2005 DOE Hydrogen, Fuel Cells & Infrastructure Technologies Program Review

Development of Polybenzimidazole-based High Temperature Membrane and Electrode Assemblies for Stationary and Automotive Applications

May 23, 2005

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Project ID #
FC8

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OVERVIEW

Timeline

- Project start date Aug 2003
- Project end date July 2006
- Percent complete 50%

Barriers

- O. Stack material and manufacturing cost
- P. Durability

Budget

- Total project funding $7.29 M
  - DOE share $ 5.84 M
  - Plug share $1.46 M
- Funding received in FY04 $1.50 M
- Funding for FY05 $2.05 M

Subcontractors

- Rensselaer Polytechnic Institute (RPI)
  - Polymer Science Laboratory
  - Fuel Cell Center (approval in progress)
- PEMEAS
- Albany Nano Tech
- Entegris
- University of South Carolina

OBJECTIVES

- To identify and demonstrate an MEA based on a high-temperature polybenzimidazole (PBI) membrane that can achieve the performance, durability and cost targets required for both stationary and automotive fuel cell applications (original)

- In August 2004, the Department of Energy modified the objective to focus only on stationary applications

APPROACH

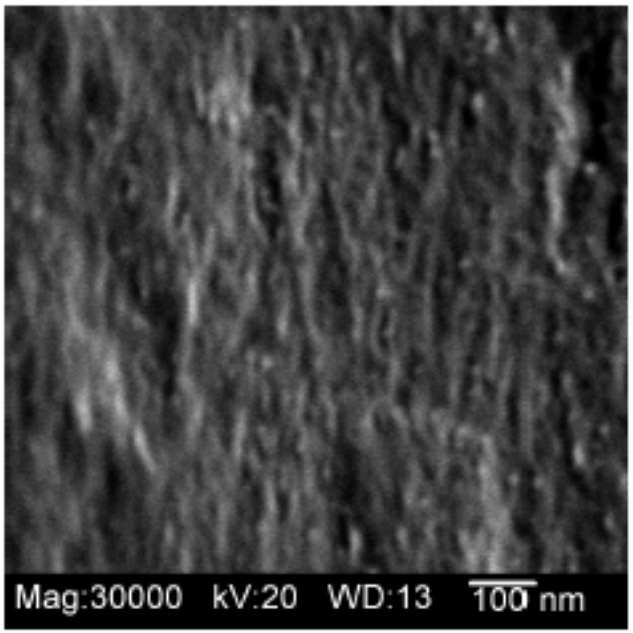
- Membrane (Task 1-4)
  - ✓ Formulate and characterize polymers
  - ✓ Improve membrane mechanical stability
  - • Scale up process and fabricate full size MEAs
- MEA (Task 5-8)
  - ✓ Conduct 50cm^2 screening tests at RPI
  - ✓ Conduct parametric tests to fully characterize MEA performance
  - • Assemble and test a full size short stack
- Stack (Task 9-12)
  - ✓ Characterize acid absorbing materials
  - ✓ Optimize flow fields and sealing
  - ✓ Develop novel electrodes using nanotechnology
  - • Cost assessment

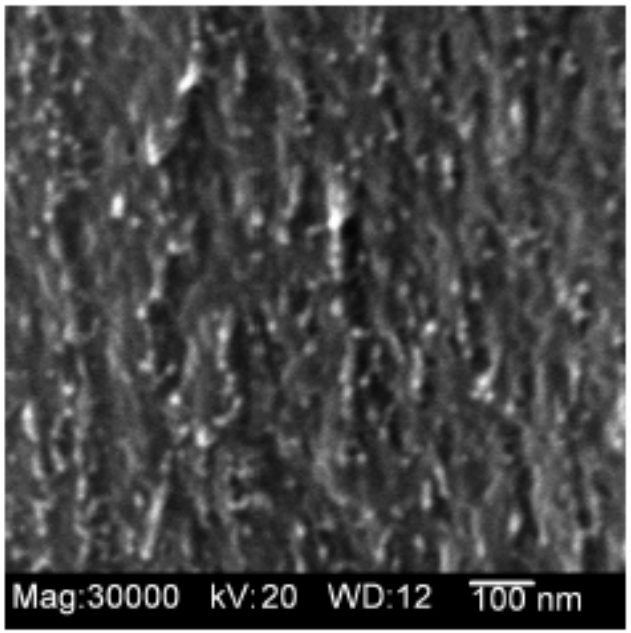
✓ indicates in progress

TECHNICAL ACCOMPLISHMENTS MEMBRANE (TASK 1)

- RPI created five membrane compositions (Type 1, 2, 3, 4 & 5)
- Down-selected to primary composition based on performance characteristics (Type 2)
- Focused polymer membrane effort on mechanical stability
  - Four reinforcing fillers scaled up and under evaluation at various loadings (Type 6, 7, 8 & 9)
- Developed techniques for uniformly dispersing fillers

Without filler

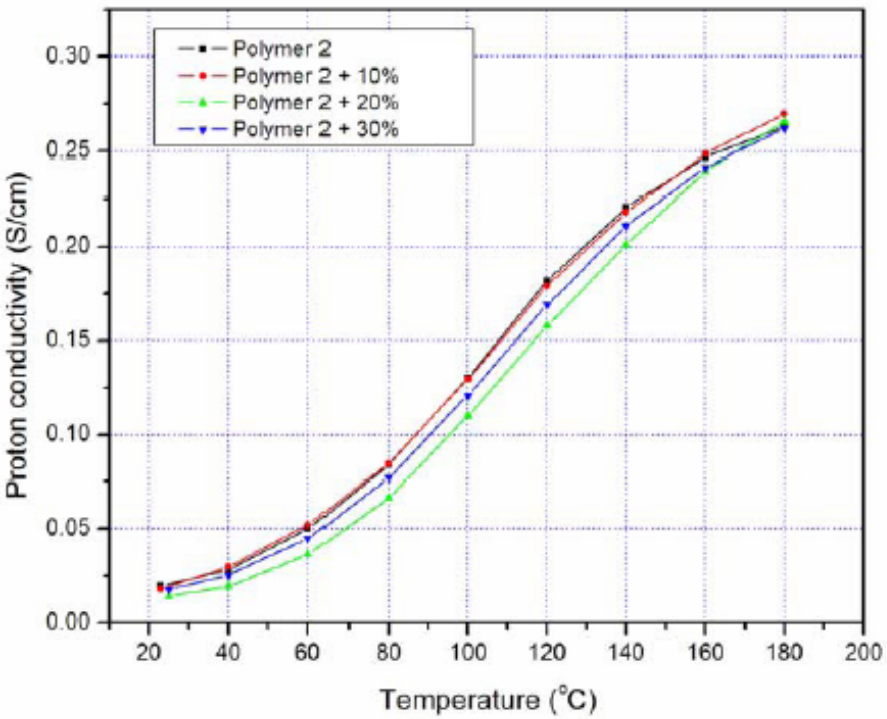


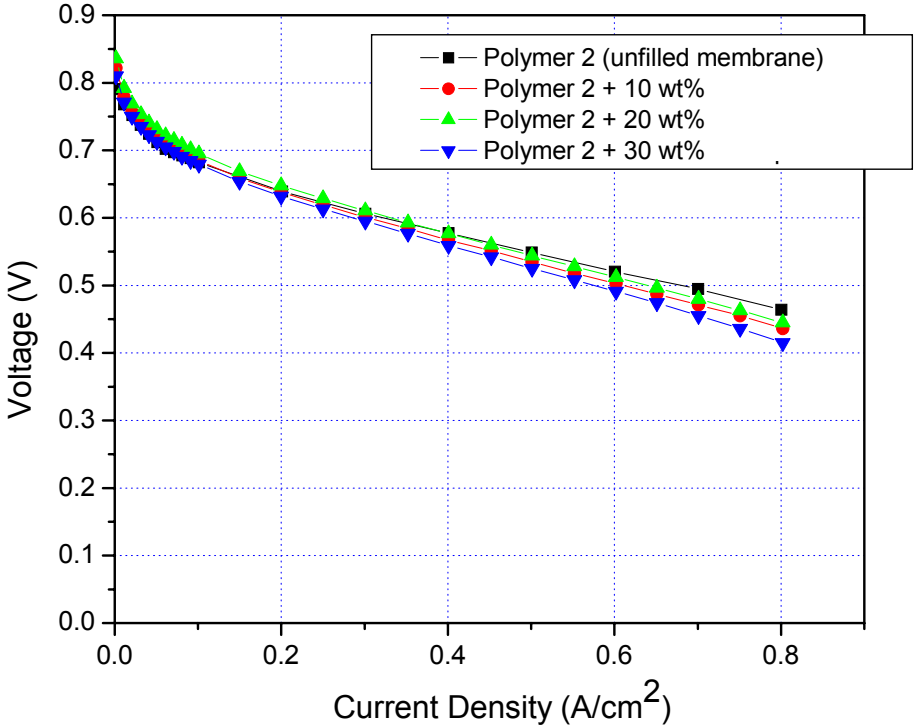


With filler

TECHNICAL ACCOMPLISHMENTS 50 CM^2 SCREENING (TASK 5)

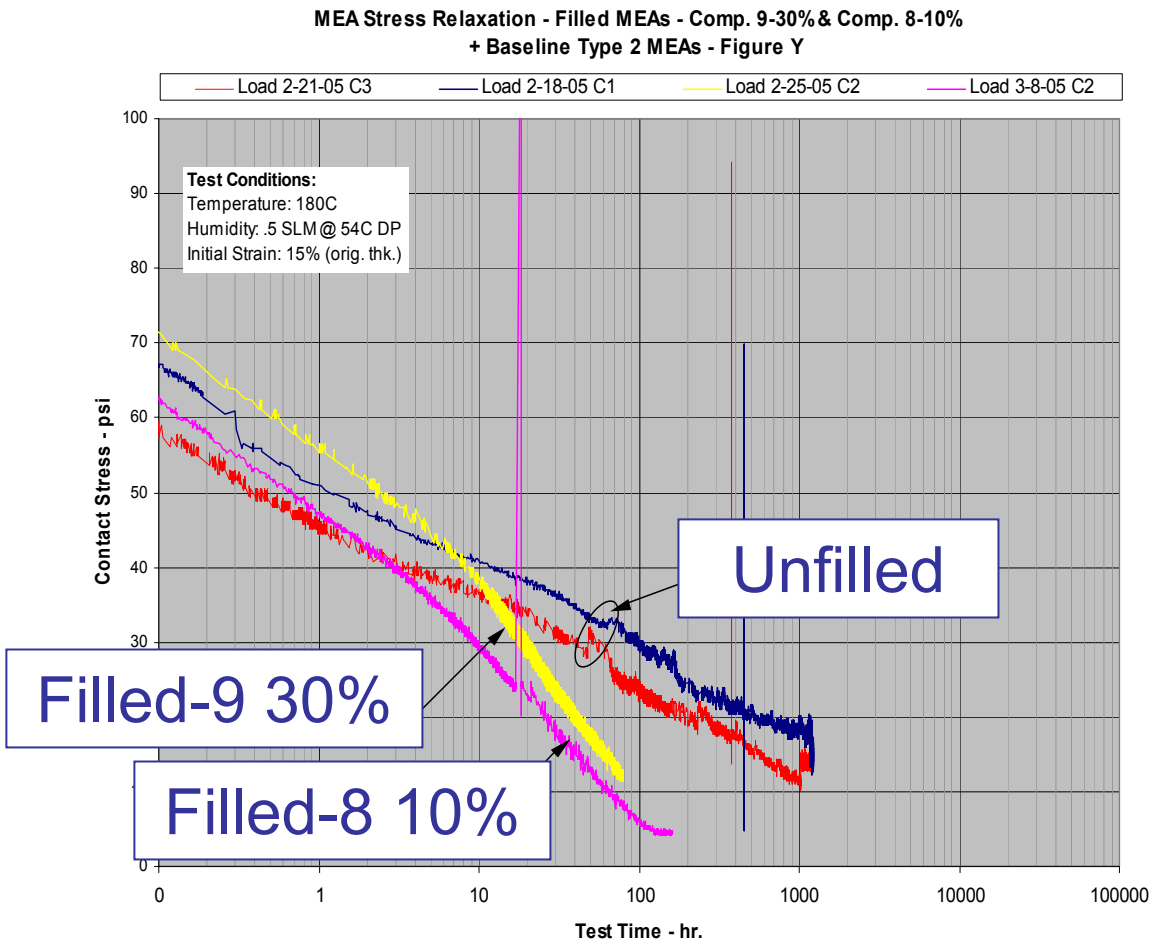
- Membrane conductivity of filled and un-filled membranes is similar
- MEA performance with filled and unfilled membranes is similar





TECHNICAL ACCOMPLISHMENTS
MEMBRANE MECHANICAL PROPERTIES (TASK 2)

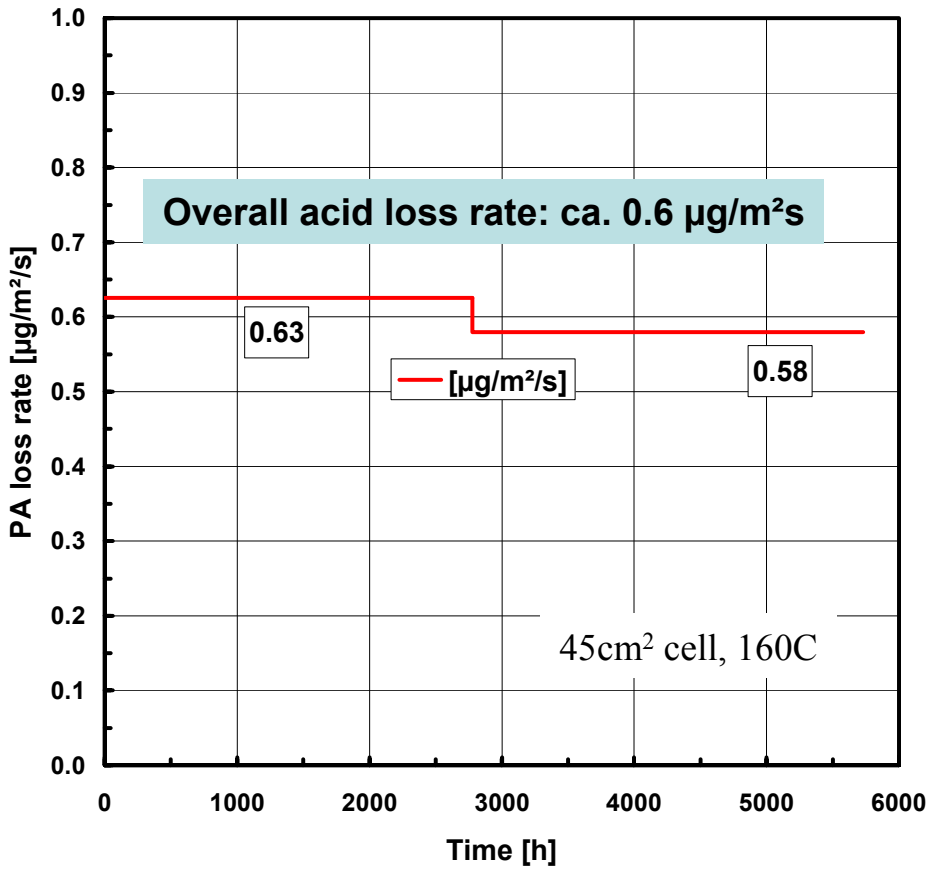
- Developed four mechanical test fixture to simulate fuel cell stack load conditions including temperature and humidity control
- Unexpectedly, the type 8 & 9 filled membranes showed a higher rate of stress relaxation than the unfilled membrane at 160°C, on different fixtures
- Further testing is underway to validate the effect with other fixtures
- Contact resistance needs to be investigated
- A mechanical model is under development at RPI to enable projection of 40,000 hour life



TECHNICAL ACCOMPLISHMENTS
ACID MANAGEMENT (TASK 9)

- Identified the factors that influence phosphoric acid loss:
  - Temperature
  - Reactant flow rates
  - Reactant water content
- Steady state phosphoric acid evaporation losses were studied on the types 1, 2, & 5 membrane and found to be less than theoretical.
- Acid loss remains unchanged during controlled startups and shutdowns.

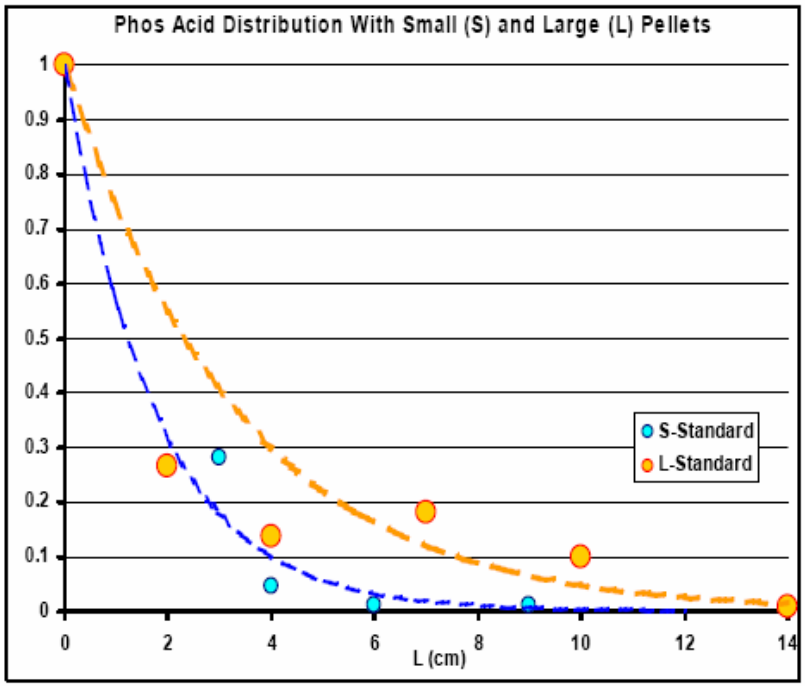
Acid Loss Rate Measurements



Projected cell life is 40,000 hours measured loss rate

TECHNICAL ACCOMPLISHMENTS
ACID MANAGEMENT (TASK 9)

- **Acid loss occurs through**
  - Diffusion
  - Capillary transport
  - Compression
  - Evaporation



- **Acid Loss Calculation**
  - Measured rate 0.6 μg/ m^{2}sec
  - At 160°C and reactant flows for 0.2 Amps/ cm^2
  - A full size 5kw stack starts with 2100 g of acid
  - Acid loss per year 83 g
  - 4% loss/ year if running constant
  - Sufficient acid is available for 40,000 hours

- **Acid Management Requirements**
  - Must prevent contamination of other system components
  - Must have low service interval
  - Must have low service cost

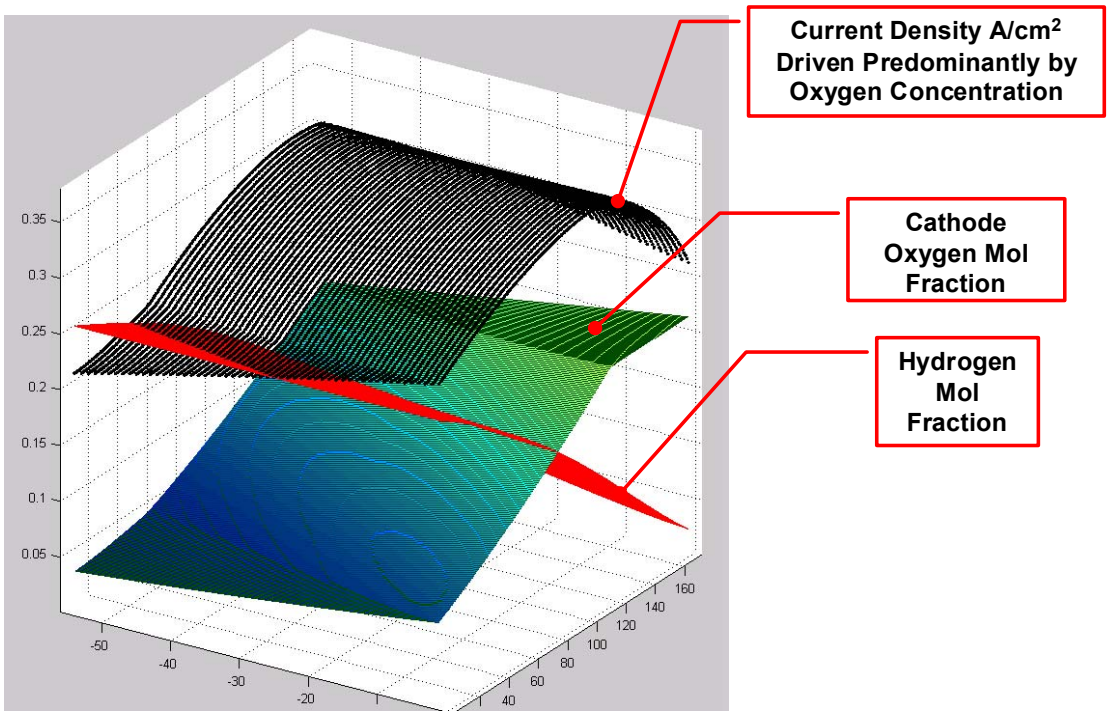
- **Acid Management Solution**
  - Stack exhaust (anode and cathode) are passed through a pelletized bed
  - Sizing studies are underway to select best size.
  - University of South Carolina engaged to develop an acid transport model

TECHNICAL ACCOMPLISHMENTS
PBI SPECIFIC FLOW FIELD DESIGN (TASK 10)

- Initiated work with Entegris for stack sealing technology development
- Modeled full size flow field to improve system efficiency
- Worked with National Renewable Energy Lab (NREL) on optimizing coolant flow

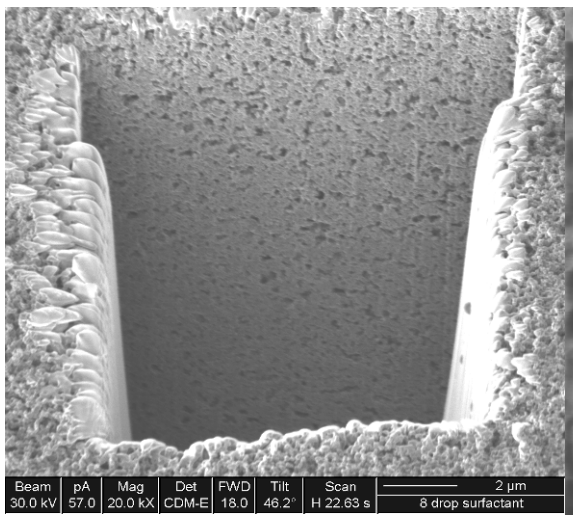
Flow field study

- 8 cathode / anode flow configuration modeled
- Co-flow vs. counter-flow and single vs. multi-pass high power vs. low power evaluated
- 2 configurations with the lowest variability and best mean performance will be built and tested to confirm model results

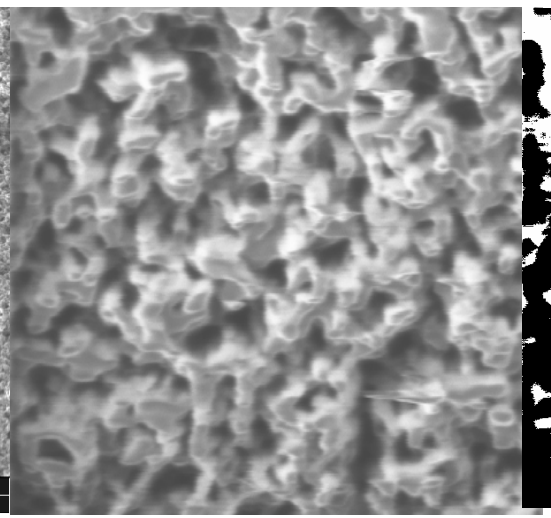


TECHNICAL ACCOMPLISHMENTS ELECTRODE (TASK 11)

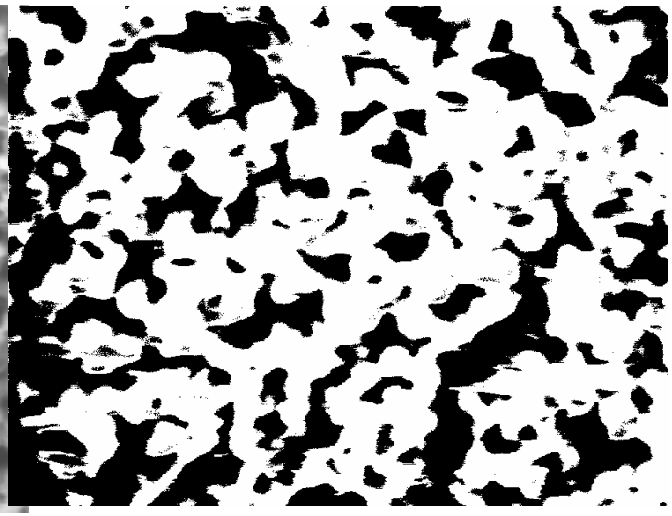
- Albany Nanotech subcontractor for nano-scale electrode development
- Investigated methods for measuring catalyst layer properties
  - Focus Ion Beam (FIB) used to look at pore structure and “connectedness” of pores below the electrode surface
- Investigating methods for creating low Pt loaded catalyst layers such as Pt sputter deposition
- RPI Fuel Cell Center added to program to explore alternate catalyst formulations to improve performance



FIB Trench



Original image



Black and white image model

TECHNICAL TARGETS- STATIONARY

Characteristics	Units	Calendar Year		
		2004 status	2005	2010
Membrane conductivity, operating temperature	Ohm-cm^2	0.1	0.1	0.1
Oxygen Crossover	mA/cm^2	5	5	2
Hydrogen Crossover	mA/cm^2	5	5	2
Cost	$/ kW		50	5
Operating temperature	^{o}C	160	160	170
Durability	Hours	5,000	>15,000	>40,000
Survivability	^{o}C	-20	-30	-40
Applicable to both stationary and automotive		Yes		
Operating Pressure	atm	0-3		

- **Conductivity:** Meets DOE target at 160°C
- **Cross over:** Need to confirm with final membrane
- **Cost:** PEMEAS to provide cost estimate in 2006.
- **Temperature:** Routinely run at 180°C
- **Durability:**14,000 hours demonstrated by PEMEAS in 50cm^2 testing
- **Survivability:** Data available from PEMEAS

RESPONSES TO REVIEWERS' COMMENTS

- **" Application to stationary is clear- not sure how this can be used in automotive, specifically… freeze and thaw cycle"**
  - In August 2004, DOE modified program objective to focus on stationary applications
  - Limited freeze thaw data is available from PEMEAS
- **"… PBI has serious acid retention and mechanical stability", "... recommend speaking/ working with a PAFC developer who can help with acid management, flow field design, cathode catalyst …"**
  - Acid retention is not the issue, acid management is the issue- our solution is at this review. A PAFC consultant helped Plug Power and PEMEAS understand acid loss
  - Agree that mechanical stability is an issue and our focus has been around the membrane mechanical properties for the past 9 months
- **"… an electrode structure that will allow performance greater than state of the art materials eg. Nafion"**
  - We are adding a major program initiative to further address the electrode structure
  - Stack size is not critical for stationary applications, however cost is
  - PBI membranes are projected to be lower cost that Nafion event though the polar curve is lower

FUTURE WORK

Remainder 2005:

- Complete filled membrane characterization
- Select primary membrane for scale-up at RPI
- Build small scale prototypes and demonstrate stack sealing concept with Entegris
- Investigate failure modes associated with starts/stops and load cycling
- Test full-size flow field and quantify efficiency improvement
- ANT will build nano-scale electrodes for Plug Power test and quantify for Pt loading reduction & performance improvement

2006:

- Build and test a full size module with improved membrane, flow field and sealing concept
- PEMEAS will deliver price estimate for MEA
- Demonstrate 1,000 hours life with low degradation rate and project 40,000 hours life

PUBLICATIONS AND PRESENTATIONS

Publications (RPI-3)

- Synthesis and Characterization of Pyridine-Based Polybenzimidazoles for High Temperature Polymer Electrolyte Membrane Fuel Cell Applications. Xiao, L.; Zhang, H.; Jana, T.; Scanlon, E.; Chen, R.; Choe, E.-W.; Ramanathan, L.S.; Yu, S.; Benicewicz, B.C. *Fuel Cells*, **2005**, xxxx.
- Synthesis, Characterization and Fuel Cell Performance of Poly(2,2'-(p-phenylene)-5,5'-bibenzimidazole) as a High Temperature Fuel Cell Membrane. Zhang, H.; Chen, R.; Ramanathan, L.S.; Scanlon, E.; Xiao, L.; Choe, E.-W.; Benicewicz, B.C. *Fuel Division Prepr.* **2004**, *49(2)*, 588-589.
- Polybenzimidazole Based Segmented Block Copolymers for High Temperature Fuel Cell Membranes. Scanlon, E.; Benicewicz, B.C. *Fuel Division Prepr.* **2004**, *49(2)*, 522-523.

Presentations/ Posters (RPI-20)

- Polybenzimidazole Based Polymers for High Temperature Fuel Cell Membranes. MACRO 2004, Paris, France, 7/6/04.
- Polybenzimidazole/Phosphoric Acid Polymer Gel Electrolytes for Fuel Cells, Novel Polbenzimidazole Membranes for High Temperature PEM Fuel Cells, Segmented Block Copolymers of Para-PBI and AB-PBI for Fuel Cell Membranes, Segmented Copolymers of Polybenzimidazole (PBI) for High Temperature Fuel Cells, Morphological Studies of PBI Membranes Made by the PPA Process. Gordon Conference on Fuel Cells, 7/25-29/04.
- Synthesis, Characterization and Fuel Cell Performance of Poly(2,2'-(p-phenylene)-5,5'-bibenzimidazole) as a High Temperature Fuel Cell Membrane, Polybenzimidazole Based Segmented Block Copolymers for High Temperature Fuel Cell Membranes. ACS Meeting, Fuel Division, Philadelphia, PA, 8/22/04.
- Advances in Polybenzimidazole Synthesis and Applications to Fuel Cells. Polycondensation 2004, Roanoke, VA, 9/28/04.
- NMR Studies of Mass Transport in High Acid Content Fuel Cell Membranes Based on PBI/Phosphoric Acid. 206th Electrochemical Society Meeting, 10/7/04.
- Polybenzimidazole Polymers for High Temperature Fuel Cells. Engelhard Corporation, Iselin, NJ, 1/11/05.
- Quantitative Investigation of Electrolyte Loss in Polymer Electrolyte Fuel Cell Membranes , Segmented Block Copolymers of Polybenzimidazole for High Temperature Fuel Cell Membranes, Segmented Block Copolymers of p-PBI and AB-PBI for Fuel Cell Membranes, Electrolyte Concentration Effects in Polybenzimidazole Fuel Cell Membranes. The Fifth New England Polymer Workshop, RPI, Troy, NY, 1/15/05.
- Electrolyte Concentration Effects in Polybenzimidazole Fuel Cell Membranes. Eastern NY ACS Undergraduate Research Symposium and 6th Robert A. Laudise Symposium, 2/5/05.
- Electrolyte Concentration Effects in Polybenzimidazole Fuel Cell Membranes. Advances in Materials for Proton Exchange Membrane Fuel Cell Systems 2005, 2/21/05.
- Quantification of Electrolyte Loss in a Phosphoric Acid/Polybenzimidazole Membrane Fuel Cell. Advances in Materials for Proton Exchange Membrane Fuel Cell Systems 2005, 2/21/05.
- Polybenzimidazole Segmented Block Copolymer Membranes for High Temperature Fuel Cells. Advances in Materials for Proton Exchange Membrane Fuel Cell Systems 2005, 2/21/05.
- Advances in Polybenzimidazole Synthesis and Applications to Fuel Cells. Advances in Materials for Proton Exchange Membrane Fuel Cell Systems 2005, 2/21/05.

HYDROGEN SAFETY

- The most significant hydrogen hazard associated with this project is:

  Hydrogen service to the lab space in order to perform the testing introduces hazards of pressurized gases, hydrogen leaks, which could serve as an ignitions source.

- Our approach to deal with this hazard is:
  - Annual safety training for every employee including hazard communication and general lab safety.
  - Lab design is expected to safe environment (100% outside ventilation, hydrogen sensors set at 18% and 25% LEL)
  - Safety reviews for all equipment prior to start-up

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