Center for Automation Technologies and Systems

Adaptive Process Controls and Ultrasonics for High Temperature PEM MEA Manufacture

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Overview

Timeline

- Project start date: 9/01/08
- Project end date: 2/28/12
- Percent complete: 15%

Budget

- Total project funding: $2,479,908
  - DOE share: $1,611,129
  - Contractor share: $868,779
- Funding received in FY08: $743,027
- Funding for FY09: $0

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

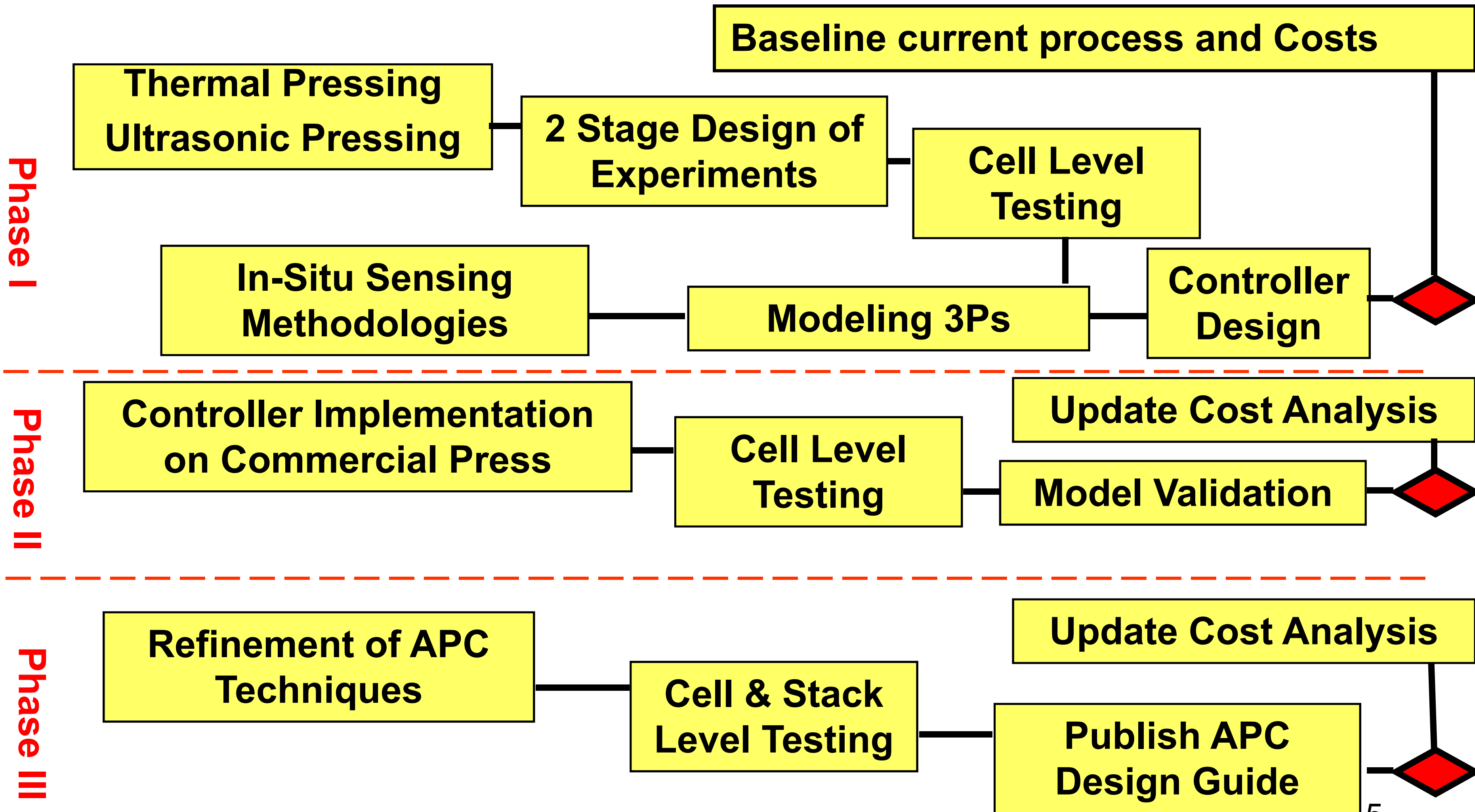
Relevance (1)
Situation and Objectives

- **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 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**

Technical Approach (1)
Adaptive Real-Time Process Controls (APC)



Technical Approach (2)

Phase I Design of Experiments

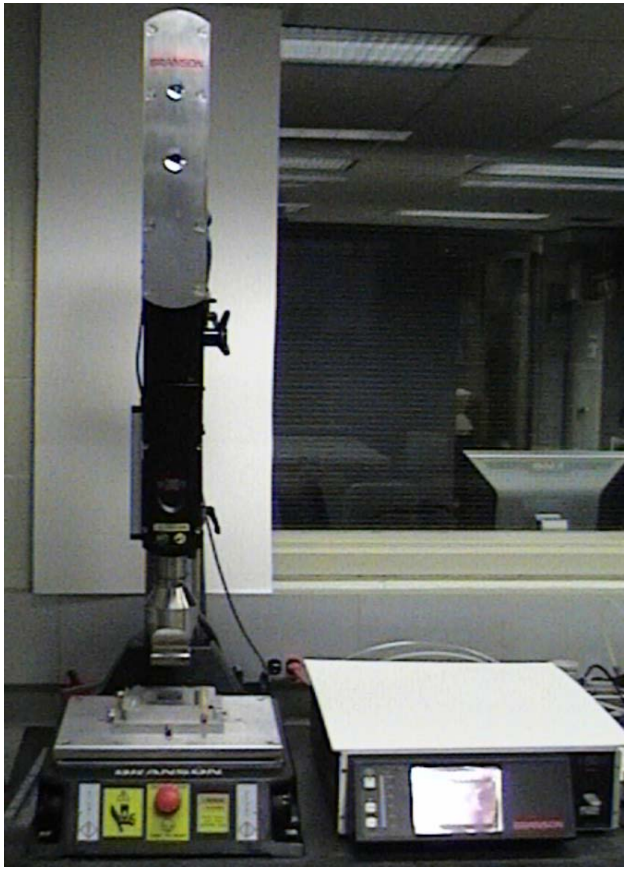
- 2^k factorial set of designed experiments (2 replicates) to determine main effects and interactions on cell performance based on ANOVA
  - $2^4 \times 2 = 32$ experiments for thermal pressing (temperature, pressure, time, annealing)
  - $2^5 \times 2 = 64$ experiments for ultrasonic pressing (energy flux, pressure, vibration amplitude, backer hardness, annealing)
- 3^k factorial set of designed experiments (2 replicates) with reduced number of factors (e.g., 2 or 3) based on main effects and interactions analysis from 2^k data. Data will be used for response surface modeling.
- Single factor experiments with >3 levels where appropriate
- Experiments involving changes in assembly process factors (e.g., cell performance vs. amount of MEA compression)

Technical Approach (3)
Modeling of Thermal and Ultrasonic MEA Sealing Processes

- Modification of existing 1-D transient (1) analytical and/or computational (e.g., FEA) models for ultrasonic pressing and (2) analytical models for thermal pressing that include thermal and structural effects to predict in-process temperature and post-process compressive stiffness through the MEA thickness
- Validation of ultrasonic and thermal pressing models using temperature data from miniature embedded thermocouples and compression testing data
- Development of response surface (empirical) models using data from designed experiments in Phase I that correlate pressing process input variables with overall cell performance.
- An assessment of which models (analytical, computational, and empirical) should be used for process control.

Technical Approach (4)
Ultrasonics for MEA Pressing

- Major challenges being addressed
  - U/S horn materials and design.
  - Anvil materials and design.
  - Allowable power/energy to prevent material damage.
  - Best methods for U/S process controls (e.g. energy mode, collapse mode, absolute mode).
  - How to meet tolerance requirements
  - Target cycle time of <1 sec.
  - Rapid sensing modes for U/S APC.

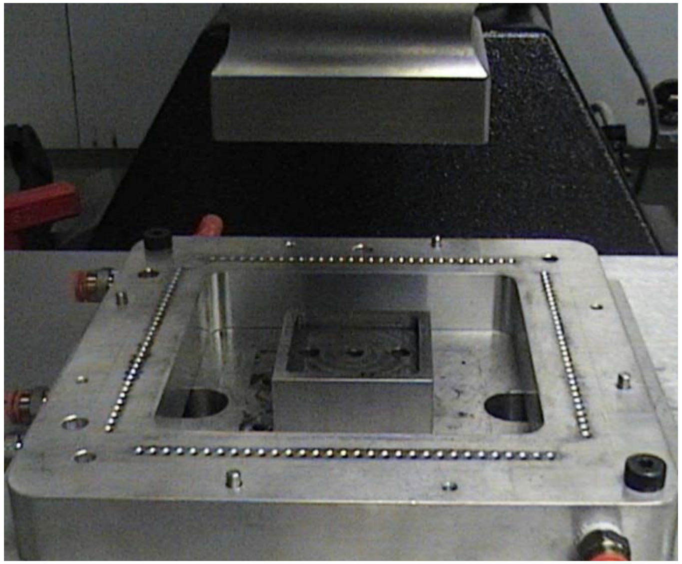


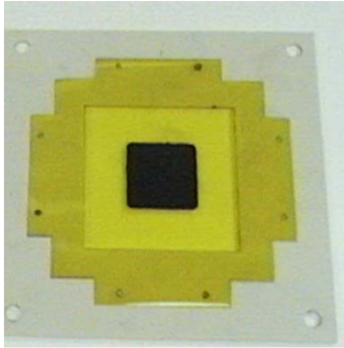
Approach/Milestones

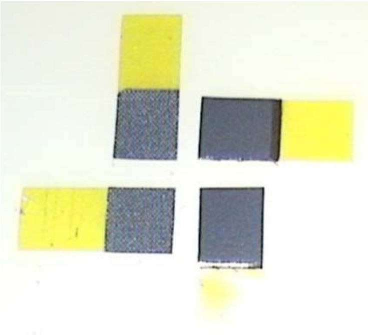
Month/Year	Milestone or Go/No-Go decision
September, 2009	Phase I Milestone: Identify relationships among properties, parameters and performance. In-situ sensing techniques that can be correlated to performance. Go/No-Go Decision: Potential of APC and Ultrasonics to reduce MEA costs.
March, 2011	Phase II Milestone: Demonstrate the ability of APC and Ultrasonics to improve the performance and uniformity of MEAs. Go/No-Go Decision: Ability to meet cost target (TBD).
February, 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 30-50%.

Technical Accomplishments (1)
Ultrasonic Welding

- **Bonding of electrodes to sub-gaskets is currently performed in a heated press. Cycle time is about 1 minute.**
- **Ultrasonic welding results:**
  - **U/S <u>cycle time of less than one second</u> has been demonstrated.**
  - **Weld <u>strength equal to or greater</u> than that of thermal process.**
  - **<u>>95% reduction</u> in energy consumed**
  - **Optimize process with full factorial design of experiments.**

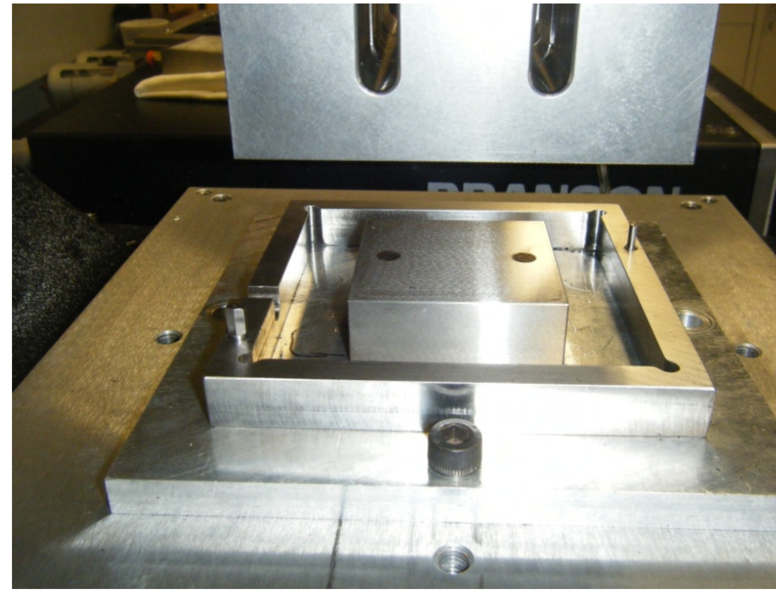


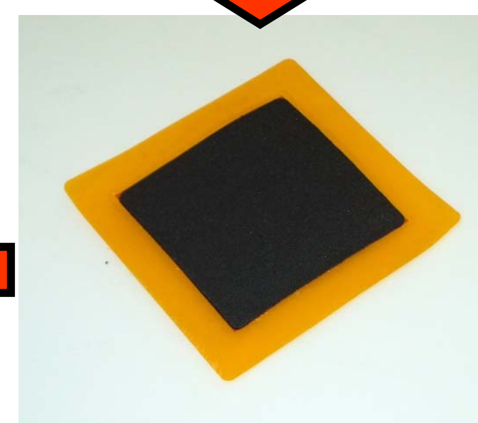


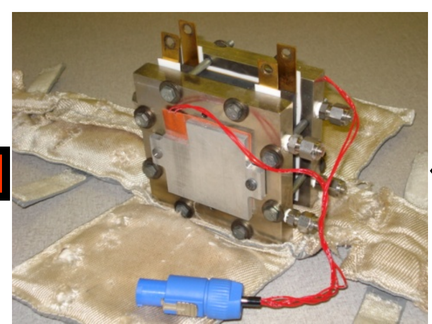


Technical Accomplishments (2)
Ultrasonic Sealing

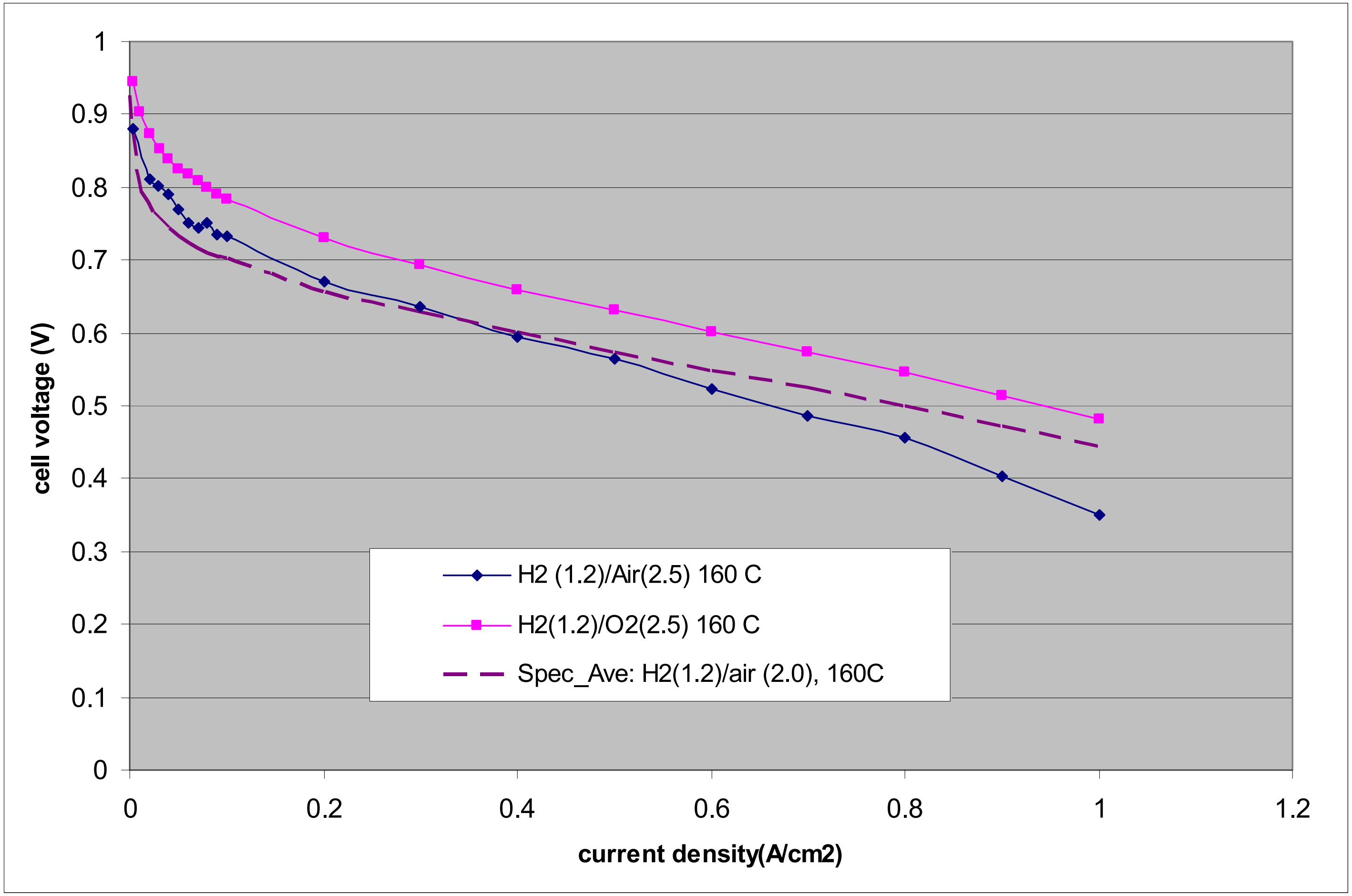
- **Initial feasibility of U/S sealing investigated.**
- **Performance of U/S welded and sealed MEA measured.**
- **No process optimization has yet been performed.**
- **Promising initial results.**







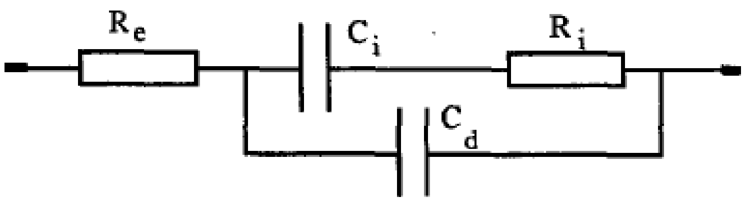
Technical Accomplishments (3)
U/S Welded and Sealed MEA IV Curve



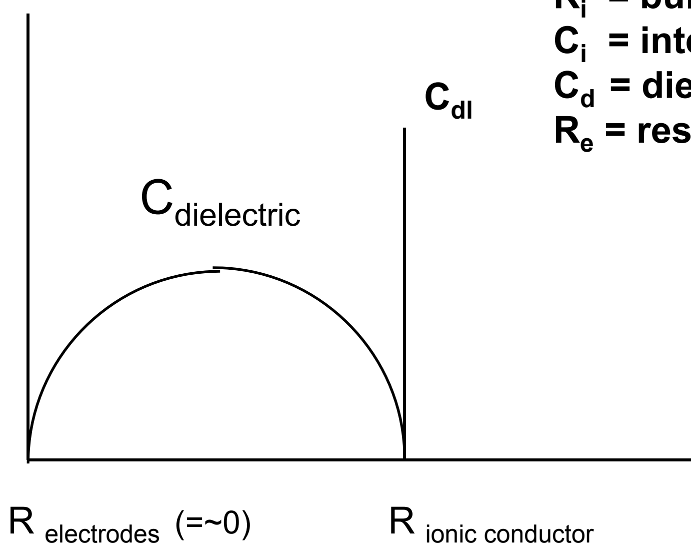
Note: No Process Optimization

Technical Accomplishments (4)
Electrochemical Impedance Spectroscopy

Simplified Blocking Electrode Circuit



"ideal cell" for perfectly symmetrical blocking electrodes



Nyquist Plot for Blocking Electrode

R_i = bulk ionic resistance
C_i = interfacial capacitance (C_{dl})
C_d = dielectric capacity,
R_e = resistance of electrodes (negligible)

Arizona State University

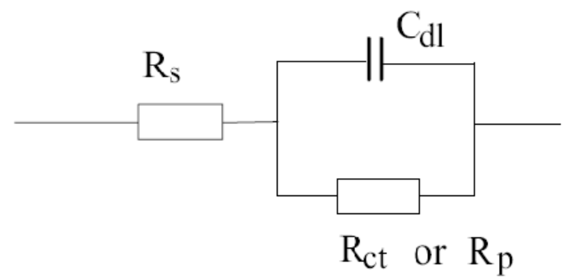
- Circuit for ac treatment of a traditional "dip cell"
- Simple method to measure ionic conductance

Problem: if electrodes "polarize" then an interfacial resistance develops and circuit is no longer valid

Technical Accomplishments (5)
Electrochemical Impedance Spectroscopy

Simplified Randles Cell, Depolarized Electrode

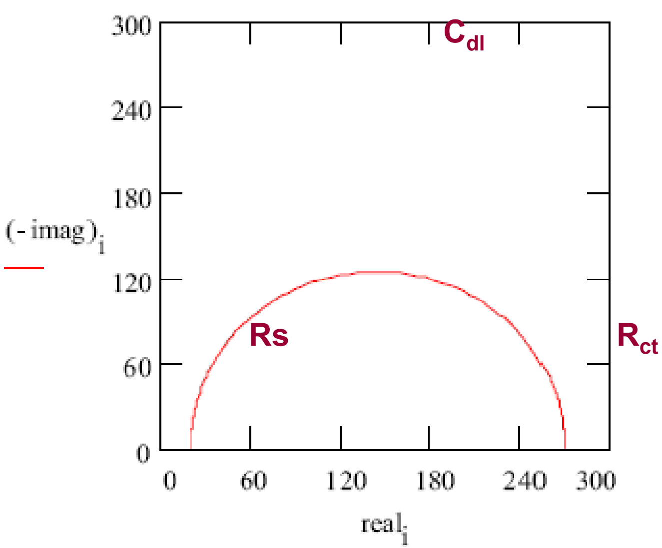
The Simplified Randles cell is one of most common cell models, and includes
- a solution resistance,
- a double layer capacitor
- a charge transfer (or polarization resistance).



The double layer capacitance is in parallel with the charge transfer resistance.

ARIZONA STATE UNIVERSITY

Nyquist Plot for Randles Cell



R_{ct}, polarization resistance, is 250 Ω,
C_{dl}, double layer capacitance: 40 µF/cm2
R_s. solution resistance: 20 Ω.

The Nyquist Plot for a simplified Randles cell is a semicircle.

The solution resistance can be found by reading the real axis value at the high frequency intercept.

The real axis value at the other (low frequency) intercept is the sum of the polarization resistance and the solution resistance.

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, evaluate stack performance.
  – National Renewable Energy Laboratory (Government Lab): independent validation of testing results.

Proposed Future Work (Phase I)

- Complete process cost analysis
- Complete two stage design of experiments and cell level testing
- Initial analytic and empirical modeling of relationships among properties, parameters and performance
- In-situ sensing development
- Initial controller development
- Phase I program review

Proposed Future Work (Phase II)

- Implementation of adaptive process controls on commercial thermal press.
- MEA performance evaluation (single cell)
- Update cost analysis
- Phase II program review

Proposed Future Work (Phase III)

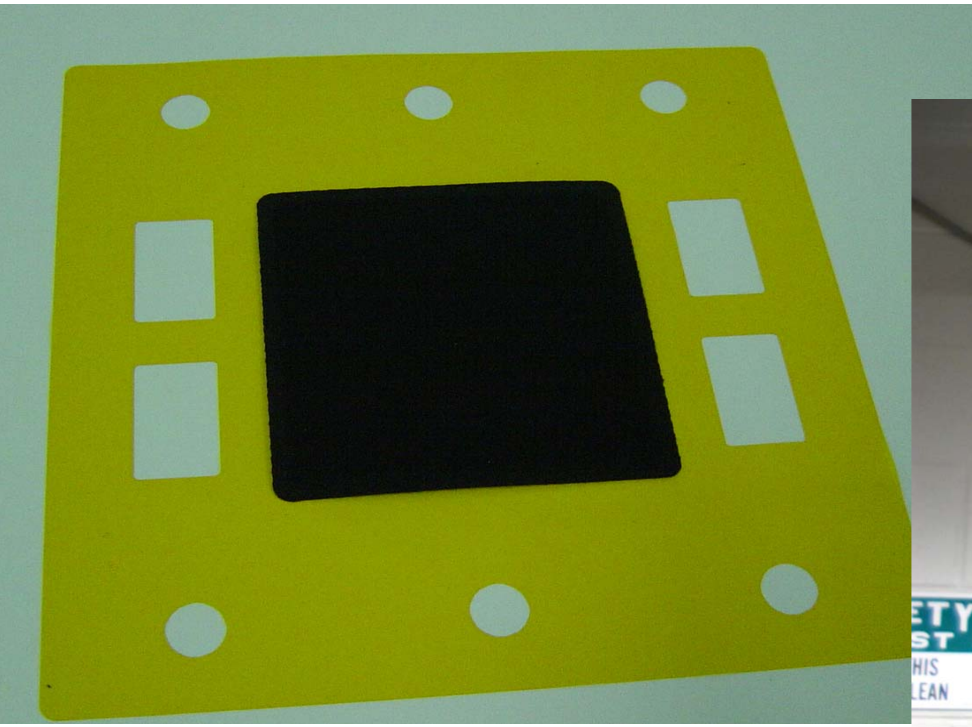
- Refine APC techniques
- APC evaluation, single cell and short stacks
- Develop design guidelines based on lessons learned.
- Phase III program review.

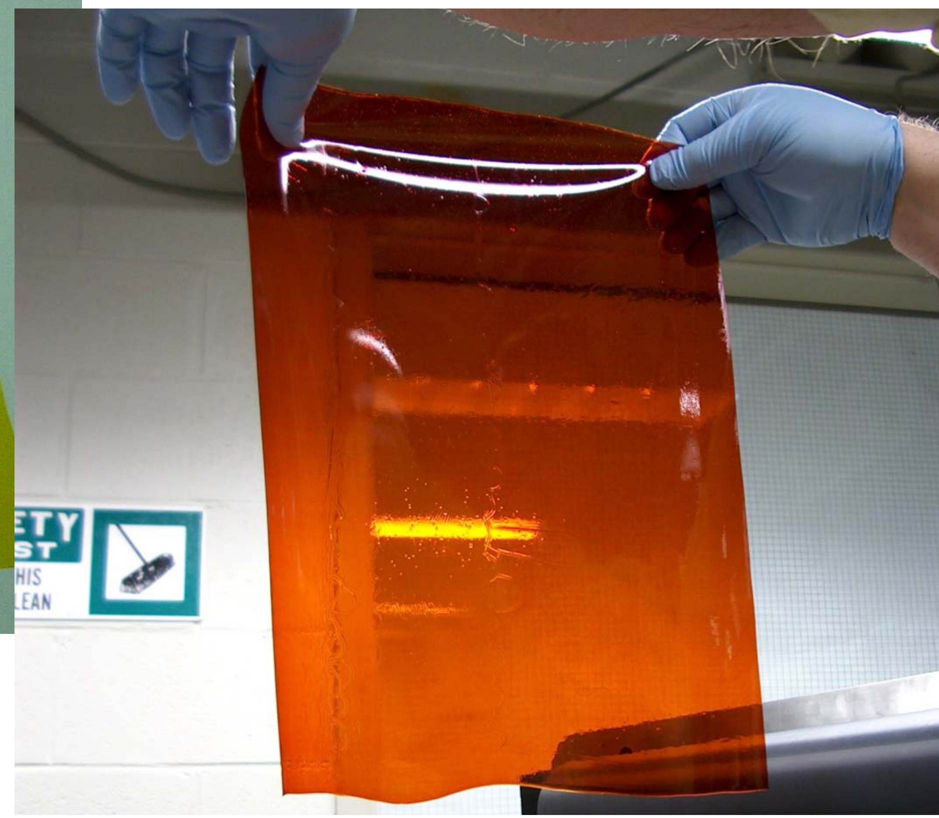
Project Summary

- **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.
- **Approach:**
  - Develop and apply adaptive, real time, process controls to improve performance and uniformity of HT PEM MEAs
  - Develop 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:** Have demonstrated feasibility of ultrasonic welding and sealing of HT PEM MEAs.
- **Proposed Future Research:** Develop models and controller, implement APC, validate via cell and stack testing

Supplemental Slides

Typical HT PEM MEA Design & PBI Membrane





BASF
The Chemical Company

LT PEM MEA Sealing Experiments

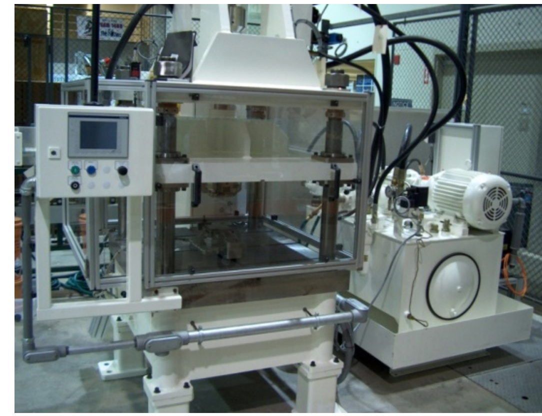
- In Fall 2008, MEAs consisting of Nafion® 117 with 10 cm^2 BASF Fuel Cell 250EWALT electrodes (0.25 mg/cm^2) were thermally and ultrasonically pressed to:
  - Demonstrate that both pressing processes worked prior to running tests with high temperature MEAs
  - Confirm published effects of thermal pressing parameters on low temperature PEMFC performance and
  - Demonstrate that functional low-temperature MEAs can be thermally pressed.

LT MEA Operating Conditions

- **Anode and cathode flowfield plates had a single serpentine channel**
- **Startup consisted of 1 hr burn-in at 23°C and 0.2 A/cm^2 with cathode and anode humidification at 35°C.**
- **H_2 and air flowrates were both 100 ml/min.**
- **Polarization curve taken at 30°C operating temperature**
- **Operating temperature raised to 80°C with anode and cathode humidifiers at 85°C.**
- **Second polarization curve taken after running at 0.2 A/cm^2 for several minutes.**

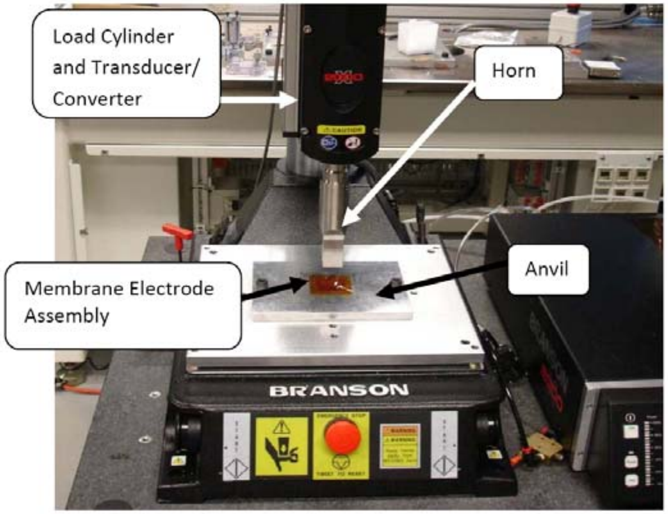
Thermal LT MEA Pressing Results

- **Increased sealing time resulted in a slight improvement in MEA performance at 30°C but essentially no improvement at 80°C.**
- **The middle sealing temperature (140°C) yielded optimum MEA performance at both 30°C and 80°C.**
- **The middle sealing pressure (50 kg/cm^2) yielded the optimum MEA performance at both 30°C and 80°C.**



Ultrasonic LT MEA Pressing Results

- **High sealing pressure yielded the best MEA performance at 30°C.**
- **There is little difference in MEA performance at 80°C.**



Comparison of Optimal Thermal Pressed and Ultrasonic Sealed MEAs

- **The thermal pressed MEA performed better at 80°C, while the ultrasonically sealed MEA performed better at 30°C.**