II.C.2 Process Intensification of Hydrogen Unit Operations Using an Electrochemical Device

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PBI Performance Products, Inc., Rock Hill, SC

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Fiscal Year (FY) 2012 Objectives

Develop and demonstrate a multi-functional hydrogen production technology based on a polybenzimidazole (PBI) membrane which exhibits:

- High efficiency (70%)
- Up to 100 scfh pumping capability
- CO_2 and CO tolerance
- 300 psig (differential) pressurization capability
- \$3/kg operating costs

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:

(A) Reformer Capital Costs
(D) Feedstock Issues
(E) Greenhouse Gas Emissions

Technical Target

This project is focused on fundamental chemical and mechanical engineering studies on PBI proton exchange membranes and electrochemical cell hardware, respectively. Learnings gained from these studies will be applied to the membrane fabrication process as well as toward the electrochemical cell architecture to meet the following key targets:

- 300 psid pressure operation at 160°C
- CO_2 tolerance
- High efficiency (70%)

FY 2012 Accomplishments

- 300 psid compression demonstrated at 160°C for over 4,000 hours on a PBI-based 50-cm^2 electrochemical pump
- Materials processing finalized at PBI Performance Products
- Large-scale membrane and electrode fabrication process developed
- Cell hardware scale up completed and validated using low temperature membranes
- Stack design demonstrated to over 2,300 pressure cycles

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Introduction

One of the barriers to fuel cell acceptance is the lack of a simple, reliable, cost-effective and robust process to purify, pump, and pressurize hydrogen. This challenge is magnified by impurities and that hydrogen generation often occurs at near ambient pressure. Technical means of pressurizing the hydrogen is especially daunting for low to moderate flow rates. If the pressurization, purification, and recovery of hydrogen from any source, including from hydrogen-intensive industrial processes, can be developed into a single unit operation, it would be an attractive and enabling option for recycling hydrogen. H2Pump plans to leverage its extensive experience in electrochemical separation and pressurization systems to meet the project objectives with a high temperature membrane-based electrochemical hydrogen pump. The solutions will be based on developing a chemically and mechanically robust membrane in conjunction with advancements in cell hardware.

The significance of a successful project would be that impure hydrogen from a variety of sources could be simply, reliably, and inexpensively processed for further use. The hydrogen could be reused in the existing process as recycle, or in new applications including as a source of hydrogen for fuel cells. It is the reduction of multiple unit operations in combination with the high temperature membrane operating at high differential pressure that will enable the benefits of this novel electrochemical approach.

Approach

H2Pump has shown that electrochemical methods to recover, purify, and pressurize hydrogen could be a viable option for low to moderate volumes of hydrogen-containing gases streams. The main challenge for this specific application is the lack of a proton conducting membrane which exhibits carbon dioxide and carbon monoxide tolerance and at the same time be able to pressurize the hydrogen from atmospheric pressure to 300 psid. Working closely with PBI Performance Products, the approach to address this challenge is to enhance the polybenzimidazole membrane properties via chemical and thermal cross-linking methods. Concurrently, H2Pump will work on cell hardware architecture to mechanically support the high temperature, CO_2 and CO tolerant membrane. The effort will focus on the structural integrity of all of the membrane and electrode components, plate materials and geometries, sealing mechanisms, and compression requirements. H2Pump will also investigate operating modes and the impact on performance of various membrane–electrode interface concepts.

Membrane modifications will be tested using 50-cm^2 single-cell hardware as a test platform. Design guidelines developed with the 50-cm^2 lab-scale pumps will be scaled up to the larger format to meet the program targets. Stack hardware development will take place concurrent to the membrane development using low-temperature (LT) membranes as a test platform. Results from the membrane and stack development effort will be combined to produce a large format high-temperature (HT) stack. This large format HT stack will be tested independently and then in combination with a reformer.

Results

During this reporting period there have been significant accomplishments in stack hardware design and scale up as well as completion of the HT membrane electrode assembly (MEA) architecture.

Stack Hardware

Stack hardware has been designed, built, and tested on the large format (920 cm^2) using LT membranes. Large-scale HT PBI membranes were not used due to their unavailability in this period. Furthermore, the cell and stack hardware were designed to be able to accommodate both types of membranes, and as such, the LT membranes provided a means to accelerate stack engineering prior to the HT membranes being available. First generation PBI 920-cm^2 membranes have been successfully prepared and assembled into a short stack and are awaiting testing and evaluation.

Stack hardware design has been improved for high differential pressure performance using the LT membrane. Figure 1 shows how the first generation large format stack hardware had pressure induced lift-off causing loss of performance at high pressures. Figure 2 data shows how the second generation design does not exhibit pressure induced performance limitations at high pressure. Pressure cycle testing was also performed as part of the qualification testing. Over 2,300 pressure cycles have been completed on this hardware without failure or degradation.

PBI MEAs

Three generations of PBI MEA architectures have been tested. Figure 3 shows the significant durability improvement

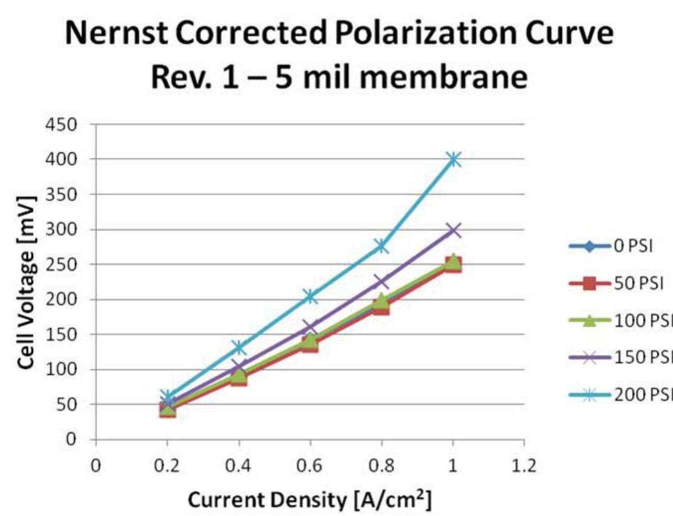

FIGURE 1. First Generation Stack Hardware

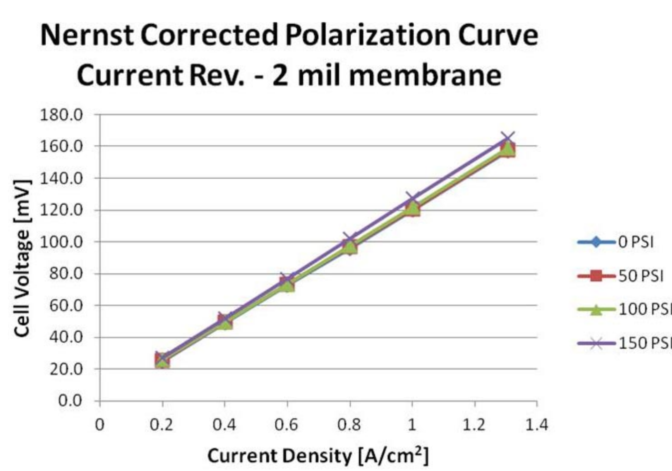

FIGURE 2. Second Generation Stack Hardware

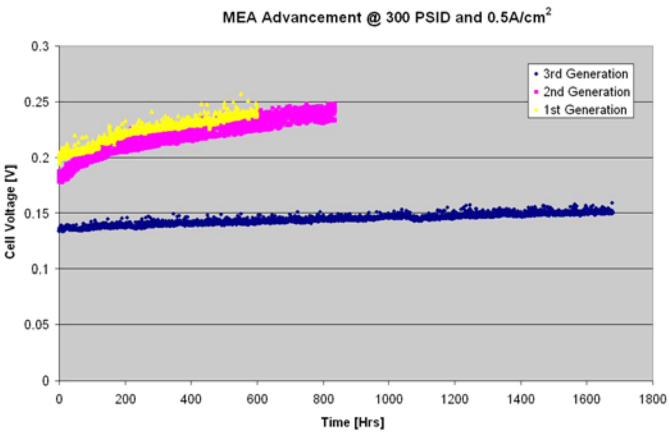

FIGURE 3. PBI MEA Architecture Advancements

the third generation has over the first two generations. Multiple 50-cm^2 PBI single cells have operated longer than 1,000 hours at 300 psid (differential pressure). One cell has even exceeded 4,000 hours at 300 psi differential pressure. Pressures of 400 psid with the PBI membrane have been achieved on a cell for short durations without damage or loss of performance.

The large format HT membrane and electrode fabrication process has been finalized and a novel, proprietary sealing method has been developed that utilizes commercially available, low cost sheet elastomer. Large format PBI-based MEAs have been produced using the selected sealing, electrode, and membrane fabrication process.

Reformer-Pump Integration

The reformer module has been built and tested independent of the pump module.

All major subsystems have competed verification testing in preparation for the sustained 36-hour test of the integrated unit. Gas analyses using gas chromatography methods have been used to determine reference gas compositions at various points within the integrated system. These results will be compared to the actual gas compositions of the integrated unit during the final demonstration.

Conclusions and Future Directions

- In collaboration with our partner, PBI Performance Products, PBI membrane has been successfully modified and is now stable in the targeted operating environment.
- 920-cm^2 stack hardware has been developed and successfully tested using LT membranes. Large-scale HT PBI membrane testing is underway.
- Enhancement of the membrane – electrode interface optimization successfully completed and has been scaled up to large format.
- 300 psid pressure operation has been demonstrated on multiple cells for up to 4,000 hours.
- Scale up of the membrane and electrode assemblies as well as the hardware has been completed.
- Gas quality and analytical tests will continue to be performed to further assess the performance of the 300 psid pump cells.
- Reformer-pump integration and preparation for final demonstration underway.
- H2A analysis to be completed.

FY 2012 Publications/Presentations

1. Eisman, G., Carlstrom, C “Process Intensification of Hydrogen Unit Operations Using an Electrochemical Device” Proceedings of the DOE Hydrogen and Fuel Cell Annual Merit Review Meeting, Crystal City, VA., May, 2012.