VI.7 High Speed, Low Cost Fabrication of Gas Diffusion Electrodes for Membrane Electrode Assemblies

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Fiscal Year (FY) 2011 Objectives

- Reduce cost in fabricating gas diffusion electrodes (GDEs) 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 (CHP) generation.
- Relate manufacturing variations to actual fuel cell performance in order to establish a cost-effective product specification.
- Develop advanced quality control methods 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.

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 GDEs, a critical component for MEA fabrication. 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 through-put and uniformity. This sub-task directly contributes to the fourth 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 2011 Accomplishments

- Reduced labor cost to manufacture GDE by 50%.
- Improved cathode catalyst utilization by ~25% through 20 mV gain in performance.
- Met key milestone of doubling throughput for GDE at production scale.
- Coated carbon cloth GDEs at >2X width – timing is ahead of milestone plan.
- Demonstrated pilot-scale fabrication of GDEs on a lower cost non-woven web.

Introduction

The basis of this project is to create GDEs 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 significantly advanced our understanding of the inherent limitations of suspensions of carbon black or carbon black catalysts mixed with a hydrophobic binder. The use of a hydrophobic binder is critical to create GDEs for high temperature MEAs. Through this project's understanding we developed improved inks that can be made more concentrated and thus reduce the number of applications needed per pass. These new inks also used substantially less time to prepare, and afforded greater utilization of the catalyst once in the MEA, improving performance. We also began to apply the ink development sequence to lower non-woven carbon paper cost.

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 highly 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 will develop a model that will predict the impact of manufacturing variations on MEA performance, and use this model to determine the level of coating quality needed to maintain consistent current and voltage. Also, we will create on-line instruments to lead development of more precise coating processes.

In this period, we combined next generation stabilizing additives with high energy dispersion techniques to greatly improve the inks, and scaled these through to the production coater.

Results

Microporous Layer

One significant problem was posed by the hydrophobic carbon used in the microporous layer (MPL). When benchmark inks and additives are used, this ink became less viscous as the shear rate increases. This shear-thinning behavior indicates strong particle-particle interactions and is shown as the trace labeled "baseline, low solids" in Figure 1. The shear-thinning becomes worse when we increased the solid content of the ink – desirable to reduce number of coating passes (see trace "baseline, high solids"). High shear-thinning indicates instability for the formulation as well as higher potential for non-uniformity under the shear forces of the applicator. By using a combination of additives and a high energy preparation method, we were able to create inks for the MPL that are Newtonian, that is, shear insensitive over a wide range. See the Figure 1 trace labeled "improved, highest solids." Note the solid content of this ink enabled >55% reduction in number of application passes needed, as well as a 50% reduction in time to prepare the ink. We scaled this MPL ink formulation to the production coater and demonstrated full width coating ahead of plan.

Cathode Electrode Layer

Last year we reported improvements in the cathode catalyst ink formulation that decreased the number of coating defects due to agglomeration and held potential to reduce the number of application passes by 30% when using our pilot-scale coater. We have now identified an

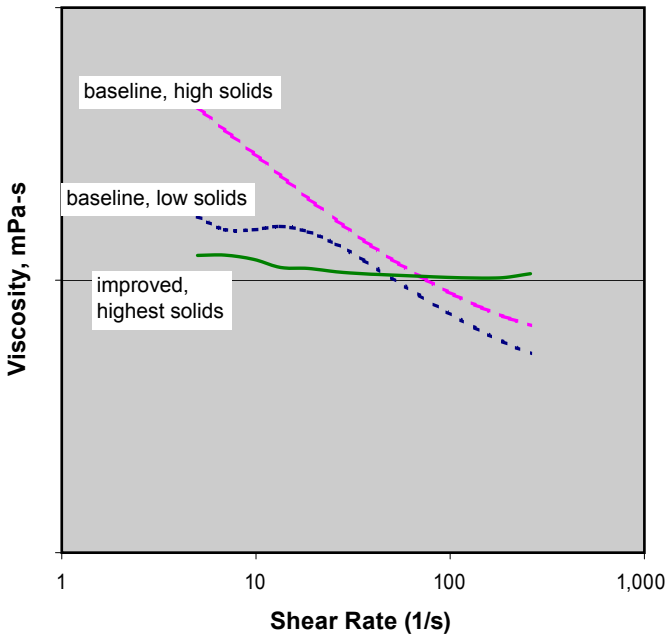

FIGURE 1. Viscosity vs. shear rate for three different ink formulations: baseline (low solids), baseline (high solids), and improved (highest solids).

improved additive system (several additives that work in concert) that not only stabilize higher concentrations of inks, but completely eliminated a labor-intensive step in ink making. We have scaled this improvement to our production scale coater as well as demonstrated a savings of >60% in labor hours for ink making and decrease in number of coating passes by 40%. Furthermore, these inks have shown a remarkable improvement in cathode performance as evidenced in Figure 2. We attribute the ~20 mV gain over baseline to an increase in platinum utilization through enhanced uniformity by 25%.

Anode Electrode Layer

A key difference between the anode and cathode electrodes is the catalyst. The cathode employs an alloy to facilitate oxygen reduction. Since the MEAs being developed under this project operate between 160°–180°C, they are highly tolerant to carbon monoxide (CO) and operate well at 1%-2% CO using only a pure platinum catalyst. Throughout this project we have adhered to the ink additive selection rules that are summarized as follows: select compounds that stabilize the ink components, leave no or minimal residue in the MPL or electrode layers, and do not disturb the intended porosity and hydrophobicity profiles. The anode catalyst is sufficiently different than the cathode that not only was a modified suite of additives employed, but extensive efforts were needed to develop a removal sequence to both minimize residues and preserve optimal hydrophobicity in the electrode layer. We were successful in developing an anode ink that also eliminated the labor-intensive step of the cathode, and also reduced ink preparation labor by 50% and decreased the time for coating passes by 40%. This ink has been scaled through to the full production coating machine.

Conversion of Lower Cost Substrates

A second major focus of this project is to develop lower cost substrates into GDEs. 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 the MPL, anode electrode layer, and cathode electrode layer. Although in the pilot-scale with limited coating lengths, we have established good progress. Figure 3 shows our status with a MPL plus cathode catalyst layer compared to the benchmark carbon cloth performance in hydrogen/air at 160°C. The inks developed for the carbon paper have been able to preserve the higher solid content of the carbon cloth inks and reduced number of application passes.

Conclusions and Future Directions

Through last year's foundation in understanding principle mechanisms behind ink destabilization and additive selection, we have been able to significantly improve the coatings of MPLs, anode catalyst layers, and cathode catalyst layers. While the objective of this project is to decrease cost by increasing throughput, we have also improved the overall process by decreasing the labor content for ink preparation by ~50%. We have scaled these new formulations to a full-

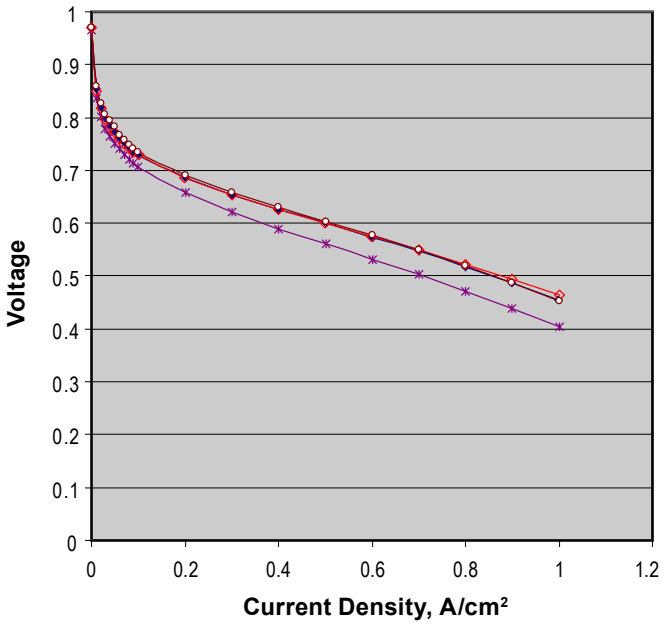

FIGURE 2. Voltage vs. current plot of baseline high temperature MEA vs. same with improved cathode catalyst layer at pilot and production coating scales. 160°C, H_2/air, 1.2/2, ambient pressure.

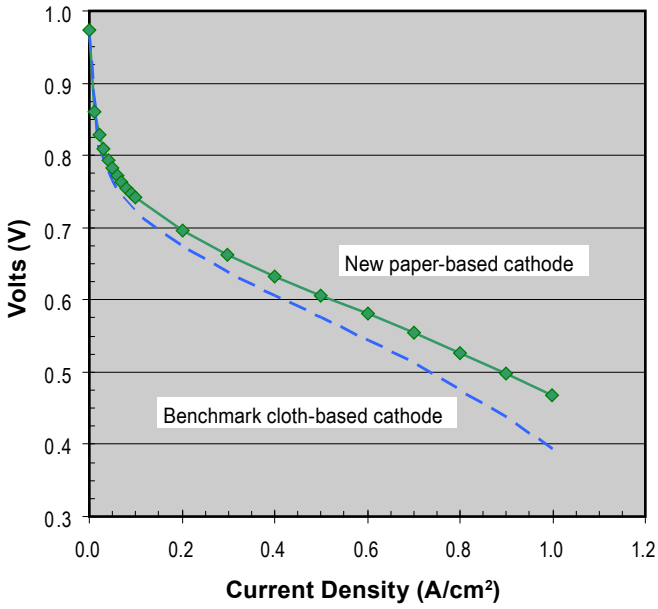

FIGURE 3. Voltage vs. current plot of baseline high temperature MEA vs. same made with carbon paper substrate at pilot coating scales. 160°C, H_2/air, 1.2/2, ambient pressure.

scale production coater, and achieved our key milestone of doubling throughput. A fringe benefit of the more uniform structures created by this approach is an improvement in performance by ~20 mV, inferring an increase in catalyst utilization by around 25%. Pilot-scale results on low cost non-woven substrates are encouraging and show a path for further decease of costs. Advances in this project are being transitioned to our current production techniques with a direct immediate impact on component cost.

Upcoming Focus

1. Develop full-width carbon cloth coating to gain another 2X throughput factor.
2. Qualify current formulations on full-width, full length production coating trials.
3. Develop formulations for non-woven carbon paper for a further reduction in cost: demonstrate on production coater.

FY 2010 Publications/Presentations

1. "Celtec®-P high temperature MEAs making a difference in power applications," F-Cell, September 2010, Stuttgart, Germany, Emory S. De Castro (Invited Speaker).