

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

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Technologies Office Multi-Year Research, Development, and Demonstration Plan (Section 3.5):

- (A) Lack of High-Volume Membrane Electrode Assembly Processes
- (K) Low Levels of Quality Control

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 Office Multi-Year Research, Development, and Demonstration Plan (Section 3.5.7):

- (1.1) Develop continuous in-line measurement for MEA and MEA component fabrication. (4Q, 2012)
- (1.3) Develop processes for direct coating of electrodes on membranes or gas diffusion media. (4Q, 2014)
- (1.6) Demonstrate processes for direct coating of electrodes on membranes or gas diffusion media. (4Q, 2016)

This project addresses coating throughput and uniformity of gas diffusion electrodes (GDEs), a critical component for MEA fabrication. Achievement of this project's objectives would satisfy milestones 1.3 and 1.6, direct coating of electrodes on gas diffusion media. One sub-task is to develop a continuous X-ray fluorescence analyzer that directly measures catalyst deposition level and distribution on rolled goods, ultimately guiding improvements in throughput and uniformity. This sub-task directly contributes to the fourth quarter 2012 goal for in-line measurement.

FY 2013 Accomplishments

- Demonstrated "one pass" micro-porous layer (MPL) and catalyst coating on carbon paper
- Reduced labor and material costs >30% for paper structures compared to best cloth GDEs
- Demonstrated 30% reduction in platinum loading compared to best cloth-based anodes without losing performance
- All customers changed over to new, more uniform, higher performing product based on this work (Celtec® P1100W)



Overall 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.
- 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.

Fiscal Year (FY) 2013 Objectives

- Further decrease cost beyond that achieved with cloth substrates by developing paper substrates and single-pass applications.
- Exploit stable inks to promote platinum thrift with cloth-based substrates.

Technical Barriers

This project addresses the following technical barriers from the Manufacturing R&D section of the Fuel Cell

INTRODUCTION

The basis of this project is to create GDEs at a far lower cost than those currently available. GDEs are critical components of MEAs 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 while also providing a “six-sigma” based manufacturing specification. 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 further reduced the cost of manufacturing GDEs as applied to paper substrates, and exploited the ink stability to reduce precious metal in the anode of a cloth-based GDE. The basis of this project is to produce 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 demonstrated that the new stability in these inks affords more concentrated suspensions and thus, for the paper system, reduces the number of applications needed per pass to a single application for the MPL and a single application for the catalyst layer. For the cloth-based materials, we used these new inks and developed an alternative anode structure that shows improved utilization and thus reduced precious metal loading without loss in performance. Both these efforts support additional cost reduction.

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 as well as a gas distributor. 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 is soluble in aqueous solutions. Aqueous solutions as solvents are much preferred in order to reduce volatile organic emissions, and, more importantly, employing 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, developing an understanding of the forces behind ink stability, and introducing solution measurement methods that relate ink performance to the quality metrics. With more stable ink formulations, we have shown ability to coat longer and wider webs at higher speeds with a lower incidence of defects. Since the 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 an on-line analyzer to track the distribution of precious metal catalysts and guide more precise coating processes.

RESULTS

Conversion of Lower Cost Substrates

Last year we presented some early results on developing 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 and are anticipated to be more uniform with lower material defects. 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 the MPL, anode electrode layer, and cathode electrode layer. This year we demonstrated production scale coating of a single-pass MPL on paper and proof of principle for a single-pass catalyst coating at the pilot scale. Figure 1 illustrates this progress compared to the new carbon cloth-based materials developed in this project. Projecting cost savings from coating time and cost of the base materials, we exceed an additional cost reduction of 30% over the best carbon cloth systems developed in this project (for 3,000 5-kW systems).

Platinum Thrift on Cloth Substrates

Reducing the precious metal content in the MEA was not part of the original scope of this project. However, based on reviews of this project, it was suggested that the improved inks may lead to more uniform structures and thus greater utilization within the catalyst layer. We exploited properties of the new inks and developed a new anode structure that contains 30% less platinum, but performs the same or better than similar carbon-based anodes from this project. Figure 2 illustrates this point. At current above 0.3 A/cm², even with 2% CO in the reformat, there appears to be an improvement in mass transport due to these new structures.

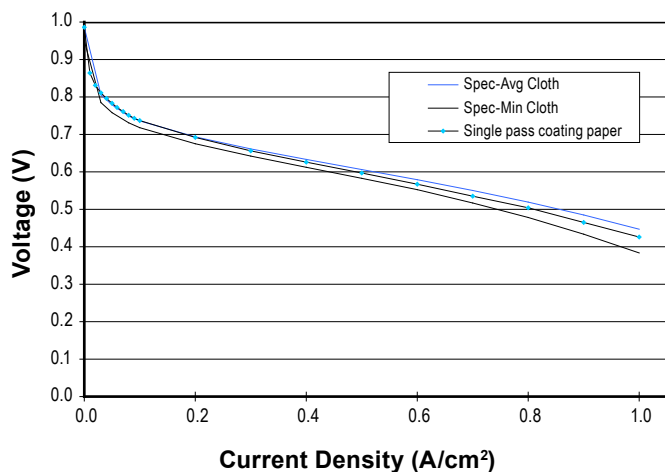


FIGURE 1. Comparison of single-pass applications (MPL and catalyst layer) on paper to multiple pass applications on woven materials from this project ("Spec Avg." & "Spec Min"), $T = 160^{\circ}\text{C}$, pressure = 1 bar_a , stoich: $1.2/2 \text{ H}_2/\text{air}$.

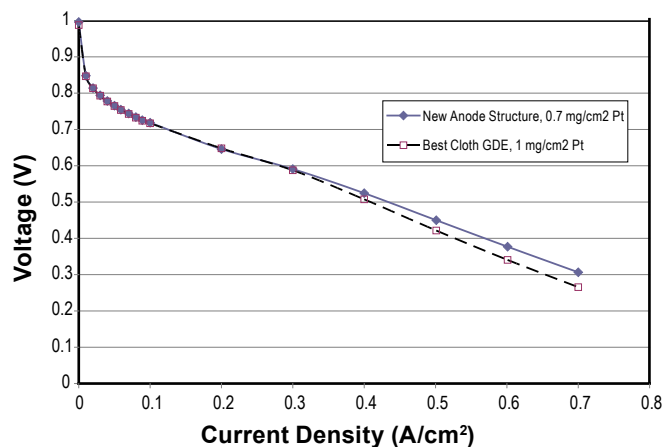


FIGURE 2. Comparison of new anode structure (lower precious metal) to best materials from this project, $T = 160^{\circ}\text{C}$, pressure = 1 bar_a , stoich: $1.2/2$ reformat air, reformat = $70\% \text{ H}_2$, $28\% \text{ CO}_2$, $2\% \text{ CO}$.

Create Specification Using "Six Sigma" Statistical Approach

One part of this project is to relate variations in performance to manufacturing variations, such as the distribution of precious metal. A basis for this approach is to test a statistically significant population of coating runs, and construct an average and process variation. Figure 3 shows the average $\pm 3\sigma$ of over 200 fuel cell tests on cloth-based cathodes developed in this project. For the first time, we are able to use statistical control processes to release the new product based on this project's advances, as opposed to selecting "high performers" at the cost of very low material yields. Figure 3 summarizes these results.

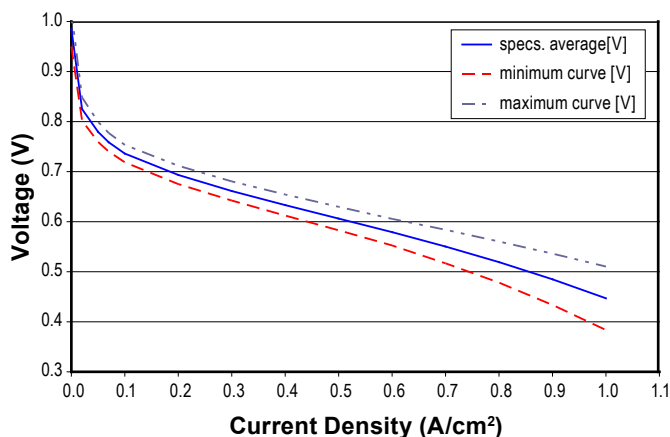


FIGURE 3. Cathode average $\pm 3\sigma$, $T = 160^{\circ}\text{C}$, pressure = 1 bar_a , stoich: $1.2/2 \text{ H}_2/\text{air}$.

CONCLUSIONS AND FUTURE DIRECTIONS

We have achieved DOE's milestone 1.6 for direct coating of catalyst on gas diffusion media and exceeded our project goal of three-fold improvement in material throughput for carbon cloth-based media. This improvement has led to a 75% reduction in labor cost to manufacture and produced very uniform, lower defect materials. A well-received new product (Celtec® P1100W) was launched based on this project. Through the gain in our understanding of formulating these materials in aqueous suspensions, we developed a new set of inks (MPL, anode catalyst, cathode catalyst) for use on carbon paper substrates and project an additional 30% savings over that achieved with cloth substrates. The development of an on-line X-ray fluorescence scanning instrument for measuring catalyst applications on gas diffusion media meets DOE milestone 1.1 – continuous in-line measurement for MEA component fabrication. Although our project concludes with this report, we will continue to work on cost reduction through developing a new product based on carbon paper substrates.

SPECIAL RECOGNITIONS & AWARDS/ PATENTS ISSUED

- 2013 DOE Hydrogen and Fuel Cells Program R&D Award for reducing the cost of manufacturing gas diffusion electrodes (May Annual Merit Review).

FY 2013 PUBLICATIONS/PRESENTATIONS

- Joerg Belak, "Improved Membrane Electrode Assemblies for High Temperature PEM Fuel Cells." ACS Meeting (Division of Polymer Chemistry): Advances in Materials for Proton Exchange Membrane Fuel Cell Systems, Feb. 17–20th, 2013, Asilomar Conference Grounds, Pacific Grove, California.