Fuel Cell MEA Manufacturing R&D

National Renewable Energy Laboratory

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Overview

Timeline
Start: July 2007
End: Project continuation and direction determined annually by DOE
% complete: N/A

Budget
Funding received in FY12
- $619,000 (includes $130,000 to partners)
Planned funding in FY13
- $575,000 (includes $160,000 to partners)

Barriers
<table>
<thead>
<tr>
<th>Barriers</th>
<th>Target</th>
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</thead>
<tbody>
<tr>
<td>E: Lack of Improved Methods of Final Inspection of MEAs</td>
<td>$21/kW (2017) at 500,000 stacks/yr</td>
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<tr>
<td>K: Low Levels of Quality Control</td>
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Funded Partners
Lawrence Berkeley National Laboratory
Colorado School of Mines
New Jersey Institute of Technology
DJW Technology
Relevance: NREL addresses most MYPP milestones

<table>
<thead>
<tr>
<th>Task 6: Quality Control and Modeling and Simulation</th>
<th>Task 1: Membrane Electrode Assemblies</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.2 Develop defect detection techniques in pilot scale applications for manufacturing MEAs and MEA components. (4Q, 2013)</td>
<td>Reduce the cost of manufacturing MEAs by 25%, relative to 2008 baseline of $126/kW (at 1,000 units/year). (4Q, 2013)</td>
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<tr>
<td>6.3 Establish models to predict the effect of manufacturing variations on MEA performance. (4Q, 2014)</td>
<td>Develop processes for direct coating of electrodes on membranes or gas diffusion media (4Q, 2014)</td>
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<tr>
<td>6.4 Demonstrate methods to inspect full MEAs and cells prior to assembly into stacks (4Q, 2014)</td>
<td>Develop processes for highly uniform continuous lamination of MEA components (4Q, 2014)</td>
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<tr>
<td>6.5 Validate and extend models to predict the effect of manufacturing variations on MEA performance. (4Q, 2014)</td>
<td>Develop cell manufacturing processes that increase throughput and efficiency and decrease complexity and waste (4Q, 2015)</td>
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<tr>
<td>6.6 Demonstrate continuous in-line measurement for MEA and MEA component fabrication. (4Q, 2015)</td>
<td>Demonstrate processes for direct coating of electrodes on membranes or gas diffusion media (4Q, 2016)</td>
</tr>
<tr>
<td>6.7 Develop methods to mark identified defects for later removal (4Q, 2015)</td>
<td>Demonstrate processes for highly uniform continuous lamination of MEA components (4Q, 2016)</td>
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<tr>
<td>6.8 Develop and demonstrate techniques and diagnostics for automated or continuous in-line measurement of high temperature cells and sub-assemblies during fabrication. (4Q, 2016)</td>
<td>Develop fabrication and assembly processes for PEM fuel cell MEA components leading to an automotive fuel cell system that cost $30/kW. (4Q, 2017)</td>
</tr>
<tr>
<td>6.9 Develop correlations between manufacturing parameters and manufacturing variability, and performance and durability of MEAs (4Q, 2017)</td>
<td>Develop fabrication and assembly processes for membranes that operate at T &gt; 150°C with a projected durability of 60,000 hours. (2Q, 2019)</td>
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</table>
Relevance: Cost of Poor Quality

• “Just four sequential process steps with 90% yields would increase costs by 30%...”


~$12/kW cost increase (at 500,000 units/yr) relative to total stack cost target of $21/kW

Preliminary Strategic Analysis Inc./NREL analysis using SA Inc’s automotive fuel cell cost model, 2012.
Collaborations


- **NREL National Center for Photovoltaics/New Jersey Institute of Technology**: diagnostics development

- **Lawrence Berkeley National Lab**: model development and integration

- **Colorado School of Mines**: diagnostic development, test method development and defect analysis

*DOE Manufacturing projects*
## Approach

- Understand quality control needs from industry partners and forums
- Develop diagnostics
  - Use modeling to guide development
  - Use in-situ testing to understand the effects of defects
- Validate diagnostics in-line
- Transfer technology

<table>
<thead>
<tr>
<th>Date</th>
<th>Milestone/Deliverable</th>
<th>Complete</th>
</tr>
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<tbody>
<tr>
<td>3/13</td>
<td>Prove feasibility of through-plane IR/DC using continuous sheet</td>
<td>100%*</td>
</tr>
<tr>
<td>6/13</td>
<td>Prove feasibility of optical reflectometry for detection of surface defects on SOFC tube cells</td>
<td>90%</td>
</tr>
<tr>
<td>8/13</td>
<td>Go/No-go decision on feasibility of implementing IR/RFT on web-line</td>
<td>75%</td>
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* Proved using CCM sheet
Approach: Prologue

• Last year we focused on web-line demonstration

• This year we’re focusing more on exploratory studies
  o Improving sensitivity of current techniques
  o Expanding techniques to new materials
  o Exploring feasibility of new diagnostic concepts
  o Developing new in situ techniques/capabilities
Technical Accomplishments:

- **Experimental electrodes with direct-coated ionomer**
  - From initial trials of Gore’s DOE-funded process improvements

- **Difficult to measure black surfaces**

- **Inspection of surface structure**
Technical Accomplishments:

• Actual defects on GDE sheet
  o From initial trials of BASF’s DOE-funded process improvements

• Black-on-black defects difficult with commercial vision systems

• Detected defects of dimension ~1 mm
Technical Accomplishments:

• Studied fired tubes (anode + electrolyte)
• Used standard line camera setup with manual rotation of tube
• Detected defects of ~1 mm dimension

Optical diagnostic demonstrated on tube cells

$\frac{1}{4}''$ diameter tube

$\frac{3}{4}''$ diameter tube
Technical Accomplishments:

- Developed dual light source methodology
  - Improved sensitivity to low reflectance materials (membrane)
  - Improved detection of defects
- Developed new conveyor-based motion system
  - Applicable for rigid plate materials
  - Leveraging NREL PV group
- Investigating use of band-pass filters to separate surface from bulk data
Technical Accomplishments:

- GDL/CCM cracks in line with electrical field hard to detect
- Investigated non-uniform excitation field
- LBNL modeling indicates improvement in detectability
- Modeling used to optimize excitation geometry

Modeling to improve IR/DC diagnostic

Uniform excitation provides very small temperature rise

Non-uniform excitation enables detection of $\Delta T > 2 \, ^\circ C$
Technical Accomplishments:

• Validated equipment and operating parameters with experimental material made on coating line
• Designed wider roller system and camera stand for deployment
• Plan to deploy on Ion Power catalyst coating line

Agreement on first industrial deployment of IR/DC
Technical Accomplishments:

Effect of $H_2$ concentration

- Initial reactive gas mix (0.4% $H_2$) chosen based on 10% LFL
- LBNL modeling predicts improvement by increasing $H_2$ concentration
- Performed experiments with 2% $H_2$ / 1% $O_2$ / $N_2$ balance (<LFL, <5% $O_2$)
  - Confirmed predicted increase in $\Delta T$
  - Showed that detection time decreases

Predictive modeling leads to improved IR/RFT detectability
**Technical Accomplishments:**

Predictive modeling leads to improved IR/RFT detectability

**Comparison case:** 1500 sccm flowrate, nominal loading 0.2 mg/cm²

**0.0625 cm² with 50% reduction defect:**

- Undetected at 0.4% H₂ concentration
- Detected ($\Delta T = 1°C$) in less than 1 second at 2% H₂

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**Graphs:***

- **Left Graph:**
  - Comparison of pristine and defect electrodes under varying H₂ concentrations.
  - Pristine electrode shows lower ΔT across all concentrations.

- **Middle Graph:**
  - Graph showing peak temperature ($T_{max}$) variation with H₂ concentration.
  - Experimentally measured values compared with model predictions.

- **Right Graph:**
  - Defect electrode peak intensity (ΔI) under different H₂ concentrations.
  - Demonstrates significantly higher ΔI at 2% H₂ concentration.

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**Defect electrode:** 1 cm², 50% reduction

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**Technical Details:**

- **Profile - Line 1**
  - Temperature profile along a line.
  - Max: 28.5°C, Min: 25.2°C, Av: 27.5°C
Technical Accomplishments:

Prototype air knife

- Home-built knife for determination of feasibility
- 115 holes, 0.5 mm diameter, 2 mm hole spacing
- Enables studies of flowrate, flux, width, knife/substrate separation
- Initial static experiment successful

Demonstrated IR/RFT in open environment
Technical Accomplishments:

Demonstrated IR/RFT in open environment

Prototype
Air Knife Temperature Profile: Static Sample

Next step to demonstrate with moving substrate
Technical Accomplishments: New Diagnostic Concept

Impedance Measurement of Ionomer:Carbon Ratio

- Does electronic capacitance change with I:C ratio?
- CCMs with 0.4/0.4, 0.4/0.8, 0.8/0.8 I:C ratios investigated (0.2/0.2 mg Pt/cm²)
- AC perturbation 0.1 – 10 kHz, 0.5 V_{pp}
- Dependence of electronic capacitance confirmed
- Ongoing work to determine if sensitivity can be improved to detect smaller variations of I:C ratio
Technical Accomplishments:

What is the threshold for measurement of electrode bare spots?
• Initial performance
• Aging

Spatial Performance at 1.0 A/cm², Cathode defect, 150/150 kPa, 1050/3500 sccm H₂/air, 100/50% RH

Studied electrode bare spot initial performance effects

0.5 cm² = 1% uncoated area

0.13 cm² = 0.26% uncoated area

Pristine

Data shown last year
Technical Accomplishments:

- No effect of bare spots on total cell performance
- Reduction of local performance at the defect location (as a function of defect size)
- Based on initial performance at selected conditions, these defects don’t need to be detected by QC diagnostics

Studied electrode bare spot initial performance effects
Technical Accomplishments:

• Can we detect and spatially resolve failure due to defects?
• Select (with LANL) and validate accelerated stress test
  - Combined mechanical/chemical AST
• Use OCV with anode overpressure as failure indicator
• Developed new hardware to spatially detect hydrogen crossover

### AST Conditions

<table>
<thead>
<tr>
<th>AST Duration</th>
<th>24 hrs/step</th>
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</thead>
<tbody>
<tr>
<td>Cell Temp</td>
<td>80°C</td>
</tr>
<tr>
<td>H₂/Air Gas</td>
<td>2000/2000 sccm</td>
</tr>
<tr>
<td>Pressures</td>
<td>150/150 kPa</td>
</tr>
<tr>
<td>Humidification</td>
<td>90/90 and 20/20°C Dew Points switching every 2 min</td>
</tr>
</tbody>
</table>

![Graph showing OCV Analysis](image-url)

- **OCV Analysis**
  - Cell Voltage during 0-24 hrs AST
  - Cell Voltage during 24-48 hrs AST

- **OCV & IR Analysis**

![Graph showing OCV & IR Analysis](image-url)

- **No failure**
- **Failure!**

Studied electrode bare spot aging effects
**Technical Accomplishments:**

- Applied AST to pristine MEAs and MEA with bare spot
- Observed failures at:
  - Edge of active area
  - Location of defect
- **Based on aging, this bare spot needs to be detected by QC diagnostics!**
- Next steps:
  - ‘Tone down’ AST to focus on onset of failure point
  - Protect edge of active area to focus on effects of defect
  - Reduce defect size to identify threshold

**Studied electrode bare spot aging effects**
Future Work

• Work toward deployment of diagnostics on partner manufacturing lines, while continuing to optimize their performance

• Complete feasibility study for deployment of IR/RFT diagnostic in-line

• Continue to study capacitance and other new diagnostics concepts, per industry inputs

• Continue to integrate modeling results to support diagnostic development

• Emphasize modeling and in situ testing of effects of defects (MYPP milestones)

• Refine protocols and techniques and continue aging and spatial failure studies of electrode and other MEA defects

• Complete specific partner studies and continue to support the industry
Summary

- Highlighted relevance of cross-cutting QC development via cost of poor quality analysis
- Optical Reflectometry:
  - Demonstrated on CCMs (Gore), GDEs (BASF), tubular SOFCs (AMI, Acumentrics)
  - Initiated modifications to improve sensitivity
- IR/DC:
  - Modeled nonuniform excitation to improve detection
  - Initiated effort to deploy on commercial coating line (Ion Power)
  - Continued development of through-plane method for MEA shorting
  - Studied potential noncontact excitation sources
  - Studied quality measurements of electrolyzer electrodes (Proton OnSite)
- IR/RFT:
  - Demonstrated improved detectability via higher H₂ concentration
  - Demonstrated open-environment operation with prototype air knife
- Explored CCM capacitance vs. I:C ratio as a potential new diagnostic
- In situ:
  - Determined local vs. overall performance effects of electrode bare spots
  - Developed new hardware and proved methods for aging and spatial failure detection
  - Completed detailed performance comparison of ultrasonic vs. thermal pressed MEAs (RPI)
Acknowledgement

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Rene Rivero

DOE
Nancy Garland
TECHNICAL BACK-UP SLIDES
PEM Electrolyzer Electrodes

- Experimental Proton OnSite electrodes made by simpler manufacturing process
- Different morphology renders current resistance QC test unstable
  - In situ testing good, but
  - QC test unusable
- Evaluated electrodes for uniformity
  - Part-to-part variability
  - Effects of orientation
  - In-plane conductive discontinuity
Noncontact Excitation

- Rollers typically used for electrical excitation
  - Contact, but not ‘Additional Contact’
- However, minimization of contact may be desired, e.g., for extremely thin layers (NSTF)
- Scoping performed with home-built Eddy Current ring
- GDL sample tested successfully at commercial eddy current supplier
- Also initial experiment and research into microwave as source
We completed in situ testing of RPI MEAs, comparing ultrasonic pressing with traditional hot-pressing

- Sidedness
- Comparison to Thermal Pressing

"NREL results and conclusions were similar to many of RPI’s own and provided deeper insight into how ultrasonic bonding of low-temperature MEA components affects cell performance. RPI will be using these results to assist in optimizing the process.”