II.D.5 Low-Cost Large-Scale PEM Electrolysis for Renewable Energy Storage

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Contract Number: DE-SC0001338

Subcontractors:
• 3M, Minneapolis, MN
• University of Wyoming, Laramie, WY

Project Start Date: June 19, 2010 (Phase 1)
Project End Date: August 18, 2013 (with Phase 2 continuation)

Fiscal Year (FY) 2012 Project Objectives

- Demonstrate optimal membrane electrode assembly (MEA) efficiency through:
  - Refinement of catalyst compositions based on identified design spaces from Phase 1.
  - Rapid screening of additional metal candidates through combinatorial methods to determine optimal anode catalyst composition.
  - Fabrication of identified materials as both traditional and nano-structured thin-film electrode (NSTF)-type electrodes.
  - Integration of new catalyst materials with advanced thinner membranes.

- Reduce catalyst loading through:
  - Formulation optimization of metal oxide based inks.
  - Improved electrode application processes for uniform distribution.
  - Application of NSTF structures.
  - Demonstration in Proton's large active area format.

- Demonstrate 1,000 hours system operation at >69% efficiency.

- Develop a concept design to scale the system to 50,000 kg/day H₂ production including:
  - Definition of the requirements for operation and maintenance.

- Updated H2A analysis based on the actual 435 psi system cost and operation projected to 50,000 kg/day.

- Analysis of greenhouse gas and petroleum reductions that will occur with the successful implementation of the proposed technology.

Technical Barriers

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

(G) Capital Cost
(H) System Efficiency
(J) Renewable Electricity Generation Integration

Technical Targets

TABLE 1. Proton Energy Systems Progress Towards Meeting Technical Targets for Distributed Water Electrolysis Hydrogen Production

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Units</th>
<th>2012 Target</th>
<th>2017 Target</th>
<th>Proton Status</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrogen Cost</td>
<td>$/gge</td>
<td>&lt;3.70</td>
<td>&lt;3.00</td>
<td>3.46</td>
</tr>
<tr>
<td>Electrolyzer Capital Cost</td>
<td>$/gge</td>
<td>0.70</td>
<td>0.30</td>
<td>0.64</td>
</tr>
<tr>
<td>Electrolyzer Energy Efficiency</td>
<td>% (LHV)</td>
<td>69</td>
<td>74</td>
<td>67</td>
</tr>
</tbody>
</table>

Note: Estimates are based on H2A v2.1, for electrolysis only (compression-storage-delivery not included). Model assumes $0.05/kWh.

Electrolyzer cost based on 1,500 kg/day capacity, 500 units/year. Efficiency based on system projections and demonstrated stack efficiency of 74% LHV efficiency.

FY 2012 Accomplishments

- Completed anode formulation optimization demonstrating >50% reduction in catalyst loading and passed internal concept review to build qualification stack.
- Synthesized initial matrix of alternate metal oxide compositions for screening based on refinement of Phase 1 results.
- Combinatorial study completed for several metal combinations and candidates downselected for powder synthesis.
- Tooling procured for large active area electrodes.
- NSTF electrodes successfully applied to alternative membranes.
- 50,000 kg/day concept design completed and quoted.
Introduction

This project addresses the DOE Hydrogen Program objective for distributed production of hydrogen from proton exchange membrane (PEM) water electrolysis. The DOE technical targets for hydrogen cost as well as electrolyzer efficiency and capital cost will be directly addressed through the advancement of key components and design parameters. For renewable applications such as grid energy storage, a continuum of options from distributed hydrogen generation to centralized production at capacities on the order of 50,000 kg/day will be needed. The majority of the electrolysis efficiency losses arise from the oxygen evolution overpotential and the membrane ionic resistance. To reach a target system operating efficiency of 69%, new catalyst and membrane materials are needed. Especially for these large scales, it is necessary to minimize the use of precious metals. Many learnings from fuel cell materials research can be applied to enable the required advancements. Design studies are also needed for PEM electrolysis at this scale to understand the capital costs, environmental impact, and operation and maintenance requirements.

Approach

This project addresses crucial elements of Proton’s technology roadmap, with a focus on improving MEA performance. The oxygen evolution catalyst and membrane are therefore the two key areas of focus. Critical issues include long-term stability of the membrane under electrolysis conditions, particularly as it relates to long-term creep with a thinner membrane, and stability of the catalyst to dissolution. Continuation of the work completed in Phase 1 includes: 1) refinement of the mixed metal oxide catalyst composition for increased activity while maintaining voltage stability, 2) initial reductions in catalyst loading of 50% or greater based on implementation of improved manufacturing processes at Proton, 3) integration of optimized catalyst compositions into nanostructured thin films to demonstrate an additional order of magnitude reduction in catalyst loading and development of high speed manufacturing capability, 4) use of reinforced membranes in order to reduce membrane thickness and ionic resistance without decreasing durability, and 5) projection of these improvements to 50,000 kg/day production levels including cost modeling, impact on reduction of greenhouse gas emissions, and conceptual system design.

Catalyst compositions were made using two techniques. Iteration on promising compositions from Phase 1 were manufactured as nanopowders through fusion of soluble metal salt precursors at high temperatures. Nanopowders will be screened through fabrication of bench-scale MEAs. New candidates were made through ink jet printing and sintering, and were screened through a fluorescence technique to determine the relative amount of oxygen evolution at a given overpotential. The approach is shown in Figure 1. The conceptual 50,000 kg/day system design was developed through sizing calculations of the major system components and consultation with relevant industries such as chlor-alkali to determine typical redundancy factors and margins. Items such as power conversion components, water treatment facilities, pressure vessels, and storage tanks were quoted through suppliers well versed in the relevant sizes. Cell stack costs were estimated based on a module similar to Proton’s largest active area platform and known design improvements validated at a prototype level.

Results

Building from initial feasibility studies under a previous DOE project, significant progress has been made towards production implementation of reduced catalyst loading. Improvements in print uniformity were accomplished through formulation optimization of the ink carrier solution. With the optimized formulation, improved mass activity was observed as shown in Figure 2. Initial cells have been placed on durability testing, with a full scale validation stack expected within the next month. NSTF electrodes have also been fabricated at 3M and are being sent to Proton for testing.

The nanopowder catalyst candidates fabricated at Proton are being fabricated into MEAs for performance testing. For the combinatorial approach, the baseline formulation was...

FIGURE 1. Ink jet printer used for combinatorial studies and sample print

FIGURE 2. Mass activity for various catalyst loadings with improved formulation
Trimetallic compositional matrices were printed for several candidates including combinations of Ir, Ru, Pt, Sn, Nb, and Mn. All compositions contained two of the catalytic metals (Ir, Pt, and Ru) and one of the structural metals (Sn, Nb, and Mn). This approach has been proven to be successful in developing catalysts for photoelectrochemical activity and was thus applied for electrolysis. Screening was completed for electrochemical activity and the fluorescence technique was demonstrated to be effective in determining areas of high activity. Polarization curves also show improvement for an initial candidate as shown in Figure 3. Final downselect is ongoing, to be completed within the next month.

The 50,000 kg/day design concept is shown in Figure 4. Power supplies were sized for voltage and current output based on optimal integration with grid input, for minimal efficiency loss. Cell stack voltage was then determined and the module size scaled to leverage Proton’s existing design principles. The number of components such as phase separators, pumps, and tanks were determined based on optimization of cost, reliability considerations, and available sizes. The overall concept was estimated to result in a capital cost of $0.49/kg.

**Conclusions and Future Directions**

- Manufacturing transition for initial reduction in catalyst loading in progress:
  - Reproducibility and manufacturability demonstrated.
- Catalyst activity and durability screening ongoing:
  - Proton compositional testing in progress at MEA level.
  - U. Wyoming compositions evaluated and undergoing final downselect.
  - Synthesis scale up to be initiated for Wyoming candidates.
- Electrode and stack scale up initiated:
  - Tooling obtained for large active area stack.
  - Reduced loading validation stack to be tested.
- 50,000 kg/day initial concept design complete:
  - Add stack balance of materials as inputs.
  - Conduct environmental impact assessment.
  - Update with MEA electrical efficiencies and operational data as testing progresses.