V.H.2 Visualization of Fuel Cell Water Transport and Performance Characterization under Freezing Conditions

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Objectives

- Improve fundamental understanding of the water transport processes in the proton exchange membrane fuel cell (PEMFC) stack components under freezing and non-freezing conditions.
- Optimize materials, design, and surface properties of gas diffusion layer (GDL) and bipolar plate to alleviate flooding and suppress regions of dehumidification.
- Develop experimental and modeling tools to evaluate ex situ and in situ performance of PEMFC stack components.

Technical Barriers

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

(A) Durability
(C) Performance
(D) Water Transport within the Stack

Technical Targets

This project is directed at developing a better water management strategy within PEMFC stacks. Insight gained from this study will be applied toward the design and demonstration of a robust fuel cell that meets the following DOE technical targets as outlined in the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

- Unassisted start from low temperature: -40°C
- Durability with cycling at operating temperature of ≤80°C: 5,000 h
- Energy density: 2 kW/L

Accomplishments

To date, we have completed characterization of the baseline system, with ex situ experiments and modeling at the component level (GDL and channels), and in situ testing to determine steady-state water distributions at low operating temperature, water content change during air purge, and start-up from freezing conditions:

- Determined the two-phase flow patterns, instantaneous flow distribution and pressure drop signatures of the baseline system on the ex situ multi-channel apparatus. Established the ex situ multi-channel performance matrix for the baseline system.
- Quantified the GDL intrusion (as much as 20% at compression of 2,068 kPa) and its effect on the flow maldistribution (flow rate variation of 70-130% from mean flow in individual channels).
- Determined the effect of contact angle on two-phase flow regime transition in flow channel.
- Developed a two-dimensional network model to investigate the water transport through the GDL and the complex physics of the liquid water interaction with the GDL.
- Demonstrated that water accumulation under low operating conditions occurs primarily within the anode channels, within the GDL on both the anode and cathode sides, at the transitions between flow field channels and plate manifolds, and at the plate edges within the manifolds.

- Determined the minimum cathode air purge for the successful freeze start. With purge times on the order of 30 seconds, a small pressure drop is observed and cell voltage can be attained. Two distinct regimes of water removal are observed for cathode air purges: convective removal of water slugs from the flow field channels and evaporative removal from the diffusion media and membrane electrode assembly (MEA).

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Introduction

Water management is critical to the successful implementation of PEMFCs in the automotive transportation sector. Water management is especially challenging under low-temperature conditions due to the low water carrying capacity of the reactant stream and the possibility of water freezing within the fuel cell assembly, which results in premature degradation. This project is directed at developing a better water management strategy within PEMFC stacks. To achieve these objectives, the project has been focused on the following technical issues:

- Two-phase (water and gas) transport in the fuel cell stack, including the GDL, flow channels, and their interfaces.
- Structural and surface properties of materials and how they change during operational events, such as freeze/thaw.
- Experimental and modeling tools to evaluate ex situ and in situ performance as well as local variations in current density and water distribution.

Approach

The objectives of this project will be accomplished through an iterative approach that starts at the component level, synthesizes this fundamental learning into combinatorial ex situ experiments with nearly full visual access, and then progresses to increasingly more complex in situ experiments that utilize advanced diagnostic methods such as current density and high-frequency resistance distributions and neutron radiography. Both experimental and modeling tools will be used to evaluate ex situ and in situ performance.

The success of the proposed project lies in new materials, improved design concepts and operating strategies. These can be combined to improve fuel cell performance through control of liquid water, mitigation of water accumulation and dehumidification, and suppression of the effects of freezing on start-up time and material degradation.

Results

In the past year, we focused on the complete characterization of the "baseline" system (GDL and flow channel), with ex situ experiments and modeling, in situ fuel cell testing, and freeze-thaw experiments. The GDL used was U-105 carbon fiber (Mitsubishi Rayon, Tokyo, Japan) which was wet proofed with polytetrafluoroethylene and coated with a miroporous layer "in-house" in General Motors Fuel Cell Activities. The cathode flow channel dimensions were 183 mm long, 0.7 mm wide, and 0.4 mm deep, with a land width of 0.5 mm between two adjacent channels. The anode channels were 183 mm long, 0.5 mm wide, and 0.4 mm deep with a land width of 1.5 mm. Both anode and cathode channels have a rectangular cross-section and a weavy shape with a 5° weaving angle to avoid mechanical shear on the GDL associated with straight channels. The GDL material and the flow channels offered excellent in situ fuel cell performance and meet the Department of Energy volumetric power density target for automotive fuel cell stack (2 kW/L).

The parametric studies on the component level were first conducted. For the flow channel, the effect of contact angle on two-phase flow regime transition was determined. In general, the transition from slug-annular flow to annular flow occurs at lower gas and liquid flow rates in non-wetting channels (contact angle = 110 degrees) than for wetting channels (contact angle <20 degrees). This result is important for understanding the pressure drop required for a successful flow field purge. For the GDL, the wettability (contact angle and contact angle hysteresis), structure (fiber diameter and orientation), and morphology (surface roughness and coating distribution) were experimentally measured. The transport property was modeled by a two-dimensional network model, which has been validated against in-plane percolation experiments. The three primary fingering patterns (stable displacement, viscous fingering and capillary fingering) generated in a GDL can be duplicated in the network model (Figure 1). Thus, the complex physics of the liquid water interaction with the GDL are captured and can be used to predict under what operating conditions flooding is expected as well as the location and migration of the water.

The multi-channel performance of the baseline system was characterized in an ex situ experimental setup. This setup enables the measurement of instantaneous flow rates in individual gas channels and the simultaneous visualization of the two-phase flow structures in flow channels. Remarkable GDL intrusion was observed and showed significant effect on the flow maldistribution. As much as 20% was obtained for the intrusion of the GDL into the flow channel (80 um out of 400 um) under compression of 2,068 kPa (300 psi), which leads to a flow rate variation of 70-130% from the

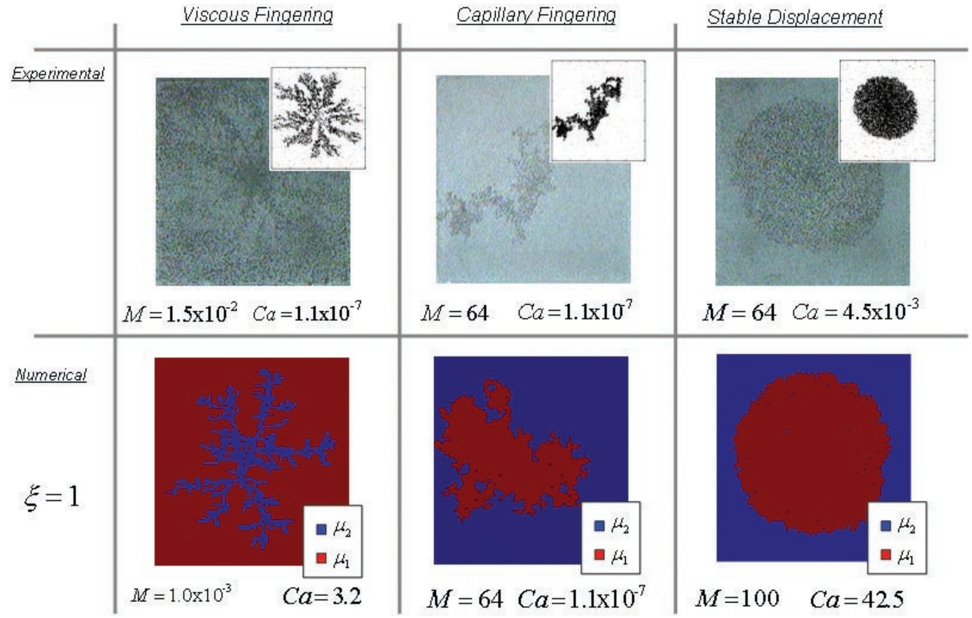

FIGURE 1. Experimental and Numerical Displacement Patterns in the GDL

mean flow in individual channels. The effect of water presence in the flow channels was then investigated. In general, two-fold effects were observed: one is to lead to severe flow maldistribution, the other is to result in a high water retention in channels. Figure 2 shows the variation of instantaneous flow rates in individual channels at a water flow rate of 0.04 mL/min and an air flow rate of 400 sccm (corresponding to a stoichiometric ratio of 3, commonly used in fuel cell operation). A flow rate sudden drop in channel six indicates the formation of a slug in this channel, which can last up to several hundreds of seconds. The flow structure visualization shows a large amount of water stays in the channels, especially at the channel end, which is difficult to remove by purging. Large fluctuations (in the magnitude of 0.05 kPa) in the pressure drop across the entire channel was observed for the slug flow. In addition to slug flow, film and mist flow are also observed in the flow channels. The flow pattern map for the baseline system is shown in Figure 3. The two-phase flow pattern, pressure drop signature, and the slug residence time are identified as important parameters to characterize the GDL-channel combination.

In situ fuel cell testing with neutron radiography was carried out to determine steady-state water distributions at low operating temperature, water content change

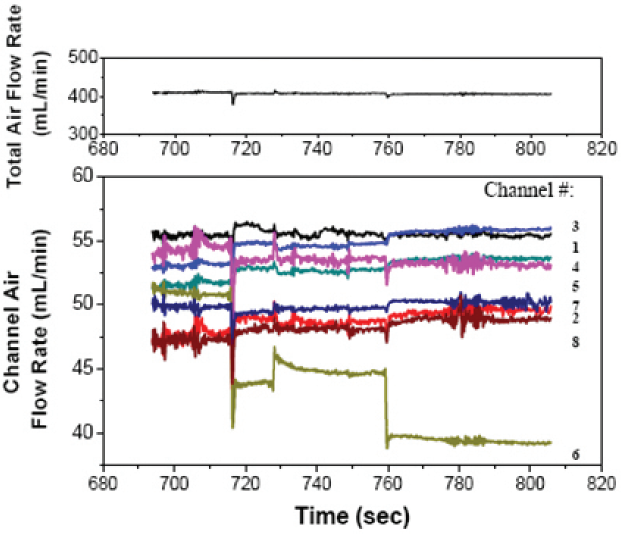

FIGURE 2. Instantaneous Flow Rate in Individual Channels at a Water Flow Rate of 0.04 mL/min and an Air Flow Rate of 400 sccm (corresponding to stoichiometric ratio of 3)

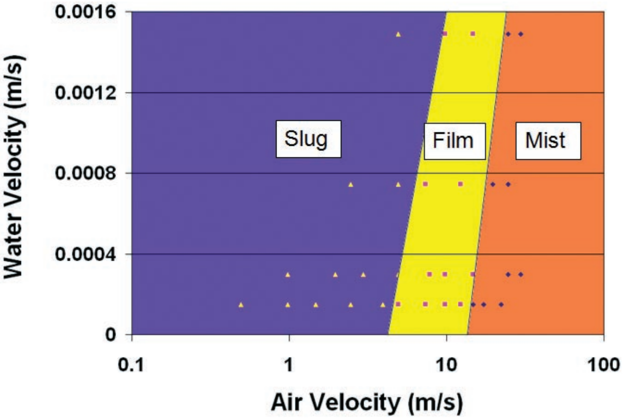

FIGURE 3. Parallel Channel Flow Pattern Map Obtained from the Ex Situ Experiment for the Baseline System

during air purge, and start-up from freezing conditions. Neutron imaging demonstrated that water accumulation under low operating conditions occurs primarily within the anode channels, within the GDL on both anode and cathode sides, at the transitions between flow field channels and plate manifolds, and at the plate edges within the manifolds (Figure 4). Cathode air purges show two distinct regimes of water removal. The first relatively short regime corresponds to convective removal of water slugs from the flow field channels. The second regime corresponds to evaporative removal from the diffusion media and MEA, with a distinct drying front propagating across the active area from cathode inlet to outlet. The evaporative regime features a linear water volume change with time, but the evaporation rate is strongly dependent on both air flow rate and temperature (Figure 5). The ability to start the fuel cell from a frozen state was evaluated by monitoring both anode/cathode pressure drop and cell voltage. With zero purge, the pressure drops are extremely high, and it is not possible to attain open circuit voltage, indicating complete blockage of reactant flow. With purge times on the order of 30 seconds, a much smaller pressure drop is observed and cell voltage can be attained.

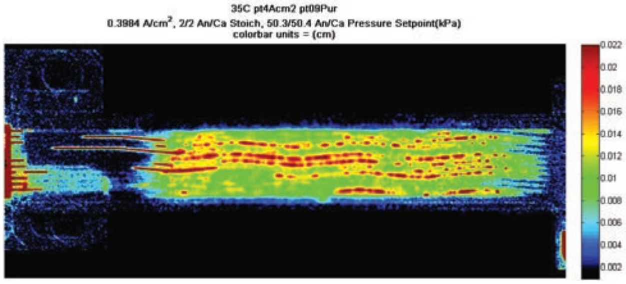

FIGURE 4. In Situ Neutron Radiograph, Showing Key Areas of Fuel Cell Water Accumulation at 35°C

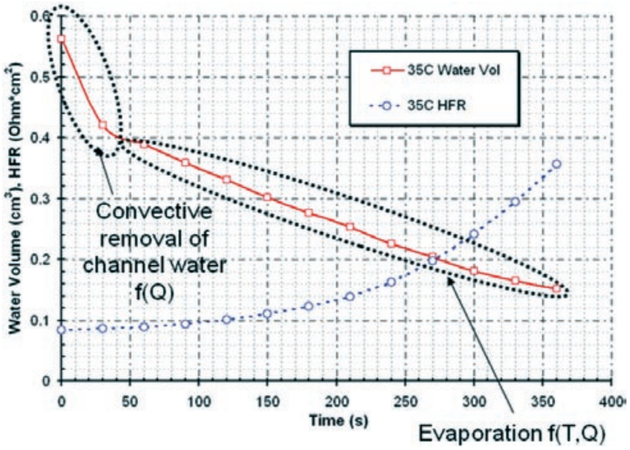

FIGURE 5. Change in Water Mass during Cathode Air Purge

Conclusions and Future Directions

The baseline system (GDL and flow channel) has been comprehensively characterized in terms of the material properties, ex situ single- and multi-channel performance, in situ fuel cell steady-state water distribution, and freezing start-up performance. A performance matrix, including pressure drop, flow pattern map, slug residence time, water accumulation locations, minimum cathode purge time for successful freeze start, has been established to characterize the GDL-flow channel combination.

On the basis of the performance matrix of the baseline system, in the future we will implement changes to the GDL and flow channel and explore various GDL/channel combinations with ex situ and in situ experiments. The most effective GDL properties (wettability, structure and morphology) and the effective channel surface treatment will be determined. Liquid water transport in the GDL and the channels under normal and freezing conditions will be mapped and quantified. From these deliverables, final recommendations will be made on the optimized combination of GDL material and design, flow channel design and surface treatment, and anode/cathode flow conditions.

FY 2008 Publications/Presentations

1. Kandlikar, S.G., "Microscale and Macroscale Aspects of Water Management Challenges in PEM Fuel Cells," Plenary Paper, ISTP-18, 18th International Symposium on Transport Phenomena, Daejeon, Korea, August 27-30, 2007. Also published in Heat Transfer Engineering, 29-7, 575-587, 2008.

2. Kandlikar, S. G., and Lu, Z., Current Status and Fundamental Research Needs in Thermal Management within A PEMFC Stack," Accepted for publication in ASME Journal of Fuel Cells Science and Technology, 2008.

3. Kandlikar, S.G., Lu, Z., and Trabold, T., "Current Status and Fundamental Research Needs in Thermal Management within a PEMFC Stack," Keynote Paper, 10th UK Heat Transfer Conference, Edinburgh, September 10-12, 2007.

4. Herescu, A., Allen, J.S., "Wetting Effects on Two-Phase Flow in Microchannel," IMECE2007-42050, ASME International Mechanical Engineering Congress and Exposition (IMECE2007), November 11-15, Seattle, Washington, USA.

5. Medici, E., Allen, J.S., "2D Parametric Study of Viscous Fingering," IMECE2007-42024, ASME International Mechanical Engineering Congress and Exposition (IMECE2007), November 11-15, Seattle, Washington, USA.

6. Lu, Z., White, A.D., Pelaez, J., Hardbarger, M., Domigan, W., Sergi, J., Kandlikar, S.G., "Investigation of Water Transport in an Ex-Situ Experimental Facility Modeled on an Actual DOE Automotive Target Compliment Fuel Cell," ICNMM2008-62200, accepted to Sixth International ASME Conference on Nanochannels, Microchannels and Minichannels, Darmstadt, Germany, June 23-25, 2008.

7. Owejan, J.P., Gagliardo, J.G., Sergi, J.M. and Trabold, T.A., "Two-Phase Flow Considerations in PEMFC Design and Operation," Proceedings of ASME ICNMM2008, 6th International Conference on Nanochannels, Microchannels and Minichannels, Paper ICNMM2008-62037, Darmstadt, Germany, June 2008.

8. LaManna, J., and Kandlikar, S. G., "A Critical Review of Water Transport Models in Gas Diffusion Media of PEM Fuel Cell," ICNMM08-62201, Sixth International Conference on Nanochannels, Microchannels and Minichannels, Darmstadt, Germany, June 23-25, 2008.

9. LaManna, J., , Underhill, R., and Kandlikar, S.G., "Simulation of Heat and Mass Transport in Gas Flow Streams of a PEMFC from Water Management Perspective," submitted to the Nineteenth International Symposium on Transport Phenomena, Reykjavik, ICELAND, 2008.

10. Stacy, R., Allen, J.S., "Automated Contact Angle Measurement Using the Laplace-Young Equation," submitted to the Nineteenth International Symposium on Transport Phenomena, Reykjavik, ICELAND, 2008.

11. Medici, E., Allen, J.S., "A Novel Technique to Characterize Gas Diffusion Layers of PEM Fuel Cell," submitted to the Nineteenth International Symposium on Transport Phenomena, Reykjavik, ICELAND, 2008.

12. Stacey, R., and Allen, J.S., "Automated Contact Angle measurement using Laplace-Young Equation," 35th Mid-Western Universities Fluid Mechanics Retreat, MUFMECH08, Rochester, IN, April 3-5, 2008.

13. Medici, E., and Allen, J.S., "A Novel Technique to Characterize Porous Media, 35th Mid-Western Universities Fluid Mechanics Retreat, MUFMECH08, Rochester, IN, April 3-5, 2008.

14. Medici, E., and Allen, J.S., "One the Gas Diffusion Layer's Ca-M Phase Diagram," Heat Transfer and Fluid Flow in Microscale III, To be presented at Whistler, BC, Canada, September 21-26, 2008.

15. Trabold, T.A., Owejan, J.P., Gagliardo, J.G., Jacobson, D.L., Hussey, D.S. and Arif, M., "Use of Neutron Imaging for PEMFC Performance Analysis and Design," Handbook of Fuel Cells - Volume 5, Prof. Dr. W. Vielstich et al. (Eds.), John Wiley & Sons Ltd., accepted for publication (2008).