VI.1 Fuel Cell Membrane Electrode Assembly Manufacturing R&D

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- 3M, St. Paul, MN
- Acumentrics, Westwood, MA
- BASF Fuel Cells, Somerset, NJ
- Delphi, Fenton, MI
- General Motors, Pontiac, MI
- Ion Power, New Castle, DE
- Proton OnSite, Wallingford, CT
- Ultra Electronics – Adaptive Materials Inc., Ann Arbor, MI
- W.L. Gore and Associates, Elkton, MD

Project Start Date: July 16, 2007
Project End Date: Project continuation and direction determined annually by DOE

Overall Objectives

- Evaluate and develop in-line diagnostics for cell and component quality control and validate diagnostics in-line.
- Investigate the effects of membrane electrode assembly (MEA) component manufacturing defects on MEA performance and durability to understand the required performance of diagnostic systems and contribute to the basis of knowledge available to functionally determine manufacturing tolerances for these materials.
- Use established models to predict the effects of local variations in MEA component properties, and integrate modeling of the operational and design characteristics of diagnostic techniques into the design and configuration of in-line measurement systems.
- These objectives have strong support from the industry. Specifically, the outcomes of the 2011 NREL/DOE Hydrogen and Fuel Cell Manufacturing R&D Workshop and the Office of Naval Research-funded Manufacturing Fuel Cell Manhattan Project confirmed the importance of continued development of in-line quality control techniques for cell manufacturing. Our specific development activities have been and will continue to be fully informed by direct input from industry. As new technologies emerge and as the needs of the industry change, the directions of this project will be adjusted.

Fiscal Year (FY) 2013 Objectives

- Prove the feasibility of the through-plane infrared/direct current (IR/DC) diagnostic using moving sheet material
- Prove the feasibility of optical reflectometry for detection of surface defects on solid oxide fuel cell (SOFC) tube cells
- Go/No-go decision for further work to implement the IR/reactive flow-through (RFT) technique on the web-line.

Technical Barriers

This project addresses the following technical barriers from the Manufacturing R&D section (3.5) of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan:

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

(E) Lack of Improved Methods of Final Inspection of MEAs

(K) Low Levels of Quality Control

Contribution to Achievement of DOE Manufacturing R&D Milestones

This project contributes to the achievement of the following DOE milestones from the Manufacturing R&D section (3.5) of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan:

- 6.2. Develop defect detection techniques in pilot scale applications for manufacturing MEAs and MEA components. (4Q, 2013)
- 6.3 Establish models to predict the effect of manufacturing variations on MEA performance. (4Q, 2014)
- 6.4. Demonstrate methods to inspect full MEAs and cells prior to assembly into stacks. (4Q, 2014)

FY 2013 Accomplishments

- Demonstrated detection of surface defects and morphology in electrode layers, in both catalyst-coated membrane (CCM) and gas diffusion electrode (GDE) configurations, using our optical reflectometry technique.
- Demonstrated the detection of surface defects in tubular SOFC cells on our bench-top motion stage using our optical reflectometry technique.
- Proved the feasibility of using our infrared/reactive flow-through (IR/RFT) technique to detect thermal responses from GDEs in open-environment, moving substrate operation with a prototype air knife.
- Demonstrated improved detectability and response time of our IR/RFT technique.
- Demonstrated the through-plane IR/DC technique to detect shorting through the membrane using two-side coated CCM sheets.
- Performed modeling of IR/DC and IR/RFT process variables and integrated modeling results into further improvement of the techniques.
- Performed feasibility studies of a new capacitance-based diagnostic aimed at measurement of ionomer-to-carbon ratio in electrodes.
- Used segmented and single cell testing to understand how defects in CCM catalyst layers affect local (spatially resolved) and total cell performance as well as failure development under accelerated aging.
- Developed a new cell hardware and methods for quasi-in situ spatial detection of failure development.
- Completed a detailed comparison of in situ performance between thermal and ultrasonically pressed proton exchange membrane MEAs.
- Continued collaboration with our industry partners, including three of DOE's competitively awarded manufacturing R&D projects, in accordance with our project charter.

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INTRODUCTION

In FY 2005–2007, NREL provided technical support to DOE in developing a new key program activity: manufacturing R&D for hydrogen and fuel cell technologies. This work included a workshop on manufacturing R&D, which gathered inputs on technical challenges and barriers from the fuel cell industry, and subsequent development of a roadmap for manufacturing R&D. In late FY 2007, NREL initiated a project to assist the fuel cell industry in addressing these barriers, initially focusing on in-line quality control of MEA components. The project is utilizing the unique and well-established capabilities of NREL's National Center for Photovoltaics for developing and transferring diagnostic and process technology to the manufacturing industry.

Defects in MEA components differ in type and extent depending on the fabrication process used. The effects of these defects also differ, depending on their size, location in the cell relative to the reactant flow-field, cell operating conditions, and which component contains the defect. Understanding the effects of these different kinds of defects is necessary in order to specify and/or develop diagnostic systems with the accuracy and data acquisition/processing rates required for the speed and size scales of high-volume continuous manufacturing methods. Furthermore, predictive capabilities for manufacturers are critical to assist in the development of tolerances and to enable assessment of the effects of material and process changes.

APPROACH

NREL and its partners are addressing the DOE manufacturing milestones listed above by evaluating, developing, and validating (in-line) diagnostics that will support the use of high-volume manufacturing processes for the production of MEAs and MEA component materials. Prioritization of this work is based on inputs from our industry partners on their critical manufacturing quality control needs. We are focusing on diagnostic capabilities not addressed by commercially available in-line systems; in particular we are evaluating methods to make areal rather than point measurements such that discrete defects can be identified. We are also developing test methodologies to study the effects of the size and/or extent of each important type of variability or defect. These results will assist our industry partners in validating manufacturing tolerances for these materials, ultimately reducing scrap rates and cost, and improving supply chain efficiency. Finally, predictive models are being used at LBNL to understand the operational and design characteristics of diagnostic techniques by simulating the behavior of MEA components in different excitation modes. These results are being fed back to our design effort in configuring the diagnostics for in-line implementation. MEA models are also being utilized to understand the in situ behavior of defect MEAs to guide and further elucidate experiments.

RESULTS

Over the past year we advanced the capabilities and applicability of previously developed techniques, explored new techniques, and continued to advance the understanding of the effects of defects in an operating cell. We proved that our optical reflectance diagnostic is very sensitive to both variability and morphology of electrode surfaces, despite the black, low contrast nature of the surfaces. Figure 1 shows an example of a GDE with undispersed fiber bundles creating high spots and creases on the electrode surface. We also extended our previous work on planar SOFC cells and showed that optical reflectance can be used to detect surface defects in tubular cells.

In work to advance our infrared-based techniques, we successfully transitioned the IR/RFT technique from a totally enclosed measurement of a small sample to a measurement in an open environment, which would be required for an in-line measurement of a sheet or continuous web of GDE. We implemented these advancements on our bench-top roller system to demonstrate the operation of the technique on a moving (10 foot per minute) sheet of GDE. Figure 2 shows the roller system with a GDE sheet and the air knife used to impinge the reactive gas mixture onto the electrode surface. In addition, using a combination of both experimental and numerical methods, we and our partners at LBNL predicted and experimentally proved increased detection sensitivity and reduced response time by modifying the hydrogen concentration in our nonflammable excitation gas mixture. LBNL also continued modeling the excitation geometry of the IR/DC technique and showed that detection sensitivity to thin cracks, which were experimentally determined to be the most difficult type of defect to detect, could be greatly improved by application of a nonuniform electrical excitation.

In more developmental work, we proved the feasibility of using an ex situ capacitance technique to indicate relative changes in the ratio of ionomer to carbon in the electrode – an important parameter to cell performance. Continuing studies will address improving the sensitivity of the technique to small ionomer-to-carbon variations. We continued our studies of the effects of electrode bare spots, identifying a threshold of size below which total cell performance is not affected. Figure 3 shows a comparison of performance curves for electrodes with and without defects as well as

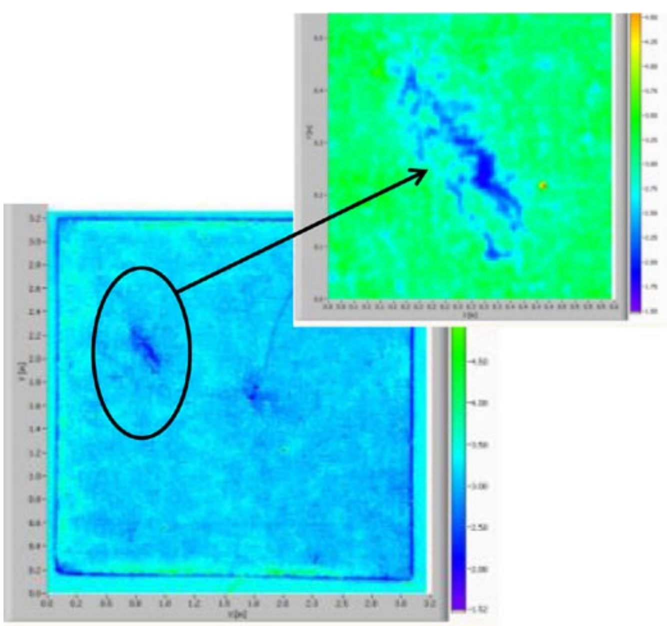


FIGURE 1. Optical reflectance images of surface irregularities in a GDE due to undispersed fiber bundles.

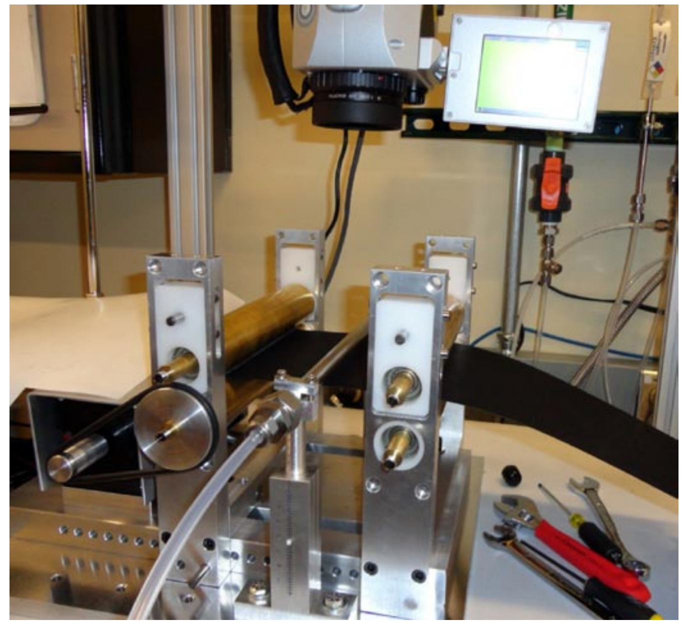
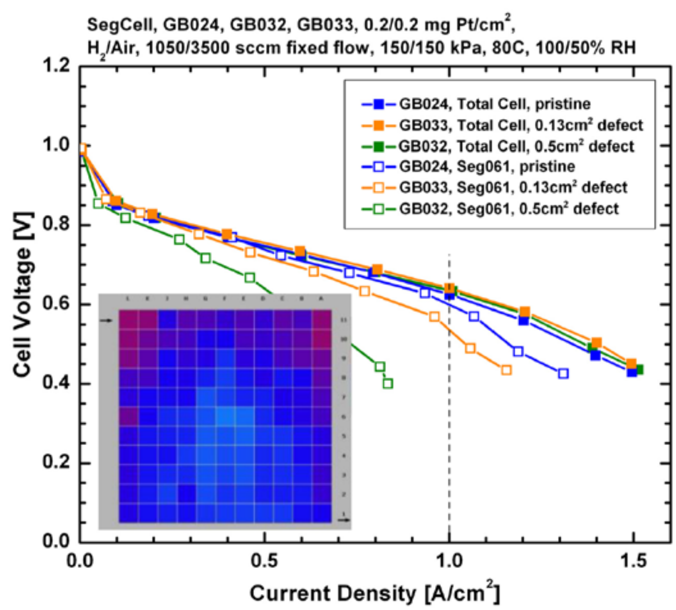


FIGURE 2. IR/RFT setup showing bench-top roller system, GDE sheet material, air knife to impinge reactive gas onto the GDE surface, and infrared charge-coupled device.



RH - relative humidity

FIGURE 3. Polarization data for a pristine and two defect MEAs showing that, while the performance at the location of the electrode bare spot (open symbols) varies greatly, the total cell performance (filled symbols) is the same; inset shows spatially resolved voltage data for the MEA with a 0.13 cm^2 defect.

spatially resolved performance data for an electrode with a small defect. In addition, we developed a novel cell hardware that enables typical cell operation and in situ electrical and electrochemical diagnostics, but also allows us, using an infrared charge-coupled device detector, to spatially observe the development of failures in the cell resulting from these defects under accelerated aging conditions. Using this hardware, we showed that a bare spot in the electrode acted as the initiation location for a mechanical failure of the cell.

FUTURE DIRECTIONS

- Demonstrate the IR/RFT technique on the research web-line
- Determine the feasibility of using the capacitance technique as a rapid, sufficiently sensitive quality measurement for electrode ionomer-to-carbon ratio
- Continue to use predictive modeling and single and segmented cell test methods to assist diagnostic development
- Study the effect of as-manufactured defects on MEA lifetime using standard or modified accelerated stress tests
- Work toward the implementation of our techniques on industry production lines

FY 2013 PUBLICATIONS AND PRESENTATIONS

1. "Fuel Cell MEA Manufacturing R&D," M. Ulsh; DOE Fuel Cell Technologies Program Annual Merit Review; Washington, D.C.; May, 2013.

2. "Challenges to High-volume Production of Fuel Cell Materials: Quality Control," M. Ulsh, B. Sopori, N. Aieta, G. Bender; ECS Fall Meeting, Honolulu, HI; October, 2012.

3. "The Spatial Performance Effect of Electrode Defects in PEMFC," G. Bender, W. Felt, M. Ulsh; ECS Fall Meeting, Honolulu, HI; October, 2012.

4. "Fuel Cell Manufacturing Diagnostic Techniques: IR Thermography with Reactive Flow Through Excitation," A. Manak, G. Bender, M. Ulsh; ACS National Meeting, Philadelphia, PA; August, 2012.

5. "Effects of local variations of the gas diffusion layer properties on PEMFC performance using a segmented cell system," T. Reshetenko, G. Bender, K. Bethune, R. Rocheleau; Electrochimica Acta, vol. 80, p. 368-376, 2012.