

II.B.3 Low-Noble-Metal-Content Catalysts/Electrodes for Hydrogen Production by Water Electrolysis

Dr. Katherine Ayers
 Proton Energy Systems d/b/a Proton OnSite
 10 Technology Drive
 Wallingford, CT 06492
 Phone: (203) 678-2190
 Email: kayers@protononsite.com

DOE Manager

David Peterson
 Phone: (720) 356-1747
 Email: David.Peterson@ee.doe.gov

Contract Number: DE-SC0008251

Subcontractor

Brookhaven National Laboratory (BNL), Upton, NY

Project Start Date: June 28, 2012 (Phase 1)
 Project End Date: August 13, 2015 (with Phase 2 continuation)

Characteristics	Units	2011 Status	2015 Target	2020 Target
Hydrogen Levelized Cost	\$/kg	4.2	3.9	2.3
Electrolyzer System Capital Cost	\$/kg	0.70	0.50	0.50
	\$/kW	430	300	300
Stack Energy Efficiency	% (LHV)	74	76	77
	kWh/kg	45	44	43

**Note: Assumptions include a plant design capacity of 1,500 kg/day of hydrogen and that production has realized economies of scale.
 LHV - lower heating value

Accomplishments

- Successfully scaled synthesis process by a factor of 10
- Showed >500 hours durability with ultra-low loaded Proton-made cathode
- Resolved early mass transport issues with deposited anode catalyst
- Demonstrated anode microporous layer does not impact cell resistance
- Demonstrated feasibility of >\$100,000 savings in capital cost for megawatt-scale electrolysis



Project Objectives

- Translate catalyst synthesis to a manufacturable process at Proton
- Develop a robust technique for manufacturable electrodes
- Demonstrate feasibility for 80% cost reduction in the anode catalyst
- Downselect promising anode electrode configurations to achieve >100 hours durability
- Achieve 500 hours of operation in production quality hardware using cost-reduced cathodes

Technical Barriers

This project addresses the following technical barrier from the Hydrogen Production section of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration (MYRDD) Plan:

(G) Capital Cost

Technical Targets

The table below shows the technical targets for distributed forecourt water electrolysis hydrogen production from the 2012 MYRDD Plan.

INTRODUCTION

The economical use of hydrogen as a transportation and stationary power fuel remains a long-term Department of Energy (DOE) objective. Energy storage applications in Europe such as wind capture and improved biogas conversion efficiency are also driving significant interest in hydrogen production from renewable sources. New and efficient catalytic processes for hydrogen generation are therefore needed to achieve production targets for hydrogen cost. In the Phase 1 project, Proton Energy Systems (d/b/a Proton OnSite), in collaboration with Brookhaven National Laboratory (BNL), demonstrated feasibility for development of low-noble-metal-content catalysts/electrodes for proton exchange membrane (PEM) electrolyzers, through design and synthesis of core-shell nanocatalysts. In Phase 2, continued development of the anode formulation is being performed for reproducible and stable electrode fabrication, while technology transfer and scale-up from BNL to Proton is occurring for the cathode electrode fabrication. The Phase 2 project is strategically important because reduction of noble metal content is a significant opportunity for cost reduction to

address large-scale opportunities for hydrogen-based energy storage and hydrogen fueling.

APPROACH

The Phase 2 project continues maturation of the catalyst structures and electrode processing initiated in Phase 1, to develop a manufacturable electrode at relevant scale and ultra-low catalyst loadings. The overall technical approach includes development of the manufacturing process for the cathode electrode as well as cell stack validation for the alternative electrode configuration. In addition, for the anode, work focuses on continued optimization of catalyst application and gas diffusion layer (GDL) structure for reproducible and durable performance equal to or exceeding the current baseline. The impact of these advancements will be quantified using the H2A model.

General steps for the cathode include technology transfer of the catalyst synthesis techniques and electrode formulations to Proton, a study to determine the best method of manufacturing the electrodes proven at the bench scale, process scale-up, cell stack design adjustments to accommodate the new configuration, and validation. For the anode, based on the promising performance demonstrated in Phase 1, catalyst support structures for stability at >1.6 V/cell are investigated, and the catalyst composition will be refined for high activity. A study of anode GDLs with varying porosity vs. alternate methods of applying the catalyst to the GDL will also be performed, to synthesize an electrode with good catalyst distribution at the GDL surface while maintaining adequate water transport at the membrane surface.

RESULTS

After core-shell nanoparticles were synthesized at Proton based on the BNL process, Proton successfully scaled the process by an order of magnitude, resulting in relevant batch sizes for membrane electrode fabrication across Proton's platforms. The process involved additional safety reviews for dealing with the larger solvent volumes, and the batch had to be divided for the reducing process based on Proton's infrastructure limitations. While the synthesis was successful, difficulties managing the solvent quantities and reduction process point out the need for additional facilities considerations or partnership with a catalyst manufacturer with appropriate equipment. The catalyst was deposited on a gas diffusion layer at 1/10th of Proton's commercial baseline loadings and tested in Proton's cell hardware. Performance was equivalent to baseline, as shown in Figure 1. The electrode fabrication process was also scaled up to large active area (700 cm^2) and tested vs. the baseline process, with good durability and performance over 1,000 hours (Figure 2).

For the anode, work at BNL focused on determining whether core-shell catalysts could provide the same benefit as the work on the cathode. Ruthenium oxide and ruthenium-iridium core-shell particles were synthesized, characterized, and electrochemically tested. As expected, the ruthenium-containing compounds provided higher initial performance. Also as expected, the pure ruthenium oxide was not stable over several days of testing. However, unfortunately, the iridium cores did not result in improved stability of the ruthenium. Therefore, iridium-only catalysts were downselected for the remainder of the program, focusing on minimizing loading. A microporous layer (MPL) approach was taken, similar to the cathode, to provide a more continuous layer for catalyst deposition on the anode

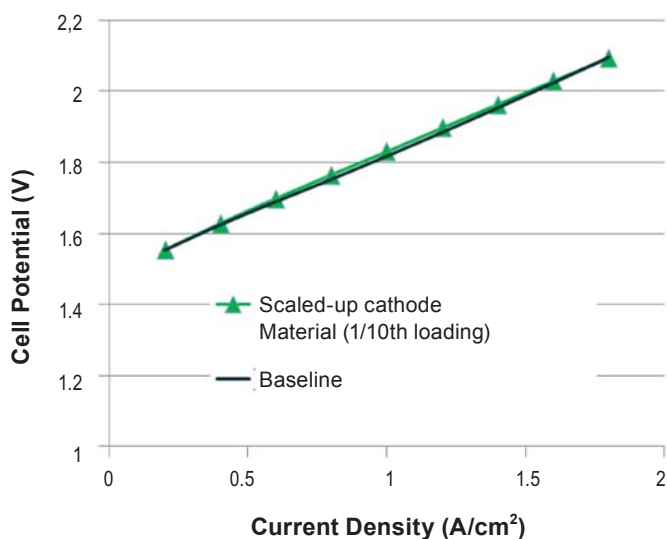


FIGURE 1. Performance data confirming scaled-up catalyst batch comparable to baseline, at 1/10th loading

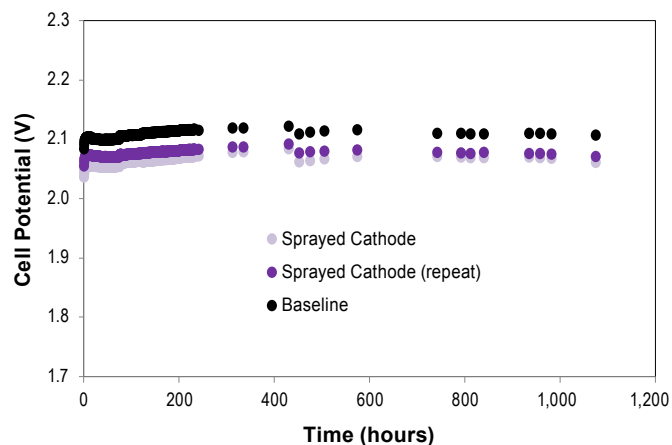


FIGURE 2. Durability performance and consistency for parts fabricated from large active area spray process

gas diffusion layer. Initially, mass transport limitations were observed, and optimization of the deposition layer had to be performed. Testing of just the GDL with an MPL against a traditional membrane electrode assembly (MEA) did not show mass transport issues, ruling out the MPL as the root cause of the issue. With adjustments in ink formulation and deposition parameters, improved performance was observed (Figure 3).

The advancements made throughout the project were combined together for a final demonstration. Anodes and cathodes with low catalyst loading were fabricated for multiple cells for Proton's 100-cm² active area configuration, and two cells were assembled with a baseline cell. The stack was tested at 400 psi and 1.8 A/cm². Performance is shown in Figure 4. While there was some variation in performance and a break-in period at the beginning, the performance stabilized after the first few hundred hours, and all three cells demonstrated stability with flat performance after the stabilization period. The variation in performance indicates that there is still optimization needed to achieve consistent electrodes, but the fact that one of the two low-loaded electrodes outperformed the baseline indicates promise for this approach with additional manufacturing development.

Based on the developments achieved over the course of the project, a cost analysis was performed for the alternate electrode configuration vs. Proton's current commercial fabrication processes. The analysis demonstrated that for megawatt-scale electrolysis, the catalyst and associated labor for the baseline deposition process represents over 25% of the stack cost. The advanced process and catalyst materials can provide over \$100/kW savings, or roughly \$0.12/kg H₂. This level of cost reduction provides a significant competitive advantage in the market, which is essential for the United States to compete in the emerging markets for energy storage.

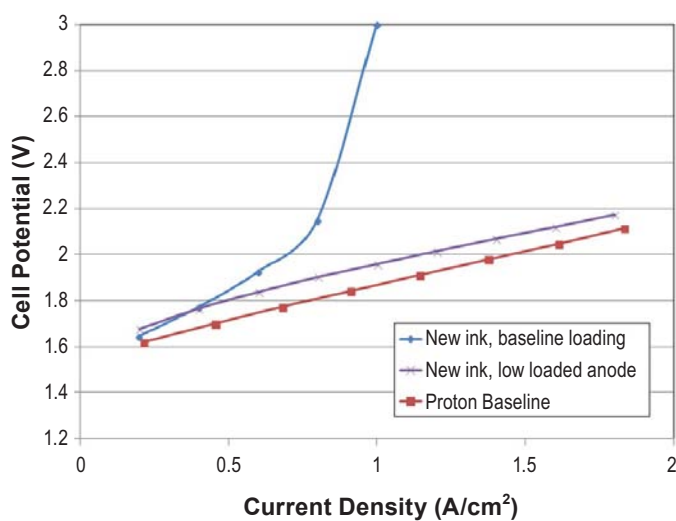


FIGURE 3. Polarization curves demonstrating resolution of anode mass transport issues

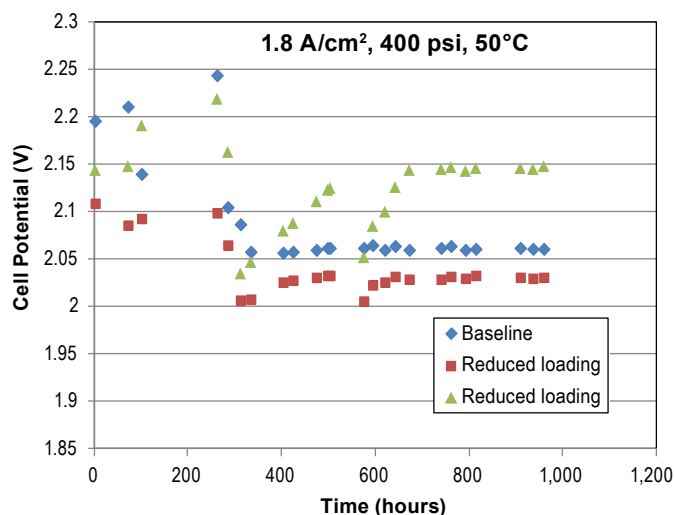


FIGURE 4. Anode and cathode performance with 90% cathode catalyst reduction and 75% anode catalyst reduction vs. baseline

CONCLUSIONS AND FUTURE DIRECTIONS

- Scale-up of core-shell synthesis and deposition process demonstrated for scales relevant to megawatt electrolysis for cathode
 - 10x catalyst batch size demonstrated
 - 90% reduction in catalyst loading
 - 7x increase in active area
- Downselected anode approach and demonstrated over 50% reduction in anode catalyst loading
 - Resolved mass transport issues at anode
 - MPL approach did not impact MEA performance
- Final demonstration completed for anode and cathode reductions combined
- Cost analysis shows \$0.12/kg savings
 - Represents several million dollars in cost savings over first few years of megawatt product sales
- Continued investment in manufacturing and catalyst needed for commercialization of demonstrated advancements

FY 2015 PUBLICATIONS/PRESENTATIONS

1. 2015 DOE Annual Merit Review and Peer Evaluation Meeting presentation: pd098_ayers_2015_o.
2. Spring Electrochemical Society meeting, "Barriers to Commercial Deployment of Advanced PEM Electrolyzers."