II.A.1 PEM Electrolyzer Incorporating an Advanced Low-Cost Membrane

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Subcontractors:

- Virginia Polytechnic Institute and University Blacksburg, VA
- Parker Hannifin Ltd domnick hunter Division, Hemel Hempstead, United Kingdom

Project Start Date: May 1, 2008
Project End Date: May 31, 2013

Overall Objective

Develop and demonstrate advanced low-cost, moderate-pressure polymer electrolyte membrane (PEM)-based water electrolyzer system to meet DOE targets for distributed electrolysis:

- Develop high-efficiency, low-cost membrane
- Develop long-life cell-separator
- Develop lower-cost prototype electrolyzer stack and system

Fiscal Year (FY) 2013 Objectives

- Deliver and demonstrate prototype electrolyzer system at the National Renewable Energy Laboratory (NREL)
- Conduct membrane evaluations under aggressive conditions:
  - High-pressure evaluation
  - High current density evaluation

Technical Barriers

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

(F) Capital Cost

(G) System Efficiency and Electricity Cost

Technical Targets

Table 1 presents the technical targets for this project.

TABLE 1. Giner Progress toward Meeting DOE Targets for Distributed Electrolysis Hydrogen Production

Characteristics	Units	2015/2020 Target[a]	2013 Giner Status
Hydrogen Levelized Cost[b]	\$/kg H_2	3.90/<2.30	3.64[c] (5.11)[d]
System Electrolyzer Energy Efficiency	% (LHV)	72/75	65
Stack Electrolyzer Energy Efficiency	% (LHV)	76/77	74

[a] 2012 Hydrogen Production MYRDD Plan.
[b] Production Only.
[c] Utilizing H2A Ver.2 (based on electricity cost of $0.039/kW).
[d] Utilizing H2A Ver.3 (based on electricity cost of $0.057/kW).
LHV – lower heating value

FY 2013 Accomplishments

- Membrane
  - Demonstrated dimensionally stable membrane (DSM™) efficiency and performance:
    - Demonstrated high membrane efficiency of 74% LHV (>87% higher heating value, HHV)
    - Demonstrated improved membrane stability at 3x nominal operating current density of 5,000 mA/cm²
    - Demonstrated membrane operation at high differential pressure of 5,000 psig
- Cell-Separator
  - Developed cell-separator with life expectancy of >60,000 hours
- Electrolyzer Stack and System Design
  - Completed fabrication of full-scale electrolyzer stack utilizing low-cost components
  - Reduced electrolyzer stack costs by 60%
  - Commercialized electrolyzer stack in 30-, 60-, and 100-cell configurations
  - Improved stack design to accommodate up to 200 cells/stack

- Delivered and demonstrated prototype electrolyzer system at NREL:
  - Demonstrated high stack voltage efficiency: 73.6% LHV (>87% HHV) @ 1,500 mA/cm²
  - Demonstrated hydrogen dryer with efficiency of 96.6%
  - Completed DOE Joule Milestone for the project

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INTRODUCTION

The DOE has identified hydrogen production by electrolysis of water at forecourt stations as a critical technology for transition to the hydrogen economy, and as the hydrogen economy matures, for hydrogen production at centralized locations using renewable energy sources. However, state-of-the-art electrolyzers are not economically competitive for forecourt hydrogen production due to their high capital and operating costs. The cost of hydrogen produced by present commercially available electrolysis systems is estimated to be \$4.20/kg-H_2, considerably higher than the DOE target of <\$2.30/kg-H_2 by 2020 [1]. Analysis of electrolyzer systems performed by Giner and others using DOE's H2A model indicate that the major cost elements are the cost of electricity, the capital costs of electrolyzer stacks and systems, and the high cost of hydrogen compression, storage, and delivery.

Giner, Inc. (Giner) has developed PEM-based electrolyzer technology that operates at differential pressure for producing hydrogen at moderate to high pressure directly in the electrolyzer stack, while oxygen is evolved at near-atmospheric pressure. The goals of the project are to reduce the cost of the stack and system, improve electrolyzer efficiency, and to demonstrate electrolyzer operation at moderate pressure.

APPROACH

To reduce the cost of producing hydrogen, Giner is improving electrolyzer stack efficiency and reducing stack cost through development of an advanced low-cost, high-strength, membrane that utilizes a perforated polyimide support imbibed with perfluorosulfonic acid (PFSA) ionomer. Giner is also reducing stack capital cost and increasing stack life through development of a long-life bipolar stack cell-separator, decreasing stack costs by initiating scale up to a larger active area, and reducing the system capital cost by applying commercial production methods to PEM-based electrolyzer systems. In each of the key development areas, Giner and its team members are conducting focused development of advanced components in laboratory-scale hardware, followed by life-testing of the most promising candidate materials.

Successful development of the advanced electrolyzer stack and system will result in a high-efficiency, low-capital-cost electrolyzer that will meet the DOE cost targets for hydrogen production, assuming high-volume production. This will provide competitively priced hydrogen for delivery at forecourt stations to enable transition to the hydrogen economy.

RESULTS

A 0.5 kg-H_2/hr prototype electrolyzer system was delivered to NREL's National Wind Technology Center for validation. The high efficiency of the electrolyzer stack and system, as measured by Giner and Parker during in-house evaluations, were confirmed by NREL. During NREL's evaluation, electrolyzer stack efficiencies were measured at 74% LHV (87%, HHV) at a nominal operating current density of 1,500 mA/cm², 72% LHV (85%, HHV) at a current density of 1,900 mA/cm², and >76% LHV (90%, HHV) at current densities less than 1,000 mA/cm². A DOE Joule Milestone was also completed with the successful validation of the electrolyzer unit.

The results in each of the key development areas of this project are summarized in the following.

<u>DSM™ Membrane Performance</u>: To improve electrolyzer efficiency, Giner has developed an advanced supported membrane having an ionic resistance comparable to that of a 0.0020 to 0.0035-inch-thick Nafion® [2] membrane, but having significantly improved mechanical properties. This advanced membrane is referred to as a DSM™ due to the membrane support that minimizes changes in dimensions (swelling/contraction) under high-pressure operation and with changes in water content. The support structure utilized in the development of the DSM™ consists of a polyimide (Kapton®) base film with a definable open pattern. The support structure is then imbibed with 1100 equivalent-weight (EW) PFSA ionomer to a thickness of 3 mil (0.003 inch). Initially, Giner fabricated the membrane support structures using a laser-drilling procedure. In 2011, a more cost-effective technique of fabricating the support structures via chemical-etching was implemented by Giner, reducing the cost of the membrane by one order of magnitude.

Polarization scans of the DSM™ were conducted in scaled-up, 0.5 kg-H_2/hr electrolyzer stack hardware, through a current density range of 0-1,900 mA/cm², a differential pressure of 300 psid, and a temperature of 80°C. The average cell voltage was measured at 1.757 V/cell corresponding to a voltage efficiency of 74% LHV (87% HHV) at a current density of 1,500 mA/cm².

Durability of the DSM™ was also demonstrated in the scaled-up, hardware via fluoride release rate (FRR) measurements at constant-current operation. Since PFSA ionomer is used as the membrane material and in the binder for the catalyst layer, the loss of fluoride is used as a measurement of membrane degradation. Based on electrolysis FRR results, the lifetime of the DSM™ is projected to be between 45,000 and 55,000 hours. Membrane lifetimes beyond this period were also demonstrated during high current density operation (5,000 mA/cm²) using an advanced DSM™ fabricated with a proprietary additive that mitigates membrane degradation. Finally, a significant improvement in membrane creep property and mechanical stability were demonstrated during high differential pressure operation. The high strength of the DSM™ membrane provided sealing up to an operating pressure of 5,000 psi.

Cell-Separator Development: The cell separator is a gas-impermeable conductive sheet that separates the hydrogen and oxygen compartments in the bipolar stack. The separator must be highly conductive, as well as resistant to hydrogen embrittlement and to corrosion in an oxidizing environment. Giner's legacy high-pressure naval electrolyzers use a complex multi-layer cell-separator incorporating a conductive compliant member and sheets of niobium and zirconium metal. Zirconium is used due to its high resistance to hydrogen embrittlement. Giner has previously evaluated a low-cost, dual-layer titanium cell-separator. Although performance was comparable to that of niobium/zirconium cell-separators, lifetimes were limited to 5,000 hours due to hydrogen embrittlement.

The most promising approach for long-term implementation has been achieved by coating titanium with a low-cost electrically conductive, embrittlement-resistant carbon coating. The challenge was the development of a pinhole-free, highly adherent coating with the required characteristics. Under the cell-separator development task, Giner demonstrated performance of a carbon/titanium cell separator in scaled-up 290-cm² electrolyzer stack hardware. Performance is comparable to that of the niobium-zirconium separator. In addition, life expectancy of the carbon/titanium separator, determined via hydrogen-uptake analysis over a 5,000-hour period, indicates lifetimes exceeding the 50,000-hour system requirement.

Electrolyzer Stack and System Fabrication: In addition to the use of chemically etched DSM™ and carbon/titanium cell-separators, the electrolyzer stack includes several modifications to Giner's legacy hardware: (1) an increase in cell active area from 160 to 290 cm², effectively reducing the number of cells required to produce a given amount of hydrogen, thus reducing the stack manufacturing labor; (2) an overall decrease in the parts count per cell (from 41 to 10); (3) a 75% reduction in anode and cathode catalyst loadings; (4) molded thermoplastic cell frames, resulting in a cost reduction of 95% as compared to machining this component; (5) a 33% reduction in cell frame thickness, thus reducing the anode and cathode support materials and costs by 33%; and (6) a low-cost carbon-steel end plate. As a result of the component and membrane development during this project, the overall projected capital cost of the electrolyzer stack (Figure 1) has decreased from greater than $1,000/kW in 2007 to <$350/kW in 2011.

The electrolyzer system, shown in Figure 2, required detailed planning with respect to system layout and fabrication sequence. Several factors, including specific codes and standards that are pertinent to hydrogen electrolyzer systems, were considered during the system layout. To meet these requirements, the system was designed with three separate compartments: the oxygen (O_2), the hydrogen (H_2), and the electrical (controller and power supply) compartments. The O_2 compartment contains the oxygen gas-phase separator, a circulating liquid pump, and the deionized water feed tank. The H_2 compartment encloses a novel high-efficiency (96.6%) hydrogen dryer assembly, high- and low-pressure hydrogen gas-phase separators, a heat exchanger, cooling fans, and various flow valves. The electrolyzer stack is powered via a direct-current (DC) power supply located in the electrical compartment.

The system build**,** undertaken at the Parker facility, was delivered and tested at NREL's National Wind Technology Center. A breakdown of the system performance is shown in Table 2. At an operating current density of 1,600 mA/cm², the electrolyzer stack exhibits an energy efficiency of 47.3 kWh/kg; the overall system (not including the H_2-dryer) is 57.5 kWh/kg, and with the H_2-dryer, 64.8 kWh/kg.

FIGURE 1. Electrolyzer Stack

FIGURE 2. Electrolyzer System

Included in the efficiency calculations are the additional power requirements for safety ventilation fans (0.7 kW) and the inefficiency of the DC power supply (4.2 kW, 80% efficient) which was below that of the manufacture's rated efficiency of 94%. Compensating for the efficiency of the DC power supply, the PEM-based electrolyzer system will operate in the range of 50.5 kWh/kg (not including the H_2-Dryer) and 54.0 kWh/kg (with the H_2-Dryer).

CONCLUSIONS AND FUTURE DIRECTIONS

Significant progress has been made in DSM™ membrane development. Giner has demonstrated membrane reproducibility and durability as well as a significant improvement in electrolyzer cell efficiency. The progress made during this program has enabled electrolyzer stack efficiencies that meet DOE's 2012 targets and that are in line with achieving DOE's 2015 efficiency goals. In addition, development efforts conducted under this project have resulted in significant cost reductions of PEM-based electrolyzer stacks and systems, an increase in the life of the cell-separators, and improved balance-of-plant components efficiency. Work on this project had been completed in May 2013, however Giner will continue to investigate methods that further reduce cost and improve the efficiency of PEM-based electrolyzer stacks and systems. Giner believes that future challenges for PEM electrolysis will include:

- Scale up of technology for large energy storage applications
- Improve pressure capabilities of stacks (6,250 and 12,688 psi) to eliminate the need for mechanical compressors

TABLE 2. Electrolyzer System Performance

Hydrogen Production & Losses	Units	1600 mA/cm²
Stack H_2-Production	kg-H_2/hr	0.468
Membrane permeation losses (-0.6%)		-0.005
Phase-Separator (-0.14%)		-0.0007
H_2-Dryer (-3.4%)		-0.015
Total H2-Production		**0.43**
Power Consumption	**Units**	**1600 mA/cm²**
Electrolyzer Stack	kW	**21.9**
DC power supply & control (~80% Efficient)		**+ 4.2**
PLC Rack		0.05
Electrolyzer Water Pump		0.30
Heat exchanger fans A & B		0.05
H_2 sensor circuit pump		0.12
Total Energy Consumption (No Dryer)		**26.6**
H_2-Dryer		0.59
Total Energy Consumption (w/Dryer)		**27.9**
Overall Efficiencies	**Units**	**1600 mA/cm²**
Electrolyzer Stack (includes permeation)	kWh/kg	**47.3**
System (No Dryer)		**57.5**
System (w/Dryer)		**64.8**

- Further improvements in membrane efficiency and costs reductions

FY 2013 PUBLICATIONS/PRESENTATIONS

1. M. Hamdan, *Evaluation and Challenges of PEM Electrolyzer for Commercial Applications. Presentation at the* NREL ESIF meeting, August 30, 2012.

2. M. Hamdan, *PEM Electrolyzer Incorporating an Advanced Low-Cost Membrane.* 2013 Hydrogen Annual Program Merit Review Meeting, Presentation #pd030, May 15, 2013.

3. M. Hamdan, *Unitized Design for Home Refueling Appliance for Hydrogen Generation to 5,000 psi.* 2012 Hydrogen Annual Program Merit Review Meeting. Presentation #PD065, May 15, 2013.

REFERENCES

1. 2012 Multi-Year Research, Development and Demonstration Plan. Hydrogen Production. DOE, Pg 3.1-14, (http://www1.eere.energy.gov/hydrogenandfuelcells/mypp/pdfs/production.pdf)

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