Overview

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

Budget
Funding received in FY14
- $920,000 (includes $330,000 to partners)
Planned funding in FY15
- $750,000 (includes $180,000 to partners)

Barriers
<table>
<thead>
<tr>
<th>Barriers</th>
<th>Target</th>
</tr>
</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>
</tr>
<tr>
<td>K: Low Levels of Quality Control</td>
<td></td>
</tr>
</tbody>
</table>

Funded Partners
Lawrence Berkeley National Laboratory
New Jersey Institute of Technology
## From MYPP Section 3.5: Manufacturing R&D

### Task 1: Membrane Electrode Assemblies

<table>
<thead>
<tr>
<th>Task</th>
<th>Description</th>
<th>Timeline</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1</td>
<td>Develop continuous in-line measurement for MEA fabrication.</td>
<td>(4Q, 2012)</td>
</tr>
<tr>
<td>1.2</td>
<td>Reduce the cost of manufacturing MEAs by 25%, relative to 2008 baseline of $126/kW (at 1,000 units/year).</td>
<td>(4Q, 2013)</td>
</tr>
<tr>
<td>1.3</td>
<td>Develop processes for direct coating of electrodes on membranes or gas diffusion media.</td>
<td>(4Q, 2014)</td>
</tr>
<tr>
<td>1.4</td>
<td>Develop processes for highly uniform continuous lamination of MEA components.</td>
<td>(4Q, 2014)</td>
</tr>
<tr>
<td>1.5</td>
<td>Develop cell manufacturing processes that increase throughput and efficiency and decrease complexity and waste.</td>
<td>(4Q, 2015)</td>
</tr>
<tr>
<td>1.6</td>
<td>Demonstrate processes for direct coating of electrodes on membranes or gas diffusion media.</td>
<td>(4Q, 2016)</td>
</tr>
<tr>
<td>1.7</td>
<td>Demonstrate processes for highly uniform continuous lamination of MEA components.</td>
<td>(4Q, 2016)</td>
</tr>
<tr>
<td>1.8</td>
<td>Develop fabrication and assembly processes for PEM fuel cell MEA components leading to an automotive fuel cell system that cost $30/kW.</td>
<td>(4Q, 2017)</td>
</tr>
<tr>
<td>1.9</td>
<td>Develop fabrication and assembly processes for membranes that operate at T &gt; 150°C with a projected durability of 60,000 hours.</td>
<td>(2Q, 2019)</td>
</tr>
</tbody>
</table>

### Task 6: Quality Control and Modeling and Simulation

<table>
<thead>
<tr>
<th>Task</th>
<th>Description</th>
<th>Timeline</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.1</td>
<td>Develop continuous in-line measurement for PEM MEA fabrication.</td>
<td>(4Q, 2012)</td>
</tr>
<tr>
<td>6.2</td>
<td>Develop defect detection techniques in pilot scale applications for manufacturing MEAs and MEA components.</td>
<td>(4Q, 2013)</td>
</tr>
<tr>
<td>6.3</td>
<td>Establish models to predict the effect of manufacturing variations on MEA performance.</td>
<td>(4Q, 2014)</td>
</tr>
<tr>
<td>6.4</td>
<td>Demonstrate methods to inspect full MEAs and cells prior to assembly into stacks.</td>
<td>(4Q, 2014)</td>
</tr>
<tr>
<td>6.5</td>
<td>Validate and extend models to predict the effect of manufacturing variations on MEA performance.</td>
<td>(4Q, 2014)</td>
</tr>
<tr>
<td>6.6</td>
<td>Demonstrate continuous in-line measurement for MEA and MEA component fabrication.</td>
<td>(4Q, 2015)</td>
</tr>
<tr>
<td>6.7</td>
<td>Develop methods to mark identified defects for later removal.</td>
<td>(4Q, 2015)</td>
</tr>
<tr>
<td>6.8</td>
<td>Develop and demonstrate techniques and diagnostics for automated or continuous in-line measurement of high temperature cells and sub-assemblies during fabrication.</td>
<td>(4Q, 2016)</td>
</tr>
<tr>
<td>6.9</td>
<td>Develop correlations between manufacturing parameters and manufacturing variability, and performance and durability of MEAs.</td>
<td>(4Q, 2017)</td>
</tr>
</tbody>
</table>

### Relevance: Project addresses MYPP milestones

- **Completed**
- **Ongoing**
- **Assisting industry**
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 (as of 4/10/15)</th>
<th>Complete</th>
</tr>
</thead>
<tbody>
<tr>
<td>8/14</td>
<td>Demonstrate IR/RIF on web-line</td>
<td>100%</td>
</tr>
<tr>
<td>12/14</td>
<td>Complete optical scanner upgrades and initial optical testing on GM samples</td>
<td>100%</td>
</tr>
<tr>
<td>3/15</td>
<td>Go/No-go decision for demonstration of through-plane IR/DC on web-line</td>
<td>100%</td>
</tr>
<tr>
<td>6/15</td>
<td>Segmented cell station operational and initial defect testing complete</td>
<td>50%</td>
</tr>
<tr>
<td>9/15</td>
<td>Go/No-go decision for demonstration of through-plane reactive excitation on web-line</td>
<td>50%</td>
</tr>
</tbody>
</table>
Collaborations

Industry Collaborators

- GM, W.L. Gore & Associates*, Ion Power: detailed input on manufacturing QC needs, prioritization of diagnostic development, feedback on technique capabilities, defect selection and sample fabrication

- Lawrence Berkeley National Lab: model development and integration

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

- Colorado School of Mines: diagnostics development and cell testing

- Georgia Tech: fabrication of as-manufactured defect samples

Labs and Academia

* DOE Manufacturing project
Technical Accomplishments:

- Demonstrated IR/RIF on NREL research web-line

Laser drilled knife holes to improve flow uniformity

- Large-plenum gas knife to improve pressure uniformity of impinging reacting flow

- Micrometer tools to set height location
Technical Accomplishments:

- Designed & fabricated web-line equipment
- Fabricated 3’ sheet of GDE with bare spots and thickness reduction defects (50% & 25%) from 1 cm² to 0.0625 cm²
- Studied effects of web speed and reactive gas flowrate
- Successfully detected both milestone defects

Demonstrated IR/RIF on NREL research web-line

Milestone 1:
10 fpm, 1.0 cm² bare spot

Milestone 2:
30 fpm, 0.25 cm² bare spot

Unintentional material defect
Technical Accomplishments:

- In addition, we exceeded the milestones, and detected all defect sizes and extents at all conditions, including line speeds up to 30 fpm.

10 fpm, 0.0625 cm², 50% thickness reduction

10 fpm, 0.0625 cm², 25% thickness reduction
Technical Accomplishments:

- **Impinging flow modeling (LBNL)**
  - Predicts operating conditions required for defect detection
  - Predicts achievable thermal response given defect size, i.e. detection limits
  - Good quantitative agreement with experiment

**Enhanced impinging flow model with web motion for RIF**

<table>
<thead>
<tr>
<th>Defect</th>
<th>Minimum detectable defect at $\Delta T_d=1^\circ$C</th>
<th>Minimum detectable defect at $\Delta T_d=2^\circ$C</th>
</tr>
</thead>
<tbody>
<tr>
<td>100% reduction</td>
<td>0.24 mm</td>
<td>0.5 mm</td>
</tr>
<tr>
<td>50% reduction</td>
<td>0.51 mm</td>
<td>1.05 mm</td>
</tr>
<tr>
<td>25% reduction</td>
<td>1.07 mm</td>
<td>2.3 mm</td>
</tr>
</tbody>
</table>
**Technical Accomplishments:**

- **Knife hole geometry**
  - Current design results in cross-web temperature variations that can affect defect detection
  - Improved geometry can decrease variation

- **Backing under GDE**
  - Potentially increases $\Delta T$, improving defect detection

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**Predicted possible improvements to RIF technique**

**Cross-section gas knife**

<table>
<thead>
<tr>
<th>$c_{H2}$ [mol/m$^3$]</th>
<th>0.6</th>
<th>0.5</th>
<th>0.4</th>
<th>0.3</th>
<th>0.2</th>
<th>0.1</th>
<th>0</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_{max}$ - $T_{min}$</td>
<td>2.0</td>
<td>1.8</td>
<td>1.6</td>
<td>1.4</td>
<td>1.2</td>
<td>1.0</td>
<td>0.8</td>
</tr>
</tbody>
</table>

- $T_{max}$ - cross-web temperature variation along CL surface

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**Knife hole geometry**

- Current design results in cross-web temperature variations that can affect defect detection
- Improved geometry can decrease variation

**Backing under GDE**

- Potentially increases $\Delta T$, improving defect detection
Technical Accomplishments: Explored image filtering to improve detection

- Low pass filter was applied to remove high frequency features
  - Specifically, we wanted to remove the effect of the individual jets in the knife
- Significant reduction in cross-web noise is achieved, while maintaining defect detection down to a cutoff frequency, $\omega_c$ of 0.1
Technical Accomplishments:

• Scale-up relevant materials
  o Gas diffusion electrodes (GDE)
  o GDEs with laminated membrane

• Intentionally created defects
  o Carbon debris deposited before (a) or after (b) electrode coating, and under membrane (c)
  o Scuffs and scratches on membrane (d)
  o Pinholes in membrane (e)

• Scanning system operated on sheet materials at 10 fpm

• Developed and demonstrated algorithms for automated detection

Debris images at 10X mag

Size distribution of debris
Technical Accomplishments:

Demonstrated Optical Reflectance for GM MEA materials

- Scanning imaging of scuffs, slits and scratches created in membrane before and after lamination

- Pinholes were laser drilled in membrane
- Yellow boxes indicate automated detection
- Pinhole images are at 10X magnification
Technical Accomplishments:

- Through-plane IR/DC to detect intentionally created shorting defects
- Defects identified on all of the MEAs with catalyst layer lumps and cuts in membrane

Applied IR/DC techniques on GM MEA materials

Slits in membrane made prior to laminating to GDE

Carbon debris applied to GDL prior to electrode coating
Technical Accomplishments:

- In-plane IR/DC
  - Detected carbon debris applied to GDE, under a laminated membrane
  - 10 fpm on bench-top roller

Applied IR/DC techniques on GM and Ion Power electrodes

- Detected electrode coating lumps on decal
- 10 fpm on research web-line
Technical Accomplishments:

- **Along-the-channel model (LBNL)**
  - Predicts current, temperature, species distribution along the channel
  - Overall performance similar for several electrode defects
    - High variability in local performance

   ![Diagram](image-url)
Technical Accomplishments:

- Along-the-channel model
  - For low humidity case, defects impact down-the-channel current
  - For higher voltage case, similar current drop for 1.5 and 0.5 cm defects

Model predicts sensitivity of defect effects to operating conditions

\[ \Delta T = 1 \, ^\circ C \]

\[ \text{RH}_{in} = 25\%, \, V=0.4V, \, 1.5 \, \text{cm defect} \]

\[ \text{RH}_{in} = 100\%, \, V=0.6V \]
Technical Accomplishments:

• Re-commissioned segmented cell test station
  o Calibration
  o Operational shake-down under different RH conditions
  o Previous work: studied local and total cell performance of small electrode bare spots
  o Current work: study electrode thin spots of different extent (%reduction) and size

• Completed setup for failure onset studies
  o Automated humidity switching for ASTs
  o NREL IR hardware for spatial cross-over
  o Previous work: studied failure behavior of large electrode bare spot under very aggressive chem/mech AST
  o Current work: study electrode bare spots of relevant size, with AST tuned to enable observation of failure onset
Barriers, Needs and Future Work

**Barriers and Needs**

- General barriers and needs are documented in the MYPP (slide 3)
  - Developing and demonstrating QC methods
  - Understanding how defects affect performance and lifetime
- We actively engage with industry to understand their needs, based on their specific processes, materials and MEA constructions
  - QC for membranes, electrodes, various sub-assemblies and full MEAs
  - Increasing interest in applicability of techniques to in-process measurement

**Future Work**

- Complete work for Go/No-go decision on through-plane reactive excitation
- Study the effects of relevant defects on cell performance and failure onset
  - Relevant electrode and as-manufactured membrane defects
- Continue to develop models, and perform modeling to optimize diagnostics and predict performance effects of defects
- Apply optical and infrared techniques, with optimal excitations, to relevant industry MEA constructions
- Develop/modify techniques to address new needs
Summary

- Addressing most of the MYPP milestones
- Continued detailed information exchange with industry partners on QC priorities
- IR/RIF:
  - Demonstrated RIF on web-line, exceeded milestone requirements
  - Expanded multi-physics model to include GDE motion, predicted detection limits and pathways for improved detection
  - Explored systemic noise reduction techniques
- Optical Reflectometry:
  - Demonstrated technique for detection of several different types of defects in scale-up relevant GDE and GDE + membrane material constructions
- IR/DC:
  - Applied through-plane IR/DC to GM MEA samples with shorting defects
  - Applied IR/DC to GM and Ion Power electrode materials
- Completed construction of through-plane reactive excitation test-bed and performed initial experiments
- Effects of defects studies:
  - Developed Along-the-channel model and began exploration of electrode defects to guide experimental work
  - Re-commissioned segmented cell and set up test station for failure onset studies
- Technical Assistance: HTAC Manufacturing Sub-committee, DOE and State of Ohio fuel cell supply chain projects, SBIR QC Topic, CEMI activities
<table>
<thead>
<tr>
<th>Description</th>
<th>Metric</th>
<th>Due Date</th>
<th>Status</th>
</tr>
</thead>
<tbody>
<tr>
<td>Expose Auto OEMs to NREL’s manufacturing QC techniques and discuss pathways for qualification and tech transfer. We will first identify companies with highest impact potential and interest in implementing QC methods, then do a selective ‘road show’ to demonstrate QC.</td>
<td>3 Auto OEMs/teams visited</td>
<td>9/30/2015</td>
<td>Leveraging relationships with OEMs in existing CRADAs. Continuing communications with a key FC supplier and an OEM. Both entities have shown strong interest, but indicate that they do not have research budget to collaborate with us. Planned discussion with another OEM during their visit to NREL in May.</td>
</tr>
<tr>
<td>2 new industrial partnerships developed</td>
<td></td>
<td>9/30/2015</td>
<td>Existing OEM partnerships (2) ongoing.</td>
</tr>
<tr>
<td>SBIR/FOA for QC system development: Seek 3rd party vendors to work with NREL to manufacture inspection systems that could be marketed to fuel cell/MEA manufacturers. We will develop a target list of companies with input from AMO, NIST, etc.</td>
<td>Identify at least 5 potential candidates</td>
<td>9/30/2015</td>
<td>The selection of appropriate 3rd parties was accomplished via competitive FOA process (per DOE direction). We provided technical input to DOE for the SBIR TTO topic. The topic was competed and is pending award.</td>
</tr>
<tr>
<td>Inspection system designed, built and marketed</td>
<td></td>
<td>9/30/2015</td>
<td>Pending awards.</td>
</tr>
</tbody>
</table>
Response to Reviewer Comments

Comment: “Techniques for on-line QC are reasonable IF it can be shown that detection limits are appropriate...flaws that cause performance or durability problems must be detectable. So far this has not been demonstrated.” (as well as several other comments similar to this)

Response: These comments are very consistent with our message and approach for several years, which have been that we need to study the effects of defects further to understand the thresholds for detection by the diagnostics we are developing. This study is optimally performed by the combination of experimental and computational work. We have performed initial studies on the effects of electrode bare spots and membrane pinholes in the past, and have reported on that work. More detailed studies, looking at a broader range of defect types and sizes, and operating conditions are still needed. NREL and LBNL have re-started work in this direction this year.

Comments: “Coordination with other [than Ion Power] industry collaborators is unclear.” “…it is unclear what CSM and LBNL are doing in the project and what value is added.”

Response: LBNL has played a foundational role since the beginning of this project, providing modeling of MEA materials with defects, either (a) in situ, or (b) during excitation for IR-based diagnostics. This work continuously guides our experimental and development work. The role of the universities we have worked with has typically been providing students that work on the project tasks. Several of the faculty have been and continue to be involved to help guide and interpret the science of the work. Coordination with our industry partners has been very much at their discretion. With some, such as GM and Ion Power, we have extremely detailed interaction on manufacturing QC needs, develop tasks and paths together, and collaborate on the fabrication and testing of representative MEA materials and demonstration of QC techniques. With others, the coordination is more ad-hoc, typically focused on establishing the feasibility of one of our techniques for one specific QC need. Others have provided generic input and guidance on critical industry needs, or have provided representative MEA materials for our development and testing, but have not actively sought assistance on QC development.
Acknowledgement

NREL
Michael Ulsh
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Adam Phillips
(SULI)

LBNL
Adam Weber
Iryna Zenyuk

NJIT
Srinivas
Devayajanam

DOE
Nancy Garland

CSM
Prof. Jason Porter
Technical Back-up Slides
Technical Accomplishments:

- 30” coated area (with 2-3 feet of uncoated GDL/MPL on either side)
- Created with ultrasonic spray system
- 50% Pt/C on GDE with MPL
- Nominal loading 0.13 mg Pt/cm²
- Three sets of localized defects
  - Defects sizes 0.0625, 0.125, 0.25, 0.5, and 1 cm²
  - Defect severity 25%, 50%, and 100% loading reduction
- Two cross-web “defects” between coated sections
  - 100% loading reduction (gap)
  - 50-100% loading increase (overlap)

Demonstrated IR/RIF on NREL research web-line
Technical Accomplishments:

- GM GDE + membrane samples
- Light angle has large impact on noise for pinhole detection (45° optimal)
- Low lighting angle potentially useful for membrane surface roughness

Studied effects of light source angle on optical reflectance imaging
Technical Accomplishments: Previously presented in situ defect study results

Spatially resolved performance studies

Spatially resolved failure onset studies

Table: AST Conditions

<table>
<thead>
<tr>
<th>Condition</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>AST Duration</td>
<td>24 hrs/stop</td>
</tr>
<tr>
<td>Cell Temp</td>
<td>80°C</td>
</tr>
<tr>
<td>H₂/Air Gas flows</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>

Spatial Performance at 1.0 A/cm², Cathode defect, 150/150 kPa, 1050/3500 sccm H₂/air, 100/50% RH
Objective: Study H₂ concentration, flowrate, and exposure time needed for detection of pinholes to determine if in-line through-plane reactive excitation is feasible

Status: Test station built and commissioned, initial experiments complete

Technical Accomplishments: Completed test-bed for through-plane reactive excitation

Controls:
- H₂/N₂ mix
- Flowrate
- Pulse duration

IR Camera
Hotspot
Pinhole
UEA
Gaskets
Cover Plate
H₂/N₂
Technical Accomplishments:

Developed along-the-channel model for effects of defects studies

**Internal Iteration loop**

- **External RH loop**
  - Computes $RH_{new}$ based on mass-balance.
  - Convergence criteria:
    $$\frac{RH_{old} - RH_{new}}{RH_{old}} < Error$$
  - **Yes**
    - Fluxes due to segment’s consumpt./gener. Species: $O_2$, $N_2$, $H_2O$
    - Conductive and convective heat flux
    - Along-channel 1D stepping
    - $RH_{new} = RH_{old}$
  - **No**
    - $RH_{new} = (RH_{old} + RH_{new}) / 2$

**2D cross-section model**

- **Channel fluxes. Species:** $O_2$, $N_2$, $H_2O$
- **Channel molar concentrations.** Species: $O_2$, $N_2$, $H_2O$
- **Enthalpy transport for gas species**
- **Channel temperature and pressure**

**Accounts for land/channel effects**

**Main iteration loop**

- **Solution for each step. Stored as .mat structure**
- **Solution for final internal iteration loop. Stored as .mph**

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**LBNL**

**MatLab**

**Comsol**

**Postprocessing with MatLab code to generate all plots.**