High Speed, Low Cost Fabrication of Gas Diffusion Electrodes for Membrane Electrode Assemblies

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16 May 2012
Overview

Timeline
- Start: 1 July 2009
- End*: 30 June 2013
- 75% complete
  (* no cost extension)

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

Budget
- Total project funding: $3.06M
  - DOE share: $1.99M
  - Contractor share: $1.07M
- Fed. funding received in FY11: $302K
- Est. Fed. funding for FY12: $250K

Partners
- Case Western Reserve University
- X-Ray Optical Systems
Membrane Electrode Assembly (MEA)
5 Layer Assembly

- Internal Gasket
- Gas Diffusion Electrode
- Catalyst
- Membrane
Overall Objective

- Reduce cost in fabricating gas diffusion electrodes (GDEs)
  - Focus on GDEs used for combined heat and power generation (CHP).
- Relate manufacturing variations to actual fuel cell performance in order to establish a cost effective product specification within six-sigma guidelines.
- Develop advanced quality control methods to guide realization of these two objectives.

Objective(s) this reporting period

- 3X throughput increase on full width/length cloth
- Scope focus: expand efforts on non-woven paper

Directly Addresses Barriers

- (A) Lack of High Volume Membrane Electrode Assembly (MEA) Processes
  - High speed or throughput coating
- (F) Low Levels of Quality Control and Inflexible Processes.
  - On-line Pt measurement.

Addresses key DOE targets

| Targets: 1–10 kW<sub>e</sub> Residential CHP FC Operating on Natural Gas |
|-----------------------------|-----------------|----------|----------|----------|
|                            | 2008 Status    | 2012     | 2015     | 2020     |
| Electrical efficiency at rated power | 34%            | 40%      | 42.5%    | 45%      |
| CHP energy efficiency | 80%             | 85%      | 87.5%    | 90%      |
| Factory cost* per kW      | $750            | $650     | $550     | $450     |

*Cost includes materials and labor costs to produce 50k/yr stacks
**Approach: Milestones and Go/No Go**

| Task          | Task 1: On-line QC to guide the process by Y1 | Task 3: full length coating by Y2 | Performance  
Defects/Uniformity  
Relate defects to performance  
Task 4: Increase line speed by Y2 (go/no go June 2011)  
Task 5: Full cloth width roll by Y2/Y3; paper>best cloth total cost  
Performance  
Defects/Uniformity  
Relate defects to performance |
|---------------|--------------------------------------------|----------------------------------|---------------------------|
| Task 2: Model impact of defects by Y1 | Task 3: full length coating by Y2 | Performance  
Defects/Uniformity  
Relate defects to performance  
Task 4: Increase line speed by Y2 (go/no go June 2011)  
Task 5: Full cloth width roll by Y2/Y3; paper>best cloth total cost  
Performance  
Defects/Uniformity  
Relate defects to performance |
| Milestone     | T1: On-line Pt measurement, on-roll porosity measurement  
T2: Verify Model, calculate defect limits  
T3: >240 lin m  
T4: 2X throughput increase (go/no go): 3X final goal  
T5: full width (>100 cm) & length at 3X throughput: paper 30% lower total cost over cloth  
Main Concept  
Use advanced dispersion and ink formulations to make aqueous solid - binder suspensions compatible | T3: >240 lin m  
T4: 2X throughput increase (go/no go): 3X final goal  
T5: full width (>100 cm) & length at 3X throughput: paper 30% lower total cost over cloth  
Main Concept  
Use advanced dispersion and ink formulations to make aqueous solid - binder suspensions compatible |
| Status        | T1: complete, modified for full width cloth. On-roll porosity canceled due to vendor failure  
T2: Base model established  
T3: complete  
T4: go/no go met. >2X throughput  
T5: full width/length cloth >3X throughput. Paper at pilot scale | T3: complete  
T4: go/no go met. >2X throughput  
T5: full width/length cloth >3X throughput. Paper at pilot scale | Base model established  
T3: complete  
T4: go/no go met. >2X throughput  
T5: full width/length cloth >3X throughput. Paper at pilot scale |
Technical Accomplishments and Progress
Tasks 3-4-5: Inks for Full Length & Width Roll Coating, 2X speed

- **Accomplishments:** Extended ink formulations to stabilizing anode catalyst suspensions that have been subject to very high energy dispersion. Scaled microporous layer (MPL) ink 15-fold. Scaled all catalyst inks 10-fold.

- **Key Challenge:** MPL ink scale up caused severe bubble formation, which led to variable viscosities. Solved through additive and process modification.

Break-through in ink-making increased capacity, decreased labor content

<table>
<thead>
<tr>
<th></th>
<th>Cost decrease vs. benchmark</th>
<th>Capacity increase vs. benchmark</th>
</tr>
</thead>
<tbody>
<tr>
<td>MPL</td>
<td>37%</td>
<td>3.0X</td>
</tr>
<tr>
<td>Anode</td>
<td>31%</td>
<td>2.1X</td>
</tr>
<tr>
<td>Cathode</td>
<td>40%</td>
<td>2.4X</td>
</tr>
</tbody>
</table>
Technical Accomplishments and Progress
Tasks 3-4-5: **Coating** Full Length & Width Roll, 3X speed

- **Accomplishments**: Quality gains preserved for length>300l.m, width 1.17m
- **Key Challenge**: Final product thinner than benchmark. Re-engineered MPL architecture to compensate for different packing densities

<table>
<thead>
<tr>
<th>Metric</th>
<th>Benchmark-start</th>
<th>This Program</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Agglomerates</strong> (avg. over roll)</td>
<td>18/m²</td>
<td>1.6/m²</td>
</tr>
<tr>
<td><strong>Pt variation</strong> (via on-line XRF, roll average)</td>
<td>+/- 2 gm Pt/m²</td>
<td>+/-0.4 gm Pt/m²</td>
</tr>
<tr>
<td><strong>Performance</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.2A/cm² H₂/air, 45cm² test cell, 160 °C</td>
<td>0.657V</td>
<td>0.683V</td>
</tr>
<tr>
<td>0.5A/cm² H₂/air, 45cm² test cell, 160 °C</td>
<td>0.573V</td>
<td>0.598V</td>
</tr>
<tr>
<td>0.2A/cm², 1.4/5 Reformate (71% H₂, 27% CO₂, 2% CO)/Air, 45cm² test cell, 180°C</td>
<td>0.668V</td>
<td>0.689V</td>
</tr>
<tr>
<td>0.5A/cm², 1.4/5 Reformate (71% H₂, 27% CO₂, 2% CO)/Air, 45cm² test cell, 180°C</td>
<td>0.571V</td>
<td>0.589V</td>
</tr>
</tbody>
</table>

Note: reformate running at 5X air stoichiometry, 180°C

Significant improvement in quality
**Technical Accomplishments and Progress**

**Task 4: Key Metrics for coating throughput**

- **Accomplishments:** Exceeded both go/no go of 2X and program goal of 3X throughput. Effective throughput boost by reducing number of application passes.

- **Key Challenge:** Applicator settings with new inks different than benchmark. Significant optimization needed.

<table>
<thead>
<tr>
<th>Throughput metric</th>
<th>Benchmark (at program start)</th>
<th>This Program (to date)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average line speed, lin.m/min</td>
<td>1</td>
<td>1.16</td>
</tr>
<tr>
<td>Sum ((S_1, S_2, \ldots, S_n))/number of applications (n), where (S=)coating rate of each step</td>
<td>1</td>
<td>1.16</td>
</tr>
<tr>
<td>Highest single line speed, one pass (lin. m/min)</td>
<td>1</td>
<td>1.67</td>
</tr>
<tr>
<td><strong>Coating throughput, m² GDE/hr</strong></td>
<td>1</td>
<td>3.94</td>
</tr>
<tr>
<td>Cost Savings (in labor hrs)</td>
<td></td>
<td>75%</td>
</tr>
</tbody>
</table>

Coating throughput, m² GDE/hr is most relevant metric
Technical Accomplishments and Progress
Task 5: Non-woven/paper substrates

- **Status:** 2011 AMR demonstrated production machine scale MPL fabrication. 2011/12 focus was on substantial reduction of application steps for MPL and catalyst layer.

- **Approach:** Increase viscosity of ink without loss of stability. Investigate new application process to handle higher viscosity inks.

- **Results:** Demonstrated potential for single-pass MPL.

Comparison of single-pass to multi-pass MPL

Target for non-woven is to reduce cost 30% over best woven configuration.
Collaborations

Task 2 Defects
- **NREL – Michael Ulsh**
  - Providing GDEs with defects for advanced defect detection

Tasks 3-4-5 (higher throughput GDEs)
- **ClearEdge Power Systems – Julia Song, Ph.D.**
  - Validated materials for use in μCHP applications at stack scale
  - Valuable feedback on material thickness specification
  - Confirmed performance gain found at subscale testing with full stacks

These collaborations were initiated outside the original team tasks in this DOE program
Proposed Future Work
Over last year of project

- Task 5: Demonstrate non-woven platform reducing total cost by additional 30% (materials + labor) over best woven scenario
  - Scale MPL to production level
  - Develop anode/cathode catalyst inks and application process that approach two or less coating passes
Reduced total GDE labor costs by ~75% due to new high energy dispersion with advanced formulations.

- Achieved June 2011 go/no go (2X throughput increase).
- Exceeded program goal (3X throughput increase at full width) to 4X improvement (by June 2012)
- Demonstrated proof of principle for one step MPL coating on non-woven non-woven.

### Summary Slide

<table>
<thead>
<tr>
<th>Platform</th>
<th>Improved Ink</th>
<th>Coating Pilot</th>
<th>Coating Production (1/2 width)</th>
<th>Sintering</th>
<th>Coating Production Full width + throughput</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cloth</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MPL</td>
<td>37%</td>
<td>&gt;55%</td>
<td>&gt;55%</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cathode</td>
<td>40%</td>
<td>&gt;40%</td>
<td>&gt;40%</td>
<td></td>
<td>This period (&gt;50%)</td>
</tr>
<tr>
<td>Anode</td>
<td>31%</td>
<td>&gt;40%</td>
<td></td>
<td></td>
<td>This period (&gt;50%)</td>
</tr>
<tr>
<td>Paper</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MPL</td>
<td></td>
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<tr>
<td>Anode</td>
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</tr>
</tbody>
</table>

% indicates reduction in labor-hours. No benchmark for paper but have achieved similar or greater results compared to cloth.
Background on high temperature MEAs
Benefits of HT PEM technology: Reduction of system complexity

High temperature PEM technology allows to simplify the fuel cell system, especially in the case of reformate feed.
MEA-components

2 x Subgasket
- Polyimide

1 x Anode / 1 x Cathode
- Gas diffusion electrode (GDE) with catalyst coating
- Catalyst is coated on top of microporous layer

1 x Membrane
- PBI based and charged with phosphoric acid
- Catalyst is not coated on the membrane
Celtec-P MEA Advantages

Technology
- Broad operating temperature window: 120 – 180 °C
- No humidification necessary
- High tolerance to CO and H₂S
- No water management system necessary for exhaust

Materials
- Membrane with Polybenzimidazole (PBI) and Phosphoric Acid (low cost)
- Gas diffusion electrodes (roll to roll processing)

Applications - end use
- Simplified overall FC system

Performance under reformate gas conditions

Unique CO-tolerance