

Chapter 1

Status of Fuel Cells and the Challenges Facing Fuel Cell Technology Today

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The Department of Energy (DOE) Hydrogen Program supports research and development that has substantially improved the state-of-the-art in fuel cell technology, especially with regard to the major technical hurdles to fuel cell commercialization - durability, performance, and cost of fuel cell components and systems. In particular, membrane and catalyst structure and composition have been found to be critical in obtaining improved performance and durability. For example, advancements in alloy catalysts, novel catalyst supports, and mechanically-stabilized membranes have led to single-cell membrane electrode assemblies (MEAs) with platinum metal group loadings approaching the DOE 2015 MEA target that have a lifetime of 7,300 hours under voltage cycling, showing the potential to meet the DOE 2010 automotive fuel cell stack target of 5,000 hours (equivalent to 150,000 miles). In addition, improvements in the performance of alloy catalysts and membranes have helped improve overall performance and reduce the modeled cost of an 80-kW direct hydrogen fuel cell system for transportation projected to a volume of 500,000 units per year to \$73/kW. While component research enabled such advances, innovation in characterization and analysis techniques has improved researchers' understanding of the processes that affect fuel cell performance and durability. An improved understanding of these processes will be key to

making further progress in eliminating cost, durability, and performance challenges that remain for fuel cell technology.

Introduction

Fuel cells offer benefits in transportation, stationary, and portable power applications. One of the major benefits is an increase in efficiency over conventional technology. Fuel cells are more than two times as efficient as internal combustion engines (ICEs), with the potential for greater than 80% efficiency in combined heat and power systems (1).

In addition to improving efficiency, fuel cells also can enhance energy security by reducing the nation's dependence on foreign oil. The United States (U.S.) imports 58% of its total petroleum, and transportation accounts for two-thirds of U.S. petroleum use (2). Projections indicate U.S. domestic oil production, even when considering biofuels and coal-to-liquids contributions, will continue to account for less than half the national demand. The U.S. Department of Energy (DOE) is investigating hydrogen and fuel cell technologies as one of a portfolio of options to reduce U.S. dependence on oil and to diminish this imbalance. In addition, fuel cell vehicles offer the potential for very low or zero emissions from the the vehicles. Emissions from the complete fuel cycle can also be substantially reduced compared to current vehicles. Recent estimates indicate a possible reduction in greenhouse gas emissions of more than a factor of two when the hydrogen is produced from natural gas reforming (3). Further reductions can be achieved when the hydrogen is made from renewable or nuclear energy.

Research has been focused on fuel cells as replacements for ICEs in light duty vehicles. For success in the marketplace, the fuel cell vehicles must offer value, performance, and benefits to the consumer that are comparable to the existing vehicles. DOE, with input from industry, has set targets for hydrogen and fuel cell technologies to achieve performance and cost comparable to competing alternatives. For example, the targets for automotive fuel cells include a cost target of \$30/kW by 2015 (\$45/kW by 2010), 5,000-hour durability (equivalent to 150,000 miles), and increased efficiency to 60% (4). The cost target is for production at manufacturing volumes of 500,000 systems per year.

In other potential applications for fuel cells, such as stationary power generation (distributed power), backup power, portable power, material handling, and other specialty applications, the life-cycle cost of the competing technology allows for a higher fuel cell cost. These applications are considered early markets for fuel cells. For example, for distributed power generation key targets include a fuel cell cost of \$750/kW and a durability of 40,000 hours (4). The cost target for distributed power is significantly higher (less aggressive) than the automotive target in order to compete with other technology in the stationary sector. While the durability target appears to be much more aggressive for distributed power generation applications than that for automotive applications, the automotive duty cycle includes much more dynamic behavior with many more cycles in power demand than the distributed power duty cycle. The 40,000 hours under the

distributed power duty cycle, therefore, is believed to be less demanding than the 5,000 hours under automotive conditions. Fuel cells are now at the point where they can begin to compete in some of these early markets. Deployment of fuel cells in these markets will help develop the manufacturing and supplier base, increase production volumes to help lower fuel cell costs, and broaden public awareness of fuel cell technology (5).

Fuel Cell Challenges

DOE has been funding research to address the technical hurdles to fuel cell commercialization. Two of the major hurdles are the cost and durability of the polymer electrolyte membrane (PEM) fuel cell.

Cost

To be competitive with entrenched technology, such as the ICE, fuel cells must provide similar benefits at a comparable cost. Fuel cells are currently more expensive and costs need to be reduced. Recent estimates indicate that at high-volume production (500,000 units), the cost of an 80-kW direct hydrogen fuel cell system for transportation would be \$73/kW (6). The DOE target for fuel cell system cost is \$30/kW by 2015. A breakdown of the cost estimate indicates that the fuel cell stack accounts for slightly more than half of the cost. To achieve the necessary activity, conventional catalysts are composed of finely-dispersed platinum (Pt) particles. Due to the high cost of Pt, the catalyst ink accounts for slightly less than half of the fuel cell stack cost (47%) at high production volumes (7). At low production volumes (1,000 systems/year), however, the membrane becomes the major contributor to the fuel cell stack cost (7). In addition, the current PEMs require humidification and limit the maximum fuel cell temperature. Membranes that could operate at low relative humidity (RH) and higher operating temperatures would allow system simplification by reducing or eliminating the need for humidification and reducing the thermal management system.

Durability

Fuel cells, especially for automotive propulsion, must operate over a wide range of operating and cyclic conditions. The desired operating range encompasses temperatures from below the freezing point to well above the boiling point of water, humidity from ambient to saturated, and half-cell potentials from 0 to >1.5 volts. The severity in operating conditions is greatly exacerbated by the transient and cyclic nature of the operating conditions. Both cell and stack conditions cycle, sometimes quite rapidly, between high and low voltages, temperatures, humidities, and gas compositions. The cycling results in physical and chemical changes, sometimes with catastrophic results. Furthermore, the

anode side of the cell may be exposed to both hydrogen and air simultaneously during start/stop cycles, leading to potentials of > 1.5 V.

DOE durability targets for stationary and transportation fuel cells are 40,000 hours and 5,000 hours, respectively, under realistic operating conditions including load cycling and start/stop. For transportation fuel cells, transient operation includes includes (8):

- 17,000 start/stop cycles

- 1,650 freeze cycles

- 1,200,000 load cycles.

The effects of the cycles are (9)

- Up-transient – hydrogen starvation

- Down-transient – differential pressure imbalance

- Dynamic operation (load cycling) – enhanced corrosion and membrane mechanical stress

- Low power – high voltage (corrosion of catalysts and/or supports)

- Off – oxygen ingress to anode, support corrosion

In addition to the foregoing cycles associated with normal operation, there is the potential for unplanned cycles associated with system failure caused by non-stack components. Such system shutdowns reportedly account for 85-90% of system failures (10). Fuel cells must be able to withstand off-specification operating conditions caused by unplanned system malfunctions.

In particular, degradation due to start-up and shut-down is an issue. Under start-stop conditions local potentials can approach 1.5 V, a potential at which carbon supports readily corrode. Catalysts and supports that can withstand fuel starvation and the mixed potentials that can result from start-up/shut-down procedures are needed.

Recent Advances

Cost

A major focus of DOE supported efforts is directed toward reducing fuel cell costs to achieve market competitiveness. Cost estimates have determined that the major contribution to stack cost at high production volumes is the catalyst. DOE investigates four strategies to improve catalysts and decrease cost: 1) lowering Pt and Pt group metals (PGM) content through improved catalyst utilization and durability, 2) use of Pt and other PGM alloys to decrease PGM content and increase activity, 3) development of non-precious metal catalysts that maintain performance and durability compared to Pt at a reduced cost, and 4) use of novel support structures to decrease corrosion and increase durability.

PGM catalyst performance or activity has been increased while reducing the Pt content through the efforts of several groups investigating Pt alloys and structured nanoparticles. The importance of the catalyst structure has been previously described by Ross et al. (11). Studies have indicated an enhanced activity for specific crystal surfaces over that for Pt particles or Pt deposited on

carbon (Pt/C). Combining control of surface structures with alloy composition, 3M has developed NSTF catalysts exhibiting transition metal alloy catalysts with an oxygen reduction reaction (ORR) rotating disc electrode (RDE) half-wave potential at least 50mV better than dispersed Pt/C, including a PtNiFe alloy having a half-wave potential ORR of 0.960 mV as measured at Argonne National Laboratory (ANL) (12). See Figure 1.

These catalysts have demonstrated 8x (PtCoMn) and 10x (PtNiFe) the activity of Pt/C. The PtCoMn catalyst has also demonstrated improved durability (13).

Other groups are investigating alternative structures to try to obtain improved activity and durability. UTC Power and Brookhaven National Laboratory (BNL) are developing core-shell structured Pt catalysts to reduce Pt content while increasing activity, while researchers at ANL are investigating core-shell structures with Pd. The catalysts consist of a core of base metal or base metal alloy, coated with a thin shell of Pt or PGM. The hopes are that by using a base metal core, the amount of PGM can be reduced. The base metal core also interacts electronically with the PGM shell, altering the d-band gap of the PGM shell, affecting reactivity. The PGM shell provides a less reactive covering, protecting the base. BNL and UTC Power have developed Pt-containing catalysts of this type with higher activity than Pt/C. Figure 2 compares the activity of Brookhaven's PtAuNi₅ core-shell catalyst with Pt/C on the basis of activity per milligram of Pt. The core-shell structure was determined by EDS. The PtAuNi₅ catalyst produced more than twice the current per gram of Pt for a given voltage at voltages of 0.8V or less, but the activity per cm² remains lower than Pt (14). ANL has been investigating Pd alloys and Pd core-shell systems. ANL has developed a Cu₃Pd catalyst with a higher activity on a surface area basis than that for Pt/C at 0.8V (15). Unfortunately, ANL has only been able to prepare this material with large particle sizes and achieve only ~ 75% of the mass activity of Pt/C per gram of PGM.

While these alloy and core-shell systems show promise, further improvements in activity are needed and durability of these systems still needs to be demonstrated.

Another strategy to reduce costs is to remove Pt and PGM altogether. Recent work at Los Alamos National Laboratory (LANL), University of South Carolina, and 3M has shown significant improvement in activity and durability of non-PGM catalysts. LANL and 3M have developed transition metal -N-C heterocyclic catalysts with impressive activity. Starting from various precursors containing C-N bonds, they have prepared active ORR catalysts, and demonstrated lifetimes on the order of hundreds to greater than 1000 hours (15, 16). RDE experiments indicate that catalysts derived from polyaniline and Fe₃Co (PANI-Fe₃Co/C) trail Pt/C reference catalyst (E-TEK) by no more than 80 mV at E_{1/2}. Peroxide generation in these catalysts is low, with H₂O₂ generation reduced to ~0.5%. MEA performance for the polypyrrole FeCo/C catalyst in oxygen is illustrated in Figure 3, below. The maximum power density was greater than 400 mW/cm² in oxygen.

The University of South Carolina has developed carbon-based metal-free catalysts and carbon composite catalysts for the ORR. Carbon catalysts have been known to catalyze peroxide production, however Popov et al. have been

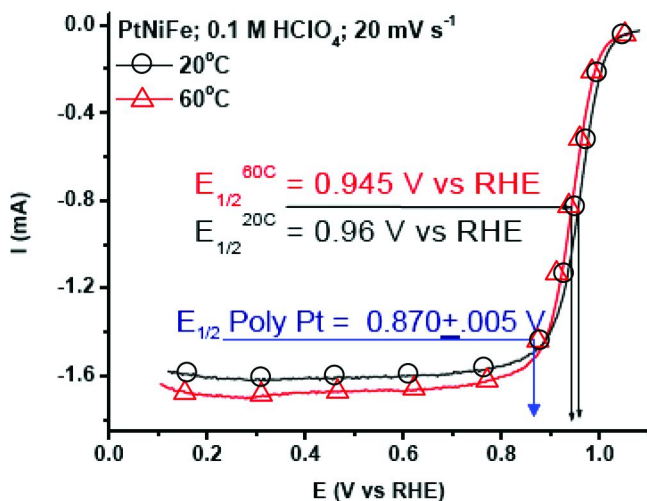


Figure 1. RDE results from ANL demonstrating >50 mV increase in $\frac{1}{2}$ wave potential for the 3M PtNiFe alloy over polycrystalline Pt.

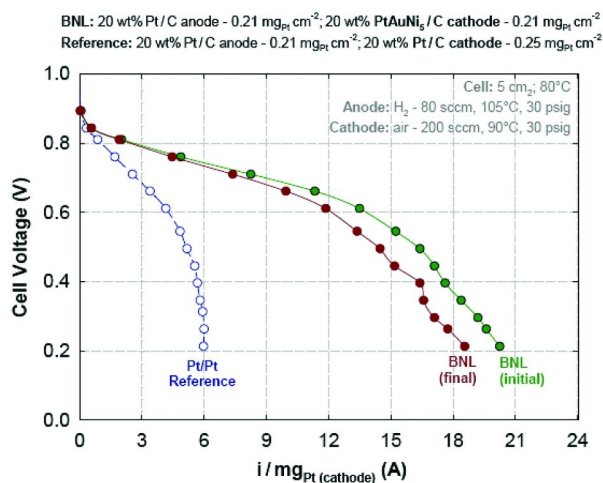


Figure 2. Comparison of PtAuNi₅ core-shell catalyst to Pt/C showing greater than twice the current per gram of Pt for the PtAuNi₅ core-shell catalyst.

able to suppress H₂O₂ formation and promote the 4 e⁻ ORR in a catalyst with no metal present (17). Activity was increased further when the carbon-free catalyst was used as a support for a carbon composite catalyst formed from pyrolysis of a CoFe-C-N non-PGM catalyst. Activity increased further upon acid leaching of this catalyst (see Figure 4). No metal atoms or particles were observable on the surface, although metal atoms covered by several graphene layers were observed (17). Significant questions remain for the non-PGM catalysts, including questions regarding the identity of the active site and the role of the transition metals and

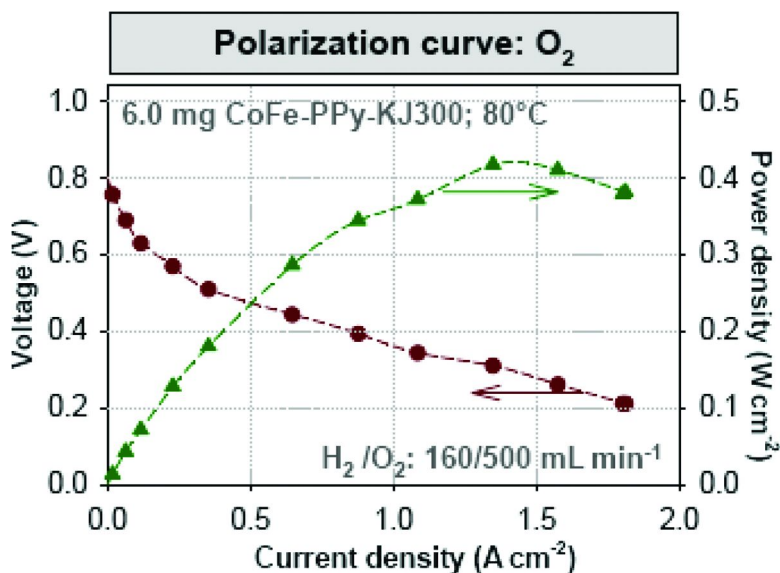


Figure 3. MEA performance of LANL FeCo/C-polypyrrole catalyst demonstrating power density of > 400 mW/cm².

nitrogen in the catalysts. The activity of non-PGM catalysts is still short of the targets.

Another area to reduce costs, especially at low manufacturing volumes, is membranes. In addition, improvements in membrane properties, particularly improved conductivity under low RH and at higher temperatures, can indirectly reduce system costs by reducing or eliminating the need for external humidifiers and reducing the size of the thermal management system. DOE has supported research to develop new membranes that can operate at high temperature (120°C) and low RH. Three main strategies and combinations of these strategies are being pursued: (1) control of polymer and membrane structure to control phase segregation and optimize the hydrophilic region where proton conduction takes place, while managing the hydrophobic region to obtain the mechanical properties needed; (2) utilizing materials such as inorganic oxides, heteropolyacids, or ionic liquids that which have potential for nonaqueous proton conduction, so the conduction mechanism does not rely on water at conditions where water may be scarce; and (3) use of hydrophilic additives to help retain water at high temperature so the water content in the membrane will be sufficient to conduct water at high temperature and low RH.

One approach for phase segregation utilizes sulfonated rigid rod liquid crystalline polymers. Bulky or angled comonomer units attached to the rigid backbone force the chains apart, creating pores lined with sulfonic acid groups. This structure creates a hydrophilic region (pores lined with sulfonic acid groups) with high concentrations of acid groups. The controlled architecture allows the polymer to hold water tightly in the regions between the hydrophobic backbones, generating high conductivity even at low RH. Case Western Reserve University

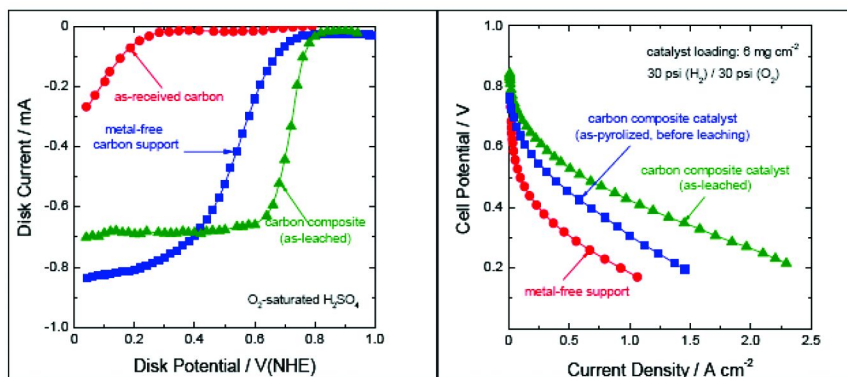


Figure 4. RDE and MEA demonstrating activity of University of South Carolina metal-free carbon and carbon composite catalyst.

achieved proton conductivity exceeding 0.1S/cm at 120°C and 50% RH using a graft copolymer of poly(p-biphenyldisulfonic acid) with di-t-butylphenol. See Figure 5 (18). However, the mechanical properties of these systems have been unsatisfactory to date.

Researchers have also developed composite membranes, which control phase segregation by providing proton conduction functionality by one polymer and the mechanical properties by another, with proton conductivity reaching ~ 0.1 S/cm at 120°C and 50% RH. Examples of these membranes include two approaches: electrospun fibers of ionomer to form a conductive mat that is filled with an inert matrix for stability, and a porous inert matrix ((either 2D grids with patterned holes or 3D mesh) filled with ionomer (19, 20). These approaches allow the use of lower equivalent weight PFSA materials with higher conductivity that would be unstable on their own. Using the dimensionally stabilized membrane, Giner achieved .093 S/cm at 120°C and 50% RH (20).

DOE has seen the most success using this first strategy (phase segregation control), but has made some progress using the second and third strategies. Colorado School of Mines (CSM) has seen some success using strategy 2, non-aqueous proton conduction. CSM's approach investigates membranes prepared by immobilizing heteropolyacids through crosslinking the heteropolyacids with organic linkers. CSM has successfully prepared new ionomers, dubbed polyPOMs, using this technique. Recent work has focused on using the lacunary heteropolyacid $\text{H}_8\text{SiW}_{11}\text{O}_{39}$. Using 80% of the heteropolyacid monomer and 20% of a comonomer, butyl acrylate, CSM has prepared a film with a conductivity of 0.1 S/cm at 120°C and 50% RH (as measured by both CSM and their partner 3M) (21), but this achievement has yet to be verified by independent testing.

The third strategy for conduction at high temperature and low RH utilizes hydrophilic additives that can provide some water for conduction at high temperature. Florida Solar Energy Center (FSEC) at the University of Central Florida has improved the conductivity of Nafion with a phosphotungstic acid (PTA) additive. FSEC has demonstrated a conductivity of 0.06S/cm at 120°C

and 50% RH (22). FSEC also demonstrated improved durability with the PTA additive (22).

Durability

Research and technical validation projects have demonstrated membrane, stack, and fuel cell system durability approximately 50% greater than previously reported. Automotive fuel cell systems are being tested in vehicles under real-world driving conditions in the Technical Validation program. Fuel cell system lifetimes have been predicted based on measured performance degradation over time, and extrapolating the results to a point of 10% voltage drop for the stack output. The maximum stack lifetime projected through this program has increased to 1900 hours (23). In addition to the maximum projected lifetime, the maximum demonstrated lifetime, average lifetime, and average projected lifetime have all increased.

To increase system durability further, DOE-sponsored efforts have focused on improving the durability of the system components. Catalyst degradation is one of the main contributors to the observed degradation in fuel cell performance. Particle growth and sintering, catalyst dissolution, and corrosion of the carbon support all contribute to performance degradation. Potential cycling conditions accelerate catalyst sintering and dissolution. Pt alloys are being investigated for improved durability, as well as increased activity.

Recent advances in membrane and catalyst technology have led to improved durability while lowering PGM loading to 0.2 mg/cm². Figure 6 presents the durability improvement using these techniques over higher-loading and unstabilized MEAs. Traditional Pt/C catalysts with traditional membranes failed in 200-600 hours of testing under load cycling conditions. 3M improved catalyst durability using its NSTF PtCoMn catalysts while still using traditional PFSA membranes without chemical stabilization, increasing the MEA durability to ~3500 hours. 3M achieved further improvements in MEA durability by combining their PFSA ionomer with mechanical stabilization to decrease swelling and shrinking during cycling. With this approach, 3M increased the durability to beyond 7300 hours (11). This feat represents significant progress in MEA durability; however these improvements must still be demonstrated in a stack and under real-world driving conditions which include start-up/shut-down cycles.

In addition to demonstrated improvements in performance and durability, there have been significant advances in characterization techniques and the fundamental understanding of degradation mechanisms. At Oak Ridge National Laboratory (ORNL), researchers using Transmission Electron Microscopy (TEM) have quantified Pt and Pt alloy particle growth in operating fuel cells, a key cause of fuel cell performance degradation. Figure 7 shows the changes in particle size for a PtCo alloy under various cycling conditions (24). Using Z-contrast Scanning TEM they have observed Pt particle coalescence during heating of a Pt/C system. From their observations they concluded that Pt particle growth did not occur by dissolution/precipitation in this system, but by Pt particles moving across the C

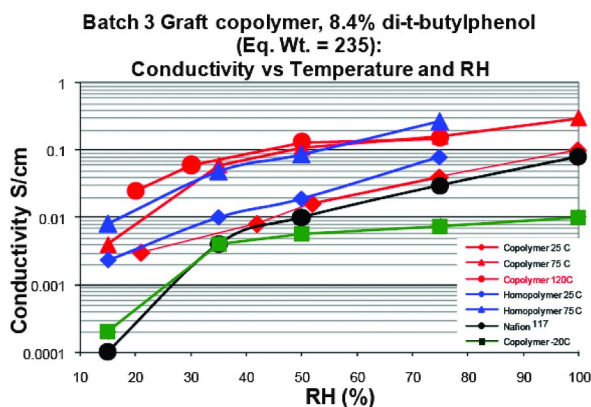


Figure 5. Conductivity of rigid rod liquid crystalline polymers at various relative humidities demonstrating high conductivity at low relative humidity.

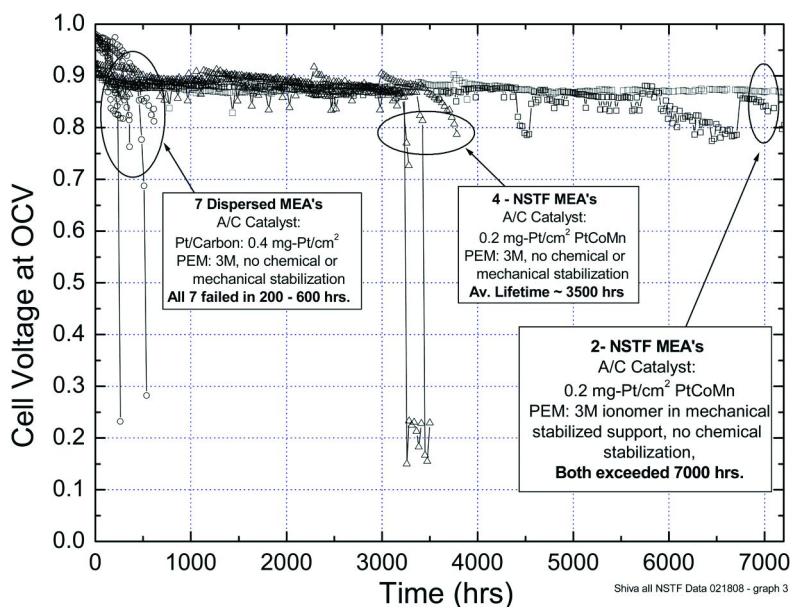


Figure 6. Accelerated durability test results demonstrating >7000 h durability under cycling conditions for 3M NSTF MEA.

surface and coalescing with nearby particles to form larger single crystal particles (25). This technique offers potential for new insight into catalyst degradation.

Using neutron imaging, researchers at Los Alamos National Laboratory (LANL) have elucidated potential pathways for fuel cell performance improvements through in situ studies of the effects of gas diffusion layer (GDL) design parameters on the water transport behavior of fuel cells (26). Neutron imaging and computational fluid dynamics (CFD) results show accumulation

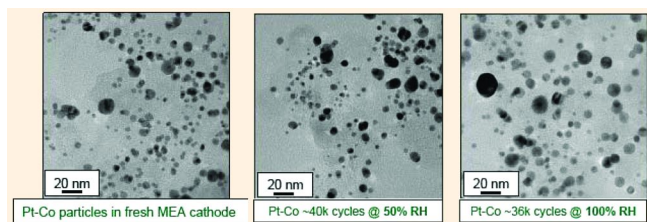


Figure 7. The effect of Relative Humidity (RH) on Pt particle size during potential cycling.

of water at the lands. Water content calculated by the CFD calculations closely matches the measured water via neutron imaging. In addition, this work has also revealed GDL degradation mechanisms.

Although the research at LANL has led to a greater understanding of material effects on water transport, more research is needed in this area to meet commercialization requirements. The stresses of freeze/thaw conditions and start-up and shut-down cycles continue to exacerbate degradation of fuel cell components (27). In addition, insufficient water transport can lead to performance degradation for MEAs. Therefore, the fuel cell research community requires a greater understanding of mass transport fundamentals, especially water transport, and new or improved MEA materials based on this fundamental understanding.

Conclusions

While recent advances have been impressive, cost, durability, and performance remain as key challenges to fuel cell technology. Catalysts remain the major cost factor at high production volumes.

At current Pt costs, the catalytic activity must be increased and Pt loading must be decreased to meet cost targets for automotive fuel cells. Recent work has been successful at reducing Pt loading using ternary alloys and by utilizing structured particles with an onion layer approach. Despite these advances, PGM loadings likely will have to be reduced further -- significantly below the current DOE targets -- if fuel cell cost targets are to be met. Consequently, future work should target ultra-low PGM or non-PGM catalysts. Recent work in the area of non-PGM catalysts at LANL, University of South Carolina and 3M has shown significant improvement in activity and durability. Further improvements are needed to make these systems viable in an automotive fuel cell.

Improvements in membrane performance, and in particular membrane performance at high temperature without external humidification, are key to reducing overall fuel cell system costs. Recent advances have improved conductivity at 120°C and reduced the humidification level needed, while still maintaining conductivity at low temperature. Several approaches exceed conductivities of 0.1 S/cm at 120°C and relative humidities below 70% RH, showing potential for achieving DOE's ultimate goals. However, durability remains to be proven.

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