V.E.5 Durability of Low Platinum Fuel Cells Operating at High Power Density

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- Los Alamos National Laboratory, Los Alamos, NM
- Argonne National Laboratory, Argonne, IL

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Fiscal Year (FY) 2011 Objectives

- The objective of this project is to study and identify strategies to assure durability of fuel cells designed to meet DOE 2015 cost targets.
- Develop a practical understanding of the degradation mechanisms impacting durability of fuel cells with low platinum loading (≤0.2 mg/cm^2) operating at high power density (≥1.0 W/cm^2).
- Develop approaches for improving the durability of low-loaded, high-power stack designs.

Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section (3.4.4) of the Fuel Cell Technologies Program Multi-Year Research, Development and Demonstration Plan:

(A) Durability
(B) Cost

Technical Targets

TABLE 1. Progress towards Meeting Technical Targets for Transportation Fuel Cell Stacks Operating on Direct Hydrogen for the Transportation Applications

Characteristics	Units	2010/2015 Stack Targets	Nuvera 2010 Status
Cost	$/kWe	25/15	~22 estimated[1]
Durability with Cycling	Hours	5,000	9,000 hrs[2] 2,000 hrs in automotive cycle conditions[3]
Performance at Rated Power	mW/cm^2	1,000	1,100[4] and 1,200[5]

[1] Cost assessment of Nuvera's architecture by Directed Technologies Inc. based on their DOE-sponsored DFMA® (Design for Manufacturing and Assembly) model [1] and comparing the results to an updated "Industry" estimate. 0.55 V/cell @2.0 A/cm^2 was obtained at 0.2 mg/cm^2 platinum loading on Orion stack by Nuvera.
[2] Demonstrated under power profile specific-to-fork truck applications in material handling market at total platinum loading of 0.5 mg/cm^2.
[3] Demonstrated in 360-cm^2 stack by Nuvera in simulated combined city and highway driving cycle at platinum loading of 0.50 mg/cm^2.
[4] Demonstrated in 250-cm^2 Orion stack by Nuvera at platinum loading of 0.2 mg/cm^2.
[5] Demonstrated in 50-cm^2 single cell with open flowfield (SCOF) by Nuvera at platinum loading of 0.2 mg/cm^2.

FY 2011 Accomplishments

- Two program milestones were completed on schedule. The first milestone comprised the development of the durability model framework by Argonne National Laboratory (ANL). The second milestone was achieved with Nuvera delivery of SCOF hardware to Los Alamos National Laboratory (LANL) for characterization and testing under accelerated stress test (AST) conditions and new stress testing representing combined power cycle (NST) protocols.
- The material sets selected for the project in standard 0.45 mg/cm^2 (A) and custom 0.2 mg/cm^2 (B) platinum loadings were delivered by W.L.Gore for various 50 cm^2 single cells including quad-serpentine, SCOF and General Motors/Rensselaer Institute of Technology architectures, as well as for 250- and 360-cm^2 cells for short stack testing. The material development scope of the project is completed.
- SCOF hardware was validated at the range of current densities up to 3 A/cm^2 and demonstrated ohmic, diffusion, and pressure drop benefits over the land-channel cells at Gore and LANL. Catalyst-coated membranes A and B were characterized at the beginning of life (BOL) in Nuvera 50-, 250- and 360-cm^2 cells and LANL 50-cm^2 hardware throughout the range of operating conditions.

- The AST campaign in LANL quad-serpentine cell was completed on all selected membrane electrode assemblies (MEAs) and test protocols. The data analysis concluded that the observed cell voltage decays resulted from the increased activation and mass transport overpotentials in the wide range of operating current densities.
- ANL evaluated platinum dissolution in aqueous media using polycrystalline and dispersed platinum on carbon electrodes, completing the ground work for further development of the platinum stability section of the durability model.
- Durability testing in short stacks continued at Nuvera under NST protocols.

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Introduction

Understanding and improving the durability of cost-competitive fuel cell stacks is imperative to successful deployment of the technology. Stacks will need to operate well beyond today's state-of-the-art rated power density with very low platinum loading in order to achieve the cost targets set forth by DOE ($15/kW) and ultimately be competitive with incumbent technologies. Little to no study of durability factors has been carried out in this area of design and operation. The industry today is focusing mostly on reduced platinum loading as it heads for the DOE target point of 0.2 mg/cm^2 platinum and 1.0 W/cm^2 power density. As demonstrated through DOE-sponsored cost modeling, this point falls short of the corresponding $15/kW stack cost target for 2015.

Approach

Nuvera proposes an accelerated cost-reduction path focused on substantially increasing power density to address non-platinum group metals material costs as well as platinum. Understanding the largely unstudied factors affecting stack durability under these high power conditions is the focus of the present project. Of specific interest is the impact of combining low platinum loading with high power density operation, as this offers the best chance of achieving long-term cost targets. The team effort is divided into two activities: modeling and experimentation.

Results

During the past year, two project milestones were completed on schedule. Development of the durability model framework by ANL concluded the first milestone. A durability model is being developed specific to the platinum stability of the selected commercially available material set (cathode 580 and 15-micron membrane M815 by W.L. Gore), independent of the cell architecture. Our modeling approach starts from platinum aqueous kinetics, defining rates for platinum dissolution and re-deposition as well as oxide coverage and continues at the electrode level for platinum ion transport. The model will utilize inputs from catalyst cycle ASTs, BOL properties of the studied material set, and prescribed use cycles to output cathode electrochemical surface area (ECSA), particle size distribution, overpotentials and cell voltage as a function of cycle time and the current density. The second milestone was achieved by delivery of SCOF hardware to LANL for characterization in ASTs and NSTs.

Development efforts were focused on further SCOF validation for performance at elevated current densities and for stability, comparing to the same diagnostics of full format area cells in Nuvera short stacks. Polarization curves of MEAs A and B in SCOF in Figure 1 showed repeatable results in multiple cell assemblies, proving control and uniformity of compression over the active area. Low resistivity and pressure drop of the open flow fields resulted into measurable performance benefits over the quad serpentine cell. High power densities above the DOE target of 1 W/cm^2 were demonstrated in both SCOF and open flowfield short stack tests at 2 A/cm^2, proving the diffusion benefits of open flowfield over the land-channel architecture, tested at Gore and LANL. While the performance of MEA B in land-channel cell was limited below 2 A/cm^2 at low pressure conditions, 1.2 W/cm^2 power density was achieved on the same MEA at 2 A/cm^2 in SCOF.

Over the past year the AST campaign in the quad serpentine cell was completed and currently moved to SCOF. ASTs focused on the electrocatalyst stability under the B1 potential cycle protocol and membrane mechanical stability under the B4 humidity cycle protocol defined by the DOE. In addition, the low platinum cell was subjected to the modified potential cycle test with the lower potential

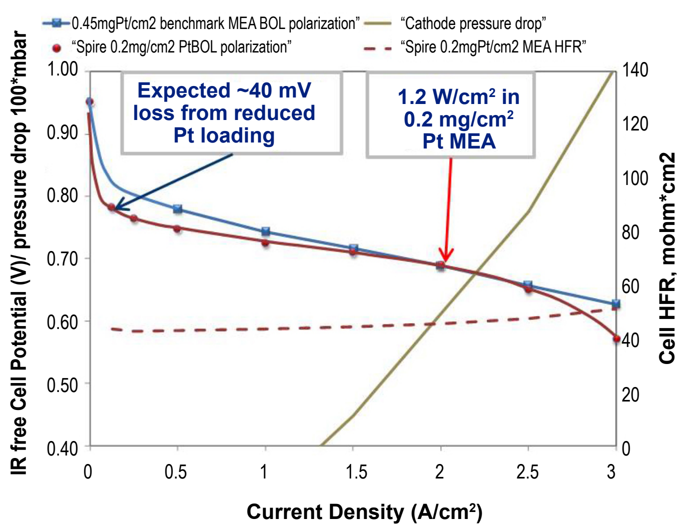

FIGURE 1. Ohmic Resistance-Free Polarization of MEAs A and B in SCOF (Dry cathode, 1.8 bara inlet pressure)

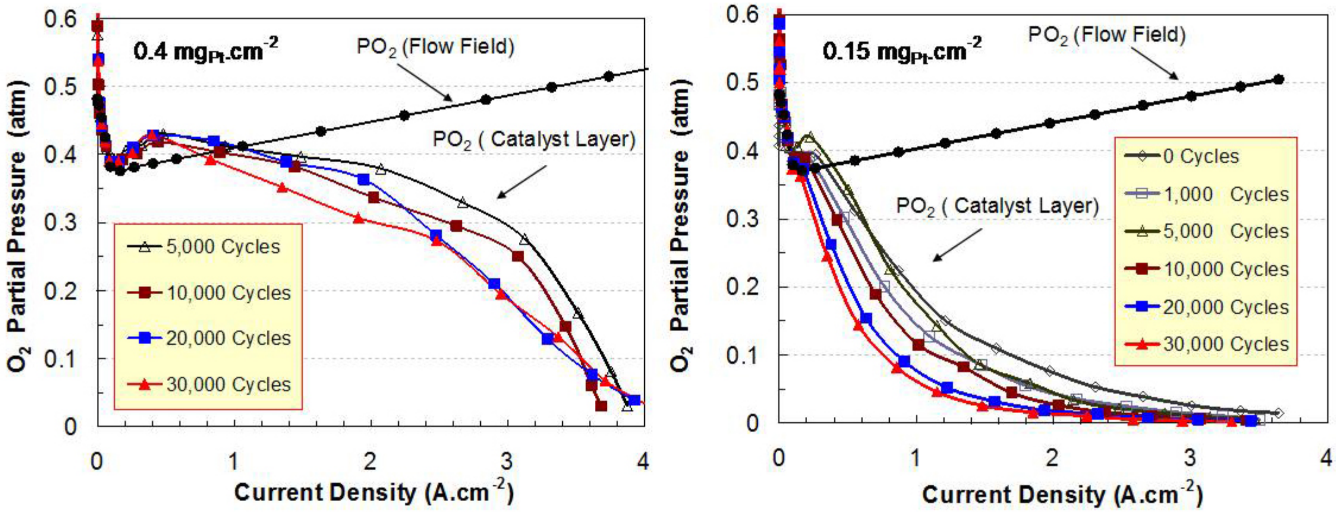

FIGURE 2. Effect of Ageing in B1 AST on the Cathode Oxygen Partial Pressure in MEAs with different Platinum Loading

range of the cycle than in the B1 test. The potentials in the modified cycle protocol correlate with cell voltages achieved in Nuvera large-area cells at 2 A/cm^2, resulting in significant improvement in cathode ECSA stability and less platinum dissolution into the membrane, confirmed by the post-mortem scanning electron microscopy and energy disperse spectroscopy analyses.

AST data, including polarization, ECSA, and impedance diagnostics from all potential cycles obtained in both normal and low-platinum cell tests were analyzed at low and high pressure conditions to determine the effect of ageing on the kinetics of the oxygen reduction reaction (ORR) and on oxygen mass transport. The model based on the electrochemical impedance spectroscopy data at different current densities was set up to distinguish oxygen mass transport coefficients between the cathode catalyst and gas diffusion layers. Observed cell voltage decays were attributed to the increase of activation and mass transport overpotentials in the ASTs and the respective decay of oxygen partial pressure in cathodes in Figure 2.

Durability testing in short stacks continued under the NST3A-2 protocol, representing a combined city and highway driving cycle designed at 2 A/cm^2 rated current density and average cell voltage response shown in Figure 3. In this NST the cathode pressure and flow conditions vary with the current density, following the operating map of the air compressor in the automotive system. Data obtained during 1,000-1,600-hour testing of three stacks were processed using ohmic resistance-corrected, polarization data at low current densities to determine the Tafel slope for the ORR kinetics. Our analysis of ECSA loss from voltage-current (V-I) curves showed consistency with direct measurements by cyclic voltammetry and correlated to the increase of average platinum particle diameter by X-ray diffraction analysis as shown in Figure 4. V-I analysis allowed calculating oxygen partial pressures in the cathodes

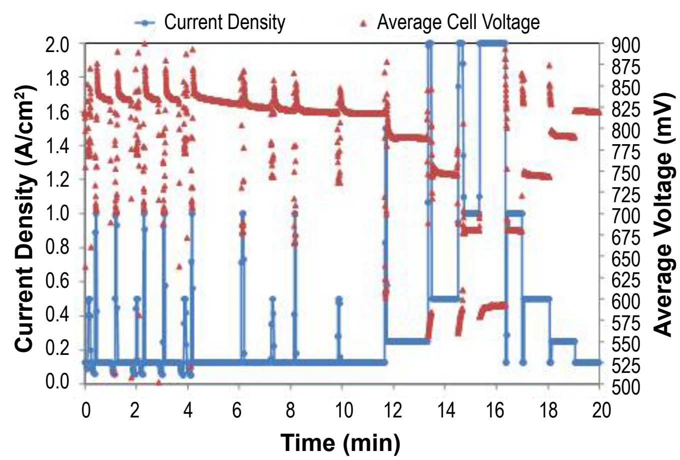

FIGURE 3. Performance of Nuvera Short Stack with MEA A under Simulated Combined City and Highway Drive Load Cycle

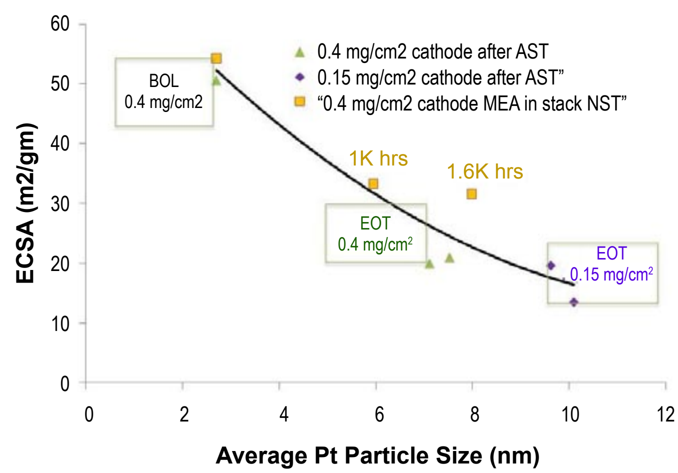

FIGURE 4. Correlation of Cathode ECSA to Average Size of Platinum Particles in ASTs and Stack NSTs

with different loadings at all operating current densities and showed decreasing values from BOL to the end of test.

Conclusions and Future Directions

- Operation at high power densities enabled by the open flowfield architecture and proven at the low platinum loading provided the ground work for accelerated cost-reduction path to the cost targets set by the DOE.
- Two milestones concluded in the reported year served as the basis for experimental and modeling activity started and to be continued in the next year.
- The AST campaign will be moved from serpentine cell to SCOF towards achieving the third project milestone.
- Durability modeling will continue, focused on the platinum stability, and will be synchronized with other durability projects covering different ageing mechanisms with the same partners, extending the current project duration by one year at no cost to the DOE.
- The NST campaign in the large-format cells in short stacks and benchmarking the results between SCOF and large area cells will continue, targeting the Go/No-Go decision.

FY 2011 Publications/Presentations

1. B. Lunt. Advanced single cell test fixture. 6th Annual International Conference on Fuel Cell Durability and Performance 2010, December 5–6, Boston, MA.
2. O. Polevaya, SPIRE Project Review with Freedom Car Technical Team, January 26, 2011, Southfield, MI.
3. S. Arisetty et. al, Effect of platinum loading on catalyst stability under cycling potentials, Abstract accepted to ECS 2011, Boston, MA.
4. F. Gambini et al., Durability of fuel cell under high power density operation, Abstract accepted to ECS 2011, Boston, MA.
5. O. Polevaya, Spire, 2011 DOE Annual Merit Review, Washington, DC, May 11, 2011.

References

1. B. James et al, Mass Production Cost Estimation for Direct H_2 PEM Fuel Cell Systems for Automotive Applications, March 26, 2009.