V.M.4 Development of Polybenzimidazole-Based High Temperature Membrane and Electrode Assemblies for Stationary Applications

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Subcontractors:

- Rensselaer Polytechnic Institute (RPI), Troy, NY
- RPI Fuel Cell Center, Troy, NY
- BASF Fuel Cell, Frankfurt, Germany
- Albany NanoTech, Albany, NY
- · Entegris, Chaska, MN
- University of South Carolina, Columbia, SC

Project Start Date: August 2003 Project End Date: December 2007

Objectives

- Select the appropriate polymer chemistry for polybenzimidazole (PBI) membrane materials optimized to meet DOE fuel cell requirements.
- Demonstrate the performance of the PBI membrane in single cells and stacks.
- Provide a cost analysis of a low-cost membrane manufacturing process to meet cost and high-volume targets.

Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section (3.4) of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

- (A) Durability
- (B) Cost

Technical Targets

This project is directed at developing hightemperature proton exchange membrane (PEM) technology (polybenzimidazole) membrane electrode assemblies (MEAs) in 45 cm² cells, 440 cm² modules and stacks operating on hydrogen and reformate to meet the DOE 2010/2015 targets for MEAs.

TABLE 1. Progress Towards Meeting Technical Targets for MEAs

Characteristic	Units	2010/2015	Project 2007 Status
Operating temperature	۵°	<120	160
Cost	\$ / kW	10 / 5	TBD
Durability with cycling At operating temp of >80°C	hours	2,000 / 5,000	6,000



Approach

Our approach to developing a MEA and stack which meet the DOE technical targets is to break the work down into three areas of focus (Figure 1).

Plug Power will act as the lead, will develop the acid traps and flowfields and will assemble and test the stack. BASF and RPI will develop the membrane and assemble and test the MEAs. BASF and Albany NanoTech will develop the electrode. Entegris will develop sealing. These focus areas were broken down into eleven tasks.



FIGURE 1. Breakdown of Tasks for International Consortium

Accomplishments

MEA

During the previous year, we continued the detailed examination of phosphoric acid movement and loss from the MEA during the separate steps of cell assembly, heat-up and operation under different conditions using temperature and time as our two major variables (cell operation temperature: 160°C and 180°C, cell operation time: 100, 500 and 2,500 hours). The phosphoric acid in the MEA and the membrane thickness were determined at each step of the assembly and operation.

We also continued the startup/shutdown cycle testing, and are looking specifically at the effects of open circuit voltage (OCV) residence time during heat-up on cell performance. We designed a series of experiments which have different times at OCV per start-up/shut-down cycle (3 hours, 2 hours, 1 hour and a few minutes) to examine the effects on the cell's performance degradation. Three finished samples (time of cell at OCV per cycle is 3 hours, 2 hours and 1 hour) show the cell performance degradation rates, measured at 0.2 A/cm^2 , decreased with decreasing the time of the cell at OCV per cycle. It appears that the effects of OCV residence time during startup/shutdown cycling are less than other effects. These tests are continuing to identify the variables that may govern the degradation processes in this type of operation.

The work on the BASF MEA has been focused on a detailed analysis of the performance of early, scaled-up versions. In particular, load cycling effects, investigation of decay rate discrepancies between Plug Power, BASF and RPI and development of approaches to higher durability. BASF and RPI have demonstrated <10 μ V/hr, H₂/air, dry, steady-state, 20,000 and 14,000 hours, respectively, >6,000 hours load cycling with a projected 14,000-hour life, and 110 start-up/shut-down cycles.

ACID TRAPS

We continue to accumulate run time on acid traps at both the module and system level. They have been exposed to both steady-state and cyclic operating conditions. At present, we have accumulated over 6,500 hours testing on module-sized traps and ~7,000 hours on stack-sized traps with no observed acid breakthrough. The traps are undergoing an extensive autopsy to characterize acid accumulation.

We continue to accumulate run time on acid traps at both the module and system level. They have been exposed to both steady-state and cyclic operating conditions. At present, we have accumulated over 8,000 hours testing on module-sized traps and 10,000 hours on stack-sized traps with no observed acid breakthrough. These exposure tests are complete due to reaching endof-life MEA voltage thresholds on their modules and stacks. The traps are scheduled to undergo extensive autopsy to characterize acid accumulation during 3Q 2007.

FLOWFIELD AND SEALING

In the area of sealing, the goal of this work has been to develop materials and methods of application to permanently seal interfaces between coolers as well as between cells in a high temperature phosphoric acid fuel cell operating environment. The decision was made to use the same nonconductive adhesive in all interfaces, designing the plate features such that suitable conductivity is achieved in the cooler interfaces, and using an insulator to keep the cell interfaces nonconductive. Work this year focused on assembling cells with machined bipolar plates and new MEAs, determining how to consistently produce a leak-proof seal, and also included continued testing on adhesive compatibility.

The first large format (440 cm²) bonded cell was delivered to Plug Power from Entegris. This cell will be the basis of our final testing.

A detailed finite element analysis (FEA) was conducted to study the bonded seal design at Entegris. In summary, the FEA determined bonding a plate this size is possible but requires careful consideration to the design of the plate geometry and bond. It also uncovered a potential source of MEA over compression in the current design. The FEA and experimental observations led Plug Power to reanalyze its tolerance stack-up with regard to pocket depth and plate compression.

This discovery could provide insight into observations made on the performance of the first short stack (16-cell module, Figure 2) using early BASF MEAs. The module ran a total of 7,200 hours and is currently undergoing autopsy. The overall degradation rate of



FIGURE 2. 16-Cell Module Endurance Run at 5,874 Hours

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the module was approximately 30 μ V/hr with most of the losses attributed to station trips. Close inspection of the individual cell data shows both a significant difference in beginning of life voltage (0.66-0.70) and a large difference in decay rates among cells (8-54 μ V/hr). The onset of this spread could coincide with the stress relaxation properties of the MEA. That is, areas of over compression could begin to see membrane thinning, gas crossover and shorts. Another area being investigated is the temperature "stack-up" and whether areas of the MEA can see operating temperatures outside of specification. The possibility of interactions between the two factors is also being investigated.