FC-PAD: Fuel Cell Performance and Durability Consortium

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Project End Date: September 30, 2021

Overall Objectives

Advance performance and durability of polymer electrolyte membrane fuel cells (PEMFCs) primarily at a *pre-competitive* level.

- Improve high-current-density performance at low Pt loadings:
  - Improved electrode structures.
  - Reduced mass transport losses.
  - Loading: ≤0.125 mg Pt/cm^2 total.
  - Performance @ 0.8 V: 300 mA/cm^2.
  - Performance @ rated power: 1,800 mW/cm^2.
- Develop knowledge base for more durable and high-performance PEMFC components:
  - Understand science of component integration, for example, ionomer interactions with carbon/interfaces between electrodes and gas diffusion layer (GDL) or membranes.
  - Improve component durability (e.g., membrane stabilization, self-healing, electrode-layer stabilization).
- Develop new diagnostics, characterization tools, and models.
- Provide support to DOE-funded FC-PAD projects from funding opportunity announcement DE-FOA-0001412 to develop a knowledge base for both industrial and academic developers.

Fiscal Year (FY) 2018 Objectives

- Understand electrode layer structures:
  - Measure effect of ionomer, catalyst inks, carbon, and cations on electrode structure, catalyst/carbon particle agglomeration, oxygen reduction reaction kinetics, and membrane conductivity and water uptake.
  - Macroscale and microstructure modeling of electrode structure—measure ionomer distribution observations as input data for model development, and define relative electrode resistive losses (e.g., local oxygen transport).
  - Define current state-of-art commercial membrane electrode assemblies (MEAs) to define materials baseline. Analysis of Toyota Mirai components provided by the United States Council for Automotive Research (USCAR).
- Define/measure degradation mechanisms:
  - Define catalyst alloy durability-related particle size agglomeration, alloy dissolution, and mass activity.
  - Define catalyst alloy effect related to carbon corrosion.
  - Define radical scavenger cation migration and diffusion.
- Novel electrode layer design and fabrication:
  - Develop MEAs—ordered arrays, nanowire catalysts, and electrospun electrodes.

Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan[1]:

A. Decrease Cost

B. Increase Power Density

C. Simultaneously Improve Durability.

Technical Targets

This project develops MEAs that meet the following targets shown in Table 1.

FY 2018 Accomplishments

- Developed new novel/experimental catalyst-layer architectures.
- Utilized characterization and experimental diagnostics to delineate catalyst-layer interactions.
- Experimentally evaluated degradation mechanisms and conditioning effects.
- Conducted analysis to set baseline and benchmark materials by analysis of Toyota Mirai components.
- Developed diagnostics and models for interpreting critical phenomena and data.
- Explored impact of ionomer and cations from inks to MEAs.

Table 1. FCTT Technical Targets: Membrane Electrode Assemblies for Transportation Applications

Characteristic	Units	2025 Targets
MEA cost	$/kW	10
Platinum group metal (PGM) total content	g/kW rated	≤0.10
Performance @ 0.8 V	mA/cm^2	300
Performance @ rated power	mA/cm^2	1,800
Durability with cycling	Hours	8,000
Loss in performance at 0.8 A/cm^2	mV	≤30
Loss in performance at 1.5 A/cm^2	mV	≤30

FCTT – Fuel Cell Tech Team

[1] https://energy.gov/eere/fuelcells/downloads/fuel-cell-technologies-office-multi-year-research-development-and-22

INTRODUCTION

Although fuel cells are being deployed in cars in limited commercialization, they still fall short of the DOE targets for this technology, which are required for widespread consumer acceptance. The FC-PAD consortium was formed to advance performance and durability of PEMFCs at a pre-competitive level to further enable their commercialization. This consortium coordinates national laboratory activities related to fuel cell performance and durability, provides technical expertise, and harmonizes activities with industrial developers. The consortium serves as a resource that amplifies the Office of Energy Efficiency and Renewable Energy's impact by leveraging the core capabilities of several labs in conducting low-technology-readiness-level research.

The major challenge addressed by this consortium is to develop the knowledge base and understanding to optimize electrode structures for more durable, high-performing PEMFC component technologies, while simultaneously reducing cost. Current research focuses on achieving high performance and durability in low-Pt-loaded PEMFCs.

APPROACH

This consortium incorporates national laboratory investigators with proven experience (developed in prior projects) related to durability, transport, and performance, and combines them into one highly coordinated effort. The consortium formalizes already existing and effective collaborations among the national laboratories that have established leadership in PEMFC performance and durability research and development. Three thrust areas are related to components: (1) Electrocatalysts and Supports, (2) Electrode Layers, and (3) Ionomers, Gas Diffusion Layers, Bipolar Plates, Interfaces; and three thrust areas are cross-cutting in nature: (4) Modeling and Validation, (5) Operando Evaluation: Benchmarking, ASTs, and Contaminants, and (6) Component Characterization and Diagnostics.

FC-PAD is an integrated five-national-laboratory consortia with a large number of contributing staff scientists, research technicians, post-docs and students. For FY 2018, FC-PAD contributors included the following.

- **Argonne National Laboratory (ANL):** Nancy Kariuki, Dennis Papadias, C. Firat Cetinbas, J-K Peng, Xiaohua Wang, A. Jeremy Kropf, Hemma Mistry, Jaehyung Park
- **Lawrence Berkeley National Laboratory (LBNL):** Lalit Pant, Meron Tesfaye, Anamika Chowdhury, Sarah Berlinger, Andrew Crothers, Peter J. Dudenas, Victoria Ehlinger, Grace Lau, Michael Tucker, Clayton Radke
- **Los Alamos National Laboratory (LANL):** Jacob Spendelow, Andrew Baker, Siddharth Komini Babu, Natalia Macauley, Sarah Stariha, David Langlois, Kavitha Chintam, Roger Lujan, Mahlon Wilson, Sarah Park, Derek Richard, Yu Seung Kim
- **National Renewable Energy Laboratory (NREL):** Guido Bender, Sadia Kabir, Jason Zack, Nihal Shah, Lawrence Anderson, Ellis Klein
- **Oak Ridge National Laboratory (ORNL):** Brian Sneed, Shawn Reeves.

Four FC-PAD projects from DE-FOA-0001412 were announced by DOE during FY 2017. Those projects are led by 3M Company, General Motors, United Technologies Research Center, and Vanderbilt University. The core national laboratory team is supporting those four projects with an equal level of effort utilizing national laboratory capabilities. National lab work for those projects is reported by those projects.

RESULTS

During FY 2018, FC-PAD conducted work in six thrust areas. Much of the work was related to fundamental understanding of the electrode structure and the relative material distribution of materials and how ionomer and ionomeric cations interact with the catalyst and catalyst support. This includes significant characterization

of the electrode structure and modeling to determine the relative impact on performance and durability. Below is a selected subset of these activities during FY 2018.

Carbon Corrosion Effect on Platinum Alloys

To examine the stability of electrode structures and further understand the combined effects of carbon corrosion on performance, we tested various types of carbon support materials, as well as platinum and platinum-alloy catalysts under accelerated stress tests (ASTs). Figure 1a shows the carbon corrosion rate during the DOE/FCTT carbon corrosion AST as measured by CO_2 evolution. The carbon corrosion rate is observed to depend only upon the weight percentage of platinum, and not on the active catalyst being Pt or PtCo. Figure 1b shows the relative carbon remaining during the cycling; while the carbon loss rate is higher for the lower-percentage-Pt catalyst, over these tests, the amount of carbon remaining is still significantly higher. Comparing Pt and PtCo on high-surface-area carbon (HSAC) versus low-surface-area carbon (LSAC), Figure 1c and 1d show that Pt and PtCo follow the same trends for HSAC. HSAC catalysts show a rapid decrease in electrochemical surface area (ECSA) and mass activity, where the Pt catalyst on LSAC has an initial lower ECSA and mass activity; however, the mass activity remains almost unchanged during the carbon corrosion AST. This agrees with the modeling analysis that demonstrates how the change in the carbon corrosion rate coordinates with local mass-transfer resistance increases.

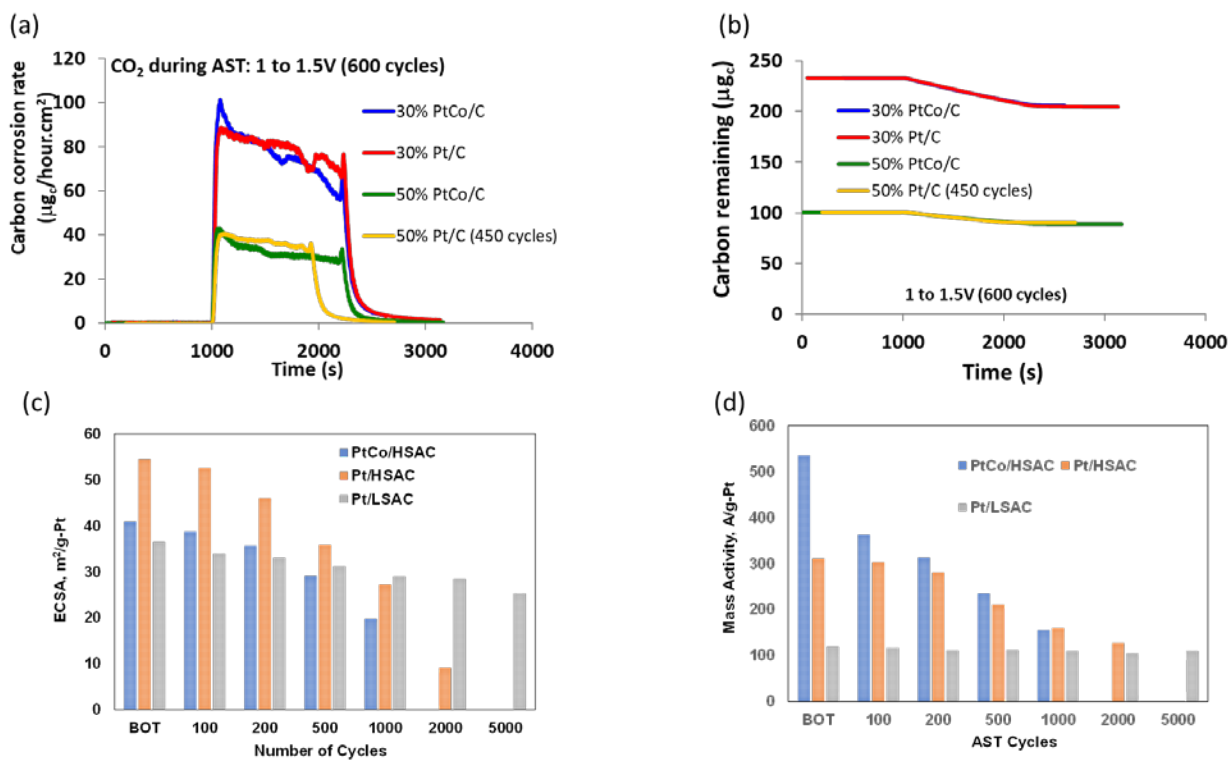

Figure 1. Carbon corrosion measurements comparing 30% and 50% Pt/C and PtCo/C during the DOE/FCTT carbon corrosion AST. (a) Carbon corrosion rate measured by direct CO_2 evolution; (b) calculated remaining carbon after the carbon corrosion and as a function of carbon corrosion cycles for catalysts: PtCo/HSAC, Pt/HSAC; (c) ECSA and (d) mass activity.

Pt and PtCo MEA Conditioning Measurements

FC-PAD has examined the relative performance of MEAs and catalysts in their initial performance compared to their eventual, beginning of life performance, which is generally termed "conditioning." The conditioning protocol was detailed in the Annual Merit Review (AMR) presentation and consists of high humidity (150% relative humidity), 40°C operation at 0.1 V, with H_2/O_2 followed by H_2/air polarization curves. Figure 2a

shows the catalyst mass activity of a 50% Pt/high-surface carbon (HSC) catalyst during a series of conditioning steps, where the maximum performance is obtained after only two or three conditioning steps. In contrast, a 30 wt% PtCo from Umicore does not reach its maximum mass activity until six to eight conditioning steps. This was measured for three different loadings of Pt: 0.05, 0.10, and 0.15 mgPt/cm^2. The loading of Pt does not make a significant difference in the conditioning time. The Pt and PtCo particle distribution size (PSD) (Figure 2c) and carbon agglomerate size (Figure 2d) also were measured both before and after conditioning. While the Pt PSD size did not significantly change, the PSD for PtCo increased significantly and the agglomerate size also increased. These measurements show different effects at work during conditioning steps, and variations between different catalysts and MEAs. Thus, care must be taken to appropriately optimize conditioning protocols separately for each material set.

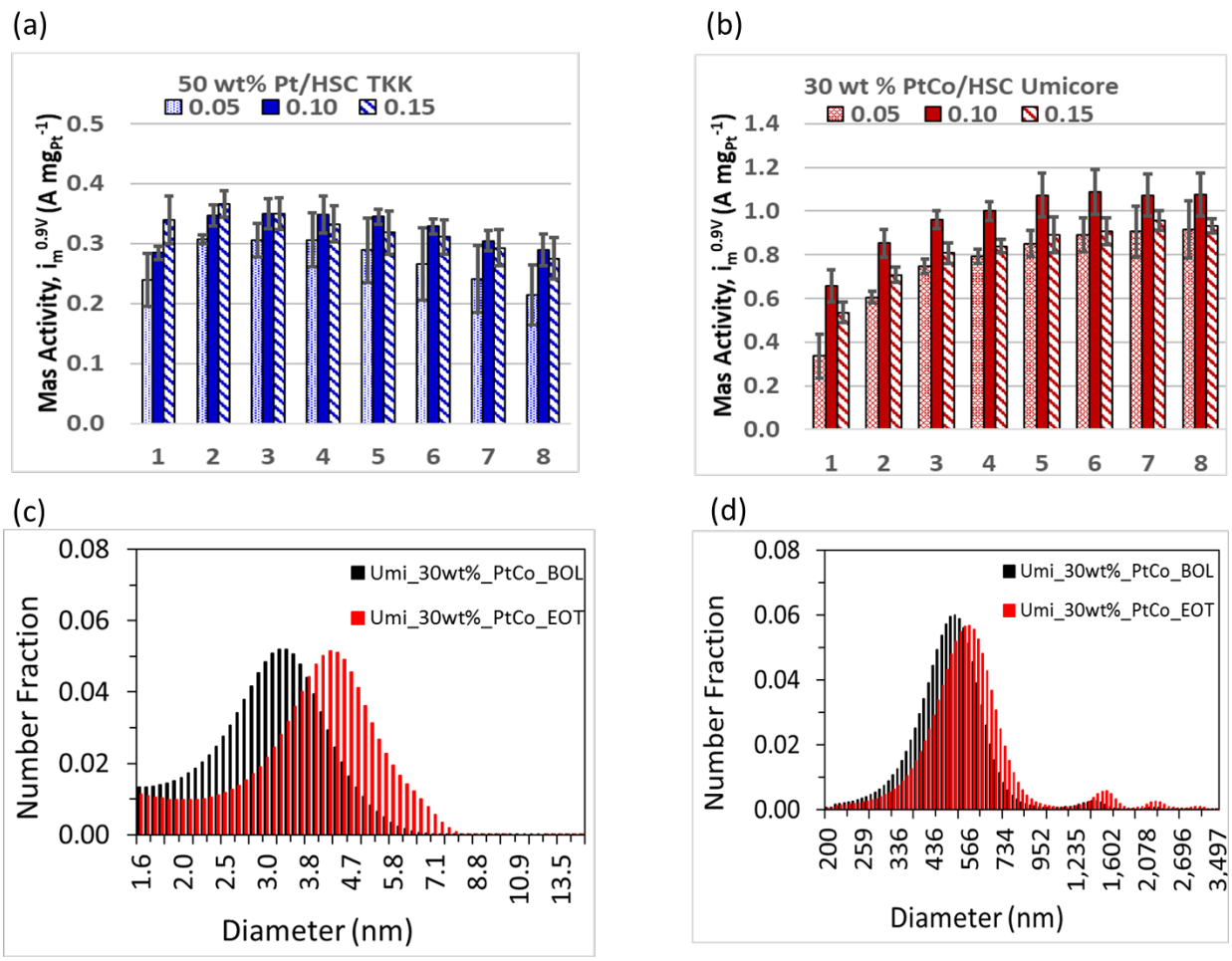

Figure 2. Effect of conditioning cycles on catalyst mass activity for (a) 50 wt% Pt/HSC from TKK and (b) 30 wt% PtCo/HSC from Umicore. Comparison during conditioning protocols at beginning of life (BOL) and end of test (EOT) of PtCo (c) catalyst particle size distribution and (d) carbon agglomerate size.

Array Electrodes

FC-PAD results have demonstrated that state-of-the-art electrode structures are hindered by severe mass-transport limitations during high-power operation at the target low-Pt loadings, resulting in a loss in performance. The ionomer within the electrode adds an additional transport resistance that becomes significant at lower Pt loadings [1]. By separating the different electrode functions into discrete electrode elements, each element can be optimized for its specific function, thereby alleviating the resultant resistances (Figure 3a). We have demonstrated this approach in array electrode through fabrication of freestanding arrays of vertically

oriented ionomer channels with different aspect ratios, and incorporation of these channels into fuel-cell electrodes where they serve as non-tortuous proton-transport highways (Figure 3b). Providing effective proton transport through these low-tortuosity percolating highways allows the catalyst domain to have a lower ionomer/catalyst (I/C) ratio, reducing transport resistance. Proof of concept is demonstrated in Figure 3c where the lower than traditional I/C = 0.5 has a higher performance than a more typical I/C = 0.9 with channels with an aspect ratio of 8. Further work is underway to increase channel aspect ratio and improve integration of channels with the surrounding catalyst layer, leading to further transport improvements and performance increases.

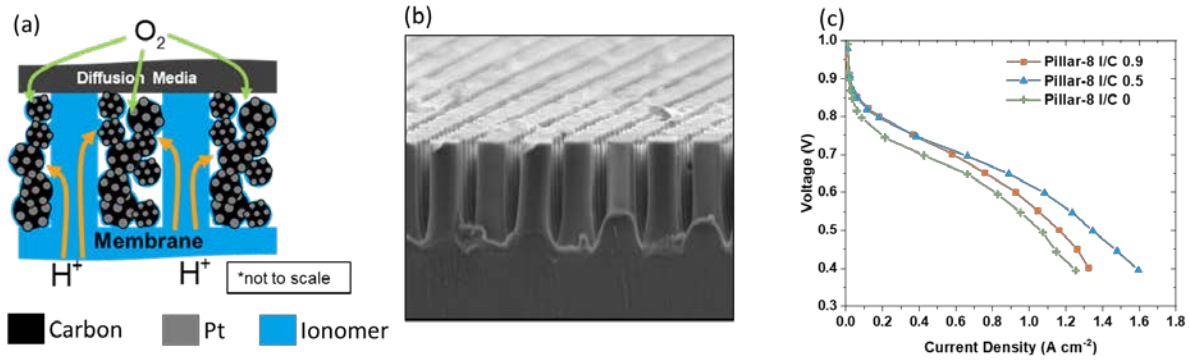

Figure 3. (a) Schematic of the array electrode design, (b) free-standing array of ionomer channels, and (c) polarization performance of array electrode structures with I/C = 0, 0.5, and 0.9

Benchmarking

With limited introduction of commercial fuel cell vehicles initiated, it is important to understand the commercialized state of the art of materials and their limitations to improve upon them. FC-PAD was provided two sets of materials by USCAR from operated Toyota Mirai fuel cell vehicles (300 h and 3,000 h operation). A summary of the MEA components is given in Table 2.

Table 2. Summary of Analysis of Toyota Mirai MEA

	Anode	Cathode
Catalyst	0.05 mgPt/cm^2 Pt/C	0.33 mgPt/cm^2 PtCo/C
GDL	Fiber substrate (150 micron) with MPL (60 micron) with 120 microgram ceria	Fiber substrate (160 micron) with MPL (40 micron) with 60 microgram seria
Membrane	10 micron reinforced Nafion	
Bipolar Plate	Carbon-coated Ti	

MPL – microporous layer

To understand the relative durability of the components, the 300-h cell materials were compared to the 3,000-h materials. Little difference was evident; nominally no membrane thinning was measured, and the cathode catalyst particle size was nearly identical at 4.86 nm and 4.96 nm for 300 h and 3,000 h respectively (by small-angle X-ray scattering). Transmission electron microscopy (TEM) analysis of 4.7 nm and 4.8 nm particle size for 300 h and 3,000 h respectively agrees closely with the X-ray measurements. TEM images are shown in Figure 4a and 4b for 300 h and 3,000 h respectively. However, when operating these cells on the DOE/FCTT recommended catalyst ASTs, measurements showed that the materials did not pass the tests. The catalyst particle size increased, and Co was measured to preferentially leach from the catalyst particles (see Figure 4c and 4d). It seems clear that the Toyota Mirai successfully implements mitigation strategies that successfully prevent the vast majority of component degradation, albeit at higher catalyst loadings than the DOE targets.

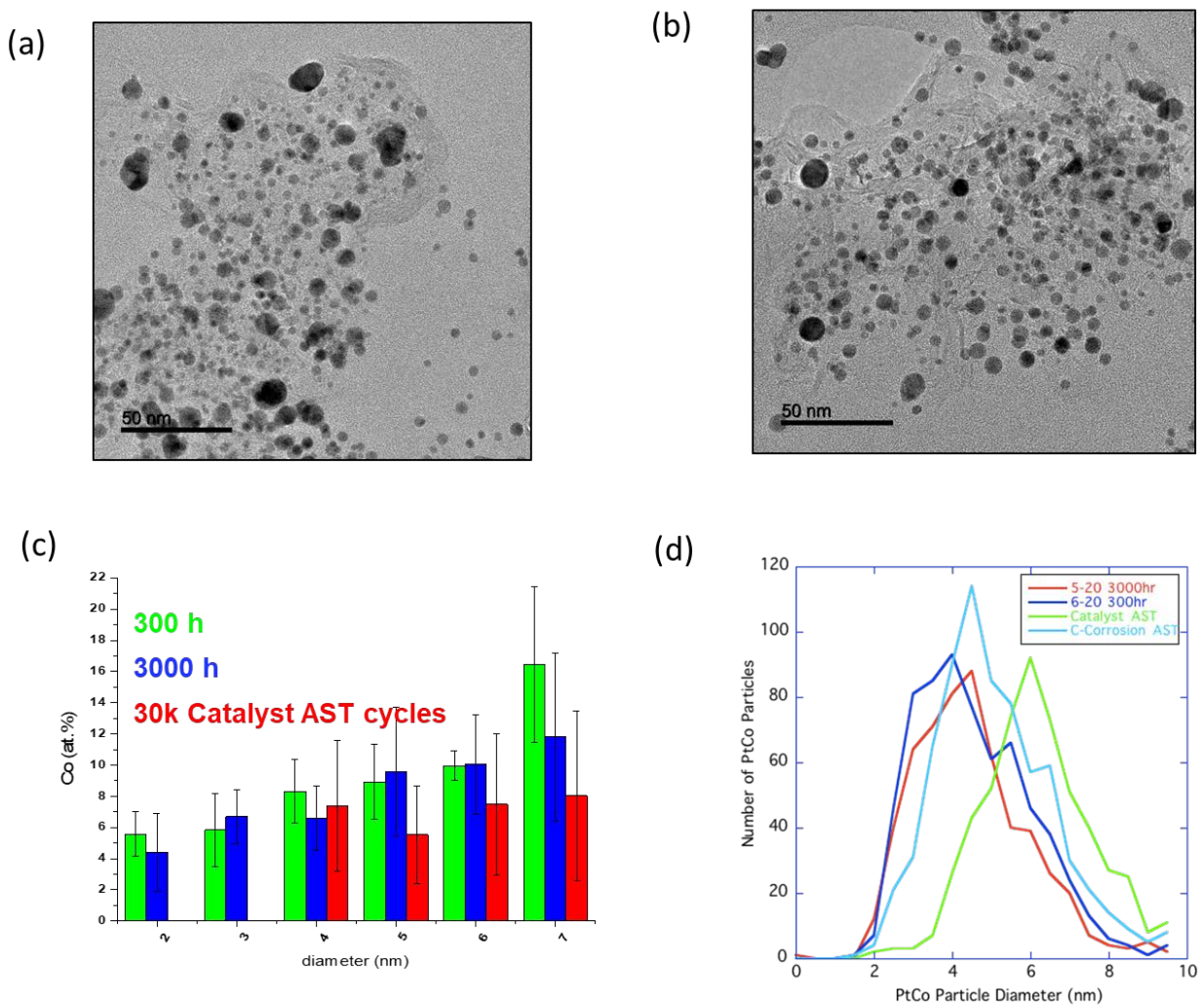

Figure 4. TEM images of the Mirai cathode catalyst after operation of (a) 300 h and (b) 3,000 h. (c) Cobalt % composition for various particle sizes for cells from vehicles operated for 300 h (green), 3,000 h (blue), and after AST (red). (d) PtCo particle size distribution for 300 h (red) and 3,000 h (dark blue), and after the DOE/FCTT recommended catalyst (green) and carbon corrosion (light blue) ASTs.

CONCLUSIONS AND UPCOMING ACTIVITIES

The FC-PAD consortium has conducted significant analysis of MEA electrode structures, analyzed the structures to understand the performance and durability losses, comprehensively defined the current commercial material baseline materials, and used this information to developed new electrode architectures that are designed to minimize existing electrode layer losses. The results and understanding from FC-PAD have been disseminated through 33 peer-reviewed publications and numerous presentations. In addition, the FC-PAD consortium national labs provided support to four DOE-funded projects related to FC-PAD as detailed in their individual project reports.

As a consortium, the future work planned is broad, but concentrates on developing the foundational science to understand MEA component performance and durability.

- Inks and thin films:
  - Examine carbon interactions with different solvents and ionomer chemistry—direct measurement of interaction forces.
  - Measure ionomer thin-film properties under applied potential.
  - Evaluate interfaces and impact of carbon type including durability protocols.
  - Examine impact of cation doping level.
  - Casting and evolution of ionomer thin-film structure.
  - Gas permeability measurements.
- Catalysts:
  - Re-examine intrinsic potential-dependent kinetics—elucidate governing binary interactions, and direct observation of dispersions.
- Catalyst-layer structure:
  - Continue exploration of different catalyst-layer structures—array, electrospun, HSC/VC layered.
  - Incorporation and characterization of novel electrocatalyst materials.
  - Microstructural reconstruction and modeling for catalyst layers including multiphase flow—impact of Pt/C agglomerates.
  - Directly measure ionomer film/carbon in operating electrodes.
  - Local resistance analysis—limiting current under variety of conditions, techniques, ionomers, gases, temperature, humidity.
- Advanced characterization:
  - Multimodal characterization of thin films.
  - New low-voltage cryo–scanning transmission electron microscopy—combinatorial electron energy loss spectroscopy and energy-dispersive X-ray spectroscopy and tomography (4-D scanning transmission electron microscopy), customized in situ cryo-holder to enable improved soft-matter imaging.
- Water and thermal management:
  - Detail model for GDL/channel interface and droplets.
  - Water visualization in various components.
  - Explore impact of carbon type in MPLs.
  - Integrate and evaluate various components to elucidate emergent phenomena.
  - Translational modeling going from ex situ property data to operando performance.

- Durability:
  - Model synergistic mechanical degradation with crossover and performance.
  - Model and measure the movement of chemical scavengers and/or other ions.
  - Examine the effect of aging on electrode microstructure.
  - Develop stabilized systems of radical scavengers.

FY 2018 PUBLICATIONS/PRESENTATIONS

Publications Relevant to FC-PAD from Consortium Members

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