Center for Automation Technologies and Systems

Adaptive Process Controls and Ultrasonics for High Temperature PEM MEA Manufacture

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MN005

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Overview

Timeline

- Project start date: 9/01/08
- Project end date: 6/30/12
- Percent complete: 35%

Budget

- Total project funding: $2,479,908
  - DOE share: $1,611,129
  - Contractor share: $868,779
- Funding received in FY09: $400,000
- Funding for FY10: $400,000

Barriers Addressed

- A. Lack of High-Volume Membrane Electrode Assembly (MEA)
- F. Low Levels of Quality Control and Inflexible Processes

Partners

- RPI CATS- Project Lead
- ASU- Subcontractor
- BASF Fuel Cell- Collaborator
- PMD- Collaborator
- UltraCell- Collaborator
- NREL- Collaborator

Relevance (1)
Situation and Objectives

- **Situation: In spite of the fact that there are variations in MEA component material properties, we use the same manufacturing process parameters. This results in variations in MEA properties and performance, and the potential for stack failures and re-work, and reduced durability.**
- **We need to develop a deeper understanding of the relationships among MEA material properties, manufacturing processes parameters, and MEA performance (3Ps).**
- **The high level objective of the proposed work is to enable cost effective, high volume manufacture of high temperature (160-180^{o}C) PEM MEAs by:**

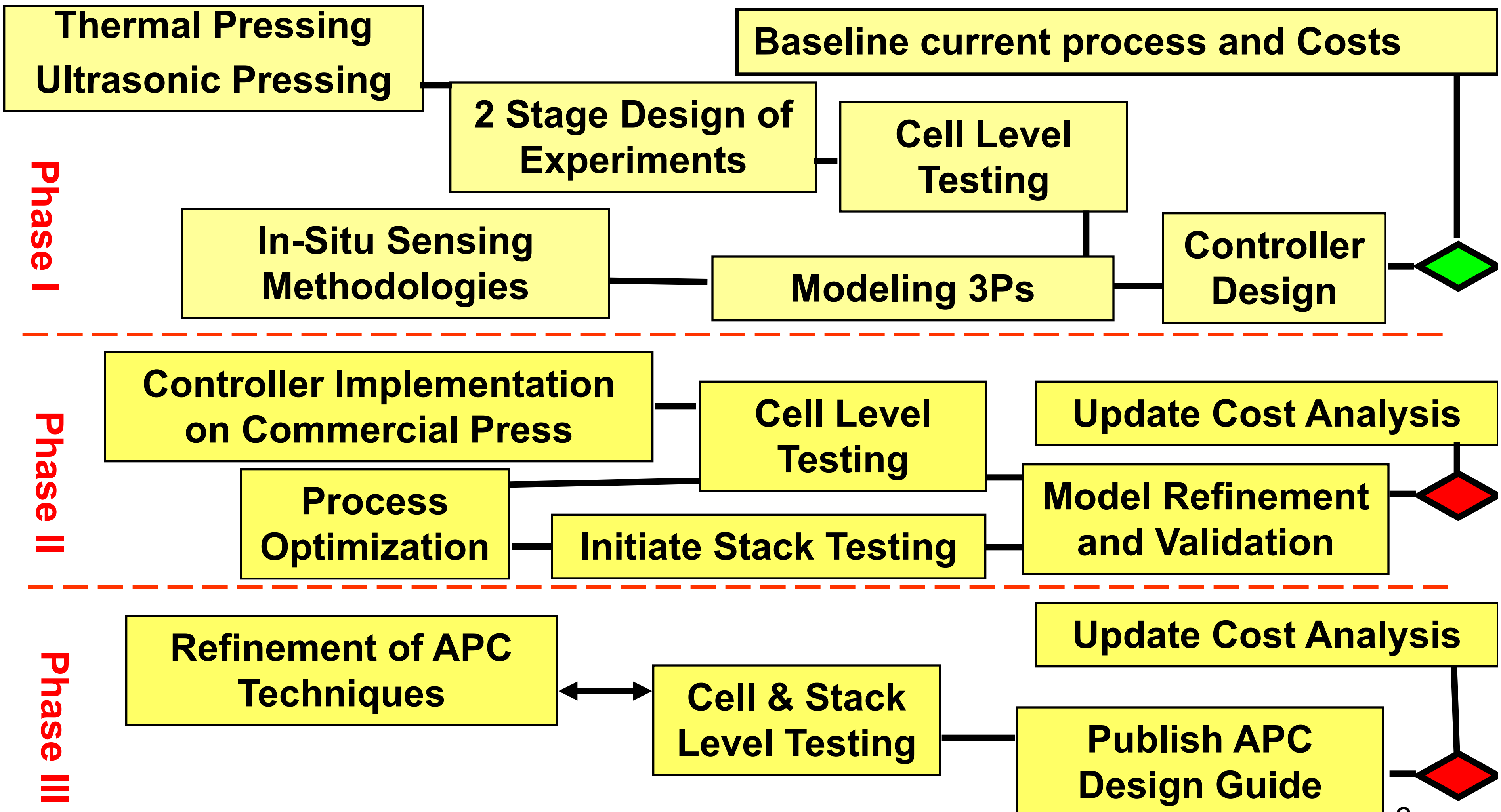
Relevance (2)
Situation and Objectives

- **(1) achieving greater uniformity and performance of high-temperature MEAs by the application of adaptive real-time process controls (APC) combined with effective in-situ property sensing to the MEA pressing process.**
  - **This objective addresses Barrier F, Low Levels of Quality Control and Inflexible Processes**
- **(2) greatly reducing MEA pressing cycle time through the development of novel, robust ultrasonic (U/S) bonding processes for high temperature (160-180^{o}C) PEM MEAs.**
  - **This objective addresses Barrier A, Lack of High-Volume Membrane Electrode Assembly (MEA) Production**
- **This year we will focus on process optimization, initial APC implementation, stack testing, and LT U/S tests.**

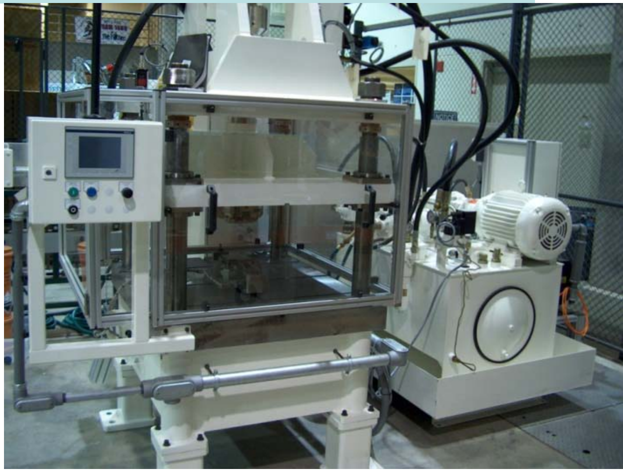
Summary of 2009 Review

- **Designed experiments for high temperature (HT) U/S welding and sealing, and thermal sealing to identify critical process parameters.**
- **Ultrasonic welding of HT MEA components stronger than thermal welds, with a cycle time less than 1 sec, and more than 95% energy savings.**
- **Promising initial results of ultrasonic sealing experiments for HT MEAs.**
- **Promising initial results of U/S sealing of LT MEAs.**

Technical Approach (1)
Project Plan

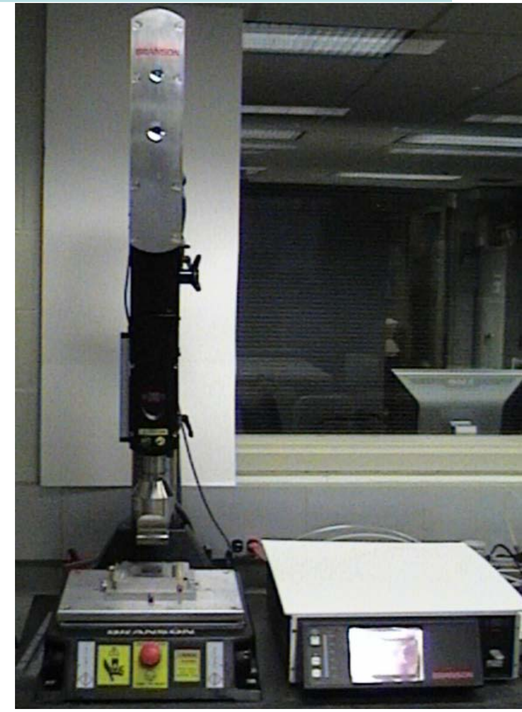


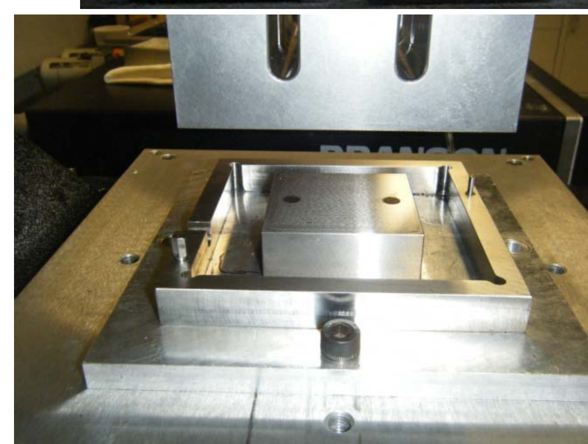
Technical Approach (2)
Phase II APC



- Design and construction of new commercial thermal press tooling to incorporate sensor(s) and electrically isolate tooling.
- Modification of press controls for real time APC using AC impedance.
- Conduct designed experiments with range of GDE and membrane material properties.
- Evaluation of APC MEA performance compared to baseline MEAs.
- Refinement of process models and control algorithms.
- Initiate stack level testing.
- Investigate APC for low temperature MEAs

Technical Approach (3)
Phase II Ultrasonic Sealing





- Ultrasonic sealing process optimization via designed experiments.
- Durability testing of U/S sealed MEAs.
- Experimentation on U/S sealing of large size MEAs.
  - Requires custom tooling and significant press re-design.
- Investigate in-situ sensing techniques for APC, compatible with process cycle time.
- Conduct designed experiments for use of U/S for sealing of low temperature MEAs.
- Stack level testing of U/S sealed MEAs compared to baseline thermal pressed MEAs.

Technical Approach (4)
Additional Phase II Tasks

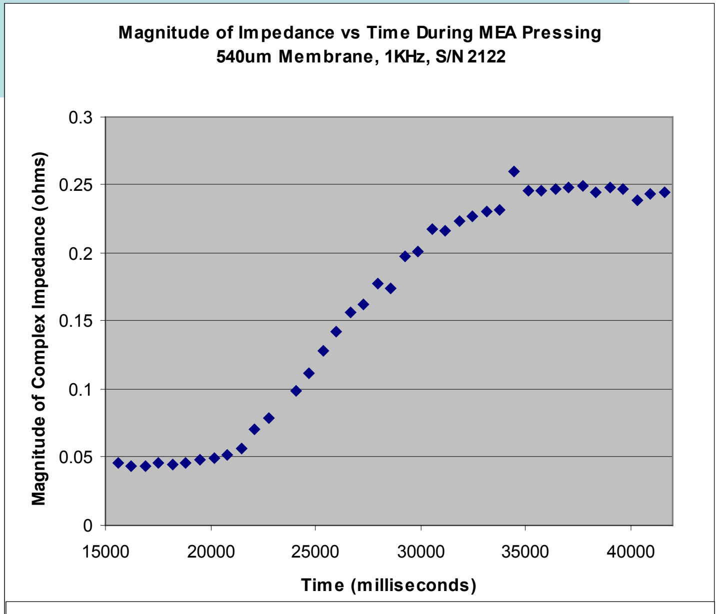
- Heat treat process optimization for both thermal and U/S sealed MEAs.
- Continue modeling efforts of relationships among material properties, manufacturing process parameters, and MEA performance.
- Manufacturing cost analysis update.

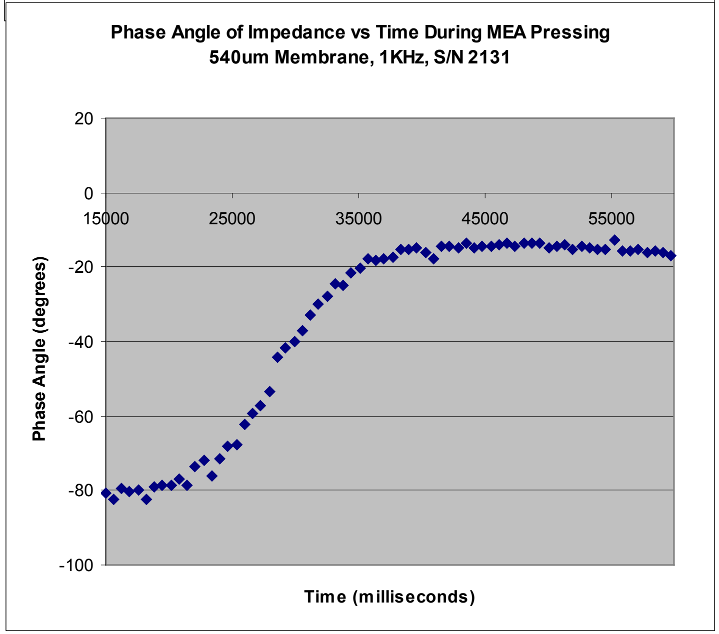
Approach/Milestones

Month/Year	Milestone or Go/No-Go decision
November, 2009	Phase I Go/No-Go Decision will be based on and initial cost analysis showing substantial reductions in PBI type MEA manufacturing costs based on the ultrasonic sealing/welding and/or in-situ adaptive process controls. Note: A Go decision was made by DOE to move into Phase II.
June, 2011	Phase II Milestone: Demonstrate the ability of APC and Ultrasonics to improve the performance and uniformity of MEAs. Go/No-Go Decision: Ability to meet target cost reductions.
June, 2012	Phase III Milestone: Analysis of benefits of APC and ultrasonics. Validation of cost analysis. Target for improvement to MEA durability is 15%, target reduction of MEA manufacturing cost for pressing is 25% for the use of APC with thermal pressing and 75% for U/S sealing.

Technical Accomplishments (1) APC

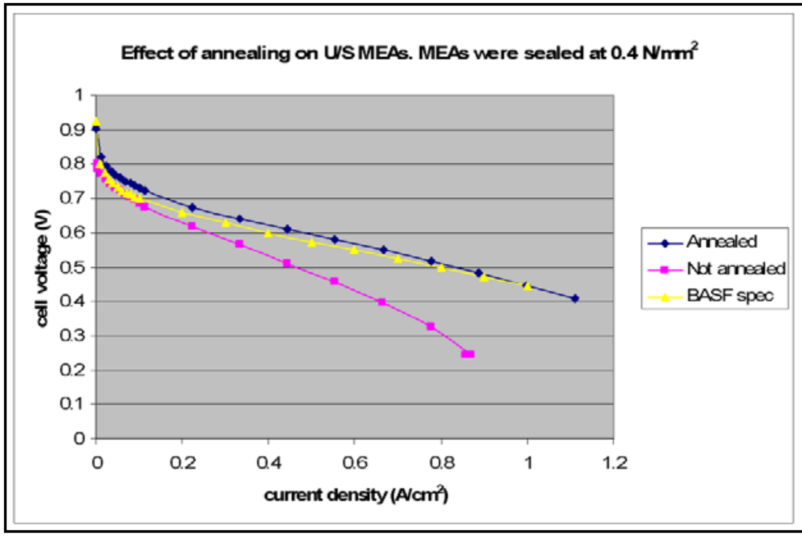
- **Down stream process benefits of APC.**
- **Potential to improve MEA uniformity and reduce process cycle time.**
- **Potential use as screening tool prior to stack assembly.**
- **Promising initial results from in-situ AC impedance measurement.**
- **Phase angle correlates to MEA performance, while impedance did not.**
- **Performed "man-in-the-loop" feedback control on commercial press.**
- **Resulting MEAs exceeded spec.**
- **Less activation loss than baseline MEAs.**
- **Greater than 50% cycle time reduction.**
- **APC concept may also be viable for LT MEAs.**

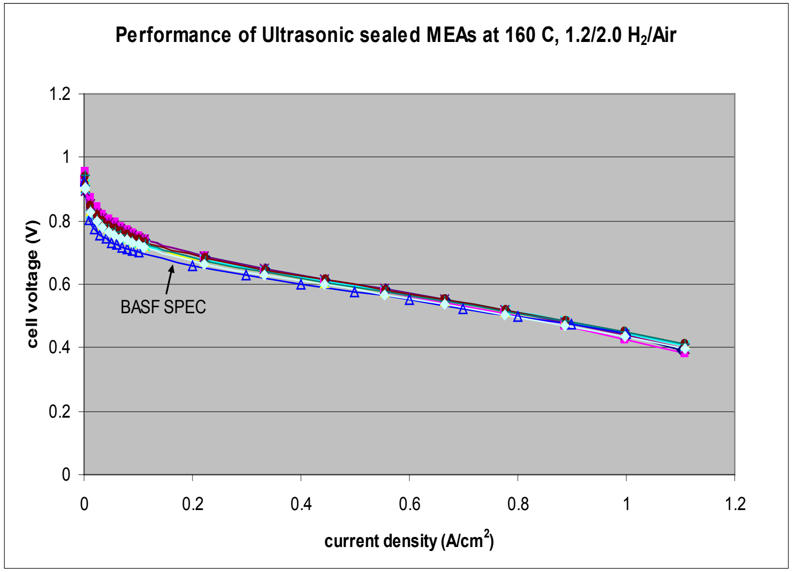




Technical Accomplishments (3) Ultrasonic Sealing

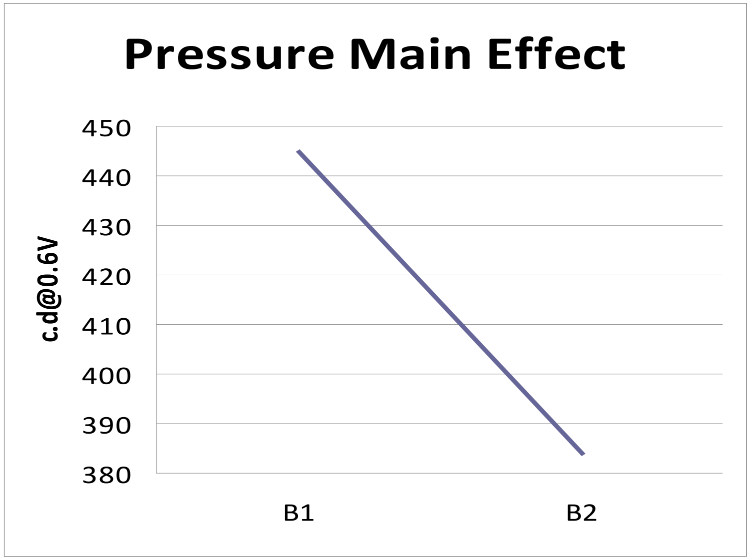
- **U/S sealing/welding has potential for significant cycle time reduction and energy savings.**
- **Two stage full factorial designed experiments completed**
- **Post-seal heat treat of MEAs was identified as dominate factor**
- **Performance of ultrasonically sealed MEAs was equal to or better than BASF MEA specifications (thermally sealed)**
- **Activation region performance improvement may be significant.**
- **Greater slope in ohmic region confirmed as the result of higher test hardware resistance.**
- **>90% cycle time reduction**
- **>95% energy savings**
- **Should also work for LT MEAs.**

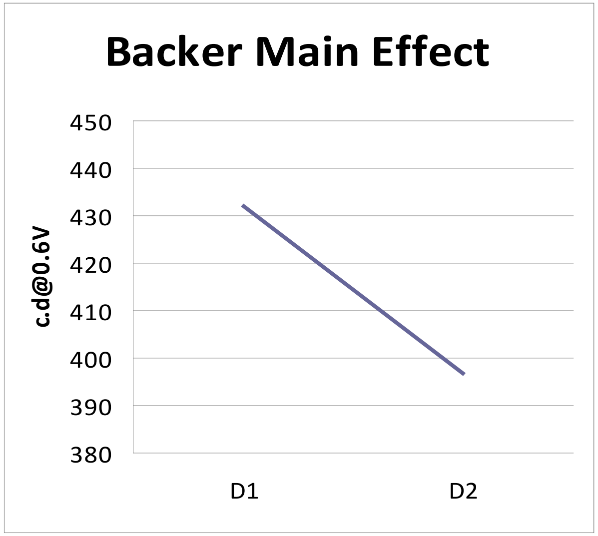




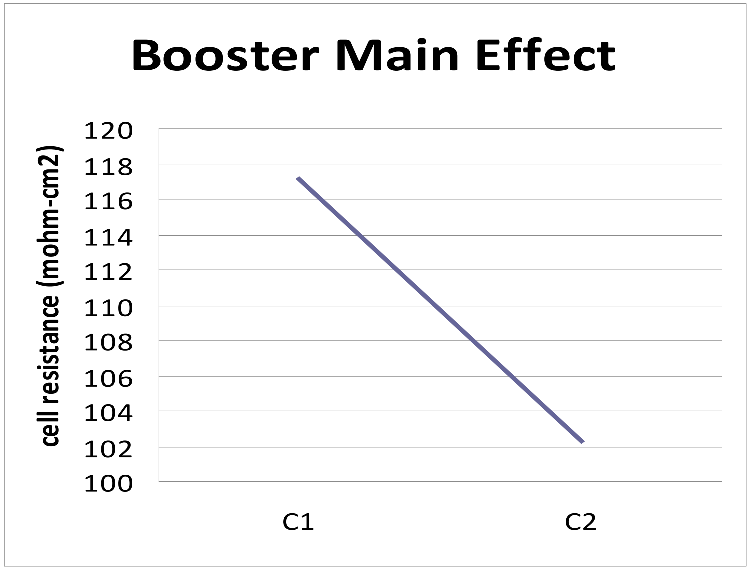
Technical Accomplishments (4)
Ultrasonic Sealing- Main Effects

- **Higher sealing pressure can result in destruction of GDE pores inhibiting gas and acid transport. This effect is also observed for current density at 0.4 V.**





- **Lower support stiffness i.e. more compliant support produces better MEA contact resulting in better performance compared to a stiff backer.**



- **Higher booster amplitude means more displacement, resulting in better MEA contact and low cell resistance**

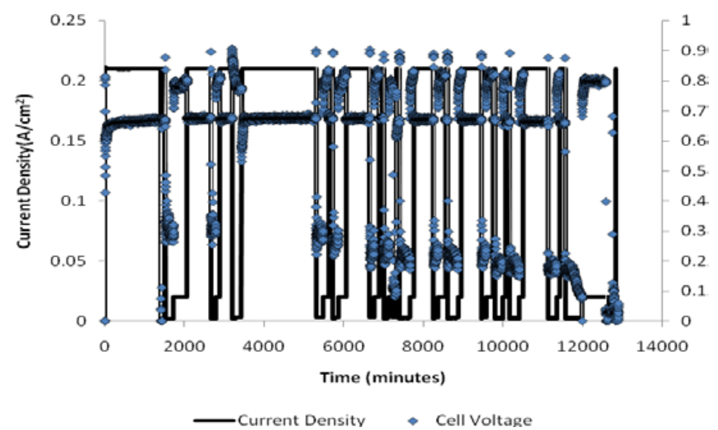
All results are "good news"

Technical Accomplishments (5) Ultrasonic Sealing – MEA Durability

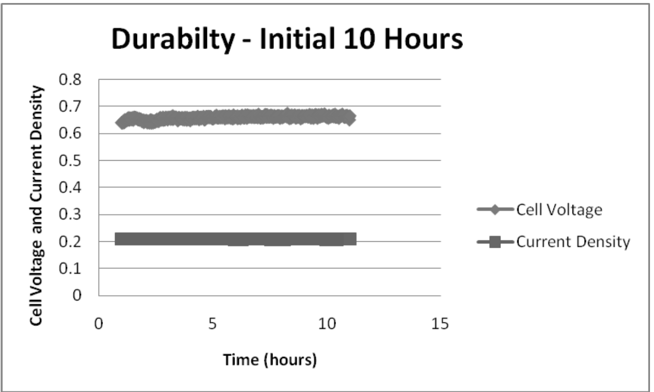
- 2 U/C sealed MEAs (not optimized) were cycled for 190 hours

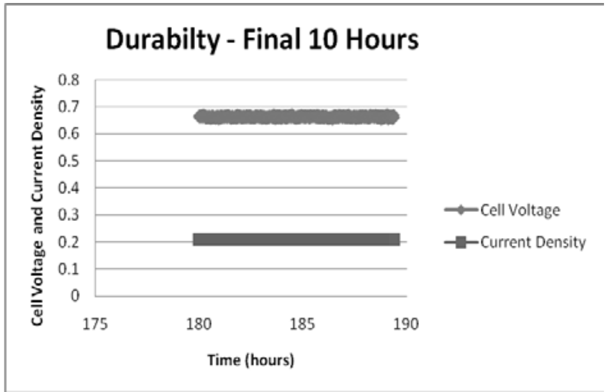
Anode Gas Flow (sccm)	Cathode Gas Flow (sccm)	Temperature (°C)	Load (A/cm²)	Time (minutes)
110	435	160	0.2	60
50	83	160	0.0	2
0	0	55	0.0	2
50	83	120	0.01	2
50	83	160	0.01	2
110	435	160	0.2	120

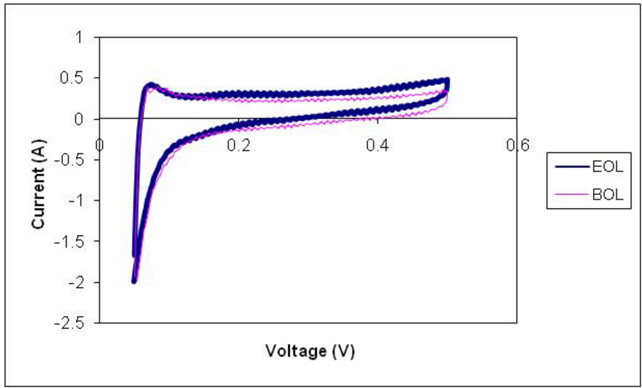
Cell durability cycling per BASF protocol



- No measureable degradation observed over life of tests
- Additional testing to be performed on optimized U/S MEAs



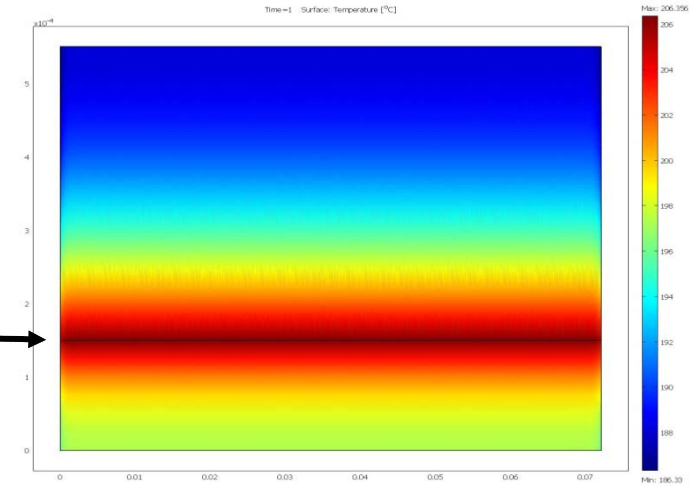




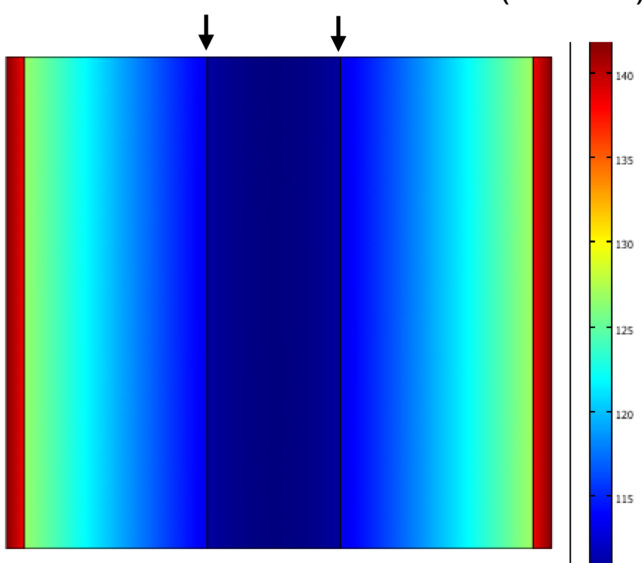
Technical Accomplishments (6) Modeling

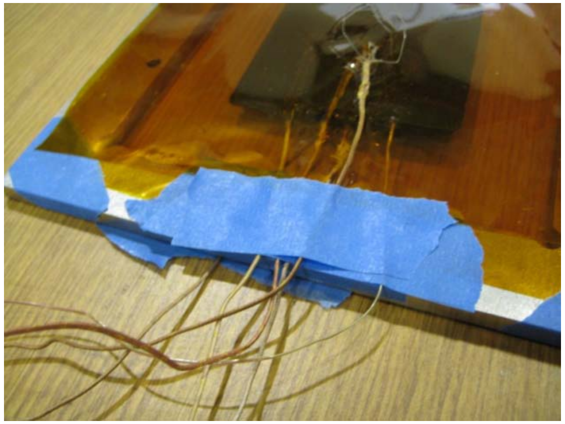
- **COMSOL thermal model of U/S sealing process**
- **COMSOL thermal model of thermal sealing process**
- **MEA compression models for both thermal and U/S sealing**
- **Validation using in-situ temperature measurements**
- **U/S heats from inside out, while thermal heats from outside in (thus longer to reach critical temperature).**

Membrane -electrode interface (U/S)



Membrane-electrode interface (thermal)





Instrumented MEA used for model validation

Technical Accomplishments (7)
Phase I Manufacturing Cost Analysis

- **Factors considered include: capital depreciation; tooling; labor; electricity; chilled water; HVAC; maintenance; space; waste disposal cost**
- **Component materials were not included in analysis**
- **Assumptions:**
  - **Baseline case is current BASF Fuel Cell process/system**
  - **Production system will be located in the U.S.**
  - **Current utilities costs**
  - **500,000 automotive stacks with 400 cells each, 80KW**
  - **2/8/5/50 operation of production facility**
  - **Phase I analysis only addresses sealing process**
- **Our results are conservative: 38% cost reduction for APC, and 84% cost reduction for U/S sealing**

Technical Accomplishments (8) Phase I Manufacturing Cost Analysis

- **Although APC will have significant manufacturing cost benefits the greatest benefit may be in the downstream stack assembly process.**
- **We have found ultrasonic sealing to be a very robust process with the potential for a major reduction in manufacturing costs.**
- **Ultrasonic welding will result in a similar cost reduction to that for ultrasonic sealing.**

Collaborations

- **Sub-contractor**
  - **Arizona State University** (Academic): application of electrochemical impedance spectroscopy (EIS), without reactant gasses, for process control.
- **Partners**
  - **BASF Fuel Cell** (Industry): HT PEM MEA expertise (materials, electrochemistry, operations, performance).

Collaborations (2)

- **Partners**
  - **Progressive Machine and Design** (Industry): expertise in industrial controls and MEA manufacturing systems design.
  - **UltraCell** (Industry): fuel cell system manufacturer, expertise on stack performance, evaluate stack performance with mEAs produced with APC.
  - **National Renewable Energy Laboratory** (Government Lab): low temperature MEA testing, independent validation of high temperature test results.

Proposed Future Work (Phase II)

- **U/S sealing process optimization**
- **U/S sealing of larger MEAs**
- **U/S MEA durability testing**
- **LT MEA U/S sealing designed experiments**
- **Initial implementation of APC on commercial thermal press**
- **Extensive designed experiments**
- **MEA performance evaluation (single cell)**
- **Model refinement and validation**
- **Initiate stack level testing**
- **Update manufacturing cost analysis**
- **Phase II program review**

Proposed Future Work (Phase III)

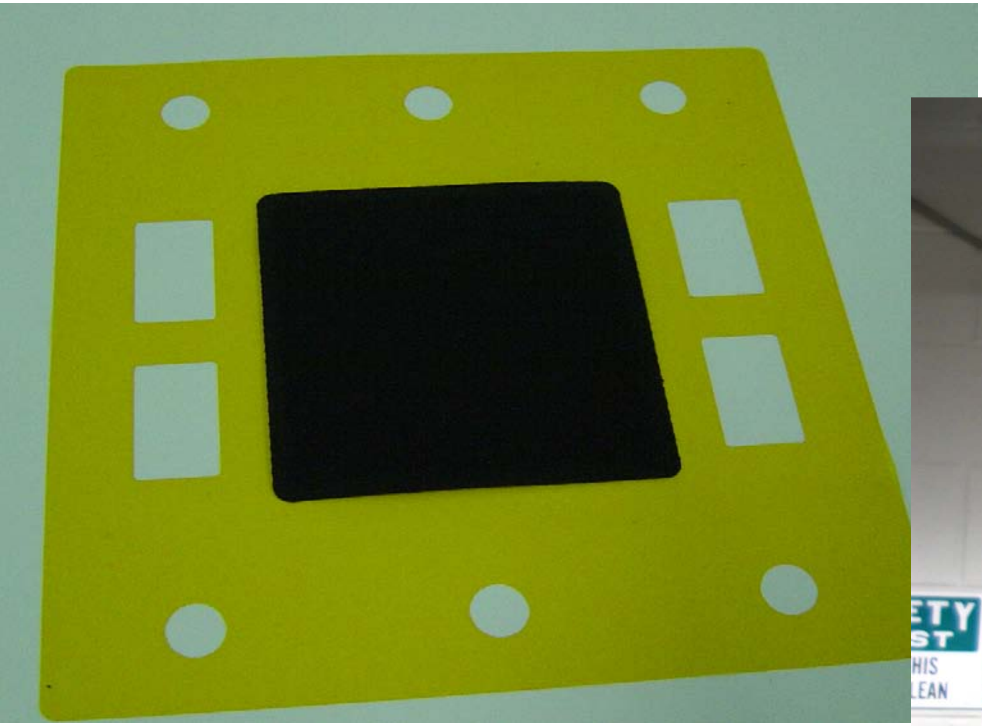
- **Refine APC techniques**
- **Model refinement**
- **APC evaluation, single cell and short stacks**
- **Develop design guidelines based on lessons learned**
- **Update manufacturing cost analysis**
- **Phase III program review.**

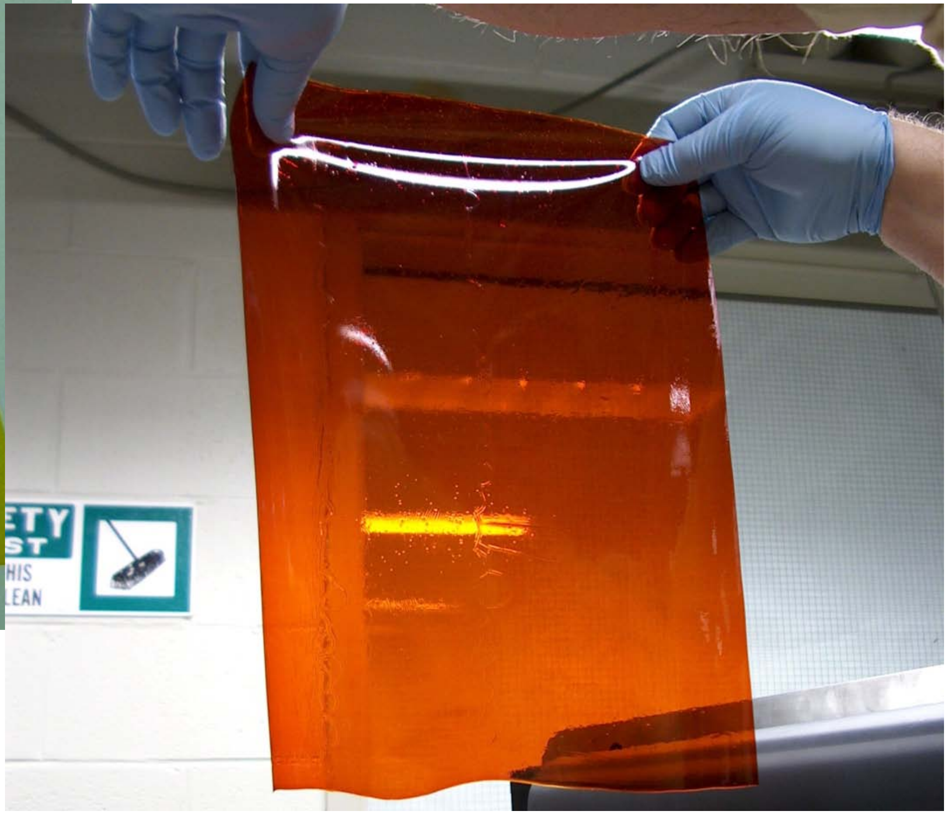
Project Summary

- **Relevance:** The proposed research addresses two critical barriers.
  - The critical need for high volume MEA manufacturing processes, and
  - The need for QC methods and process flexibility.
  - Additional ultrasonic sealing investigations for low temperature MEAs
- **Approach:**
  - Develop and apply adaptive, real time, process controls to improve performance and uniformity of HT PEM MEAs
  - Novel ultrasonic bonding methods to achieve significant productivity increases
- **Collaborations:** Strong team of RPI, ASU, BASF Fuel Cell, PMD, UltraCell and NREL with expertise in all critical elements of HT PEM fuel cell technologies.
- **Technical Accomplishments/status:** Demonstrated benefits of U/S sealing; modeling of processes; encouraging early APC results; significant cost savings projected.
- **Proposed Future Research:** Continue development of process and control models, U/S process optimization; implement and validate APC via cell and stack testing; durability testing; larger size MEAs; update cost models; LT U/S sealing investigation.

Supplemental Slides

Typical HT PEM MEA Design & PBI Membrane





BASF
The Chemical Company

Details of Phase I Manufacturing Cost Analysis

Cost Element	Current Technology	APC	Ultrasonics
Capital Depreciation	$.0896	$.0517	$.0055
Tooling	$.0521	$.0463	$.0416
Labor	$.0386	$.0223	$.0062
Electricity	$.0579	$.0334	$.0001
Chilled Water	$.0293	$.0169	$.0000
HVAC	$.0009	$.0007	$.0000
Maintenance	$.0121	$.0070	$.0008
Space	$.0041	$.0024	$.0003
Disposal	$.0896	$.0517	$.0066
Cost per MEA	**$.3741**	**$.2324**	**$.0610**
Cost per KW	**$1.4965**	**$.9295**	**$0.2440**
Percent Reduction	**--**	**37.89%**	**83.70%**

Summary of Modeling Efforts

Modeling of Thermal and Ultrasonically Sealed MEAs, Individually and in Stacks

- Identified critical sealing process output variables that affect MEA performance including through-thickness temperature distribution, amount of phosphoric acid (PA) permeating GDE during pressing and the amount of water boiled off during the sealing cycle.
- Developed 2-D, transient, thermo-mechanical FEA models (Comsol) of the MEA component stackup (anode GDE, membrane, cathode GDE) during thermal and ultrasonic sealing to predict critical temperatures throughout MEA thickness.

- Measured key thermal properties of GDE and membrane including thermal conductivity (*k*) and specific heat (c_s) using Differential Scanning Calorimetry (DSC) and a device that measures 1-D steady-state thermal conductivity, respectively.
- Measured compressive modulus of MEA components (GDE, membrane) and the assembled MEA using a low-force Instron
- Validated thermal models of thermal and ultrasonic sealing by measuring transient temperature at all critical MEA interfaces
- Developed porosity and permeability models and experiments for describing behavior of acid (PA) squeezed from membrane into GDEs during compression.

- Proposed analytical material model (coupled Kelvin-Voigt and Maxwell) to describe how internal heat generation is distributed through the MEA thickness during ultrasonic sealing as a function of time and vibrational amplitude and frequency.
- Proposed mechanical model of FC stack to explain MEA performance variation as a function of MEA mechanical properties, MEA thickness and the amount of compression.

Proposed Future Work (Phase II) - Modeling

- Perform more through-thickness temperature measurement experiments for both thermal and ultrasonic sealing
- Further refine thermomechanical models of thermal and ultrasonic sealing based on experimental data
- Measure thermal properties of PA-soaked GDEs
- Measure GDE porosity and permeability properties for PA flow
- Perform variable compression experiments on single MEAs for thermal and ultrasonic sealing to validate porosity and permeability models. The goal is to accurately predict PA content in the pressed MEA.

- Measure visco-elastic, elastic and damping properties of GDE and membrane to use in material models to predict internal heat generation during ultrasonic sealing
- Develop an MEA/bipolar plate structural model to predict the effect of MEA compression on reactant gas flow through the flow field, and run experiments using the 10-cell stack with variable compression to validate the model.

- RPI Modeling/Experimental Approach
  - Create a structural FEA model of an MEA unit cell (single flow channel and half MEA from centerline) using previously measured GDE and membrane properties
  - Accurately measure deformation of cross section of GDE compressed between two bipolar plates for various compression levels using Micro Vu Optical Measuring Machine (single micron accuracy)
  - Calculate reduction in flow due to the change in cross-sectional shape of the flow channel using simple analysis or CFD.
  - Measure the pressure drop for various reactant gas flowrates (controlled by FC test stand) with a single MEA subjected to different compression levels at RT and at operating temperature (160°C). Correlate this data with analytical model.
  - Measure pressure drop in an entire 10-cell FC stack with varying levels of stack compression and compare data to model predictions.