III.11 Electrochemical Compression

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- National Renewable Energy Laboratory, Golden, CO
- Rensselaer Polytechnic Institute, Troy, NY
- Gaia Energy Research Institute LLC, Arlington, VA

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Project End Date: September 30, 2019

Overall Objective

- Develop and demonstrate an electrochemical hydrogen compressor (EHC) to address critical needs of lower-cost, higher efficiency, and improved durability.

Fiscal Year (FY) 2017 Objectives

- Improve EHC water and thermal management .
  - Development, optimize, and demonstrate water management membranes (WaMM).
  - Engineer flow distributors for thermal management in high pressure cells.
- Optimize stack hardware and demonstrate cell performance ≤0.250 V/cell at current densities ≥1,000 mA/cm².
  - Synthesize hydrocarbon (HC) membranes for high pressure, efficient EHC operation.

Technical Barriers

This project addresses the following technical barriers from the Hydrogen Delivery section of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan.

(B) Reliability and Costs of Gaseous Hydrogen Compression

Technical Targets

TABLE 1. Progress towards Meeting Technical Targets for Hydrogen Compressors for Fueling Sites

Characteristics	Units	2020 Target[1]	2017 Giner Status
Availability	%	85	--
Compressor Specific Energy	kWh/kg	1.6[2]	2.72[3] (<1.2)[4]
Uninstalled Cap. Cost[2]	$	275,000	<450,000
Annual Maintenance	% of Capital Cost	4	--
Lifetime	Years	10	--
Outlet Pressure Capability	bar	950	350

[1] Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan/Delivery Section.
[2] 100-bar delivery/Commercial mechanical compressors are >6-8 kWh/kg (@700-bar delivery).
[3] Operation at 2-bar delivery
[4] Projected at 100-bar delivery

FY 2017 Accomplishments

Synthesized HC membranes with high ion exchange capacities (IEC), high protonic conductivity, and low electro-osmotic drag (EOD) compared to conventional perfluorosulfonic acid (PFSA) proton exchange membranes (PEM).

- Conductivity of synthesized HC membranes = 0.106 S/cm (target ≥0.100 S/cm)
- EOD: <60% of PFSA (target <50%)
- IEC: up to 2.0 mmol/g (target >1.8 mmol/g)
- Improved EHC cell voltage performance: 0.110 V/cell at 1,000 mA/cm² (target 0.25 V/cell)

- Synthesized WaMM with high water flux and high electrical conductivity.
  - Water flux: ≥0.1 g/min-cm^2 (target of ≥0.039 g/min-cm^2)
  - Conductivity: Through-plane = 1.0 S/cm, in-plane >10 S/cm (target 1.0 S/cm)
  - Demonstrated WaMM that enables passive water feed in operational EHC, that significantly improves water management, and stabilizes EHC cell voltage performance
- Successfully demonstrated >350 bar (5,000 psi) EHC operation; highest efficiency in a single-stage EHC operating at 5,000 psi.

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INTRODUCTION

Hydrogen compression represents a key technical challenge for the widespread commercialization of fuel cell electric vehicles. To dispense hydrogen to fuel cell electric vehicles, hydrogen must be compressed to a minimum of 875 bar. Conventional compressors account for over half of the refueling station's cost, have poor reliability, and insufficient flow rates [1–3]. In addition, the compressed hydrogen delivered by a mechanical compressor requires extensive purification to remove small amounts of contaminates that can degrade the performance of fuel cells receiving the hydrogen.

EHCs utilize direct current to electrochemically compress hydrogen to high pressures. Recent developments in membrane technology promise a new generation of very efficient low-cost EHCs. The emergence of PEM-based solid-state EHCs eliminates many of the issues associated with mechanical compression, however, current state-of-the-art EHCs are challenged by issues related to membrane sealing, and low operating current density – attributed to poor water and heat management.

Giner, Inc. (Giner) is a leading developer of PEM-based stack technology with over 40 years experience in advanced electrochemistry. Giner's technology ranges from highly-reliable high-powered electrolyzer stacks operating onboard U.S. Navy submarines to lower-cost electrolyzer systems operating at differential pressures of up to 1,000 psi for commercial applications. Giner has made significant advancements developing and demonstrating novel membrane technologies that enable zero-leakage, high-pressure, and high-efficiency for use in PEM-based electrolyzers and EHCs that operate >350 bar.

APPROACH

The work conducted in this program exploits the use of three novel technologies that include (1) dimensionally stable HC membranes engineered with low electro-osmatic drag and low hydrogen diffusivity that also exhibit high durability and sealing properties, (2) a WaMM that enables passive water feed and performance stability, and (3) an advanced high-pressure stack design optimized for safe high-pressure gas compression. Successful development and implementation of these technologies are essential to improving water and thermal management within the EHC and will enable high current density operation resulting in a low-cost hydrogen compressor.

RESULTS

<u>Hydrocarbon Membrane Development:</u> Water management within an EHC is significantly improved with PEMs that have a low EOD coefficient. Biphenyl based-HC membranes exhibit EOD rates as low as ~1.2 H_2O/H^+ [4]. In collaboration with Rensselaer Polytechnic Institute, high-molecular-weight, partially-fluorinated, perfluoroalkylsulfonte (BP-Ar) and bromoalkyl-tethered aromatic polymer biphenyl membranes have been synthesized. In comparison to commercially available PFSA, the biphenyl based-HC membranes have an EOD that is 60% less. Furthermore, the HC membranes have demonstrated improved EHC cell performance that is attributed to (1) higher molecular weights leading to better mechanical stability, particularly under low humidified conditions, (2) low swelling related to a hydrophobic backbone owed to partial substitutions of fluorinated moieties, and (3) higher IEC and protonic conductivity enabling higher cell performance. The achievements related to the HC membrane development are summarized below:

- Conductivity: 0.106 S/cm
- EOD of HC membrane: <1.0 H_2O/H^+
- IEC of BP-Ar: 1.45-2.0 mmol/g

<u>WaMM Development:</u> Water management within the EHC is significantly increased when low-EOD HC membranes are combined with a WaMM. The WaMM is a composite membrane that combines an ionomer with an electrical conductor composed primarily of carbon nanotubes. The ionomer allows water transport while the carbon completes the electrical circuit and minimizes ohmic losses. The ionomer-based WaMMs have a high affinity for water. Water fed to one side of the WaMM permeates through the membrane until it reaches the adjacent PEM. Water in the PEM is auto-replenished when a concentration gradient is created between the WaMM and the PEM. The efficacy of the WaMM to transport water is dependent on several variables including water temperature and WaMM thickness. Based on an EOD rate of ~3.5 H_2O/H^+ (in PFSA PEMs), a water flux of 0.04 g/min-cm^2 is required to satisfy operation in the EHC at a current density of 1,000 mA/cm^2. The water flux through the WaMM has been measured at >0.10 g/min-cm^2 which is sufficient to operate an EHC cell up to 3,000 mA/cm^2. In addition to high water flux, the WaMMs have been optimized for electrical conductivity. The through-plane conductivity of the WaMM has been measured at 1.0 S/cm. When assembled in a bipolar stack configuration, the WaMM introduces an insignificant cell voltage loss of <6 mV/cell at an operating current density of 1,000 mA/cm^2.

<u>Membrane Performance:</u> Membrane evaluations were conducted in low-pressure test hardware operating at a pressure of 300 psi (20.7 bar). The BP-Ar HC membranes were evaluated against commercially available PFSA

membranes. A modified flow distributor, used for water transport and thermal management within the cell was utilized. Polarization scans were conducted up to a current density range of 2,000 mA/cm². EHC cells with BP-Ar, membranes exhibit significantly lower cell voltages (higher cell efficiency) than EHC cells assembled with PFSA membranes. At an operating current density of 1,000 mA/cm², the cell voltage of the BP-Ar membrane is 0.110 V/cell; a 60-mV improvement compared to PFSA (Figure 1). In addition to polarization scans, short term duration testing was conducted on BP-Ar and PFSA membranes with and without the assistance of a WaMM. During duration testing, the EHC is held at a constant current density of 1,000 mA/cm². Results indicated that the PFSA membranes (with high EOD) tend to dry out at elevated current densities. When assisted with a WaMM, the PFSA membranes operated continuously without dehydrating. The BP-Ar membranes, however, exhibited stable performance with and without the use of a WaMM (Figure 2). The additional water contribution provided by a WaMM is not required for HC membranes with low EOD; albeit a WaMM may be required at elevated operated pressures when water losses related to hydraulic permeability have a larger effect on the water content within the PEM.

High Pressure EHC Stack Evaluation: A high-pressure EHC stack, designed with the modified flow distributors and WaMM, was used to demonstrate 5,000 psi (350 bar) EHC operation, Figure 3. Giner consumed all HC membranes in previous optimization testing; therefore, high-pressure testing was limited to PFSA membranes. During operation, the anode (inlet) of the EHC stack is fed with dry hydrogen from a cylinder tank at a pressure of 35 psi (~2 bar). The hydrogen feed is dead-ended, i.e., not recirculated and the anode is occasionally purged to remove impurities that can accumulate in the anode chamber over time. At the cathode (outlet) of the EHC stack, hydrogen flow is restricted using a back-pressure regulator to attain the desired operating pressure.

PFSA membranes were evaluated up to a pressure of 5,200 psig (~360 bar), Figure 4. During operation, the hydrogen pressure was gradually increased to 5,200 psig while monitoring cell voltage. A cell voltage of 0.25 V at a current density of ~900 mA/cm² was measured using PFSA membranes assisted with WaMM: **The highest efficiency demonstrated by a single-stage EHC operating at >5,000 psi (>350 bar)**. Future optimization of the high-pressure EHC includes the use of HC membranes with lower EODs (improved water management). Correspondingly, the EHC stack will be designed to operate at the target inlet pressure of 1,450 psi (100 bar), leading to a reduction in the Nernstian voltage penalty and a further improvement in EHC cell efficiency.

CONCLUSIONS AND UPCOMING ACTIVITIES

The developments made during this program have enabled EHC stack voltages that are in line with achieving DOE's 2020 efficiency goals. The highest efficiency in a single-stage EHC, operating at >5,000 psi (>350 bar),

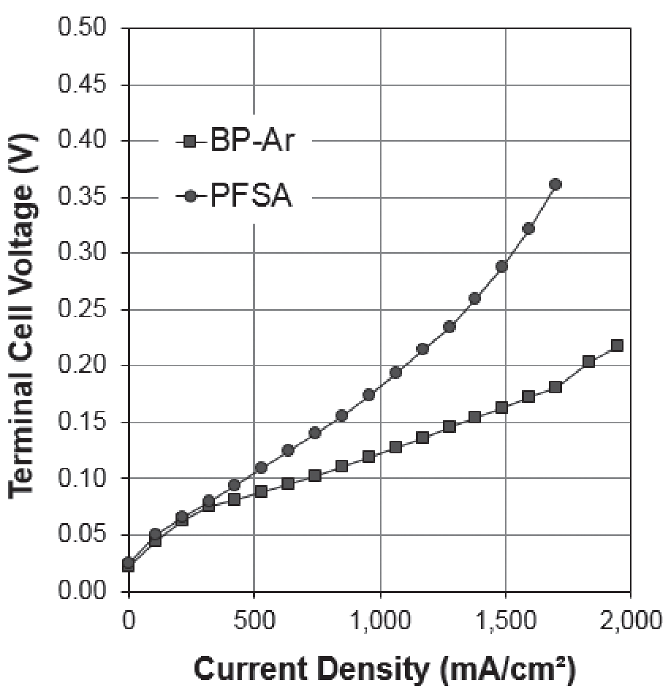

FIGURE 1. BP-Ar vs. PFSA membrane

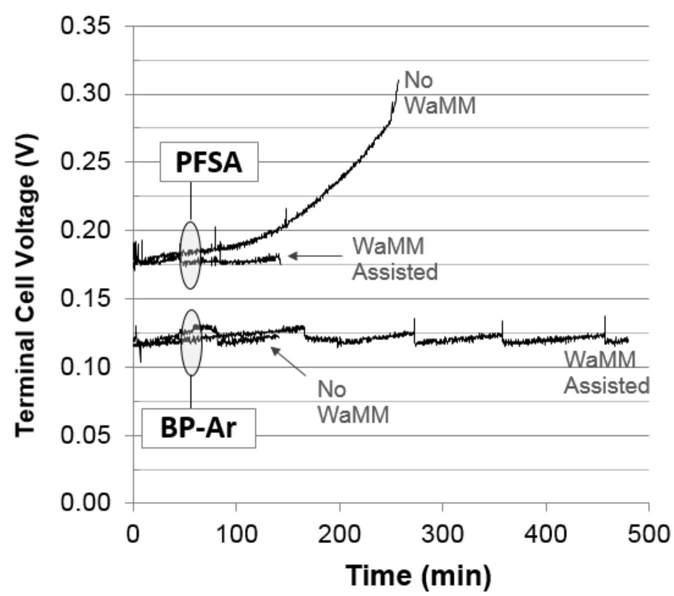

FIGURE 2. Comparison of BP-Ar and PFSA membranes held at 1,000 mA/cm²

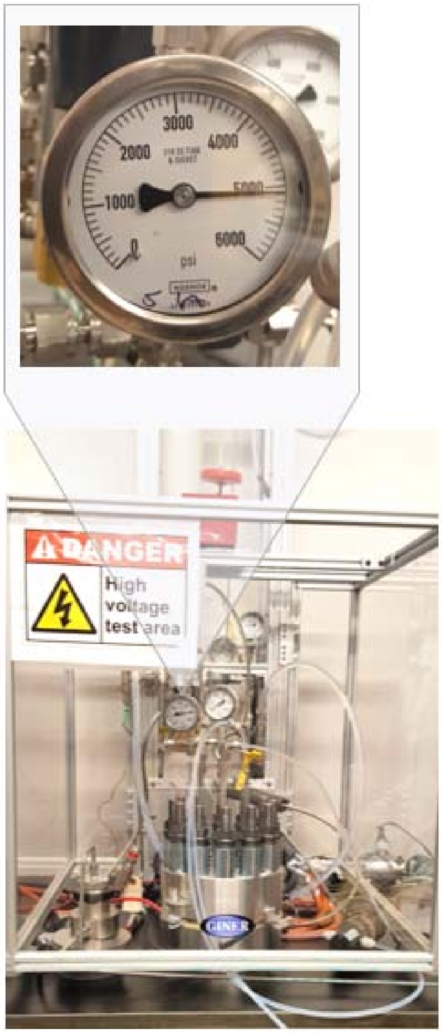

FIGURE 3. EHC stack, 5,000 psi operation

was successfully demonstrated utilizing engineered flow distributors that eliminate thermal management issues, and the development of a WaMM that enables passive water management. In addition, significant improvements in EHC cell voltage, utilizing synthesized HC membranes with low EOD coefficients, has been demonstrated. Future work includes the design, fabrication, scale-up, and testing of a high-pressure 12,688 psi (875 bar) stack and completion of a prototype system design.

FY 2017 PUBLICATIONS/PRESENTATIONS

1. M. Hamdan, *Electrochemical Compression.* 2017 Hydrogen Annual Program Merit Review Meeting, Presentation PD136, June 6, 2017.

2. W. Colella, M. Hamdan, *Techno-economic Analysis of State-of-the-Art Electrochemical Hydrogen Compressors (EHCs)*, 6th European PEFC and Electrolyser Forum, July 4–7, 2017.

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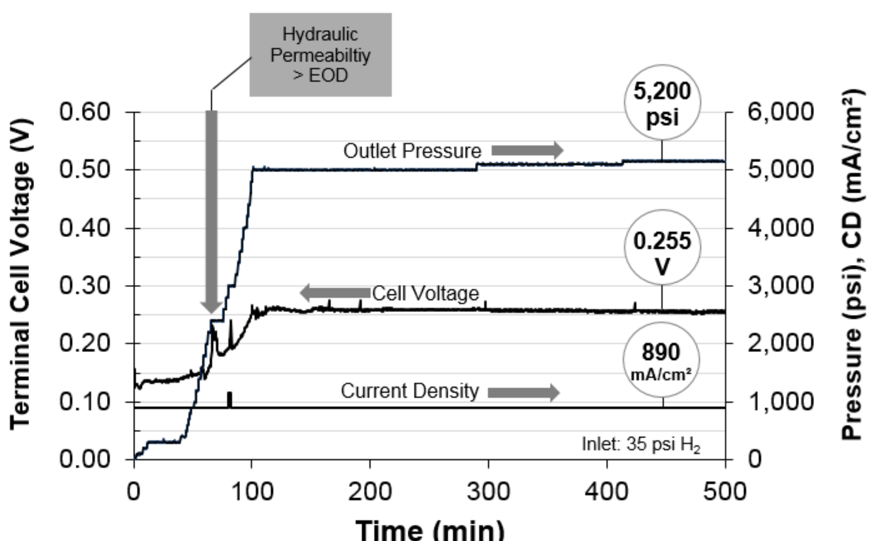
2. G. Parks et al., Hydrogen Station Compression, Storage, and Dispensing Technical Status and Costs (NREL Independent Review: May 2014).

3. http://www.nrel.gov/hydrogen/cdp_topic.html#infrastructure

4. Michael Anthony Hickner et al., Transport and Structure in Fuel Cell Proton Exchange Membranes (Dissertation submitted to the faculty of Virginia Polytechnic Institute, 2003).



CD - current density

FIGURE 4. EHC operation at 5,000 psi with PFSA membrane