V.B.2 Water Transport in PEM Fuel Cells: Advanced Modeling, Material Selection, Testing, and Design Optimization

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Objectives

- Develop advanced physical models for water transport and generation, and conduct material and cell characterization experiments.
- Improve understanding of the effect of various cell component properties and structure on the gas and water transport in a proton exchange membrane (PEM) fuel cell.
- Encapsulate the developed models in a modeling and analysis tool for cell design and future application.
- Demonstrate improvements in water management in cells and short stacks.

Technical Barriers

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

(A) Durability
(D) Water Transport within Stack
(E) System Thermal and Water Management
(G) Start-up and Shut-down Time and Energy/Transient Operation

Technical Targets

This project is addressing fundamental issues in water transport within the fuel cell stack. The resulting understanding will be applied toward the design of stack components and operating strategies that enable meeting the 2010/2015 targets for transportation fuel cell stacks operating on direct hydrogen:

- Stack power density: 2,000 W/L
- Cold start-up time to 50% rated power @ 20°C: 5 secs
- Unassisted start from low temperature: -40°C

Accomplishments

- Generated flow regime map from experimental studies of droplet emergence at a gas diffusion layer (GDL)-channel interface studies; demonstrated improved model agreement with the transition from film flow to droplet regime;
- Implemented experimental setup for collecting wet pressure drop and transient pressure signatures in two-phase flows in channels and cells; improved agreement of model predictions and observed wet pressure drop;
- Integrated electrochemistry, heat transfer, and phase change with the computational fluid dynamics (CFD) two-phase flow models and demonstrated improvement in current density distribution for initial validation test case; and
- Identified materials and design modifications to focus on for improved water management.

Introduction

Water management in PEM fuel cells is challenging because of the inherent conflicts among the needs for: (1) supplying adequate water to establish and maintain the membrane electrical conductivity, (2) removing the water produced by the electrochemical reactions at the cathode, and (3) uniformly distributing the gaseous reactants at catalyst surfaces near the membrane to effectively utilize these costly catalysts. As power density of the cells increases, more water will be generated within the same cell volume. Therefore, increasing power density requirements will drive a greater need for design tools incorporating an improved understanding of how liquid water is transported within fuel cells. An additional barrier to widespread use of fuel cells for automotive power is the performance degradation caused when liquid water freezes within the cells. Optimizing water management to influence where the liquid water remains at shutdown is a promising path to improving cold starting capabilities and freeze-thaw reliability.

This project is intended to improve the fundamental understanding of water transport within a PEM fuel cell, and capture that knowledge in design tools capable of assisting the industry to meet targets for increased power densities and improved cold-start performance. To achieve these objectives, the project is focused on developing predictive models for water transport in GDL materials, characterizing materials for model inputs and verification, implementing the resulting understanding in engineering design tools, and validating the resulting design tools against fuel cell performance data and in situ diagnostics of water distribution within operating fuel cells.

Approach

The overall approach of the project team is integrated experimental characterization with model development and application to meet the high level objectives of improving fundamental understanding of water transport in PEM fuel cells and demonstrating improved performance. The initial focus of the experimental characterization was on measuring relevant physical and transport properties of the GDL materials typically placed between the catalyst and reactant flow channels. Diagnostic and characterization studies progressed from water and two-phase (water and air) fluid transport properties of GDL materials, to analysis of water transport in non-operational and operational fuel cells. The related modeling studies have followed a similar progression, with initial emphasis on microscale simulations of single fluid and two-phase transport within GDL materials. The knowledge gained from the materials characterization and microscale simulations was used to develop models suitable for incorporation into an engineering design tool for fuel cell scale analysis of reactant and water transport coupled with power generation. The verification of these models, and the resulting design tool, is in progress and will be accomplished by comparing predicted and measured effects of material and operating conditions on cell performance and water distribution within the cell. Applying our models to screen and improve water management strategies, then testing the resulting concepts in prototype fuel cells, will further demonstrate our improved fundamental understanding and validate the resulting design tools.

Results

In this third year of research, the emphasis has been on completing experimental studies and model evaluation for water transport in fuel cell components such as GDLs and channels, and on completing integration of the water transport models with electrochemistry and heat transfer for modeling of operational cells. We have also begun to apply the gathered data and developed models to screening concepts for improving fuel cell performance through better water management.

Ex situ studies completed this year included two-phase, i.e. liquid water and air, experiments in fuel cell microchannels and channel-GDL assemblies. A flow regime map for the two-phase system arising from liquid water injection into microchannels was generated from controlled experiments designed to model water emergence phenomena in a PEM fuel cell cathode gas microchannel. The microchannel was 250 μm square in cross-section, with a single 50 μm square opening in the bottom for water injection. In general, higher air flow velocities resulted in decreased contact angle hysteresis so that the leading and trailing edges of the droplets had similar contact angles. Higher air flows also promoted droplet detachment in surface tension controlled regimes, as shown in Figure 1. The Weber number scales on the top and right axes of Figure 1 correspond to the relative values of the droplet inertial forces and surface tension forces. With increasing water velocity, the region corresponding to droplet flow regime becomes smaller. At some point, while increasing the superficial water velocity, the slug and film regime will intersect and thus the flow pattern might turn into an intermittent quasi single-phase liquid flow. The desirable operating states are expected to be at the higher air flows within the droplet regime. In this operating region, droplets are formed and more easily removed from the GDL surface, as opposed to the water forming liquid films that would prevent reactants from reaching the fuel cell catalyst or the water accumulating in slugs that would large pressure drops and pressure variations along the channels.

The capabilities of the developed two-phase flow models to predict trends from the droplet injection in microchannel experiments have been improved by

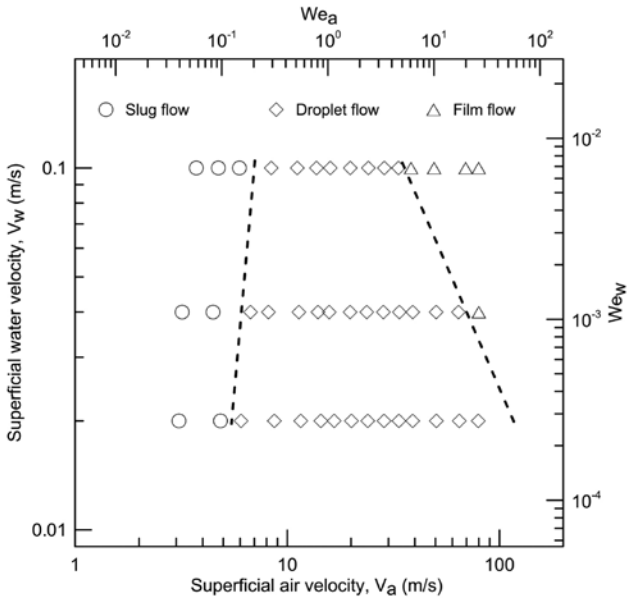

FIGURE 1. Flow Map Based on Controlled Experiments Simulating Water Droplet Detachment in Cathode Microchannels, Water Flow Regime as a Function of Velocities and Weber Numbers

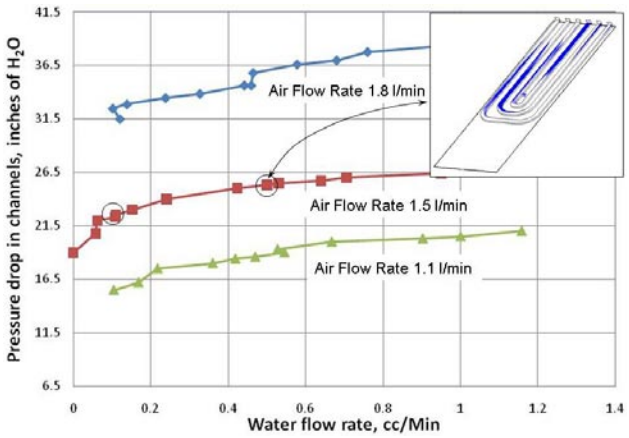

FIGURE 2. Measured Two-Phase Pressure Drop in a Serpentine Channel Cell, Lines; Predicted Pressure Drop, Circles, and Water Volume Fraction Distribution (Inset) at the Indicated Operating Condition Showing Film Formation Consistent with the Experiments

refining the treatment of surface tension. Simulations of experiments such as these are typically performed using the volume of fluid approach, VOF, which provides better resolution of liquid-vapor interface movement and better captures surface tension effects but assumes a no-slip interface at the liquid-vapor surface. The principal disadvantage of the VOF technique is the computational time requirement, since it is inherently a transient technique requiring limited time steps and a relatively fine computational grid to capture surface tension effects accurately. The computation of surface tension forces for liquid-vapor interfaces in channels has recently been improved in the developed two-fluid, two momentum equation solution approach, and initial tests indicate that this new algorithm improves the ability to identify the onset of droplet formation and surface-tension dominated flows. Although the two-phase multiplier is still slightly underestimated for 'wet' to dry pressure drop ratios, the improved surface tension algorithm does result in droplet and slug formation for steady state simulations enabling faster screening of design concepts.

Techverse experimentally studied two-phase flow regimes in the cathode channels of a non-operational cell as a function of air flow rates, water flow rates, and cell orientation to analyze transport in channel-GDL assemblies. The apparatus was prepared by using a GDL to separate a liquid water reservoir from transparent serpentine cathode channels. Visualization and image analysis were used to identify the flow regimes, and pressure drop measurements were performed. In general, a horizontal cell with water entering from above the flow channels exhibited the least two-phase flow pressure drop, while a vertical cell with air flowing up had the largest pressure drop for the same air and water flow rates. Analysis of photographs at steady operation indicated that a horizontal cell orientation with water flowing up into the channels, and a vertical cell orientation with air flowing up, were most likely to form water films blocking open GDL surface area. Several operating points from the experiments for the vertical orientation of the channels, Figure 2, were simulated using the developed two-fluid models for water transport through porous GDL materials and in the channels. The model predictions were qualitatively consistent with the experimental observation of film formation at these conditions, and the predicted variation in channel pressure drop with water flow rate was in good agreement with the experimental measurements.

The integration of two-phase flow models with the key effects necessary for simulation of an operational cell at typical conditions of temperature, pressure, and inlet states has continued. Initial testing and validation simulations were for the Ballard MK902 cell. This cell and operating conditions were selected for the detailed diagnostic data and past modeling results available for comparison [1,2]. The simulation utilized two-phase flow in the channels and porous media, electrochemistry, and heat transfer. The coupling between gas phase chemical species transport and liquid water formation was primarily through the treatment of the oxygen reduction reaction. For this reaction, the water product is formed as either vapor or liquid phase water based on the local thermodynamic state. The resulting liquid water fraction distribution at the level of the cathode GDL for an average current density of 0.85 A/cm^2 indicated that liquid water is predominantly under the lands. The predicted current density distribution, Figure 3, is below the experimental

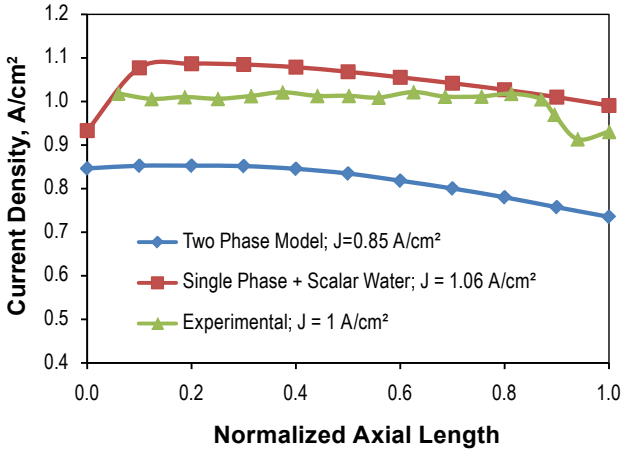

FIGURE 3. Predicted and Measured Current Density versus Distance along Cathode Channel for Ballard MK902 Cell

curve because we were unable to obtain a converged solution at the desired 1 A/cm^2 average current. The two-phase model does show improved agreement with the experimentally measured trends compared to simpler models, particularly near the inlet where the current density starts low and rapidly increases in the single phase solution. During these tests, improvement of the numerical stability and accuracy of the heat transfer solution was identified as the highest priority for further model improvements.

Two main thrusts have emerged for concepts to improve cell performance, GDL materials modification and channel design. In the area of GDL material properties, during the second year of this project BCS Fuel Cells demonstrated improved performance in a self-humidified cell operated under dead-end conditions for an intermediate loading of poly-tetrafluoroethylene (PTFE) in the microporous layer. Within the last year, these results have been confirmed, and the initial interpretation of the basis for improved performance has been supported by improved diagnostics using impedance spectroscopy. The best performance is observed at the intermediate PTFE loading because less hydrophilic GDL materials do not maintain adequate membrane ionomer hydration, while additional loading blocks pores and introduces mass transfer limitations for the gaseous reactants between the channels and the catalyst. For channel designs, the specific objectives are to minimize the pressure drop variation between 'wet' and 'dry' operation and the amplitude of pressure drop transients caused by liquid water in the cells while maintaining or improving cell power density. Initial experiments at Ballard have identified wicking channel designs, in conjunction with channel surface treatments to control the water contact angle, as promising approaches to meet these objectives. In those experiments, several channel design concepts either achieved the wet pressure drop performance improvements, or had promising cell power output, but none met both objectives.

Conclusions and Future Directions

In the past year, we have identified advantageous operating regimes while completing the planned characterization of two-phase transport in cell components and assemblies. The predictive capabilities of the cell-scale CFD models for two-phase flow in fuel cell microchannels, and in porous components such as GDLs and catalyst layers, have been improved and are integrated with electrochemistry and heat transfer to enable analysis of operational cells. Specific accomplishments for the past year include:

- Generated flow regime map from experimental studies of droplet emergence at a GDL-channel interface studies; demonstrated improved model agreement with the transition from film flow to droplet regime;
- Implemented experimental setup for collecting wet pressure drop and transient pressure signatures in two-phase flows in channels and cells; improved agreement of model predictions and observed wet pressure drop;
- Integrated electrochemistry, heat transfer, and phase change with the CFD two-phase flow models and demonstrated improvement in current density distribution for initial validation test case; and
- Identified materials and design modifications to focus on for improved water management.

Key activities planned for the coming year include:

- Improve numerical stability of electrochemistry, heat transfer and phase change models integrated with the two-phase CFD models; test and validate the developed integrated models using operational cell-scale steady and transient data.
- Apply validated measurements and simulation tools to the identified optimization strategies: channel design, surface finish, and GDL design for effective water removal with low pressure drop and minimal transients; GDL design and treatment for improved cell performance.

Future work will consist primarily of testing and improving the developed models, while applying validated capabilities to support the development and demonstration of improved water management approaches.

FY 2010 Publications/Presentations

1. "Water Management in PEM Fuel Cell - A Lattice-Boltzmann Modeling Approach," S. Mukherjee, J. Vernon

Cole, K. Jain, and A. Gidwani, FuelCell2009- 85182, Proceedings of FuelCell2009, ASME 7th International Fuel Cell Science, Engineering & Technology Conference, June 8–10, 2009.

2. "Measurements of Fuel Cell Internal Resistances for the Detection of Electrode Flooding," H.P. Dhar and S.K. Chaudhuri, J. Solid State Electrochemistry 13, (7) 999 (2009).

3. "A Two-Fluid Model for Hydrogen PEM Fuel Cell Performance Integrating Gas Phase and Water Transport with Porous Media Capillary Effects, Heat Transfer, and Electrochemistry," S. Mukherjee, A. Gidwani, A. Roy, J.V. Cole, K. Jain, C. Bapat, and R. Thoms, Presented at 217th Meeting of the Electrochemical Society, April 2010.

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1. P.C. Sui, S. Kumar, N. Djilali, "Advanced Computational Tools For PEM Fuel Cell Design Part 1. Development and Base Case Simulations," *J. Power Sources*, **180**, pp. 410-422 (2008).

2. P.C. Sui, S. Kumar, N. Djilali, "Advanced Computational Tools For PEM Fuel Cell Design Part 2. Detailed Experimental Validation and Parametric Study,"*J. Power Sources*, **180**, pp. 423-432 (2008).