VI.5 High Speed, Low Cost Fabrication of Gas Diffusion Electrodes for Membrane Electrode Assemblies

Emory S. De Castro
BASF Fuel Cell, Inc.
39 Veronica Avenue
Somerset, NJ 08873
Phone: (732) 545-5100 ext 4114
Email: Emory.DeCastro@BASF.com

DOE Managers
HQ: Nancy Garland
Phone: (202) 586-5673
Email: Nancy.Garland@ee.doe.gov
GO: Jesse Adams
Phone: (720) 356-1421
Email: Jesse.Adams@go.doe.gov

Contract Number: DE-EE0000384

Subcontractor:
Dr. Vladimir Gurau
Case Western Reserve University, Cleveland, Ohio

Project Start Date: July 1, 2009
Project End Date: June 30, 2013

Fiscal Year (FY) 2012 Objectives

- Reduce cost in fabricating gas diffusion electrodes through the introduction of high speed coating technology, with a focus on materials used for the high-temperature membrane electrode assemblies (MEAs) that are used in combined heat and power generation (CHP).
- Relate manufacturing variations to actual fuel cell performance in order to establish a cost-effective product specification.
- Use advanced quality control methods previously developed to guide realization of these two objectives.

Technical Barriers

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

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

(F) Low Levels of Quality Control and Inflexible Processes

Contribution to Achievement of DOE Manufacturing R&D Milestones

This project will contribute to achievement of the following DOE milestones from the Manufacturing R&D section of the Fuel Cell Technologies Program Multi-Year Research, Development and Demonstration Plan (Section 3.5.7):

- Develop continuous in-line measurement for MEA fabrication. (4Q, 2012)
- Establish models to predict the effect of manufacturing variations on MEA performance. (4Q, 2013)

This project addresses coating speed and uniformity of gas diffusion electrodes (GDEs), a critical component for MEA fabrication. One sub-task is to develop a continuous X-ray fluorescence (XRF) analyzer that directly measures catalyst deposition level and distribution on rolled goods, ultimately guiding improvements in through-put and uniformity. This sub-task directly contributes to the forth quarter 2012 goal for in-line measurement. Another sub-task is to develop models that predict the effect of manufacturing variations in catalyst distribution and porosity in GDEs, and relate these variations as six-sigma limits for a component specification. The establishment of a model that predicts MEA performance based on manufacturing variations in GDEs contributes to improving the quality of the component as well as achieving the fourth quarter 2013 DOE milestone above.

FY 2012 Accomplishments

- Reduced labor cost to manufacture GDE further from last year's 50% to 75%.
- Scaled up all ink preparations >10-fold.
- Exceeded project goal of three-fold increase of throughput for cloth-based GDEs at production scale.
- Demonstrated path to further cost reductions for GDEs on a lower cost non-woven web.

Introduction

The basis of this project is to create gas diffusion electrodes at a far lower cost than those currently available. GDEs are critical components of membrane electrode assemblies and represent the highest cost subcomponent

of the MEA. Cost reduction will be accomplished through development of a higher throughput coating process, modeling the impact of defects due to the higher speed process, and overcoming these limitations and providing a six-sigma manufacturing specification that relates performance to defects. The main focus of the effort is creating next-generation inks through advanced additives and processing methodologies. As part of our approach, we will also develop on-line quality control methods such as determination of platinum concentration and distribution during the coating process. The on-line mapping of platinum will guide the ink development process and provide feedback on uniformity.

For this reporting period we applied last year's understanding to scale up our approach to preparing stable inks of carbon black or catalyzed carbon black and hydrophobic binder. The use of a hydrophobic binder is critical to create GDEs for high temperature MEAs. In this period we significantly scaled up and further improved the inks. We demonstrated that these more concentrated inks are able to reduce the number of applications needed per pass. The improvements noted last year (increase of throughput, superior catalyst utilization, improved surface quality) were maintained in this period's scale up efforts. We also continued our efforts at ink development for the even lower cost non-woven carbon substrates.

Approach

GDEs are comprised of a gas diffusion layer coated with catalyst. The gas diffusion layer is simply carbon cloth or a non-woven carbon that has been coated with carbon black and serves as a current collector for the catalyst. For both the carbon black and catalyst, a hydrophobic binder is added to achieve critical porosity and hydrophobicity in the final structure. Of the carbon black, catalyst, or hydrophobic binder none are soluble in aqueous solutions. Aqueous solutions must be used as solvents since the use of organic solvents with a highly active catalyst is too dangerous in a production environment. Also, the hydrophobic binder is shear-sensitive, meaning it becomes less stable when pumped or subjected to shear forces in the coating applicator. Thus, the challenge in this project is overcoming the inherent physical limitations in these materials through advanced formulations and processing.

Our approach to solving this challenge begins with identifying key quality GDE metrics that relate directly to ink performance, develop an understanding of the forces behind ink stability, and introduce solution measurement methods that relate ink performance to the quality metrics. With more stable ink formulations, we anticipate being able to coat longer and wider webs at higher speeds. If an ink can be made more concentrated and remain stable we can use less application passes and save cost. The ink development process is supplemented by two other activities that ultimately lead to lower cost GDEs. We developed a model that will predict the impact of manufacturing variations on MEA performance, and used this model to determine the level of coating quality needed to maintain consistent current and voltage. Also, we created on-line instruments to lead development of more precise coating processes.

Results

Ink Scale Up

Last year we reported on a combination of additives and processing that allowed us to create a superior carbon-binder ink for the microporous layer (MPL). We were able to reduce the number of passes needed to form the MPL and created an improved surface. Upon scaling this preparation to support our final goal of coating cloth widths >1 meter and lengths ~300 linear meters, we developed new methods to safely introduce high volumes of carbon that led to severe bubble formation. This increase in bubbles led to variations in viscosity as the bubbles slowly degassed. We identified a defoaming approach that solved this issue and thus increased the preparation scale. Table 1 summarizes our efforts on ink scale up.

TABLE 1. Ink Scale-Up Metrics

	Cost Decrease vs. benchmark	Capacity increase vs. benchmark
MPL	37%	3.0X
Anode	31%	2.1X
Cathode	40%	2.4X

In Table 1, "Cost Decrease" represents the reduction of labor-hours to make the ink. "Capacity increase" indicates the total time saved to create a batch of ink now using larger scale preparation equipment.

Cathode and Anode Electrode Layers

Last year we reported on improvements for both the anode and cathode ink preparations. At that time, these improvements had not been evaluated at a full length of carbon cloth nor a full width. These two conditions place the maximum challenge on ink stability. Also, materials made under this project were sent as MEAs to a major supplier of micro-combined heat and power (μ-CHP) stationary power systems. From this evaluation we confirmed with large-scale format MEAs the performance and quality gains shown in our lab, but also learned the overall thickness was less than the commercially accepted product. We modified the MPL architecture based on this feedback to move the average thickness within range. Table 2 provides an overview of key metrics achieved at the full length-width scale. These metrics

were also achieved with a 4-fold increase in throughput due to: increase in solids content of inks and reduction in number of coating passes, more than doubling the width of the carbon cloth, and increasing the speed of some of the application steps.

TABLE 2. Performance Metrics at >300 Linear Meter Length and >1 Meter Width Carbon Cloth

	Benchmark at Start	This Project
Agglomerates (avg. over roll)	18/m^2	1.6/m^2
Pt variation (via on-line XRF, roll average)	+/- 2.0 gm Pt/ m^2	+/-0.4 gm Pt/ m^2
Performance		
0.2 A/cm^2 H_2/air, 45 cm^2 test cell, 160°C	0.657 V	0.683 V
0.5 A/cm^2 H_2/air, 45 cm^2 test cell, 160°C	0.573 V	0.598 V
0.2 A/cm^2, 1.4/5 Reformate (71% H_2, 27% CO_2, 2% CO)/Air, 45 cm^2 test cell, 180°C	0.668 V	0.689 V
0.5 A/cm^2, 1.4/5 Reformate (71% H_2, 27% CO_2, 2% CO)/Air, 45 cm^2 test cell, 180°C	0.571 V	0.589 V

Conversion of Lower Cost Substrates

A second major focus of this project is to develop lower cost substrates into gas diffusion electrodes. Non-woven carbon fiber materials ("carbon paper") are believed to be ~30% lower in cost compared to the carbon cloth at higher volumes. The porosity, hydrophobicity, and absorption properties of the carbon papers are totally different than carbon cloth, and we had to develop an entirely new class of inks for MPL, anode electrode layer, and cathode electrode layer. We continue our work in this area with a goal of further increasing the solid content beyond that of the carbon cloth inks and subsequently reducing again the number of application passes.

Conclusions and Future Directions

We have successfully exceeded the project's target for carbon cloth substrates by gaining a four-fold increase in material throughput and a labor savings of ~75%. Advances from this project are being transitioned to our current production with a direct immediate impact on component cost and a release of a new product. Future directions for the remaining time will be on the non-woven substrates and a further reduction in cost.

Upcoming Focus

1. Demonstrate on the pilot scale a further 30% reduction in total cost of gas diffusion electrodes when using non-woven substrates.