VI.3 Reduction in Fabrication Costs of Gas Diffusion Layers

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- Ballard Power Systems, Burnaby, BC

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Objectives

- Reduce the fabrication costs of gas diffusion layer (GDL) products, and verify the design of a new production facility incorporating process technologies that meet automotive volume requirements at the DOE 2015 cost target.
- Demonstrate the means of achieving six sigma quality standards at high volume manufacturing.
- Develop and implement new, high volume GDL process technologies.
- Produce high performance GDLs for backup power fuel cells at lower cost in the near term.
- Research, develop, and implement new on-line process control and measurement tools consistent with high volume manufacturing.
- Advance the understanding of the relationships between process capabilities, GDL product properties, and fuel cell performance to direct process specifications that maximize production yields of high-performance, low-cost GDLs.

Technical Barriers

This project addresses the following technical barriers from the Manufacturing R&D section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

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

(F) Low Levels of Quality Control and Inflexible Processes

Contribution to Achievement of DOE Manufacturing Milestones

This project will contribute to achievement of the following DOE milestones from the Manufacturing R&D section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

- **Milestone 1:** Develop prototype sensors for quality control of MEA manufacturing. (4Q, 2011)
- **Milestone 2:** Develop continuous in-line measurement for MEA fabrication. (4Q, 2012)
- **Milestone 3:** Demonstrate sensors in pilot scale applications for manufacturing MEAs. (4Q, 2013)

Accomplishments

- Completed the design feasibility and scalability analysis of the new process technologies.
- Completed multi-layer coating pilot trials:
  - Identified the major process elements for control and measurement.
  - Established design parameters for the coating module.
  - Ascertained process modifications required for integration into the current manufacturing line.
  - Determined design specifications for the coating ink delivery system.
- Completed in-line mixing lab-scale trials:
  - Established the major process variables for control and measurement.
  - Determined major design parameters for the in-line mixer.
  - Verified continuous liquid and solid feed material delivery requirements.
- Identified strategic in-line ink mixing and delivery process control and measurement tools and established their respective evaluation criteria:
  - Down-selected sensor technologies for capability demonstration of in-line ink viscosity measurement.

  - Evaluated in-line particle size distribution technologies and completed lab-scale capability testing.
  - Identified three continuous solids feeder technologies and performed capability testing.
  - Evaluated feasibility of commercial in-line surface tension and bubble content technologies.
- Down-selected in-line substrate and GDL coating process control and measurement tools for assessment and development:
  - Collected first run, machine direction and cross-web scanning data using on-line coating weight tool.
  - Completed assessment of non-contact surface profilometry techniques and identified two candidate sensors potentially capable of in-line scanning substrate paper and coated GDL.
  - Established evaluation criteria and identified three systems for capability demonstration of on-line machine vision quality inspection, down-selecting one.
  - Established evaluation criteria and initiated assessment of in-line substrate and GDL thickness sensors.
- Assessed feasible techniques for on-line characterization of GDL chemical homogeneity and down-selected Raman and Fourier transform infrared (FTIR) spectroscopy for further research and development.
- Identified a high-resolution 3-dimensional (3D) imaging technique to provide information on the topology of the pore space and its changes during processing.
- Identified carbonization process parameters affecting final substrate paper morphology and requiring tighter control to reduce product variability.

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Introduction

This project addresses the Manufacturing R&D subprogram's goal of research, development and demonstration of technologies and processes that reduce the manufacturing cost of proton exchange membrane fuel cell systems. Specifically, this project reduces the fabrication costs of high-performance GDL products, increases their manufacturing rate and demonstrates the means of achieving six sigma quality standards at high volume manufacturing.

In this first year, the team began development of new, high volume process technologies for integration into the existing GDL manufacturing line. Under development are multilayer coating and in-line mixing and delivery processes. For the carbon paper substrate, introducing in-line mixing and delivery technology will increase carbon paper substrate product volumes and yields, and reduce raw material waste and product cost. For the GDL products, the incorporation of both multilayer coating and in-line mixing and delivery technologies will reduce the number of process steps, increase product volumes and yields, and reduce costs.

Research and development (R&D) of advanced in-line process control tools and measurement systems, consistent with high volume manufacturing, was also started. These tools are essential for ensuring product quality, integrating the new process technologies, decreasing overall scrap rates, and eliminating costly ex situ testing. To direct process specifications that reduce product variability, a study of the relationships between process capabilities, GDL product properties, and fuel cell performance was also initiated.

Approach

This project consists of three integrated technology development paths, conducted over a period of three years. Together, these technologies address the objective of high-quality, low-cost GDL roll goods near term, and form the foundation for verification of a new production facility incorporating new GDL manufacturing technologies that meet automotive volume requirements at the DOE 2015 cost target.

The primary path consists of the development and integration of new process technologies; specifically, multilayer coating and in-line mixing and delivery. The GDL manufacturing process at BMP is modular; thus, it is possible to replace effectively process components with improved equipment, add new functions, and add new technology. Initial multilayer coating and in-line mixing pilot trials are followed by process development on the manufacturing line. With manufacturing involvement on the development team, this approach ensures the manufacturability and compatibility of the new technologies, reduces the time to bring to full manufacturing capacity and provides for a seamless transfer of the technologies to production.

Key to the success of both new and existing GDL process technologies is that all of the process steps and support functions operate correctly in order to achieve high quality GDL products at low cost. On-line process control tools and measurement systems are critical to ensuring the process variables and product properties are on target. This project includes R&D of new in-line process control and measurement systems that are either: (1) required for the new process technologies; (2) required for improved process specifications to achieve six sigma or better quality standards; and/or (3) required for new control strategies arising from continued development of our understanding of the relationships between process variables, GDL properties and fuel cell performance.

To date, the linkage of GDL ex situ properties to predict fuel cell performance is an on-going area of development for the fuel cell industry. Many customers still require fuel cell performance benchmark testing to establish the performance window of GDL products. Given this gap in property-performance understanding, introducing new process technology presents a challenge in ensuring all critical process variables are identified. Consequently, work will be undertaken to develop new process specifications and establish critical-to-quality and critical-to-performance characteristics. Fuel cell screening performance testing will be conducted in combination with the development of new ex situ material characterization techniques to link process variables with product microstructure, properties and performance.

Results

1. High-Volume, Low-Cost Process Technology Development

A design feasibility and scalability analysis of the new process technologies was completed in Fiscal Year 2009. Analysis benchmarks established by BMP included product specification requirements, performance, cost, quality, timing/delivery, flexibility, equipment and maintenance requirements, automation techniques, spatial layout, and waste minimization strategies. The criteria addressed strategic, organizational, and technological factors.

The design of essential equipment and systems for the new process technologies was completed in FY 2009. Design specifications for the multilayer coating equipment were established, followed by integration requirements for the coating module placement into the manufacturing line. Coating ink requirements by the coater were cascaded to the ink delivery system. In conjunction with the ink delivery system requirements, the in-line ink mixer specifications were established. Finally, the continuous liquid and solids feed-rates were specified from the in-line mixer raw material demand.

To establish the design requirements of the multilayer coater, coating module and ink delivery system, a pilot trial was conducted at the coating equipment manufacturer's laboratory. The major process elements that affect material variability and process accuracy were determined and translated into design specifications for the module integration, process variables requiring on-line control and measurement, and design specifications for the ink delivery system. The basic solution coating parameters, and the need to combine drying process optimization with solution modification were established. The coating inks are non-Newtonian fluids thus the design of both the coater and ink delivery system must accommodate this behavior.

An evaluation of in-line mixing technology was completed in FY 2009. Important considerations for the evaluation of the mixer design included: incorporation and wetting out of powders, ink holdup, mixing shear rate range and control, bubble generation, and ink flow pulsation. Key selection criteria were the design of the mixing chamber and the speed and configuration of the mixing elements being easily modified to fine-tune the mixing process. The ability to modify the mixing setup is important for processing a variety of products, consistent with delivering flexibility to current GDL manufacturing practice. A pilot trial was performed in collaboration with an in-line mixer manufacturer. Basic process parameters were examined during the trial, and process variables requiring control and measurement were identified. Liquid and solids delivery requirements were established with consideration to the ink holdup/mixer residence time for variable production rates.

The evaluation of continuous solid feeders is continuing, with the design of the feeders driven by the properties of each solid raw material. These include volumetric and gravimetric screw, vibrating, rotary valve, belt, agitator, and disc feeders. Different delivery system configurations have been evaluated to reduce cost and improve efficiency. Physical and rheological properties of the liquid feed materials were measured under various temperatures and concentrations. Accurate, low shear, and pulse-less delivery of the liquid raw materials is directing equipment selection.

2. Development of In-line Process Control Tools and Measurement Systems

The selection of on-line process control and measurement tools and techniques for research and development was initiated in FY 2009. The tools selected were identified as being required to: ensure process variables and product properties of the new process technologies are on target; improve process specifications at high volume to achieve six sigma or better quality standards; reduce/eliminate costly ex situ characterization sampling and testing; and/or address new control strategies arising from continued development of our understanding of the relationships between process variables, GDL properties, and fuel cell performance. The primary selection requirement for commercial tool evaluation was their ability to be developed and adapted for high volume GDL manufacturing and to be verified and validated with known GDL ex situ test techniques.

A survey of measurement techniques that have the potential to characterize GDL homogeneity on-line was conducted. Early challenges for these techniques with respect to GDL materials characterization include: GDL surface roughness and subsequent errors in quantitative assignment, micro- versus macro-spatial resolution, bulk versus surface measurement characteristics, and interference/noise from other constituents in the substrate and sublayers not under detection. Raman and FTIR spectroscopy were selected as viable on-line techniques to measure the chemical composition of GDLs. Raman spectroscopy was capable of detecting poly-tetrafluoroethylene (PTFE) and carbon in the GDL while infrared (IR) spectroscopy was only capable of detecting PTFE. Figure 1 shows the spatial distribution of PTFE on the surface of a GDL obtained with Raman spectroscopy. Chemical mapping of carbon was possible through Raman spectroscopy but not through FTIR because of the absorbance of IR radiation by carbon. Both of these chemical surface-mapping techniques will be pursued because it is possible for FTIR spectroscopy to provide complementary information that Raman spectroscopy cannot. Specifically, water can be detected easily with FTIR but not with Raman.

Four classes of non-contact surface topography measurement techniques were evaluated for the on-line surface profile analysis of substrate paper and coated GDLs. The assessed techniques included confocal microscopy, interferometry, triangulation laser, and scattered light. A comprehensive review was conducted that examined the merits and limitations of the techniques. Capability testing of three down-selected instruments was initiated, with one sensor identified to date. The tool delivered International Organization for Standardization (ISO)-defined 2-dimensional (2D) profiles, 3D surface topography, and 2D and 3D parameters in accordance with international standards using advanced ISO 16610 filtering techniques. Long run scan 2D profiles at scanning speeds representative of the web velocity were also demonstrated. Figure 2 exemplifies the measurement data received from this tool. A 2D roughness profile obtained by scanning the surface of a GDL is shown in 2(a). The ability to resolve a rough surface is shown in 2(b), the 3D surface topology of a substrate paper.

Evaluation and procurement of an on-line GDL coating weight measurement system was completed in FY 2009. The system was installed and first run coating weight data was collected down- and cross-web using the scanning sensors. Cross-web scanning at target web speed confirmed in-line measurement capability.

A key in-line product quality tool under development in FY 2009 was a machine vision inspection system capable of real-time monitoring of the coating processes. The system is expected to monitor line start-up and changeover, detect process upsets and catastrophic defects in-process, provide input signals for process control, and enable higher line speeds. Sets of representative defective samples were assembled and distributed to system manufacturers for capability testing and identification of system hardware configurations.

Assessment of commercial non-contact thickness sensors and moisture content tools continues. Thickness measurement methods that are under evaluation include: optical, single side laser triangulation, and dual laser triangulation. Available sensors will be tested on-line for robustness, resolution, accuracy, repeatability, range, and sampling speed. A potential moisture sensor technology employing near IR reflectance/transmittance

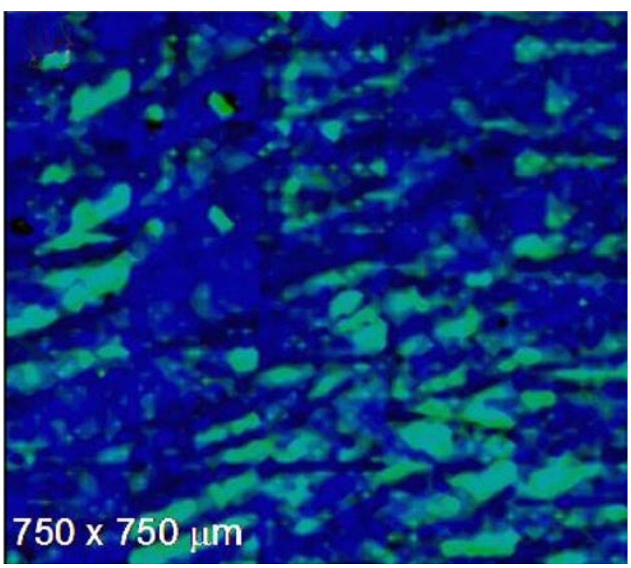

FIGURE 1. 2D spatial distribution of PTFE on the surface of a GDL, with lighter areas showing higher concentrations.

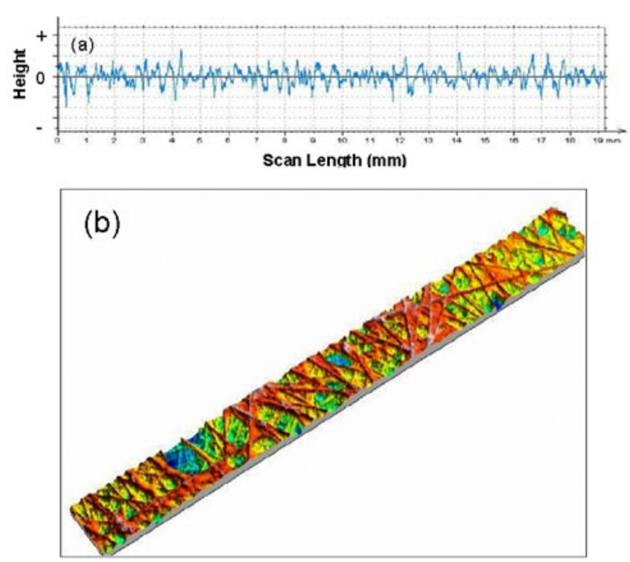

FIGURE 2. (a) Representative 2D roughness profile; and (b) 3D surface topology of rough substrate paper.

spectroscopy was identified. Due to the large amount of carbon in the materials, the resulting absorbance of IR radiation by carbon made it difficult to measure the moisture content accurately. Signal modifiers that have previously been used with carbon are being evaluated.

Particle size distribution measurement techniques utilizing laser light scattering, electrical sensing zone method, and ultrasonic spectroscopy (acoustic scattering) were evaluated. The difficulty in using a laser light scattering system was the need to dilute the inks, risk of particle agglomeration and potentially skewed distributions. Similarly, the electrical sensing zone method required the dilution of the inks and is subject to agglomeration. Ultrasonic spectroscopy, a new technology capable of analyzing concentrated multi-component mixtures in-line without the need for dilution, was selected for capability testing. The accuracy of the resulting size distributions was dependent upon the accuracy of the volume concentration of solids. Consequently, assessment of the particle size distribution accuracy will be evaluated in conjunction with the accuracy of ink solids content arising from the solids and liquid delivery and in-line mixing processes.

In-line viscosity measurement techniques under evaluation include rotational, falling piston, acoustic, and electromagnetic viscometers. The shear rates of the in-line sensors are generally constant; however, the shear rates in the coating processes are variable and the viscosity-shear rate relationship of the coating inks is non-linear. Although variable shear rate viscometers measure viscosities at the shear rates of interest over a wide range of temperatures, they are complex and not adaptable for in-line operation in this application. Consequently, variable shear rate instruments will be utilized for process development and fixed shear rate viscometers will be installed on-line, and the sensors strategically placed with a temperature control unit.

3. Development of GDL Process Parameter, Product Property, and Fuel Cell Performance Relationships

A substrate carbonization process study combining thermogravimetric analysis with lab-scale experiments and production plant trials was conducted in FY 2009 to investigate the influence of process steps on the final substrate carbon morphology, conditions for increasing thermo-oxidation/stabilization and carbonization line speeds, and to optimize current process conditions. Process parameters affecting the final substrate morphology were identified and procedural measures have been instituted to reduce product variability. During carbonization, the heating rate determines the evolution rate of volatiles, which affects the formation of pores. Lower heating rates are preferable to produce small pores and increase carbon crystallinity. Higher heating rates may lead to formation of pinholes, cracks, blisters and distortions. In addition, higher gas flow rates increase pore size by facilitating removal of volatiles and preventing condensation/sooting reactions. To monitor the topology of the pore space and its changes during processing, high-resolution X-ray tomography (HRXRT) scanning was investigated. Figure 3 shows the results of the 3D imaging of P75 substrate containing PTFE. Figure 3(a) shows the raw image of a 2.51 μm HRXRT slice; and 3(b) is representative of the 3D reconstruction using an advanced visualization program. High-resolution permits the detection of small features required to correlate process parameters, pore structure characteristics and fuel cell performance. The development of quantitative processing steps to describe the porous structure, quantify PTFE and carbon in the solid phase, and introduce and quantify water distribution is continuing.

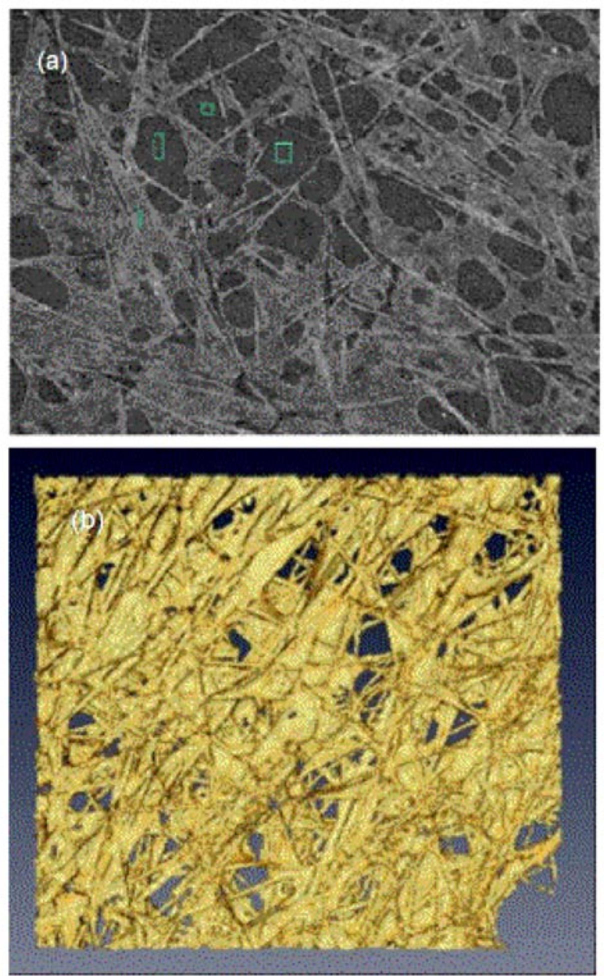

FIGURE 3. (a) HRXRT raw image slice of P75 substrate paper containing PTFE; and (b) Subsequent 3D reconstruction using advanced visualization program.

Conclusions and Future Directions

In FY 2009, this project launched three integrated technology development paths, namely (1) the development and integration of new, high volume process technologies; (2) the research and development of advanced in-line process control tools and measurement systems, consistent with high volume manufacturing; and (3) the development of process control strategies resulting from continued understanding of the relationships between process variables, GDL properties and fuel cell performance. In this first year of this project, the team:

- Completed multilayer coating and in-line mixing pilot trials, and established key design parameters for the equipment and process integration.
- Identified strategic in-line ink mixing and delivery process control and measurement tools, and established their respective evaluation criteria.
- Down-selected in-line substrate and GDL coating process control and measurement tools for assessment and development.
- Down-selected Raman and FTIR spectroscopy for further R&D of on-line characterization of GDL chemical homogeneity.
- Identified a high-resolution 3D imaging technique to provide information of the topology of the pore space during processing.
- Identified carbonization process parameters affecting final substrate paper morphology and requiring tighter control to reduce product variability.

In the next year, the future directions of this project include:

- Complete fabrication, installation and commissioning of new process technologies that include: multilayer coating module, coating ink delivery systems, in-line mixer and continuous solids and liquid delivery equipment.
- Complete evaluation, purchase, and on-line implementation of advanced process control tools and measurement systems that are under development.
- Complete substrate carbonization process study and introduce new process control procedures.
- Continue development of quantitative processing steps for HRXRT data to link GDL microstructure with material properties and fuel cell performance.
- Continue research and development of Raman and FTIR spectroscopy for on-line chemical homogeneity measurement of GDLs.
- Continue development of process variable, product property, and fuel cell performance relationships to direct new process specifications:
  - Build product process parameters-product properties relationships.
  - Conduct single cell fuel cell GDL screening performance testing.
  - Establish critical-to quality and critical-to-performance characteristics.

FY 2009 Publications/Presentations

DOE Hydrogen Program and Vehicle Technologies Program Annual Merit Review and Peer Evaluation Meeting, Washington, D.C., May 20, 2009.

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