VI.2 Manufacturing of Low-Cost, Durable Membrane Electrode Assemblies Engineered for Rapid Conditioning

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Subcontractors
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- University of Delaware, Newark, DE
- University of Tennessee, Knoxville, TN

Project Start Date: October 1, 2008
Project End Date: December 30, 2014

Objectives

- The overall objective of this project is to develop a unique, high-volume manufacturing processes that will produce low-cost, durable, high-power density 5-layer (5-L) membrane electrode assemblies (MEAs) that minimize stack conditioning:
- Manufacturing process scalable to fuel cell industry MEA volumes of at least 500k systems/year.
- Manufacturing process consistent with achieving $9/kW$_e$ DOE 2017 transportation MEA cost target.
- The product made in the manufacturing process should be at least as durable as the MEA made in the current process for relevant automotive duty cycling test protocols.
- The product developed using the new process must demonstrate power density greater or equal to that of the MEA made by the current process for relevant automotive operating conditions.
- Product form is designed to be compatible with high-volume stack assembly processes: 3-layer (3-L) MEA roll-good (anode electrode + membrane + cathode electrode) with separate rolls of gas diffusion media.
- The stack break-in time should be reduced to 4 hours or less.

Phase 2 Objectives

- Low-cost MEA R&D
  - New 3-L MEA Process Exploration
    - Investigate equipment configuration for low-cost MEA production
    - Investigate raw material formulations
    - Map out process windows for each layer of the MEA
  - Mechanical Modeling of Reinforced 3-L MEA
    - Use model to optimize membrane reinforcement for 5,000+ hour durability and maximum performance
    - Develop a deeper understanding of MEA failure mechanisms
  - 5-L Heat and Water Management Modeling
    - Optimization of gas diffusion membrane thermal, thickness, and transport properties to enhance the performance of thin, reinforced membranes and unique properties of direct-coated electrodes using a validated model
  - MEA Conditioning
  - Evaluate potential for new process to achieve DOE cost targets prior to process scale up (Go/No-Go Decision)
- Scale Up
  - Equipment setup
  - Optimization
    - Execute designed experiments which fully utilize University of Delaware and University of Tennessee modeling results to improve the new MEA process and achieve the highest possible performance and durability
- Stack Validation

Technical Barriers

This project addresses the following technical barriers from the Manufacturing section of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan:

(A) Lack of High-Volume Membrane Electrode Assembly Processes

Contribution to Achievement of DOE Manufacturing Milestones

This project will contribute to achievement of the following DOE milestones from the Fuel Cells section of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan:

- RD&D Plan Section 3.4, Task 10.1: Test and evaluate fuel cell systems and components such as MEAs, short stacks, bipolar plates, catalysts, membranes, etc. and compare to targets. (3Q, 2011 thru 3Q, 2020)
- RD&D Plan Section 3.4, Task 10.2: Update fuel cell technology cost estimate for 80 kW transportation systems and compare it to targeted values. (3Q, 2011 thru 3Q, 2020)

FY 2014 Accomplishments

- Direct Coating Process Development
  - The primary path for the new 3-L MEA process has succeeded in incorporating the previously modeled process improvements which indicated potential for a 25% reduction in high-volume 3-L MEA cost.
  - Pilot-scale demonstration of the new 3-L MEA process is nearing completion:
    - Current density of un-optimized direct-coated electrodes is equivalent to or better than current commercial electrodes over a robust range of automotive operating conditions.
    - Gore has demonstrated mechanical durability of an 8-micron expanded polytetrafluoroethylene (ePTFE) reinforced membrane. In previous testing, GORE™ MEAs exceeded 2,000 hours of accelerated mechanical durability testing, which has been equated to achieving 9,000 hours of membrane durability in an 80°C automotive duty cycle. This exceeds the DOE 2015 membrane durability target of 5,000 hours. Gore's 8-micron ePTFE reinforced membrane technology has been successfully incorporated into the lab-scale new 3-L MEA process.
- Modeling tasks at the University of Delaware and University of Tennessee are complete.

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INTRODUCTION

Over the past 20 years, great technical progress has been made in the area of improving power density and durability of fuel cell stacks, so much so that most of the requisite technical targets are now within reach. Yet, three major technical challenges remain. First and foremost is meeting the cost targets. The second challenge is producing components that are amenable for use in a high-speed, automotive assembly line. One impediment to this latter goal is that stack components must currently go through a long and tedious conditioning procedure before they produce optimal power. This so-called "break-in" can take many hours, and can involve quite complex voltage, temperature and/or pressure steps. These break-in procedures must be simplified and the time required reduced, if fuel cells are to become a viable power source. The third challenge is to achieve the durability targets in real-world operation. This project addresses all three challenges: cost, break-in time, and durability for the key component of fuel cell stacks: MEAs.

APPROACH

The overall objective of this project is to develop unique, high-volume manufacturing processes for low-cost, durable, high-power density 3-L MEAs that require little or no stack conditioning. In order to reduce MEA and stack costs, a new process will be engineered to reduce the cost of intermediate backer materials, reduce the number and cost of coating passes, improve safety and reduce process cost by minimizing solvent use, and reduce required conditioning time and costs. MEA mechanical durability will be studied and optimized using a combination of ex situ mechanical property testing, non-linear mechanical model optimization, and in situ accelerated mechanical durability testing. Fuel cell heat and water management will be modeled to optimize electrode and gas diffusion membrane thermal, geometric, and transport properties and interactions. Unique enabling technologies that will be employed in new process development include:

- Direct coating which will be used to form at least one membrane–electrode interface.
- Gore's advanced ePTFE membrane reinforcement and advanced perfluorinated sulfonic acid ionomers which enable durable high-performance MEAs.
- Advanced fuel cell testing and diagnostics.

RESULTS

Low-Cost MEA Process Development

- Primary path (Figure 1)
  - Process step 1: Coat bottom electrode on low-cost, non-porous backer
  - Process step 2: Direct coat reinforced membrane on top of the bottom electrode

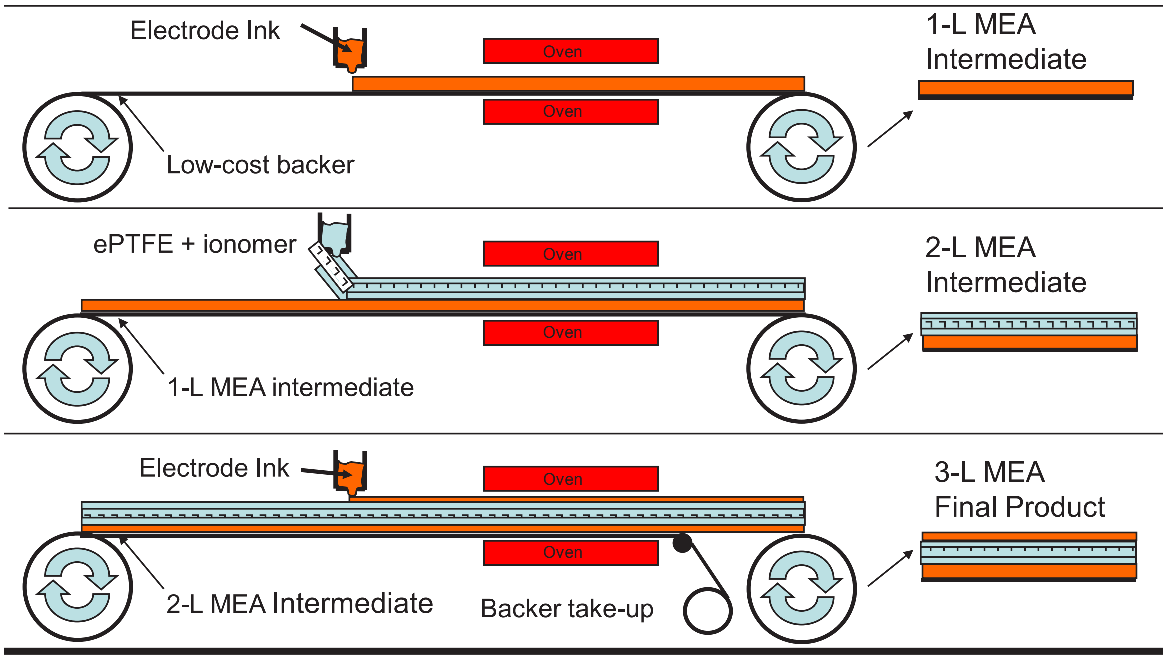
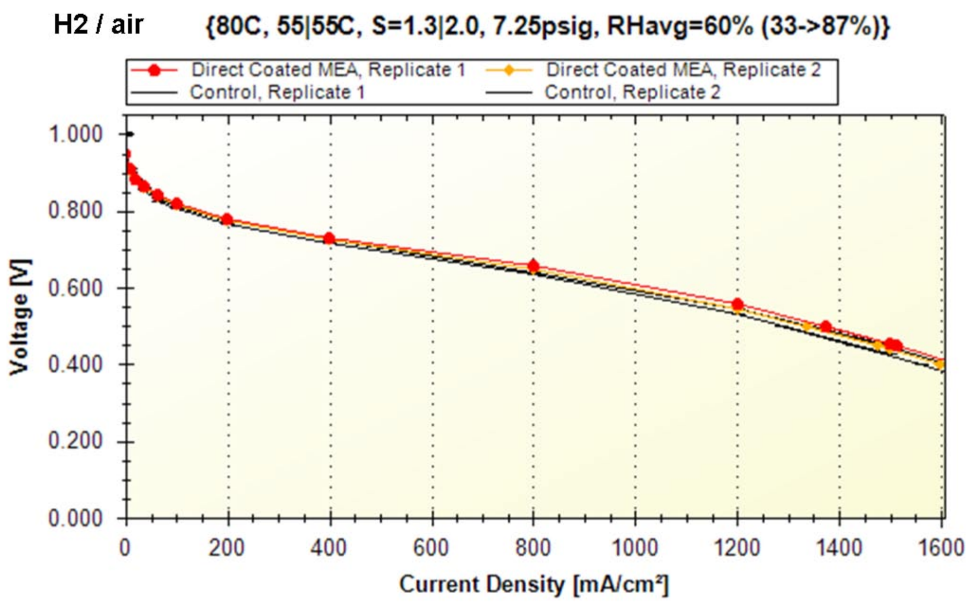


FIGURE 1. Low-Cost MEA Manufacturing Process



RH - relative humidity

FIGURE 2. Direct-Coated MEA Performance

  – Process step 3: Direct coat top-side electrode on top of the reinforced membrane
- Figure 2 indicates performance of a direct coated MEA compared with a control MEA made with the commercial process.
- Figure 3 demonstrates the advantage of thin Gore membranes in hot, dry operating conditions.

Mechanically Durable 8-µm Reinforced Membrane

Gore has successfully incorporated a mechanically durable 8-µm reinforced membrane into the current primary path process. The 8-µm membrane construction has demonstrated high performance due to reduced resistance and increased water back-diffusion (see Figure 3). In previous testing, GORE™ MEAs exceeded 2,000 hours of

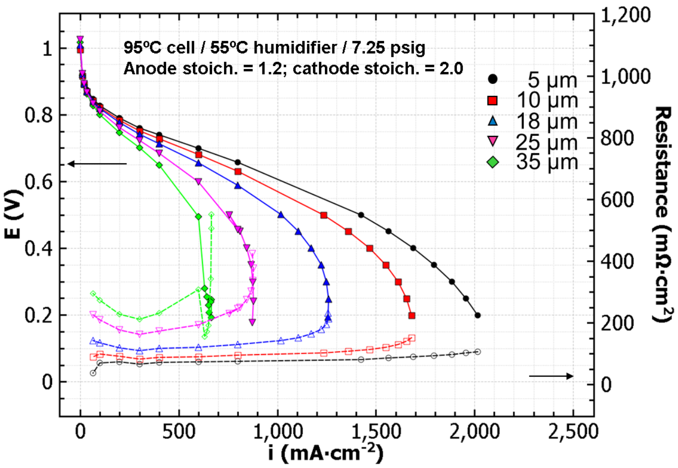


FIGURE 3. Performance Comparison of Thin, Mechanically Durable Reinforced Membranes

accelerated mechanical durability testing, which has been equated to achieving 9,000 hours of membrane durability in an 80°C automotive duty cycle. This exceeds the DOE 2015 membrane durability target of 5,000 hours. The accelerated mechanical durability testing protocol is summarized below:

Tcell (C)	Pressure (kPa)	Flow (Anode/Cathode, cc/min)
80	270	500 N_2/1,000 N_2

Cycle between dry feed gas and humidified feed gas (sparger bottle temp = 94˚C)
Dry feed gas hold time: 15 seconds
Humidified feed gas hold time: 5 seconds
For further protocol information, see: W. Liu, M. Crum, ECS Transactions **3**, 531-540 (2007)

CONCLUSIONS AND FUTURE DIRECTIONS

The combination of Gore's advanced materials, expertise in MEA manufacturing, and fuel cell testing with the mechanical modeling experience of University of Delaware and the heat and water management experience of University of Tennessee enables a robust approach to development of a new low-cost MEA manufacturing process.

Future work will focus on stack validation as well as accelerated stress testing to ensure that durability of the new, direct-coated MEAs is equivalent to or better than the current commercial control MEA.

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FY 2014 PUBLICATIONS/PRESENTATIONS

1. 2014 Hydrogen Program Annual Merit Review: mn004_busby_2014_o.pdf.

2. "Time-Dependent Mechanical Behavior of Proton Exchange Membrane Fuel Cell Electrodes" Z. Lu, M.H. Santare, A.M. Karlsson, F.C. Busby, P. Walsh, *J. Power Sources*, 245, p. 543-552 (2014).