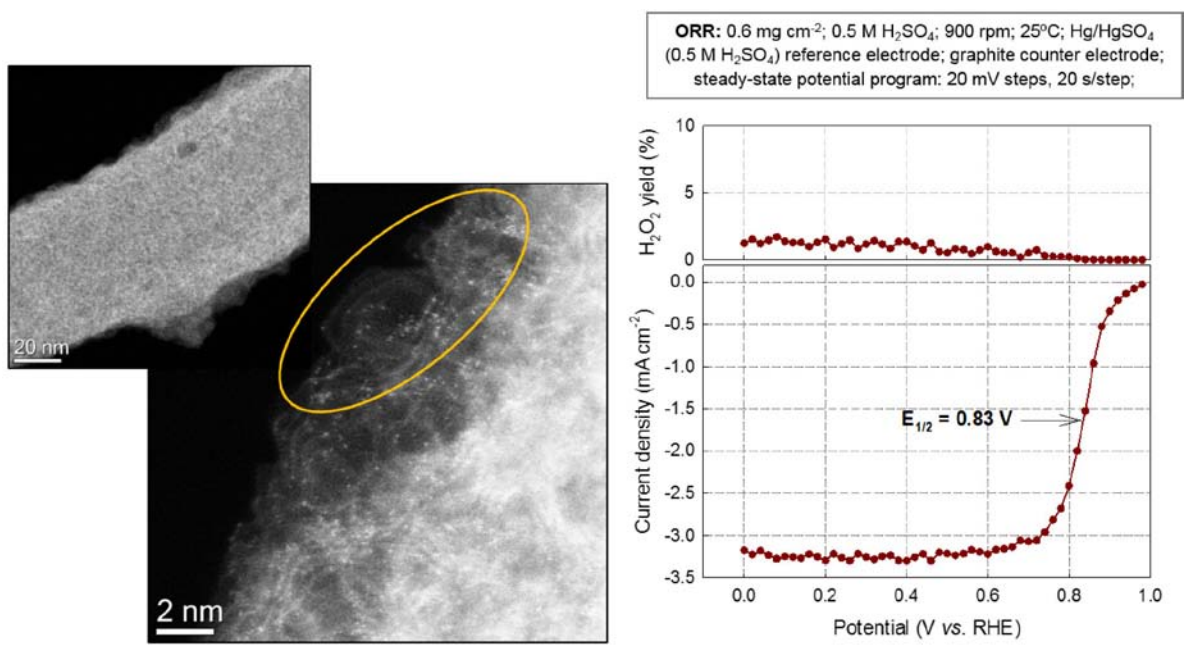


FIGURE 3. Characterization and testing of (AD)Fe-N-C catalyst: (a) high-angle annular dark field scanning transmission electron microscopy images (left); (b) H_2O_2 yield and ORR activity determined by rotating ring disc electrode testing (right)

The performance of the (CM+PANI)-Fe-C catalyst was improved by using Zn salts during preparation as pore-forming agents, by using magnetic separation to remove Fe clusters and nanoparticles, and by eliminating the hot-pressing step of MEA fabrication. These changes increase the H_2-air cathode performance by 25% over previous versions of the catalyst, up to 0.83 mA/cm^2 at 0.8 V (120 mA/cm^2 at 0.8 $V_{IR\text{-}free}$) (Figure 4).

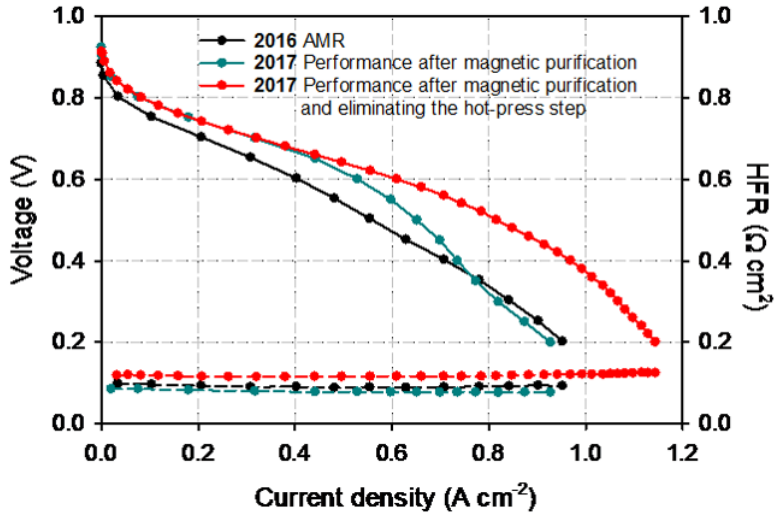


FIGURE 4. Polarization curve and high frequency resistance (HFR) measurements of (CM+PANI)-Fe-C(Zn) catalyst developed by LANL displaying improvement in performance over catalyst featured in 2016 Annual Merit Review status

High-throughput, combinatorial methods for PGM-free catalyst R&D are among the core competencies of ElectroCat. The high-throughput robotic catalyst synthesis system at ANL enhances foundational understanding of promising materials and expedites materials optimization by efficient, automated exploration of the relevant variable space. The rapid characterization of these catalysts is key to understanding structure-function relationships, which will inform further advances in catalyst development. In FY 2017, the utility of the system was demonstrated by the preparation of 40 samples of the (AD)Fe-N-C catalyst developed at Los Alamos National Laboratory (LANL) with varying composition, precursor, and pyrolysis temperature. These catalysts were then examined by high-throughput structural characterization (X-ray diffraction and X-ray absorption fine-structure spectroscopy) and ORR activity testing via rotating disk electrode. Analysis of the metadata collected from these systems is ongoing.

Pioneering results in the area of computational approaches to PGM-free catalysts were also demonstrated. At LANL, a durability descriptor calculation, based on an electron beam damage model, was applied to all atoms in 30 putative M_xN_y active sites. By examining the elastic collision energy of a potential-accelerated electron required to eject atoms from each site, and compiling these knock-on displacement threshold energies (E_{KODTE}) into a database, relative stabilities of these sites could be elucidated. These results suggest that nitrogen atoms are the most susceptible to damage for all of the structures and that edge sites, particularly those in an "armchair" configuration, were least stable. These calculations are amenable to high-throughput screening of possible active sites and fit with the consortium's goal of increasing foundational understanding of PGM-free catalysts through advanced computational capabilities at the national laboratories. Late in FY 2017, DOE organized a PGM-free modeling workshop to discuss ongoing challenges and capability needs in the area and to increase collaboration among computation experts and experimentalists. A report will be prepared based on the proceedings and released in FY 2018.

In FY 2017, the first group of FOA projects were announced, and their goals address the preeminent challenges in PGM-free catalyst development mentioned above. The first of these projects is led by Giner, Inc., and proposes to develop Mn-based, PGM-free catalysts that will not suffer from the durability issues of Fe-containing catalysts due to the Fenton activity of Fe. A Carnegie Mellon University-led project focuses on increasing power density and durability through control of electrode microstructure. Pacific Northwest National Laboratory is leading an effort to increase ORR electrocatalyst durability through synergetic active sites to decrease selectivity to peroxide formation. The fourth project, led by GreenWay Energy, LLC, will attempt rational design of PGM-free, ORR framework nano-structure catalysts.

Low-PGM Catalysts

In FY 2017, two catalysts were developed that surpassed the FCTO 2020 technical target for specific power output of 8.0 kW/g_{PGM} at the stipulated Q/ΔT of 1.45 kW/°C.

The first of these catalysts was developed by GM as part of a project that focuses on the need to develop catalysts with high performance and durability at both low and high current densities. The project aims to understand and overcome oxygen and proton transport limitations at high current density with low Pt loadings (<0.100 mg_{Pt}/cm^2). The highest specific activity to date was achieved using PtCo alloy particles supported on HSC. A PtCo/HSC catalyst (-f in

Figure 5) with a Pt loading of 0.063 mg_{Pt}/cm^2 showed the highest PGM utilization of any catalyst to date: 10.6 kW/g_{PGM} at 150 kPa and 94°C (14.1 kW/g_{PGM} at 250 kPa and 94°C), meeting the Q/ΔT requirement imposed by DOE targets. The two HSC catalysts developed in 2017 (PtCo/HSC-e and -f) show similar high activity at low current density compared to the 2016 PtCo/HSC-a catalyst, but with decreased transport losses at high current density. This improvement in performance is attributed to improved understanding of Pt and Co dissolution and to the selection of the carbon support based on improved understanding of support degradation and resistance to mass transport in nanopores.

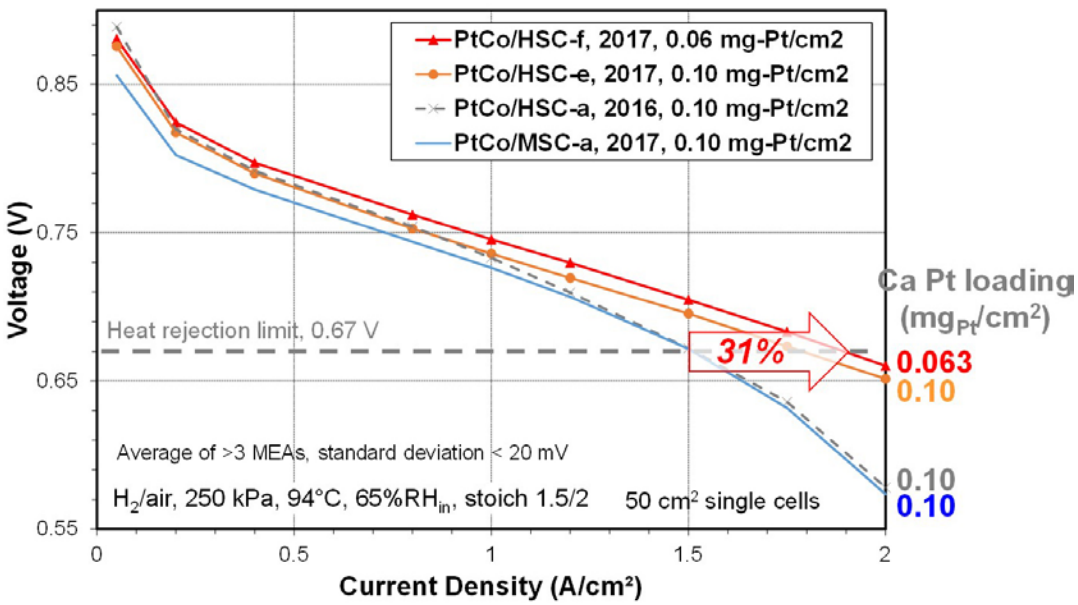


FIGURE 5. Fuel cell polarization curves of PtCo/C catalyst at different cathode Pt loadings showing improved high current density performance in recently developed catalysts over medium surface area carbon catalyst and the highest-performing HSC catalyst from 2016

The FCTO 2020 technical target for specific power output was also surpassed by a catalyst developed by 3M. The 3M project focuses on a thin film catalyst layer approach using Pt alloy (PtNi, PtNiIr) nanoporous thin film and ultra-thin film (UTF) catalysts, two distinct catalyst layer morphologies based on 3M technology. The catalysts have been improved in the last year by modifying film preparation methods, including annealing and leaching steps to control composition and morphology. Several catalysts with a total PGM loading of 0.077 mg_{PGM}/cm^2 exceed the mass activity target of 0.044 A/mg_{Pt}. One of these catalysts, PtNi UTF-f, achieves a specific power output of 8.1 kW/g_{PGM}. The improvement in high current density performance is demonstrated in Figure 6. The addition of a relatively small amount of Ir to these catalysts was found to improve their durability.

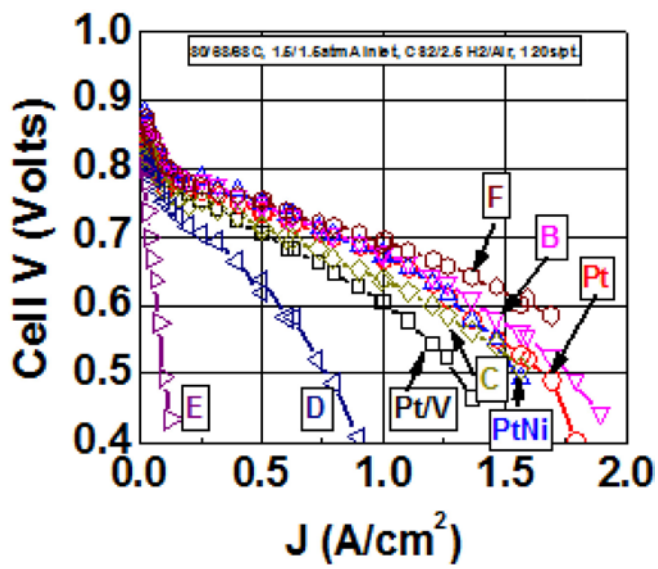


FIGURE 6. Fuel cell polarization curves of PtNi and PtNilr nanoporous thin film and ultra-thin film catalysts demonstrating the improvement in performance at high current density

Electrolytes

Alkaline Membranes

Significant improvements in the performance and durability of alkaline electrolyte membranes (AEMs) were demonstrated in FY 2017. Two areas in particular are highlighted: perfluoro-based and hydrocarbon-based alkaline membranes.

The performance of functionalized polyphenylene membrane AEMs at LANL was improved significantly this year. The hexamethyl ammonium side chains were down-selected on the basis of low area specific resistance (~0.05 Ω cm^2) and improved chemical stability. The achieved area specific resistance meets the FCTO technical target for AEM area specific resistance of <0.1 Ω cm^2. These membranes exhibited no chemical degradation at 80°C during 3,600 h in 0.5 M NaOH (1,800 h in 4 M NaOH) in ex situ testing and >300 h in in situ testing. A large increase in power density was also made possible by further exploration of the anode (hydrogen oxidation reaction) catalyst deactivation mechanism first cited in FY 2016 in which phenyl rings within the ionomer adsorb strongly on Pt catalyst surfaces. Using a commercially available PtRu catalyst, which does not adsorb these groups as strongly, and switching to an electron-withdrawing fluorene-based polymer made an increase in power from 0.4 W/cm^2 to ~1 W/cm^2 possible (Figure 7). Furthermore, the low HFR (0.07 cm^2) of this catalyst is commensurate with sufficient hydroxide conductivity for high-performing AEM fuel cells.

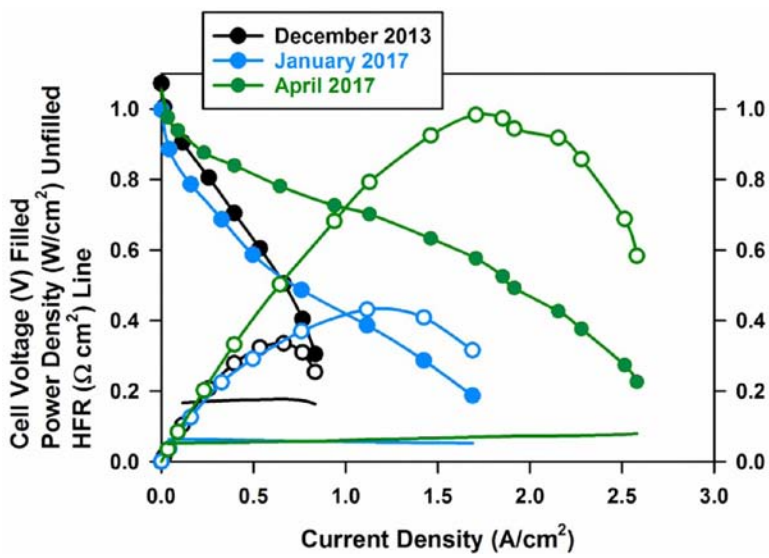


FIGURE 7. Polarization curve (closed symbols), power density (open symbols), and HFR (no symbols) demonstrating recent progress performance in hydrocarbon-based alkaline membranes

Advances in perfluoro-based AEMs at the National Renewable Energy Laboratory were demonstrated through MEA testing using Pt/C cathode and PtRu/C anode catalysts (Figure 8). The sulfonamide-linked chemistry was further optimized, achieving hydroxide ion conductivity of to up to 52 mS/cm and up to 90% of theoretical ion exchange capacity. Improved processing has enabled the synthesis of over 200 g, which was distributed to 10 other laboratories. A University of Connecticut (UConn)-prepared gas diffusion electrode (GDE) applied to the perfluoro AEM demonstrated enhanced performance at 60°C, meeting the FCTO FY 2017 area specific resistance target of <0.1 Ω cm^2. Unlike the perfluoro AEM catalyst-coated membrane (PFAEM CCM), which lost all performance within 15 h, the UConn GDE retained reasonable performance after 500 h of testing at 600 mA/cm^2.

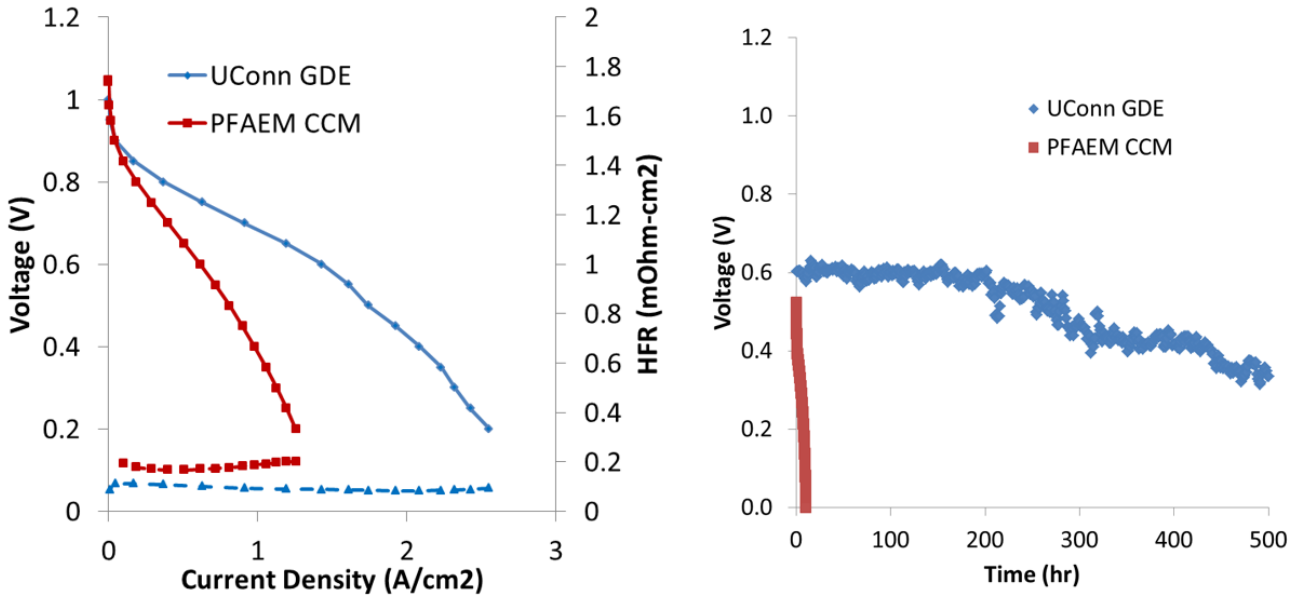


FIGURE 8. Polarization curves at 95°C and 50% inlet humidity for perfluoroimide-acid-based membranes (red lines) and a perfluorosulfonic acid control (blue lines). Voltage versus current density data are shown by lines with symbols, HFR is shown with solid lines, and HFR corrected data are shown with dashed lines.

Rotating Disc Electrode (RDE) Testing Protocols and Best Practices Disseminated

Accurate benchmarking of baseline Pt/C catalysts is necessary to expedite the ultra-low PGM catalyst development crucial for commercialization of FCEVs. It has been shown that measured ORR activity and reproducibility are correlated with system cleanliness, evaluation protocol, operating conditions, and the RDE catalyst film thickness and uniformity resulting from ink formulation, composition, and film drying. In order to establish baseline performance, a best practices and common protocol for RDE testing was developed by three independent laboratories and used to test three commercially available Pt/C catalysts (from Umicore, Johnson Matthey, and TKK). The performance of these catalysts and the reliability of the protocols for RDE testing used at the three laboratories were verified, and these results and best practices were published this year in a journal article[2]. This publication provides the standard test protocol and best practices to assist the scientific community by enabling procedural consistency and less variability.

L'Innovator

The L'Innovator (for "Lab innovator") aims to accelerate the commercialization of innovative hydrogen and fuel cell technologies developed at national labs and enable a robust domestic industry and supply base in the emerging area of hydrogen and fuel cells. In FY 2017, FCTO successfully launched the first L'Innovator pilot, building upon the due diligence carried out in FY 2016 to develop the concept. The national laboratories involved in the pilot, LANL and Brookhaven National Laboratory, executed an interlab agreement and successfully completed a cooperative research and development agreement call for a commercialization partner, run by LANL. In FY 2018, the commercial partner will work with LANL and Brookhaven National Laboratory to develop a pathway towards buildout of a working prototype MEA that can be made via a roll-to-roll process by the end of the year. Specifically, LANL's MEA technology will be optimized to integrate core-shell catalyst technology developed at Brookhaven National Laboratory, and National Renewable Energy Laboratory will be involved in developing and applying roll-to-roll processing methods to demonstrate manufacturability.

[2] S.S. Kocha, K. Shinozaki, J.W. Zack, D.J. Myers, N.N. Kariuki, T. Nowicki, V. Stamenkovic, Y. Kang, D. Li, D. Papageorgopoulos. Electrocatalysis. 8 (2017) 366.

BUDGET

The FY 2017 appropriation is $32.0 million for the Fuel Cells sub-program. In FY 2017 the sub-program funded early-stage R&D efforts focused on fuel cell stack components to increase performance and durability while reducing cost, broken down into the four key areas shown in Figure 9. FY 2017 funding was primarily directed towards improving catalysts and electrodes in order to increase performance and reduce catalyst cost by developing ultra-low PGM or PGM-free catalysts for oxygen reduction; ElectroCat and the four newly awarded FOA projects are included in this area. Slightly more than a quarter of the funding total was dedicated to the FC-PAD consortium, including the core national lab membership and its four associated industry/university-led projects. The sub-program also continued to fund innovative projects for resilient, lower-cost membrane materials, with approximately $3.2 million in funding.

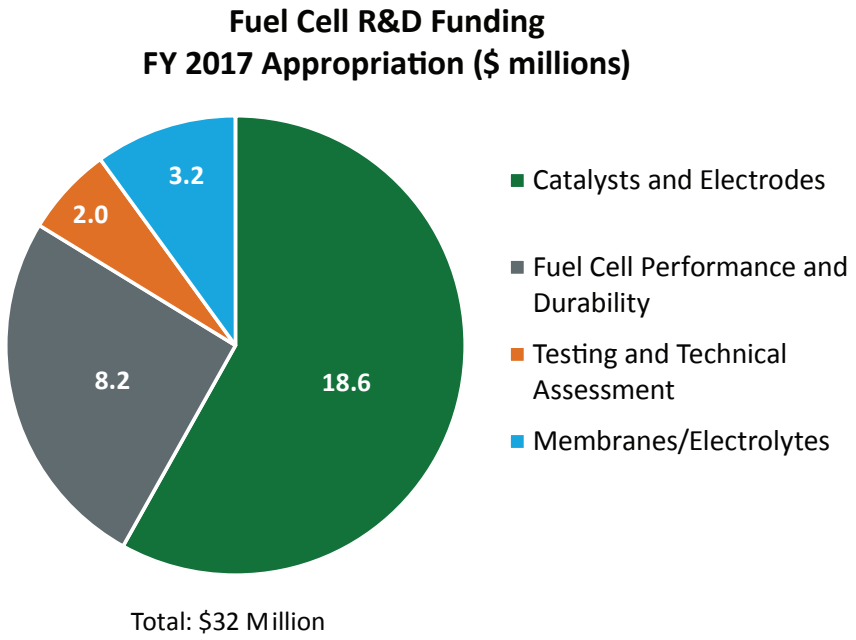


FIGURE 9. FY 2017 Appropriations

UPCOMING ACTIVITIES AND PLANS

In FY 2018, the Fuel Cells sub-program will continue early-stage applied fuel cell R&D for diverse applications that employ a variety of technologies (including PEM and AEM fuel cells) and a range of fuels (including hydrogen, natural gas, and liquid fuels). Fuel Cells R&D will focus on 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 sub-program's consortia will continue fostering national lab capabilities and collaborations with stakeholders and the research community. The sub-program will place particular emphasis on expediting the development of PGM-free catalysts and electrodes through ElectroCat, utilizing unique experimental and advanced computing capabilities at the national labs. Fuel Cells R&D will also continue efforts to advance fuel cell performance and durability through FC-PAD. Both consortia are now working with their first set of FOA projects, which will further increase the cooperativity and effectiveness of the respective communities. Ongoing support of modeling will guide component R&D, benchmarking complete systems before they are built and enabling exploration of alternate system components and configurations. The sub-program will also continue to pursue efforts to expand L'Innovator to increase adoption of the groundbreaking early-stage fuel cell technologies being developed by the national labs.

Future activities are subject to appropriations.

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