
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 Hydrogen, Fuel Cells and Infrastructure 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

- Completed characterization of key physical and transport properties for SGL, Ballard Materials Products (BMP) and Toray gas diffusion layer (GDL) materials, including variation with polytetrafluoroethylene (PTFE) loading;
- Analyzed impact of GDL microstructure and PTFE loading on GDL permeability, wetting characteristics and water breakthrough, and capillary pressure behavior with validated micro-scale multiphase Lattice Boltzmann Method (LBM) transport models;
- Measured water droplet growth rate, dynamic contact angles, and break-away frequency during water emergence at model GDL-channel interfaces;
- Measured wet pressure drop and transient pressure signatures of two-phase flows in channels and cells to evaluate design sensitivity and support computational fluid dynamics (CFD) model development and validation; and
- Demonstrated the sensitivity of cell performance to modifications in materials and design that impact water management.



Introduction

Water management in PEM fuel cells is challenging because of the inherent conflicts between: supplying adequate water to establish and maintain the membrane electrical conductivity, removing the water produced by the electrochemical reactions at the cathode, and 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 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 have transitioned to water and two-phase (water and air) fluid transport properties of GDL materials, and analysis of water transport across material interfaces and in fuel cell channels. The related modeling studies are following a similar progression, with initial emphasis on microscale simulations of single fluid and two-phase transport within GDL materials. The simulations allow us to analyze key effects such as the impact of the

microstructure and surface treatment of the solids within porous GDL materials on the two-phase water and gas transport. The knowledge gained from the materials characterization and microscale simulations is being 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 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 second year of the research, the emphasis has been on completing analysis and model formulation for water transport in isolated cell components such as GDLs and channels, and transitioning to address transport across interfaces and in operational cells. We have gathered experimental data under controlled conditions while developing, testing, and applying related models.

We have completed the measurement of key physical and transport properties for a set of GDL materials including SGL, Toray, and BMP papers initiated in the first year of the project. The resulting database includes porosity, pore size distribution, in- and through-plane gas permeability, effective diffusivity, and electrical and thermal conductivity. For the SGL and Toray materials, a range of PTFE loadings were used to identify the response of measured properties to this common treatment. LBM simulations of gas transport through numerically generated GDL microstructures with a range of PTFE loadings, Figure 1, demonstrate good agreement with the measured variation in gas permeability. The key finding from the simulations was that the relatively simple, stochastic approach we developed for incorporating the PTFE distribution in the model structures is adequate for describing the impact of PTFE loading on transport properties in carbon paper GDL materials. The other key GDL properties utilized in continuum water transport models, capillary pressure and relative permeability, were measured to determine their dependence on water saturation. The capillary pressure measurements, Figure 2, were performed using a positive displacement technique, and in addition to capillary pressure as a function of water saturation we also were able to quantify the pressure required for water breakthrough and residual water saturation after wetting. Relative permeability is a key input property for continuum CFD-based analysis of water

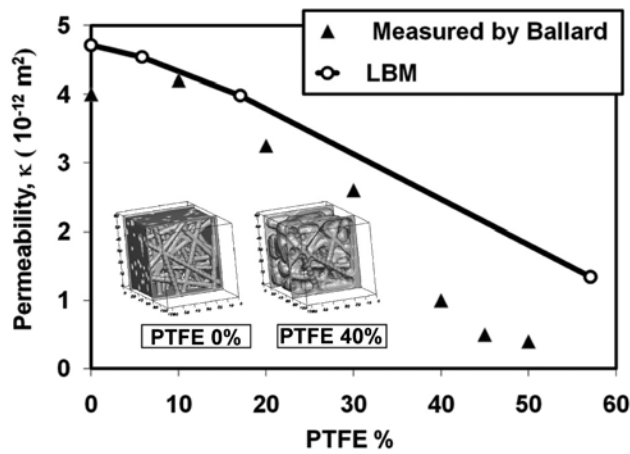


FIGURE 1. Predicted and Measured Dependence of GDL Permeability on PTFE Loading, with Numerically Generated GDL Structures

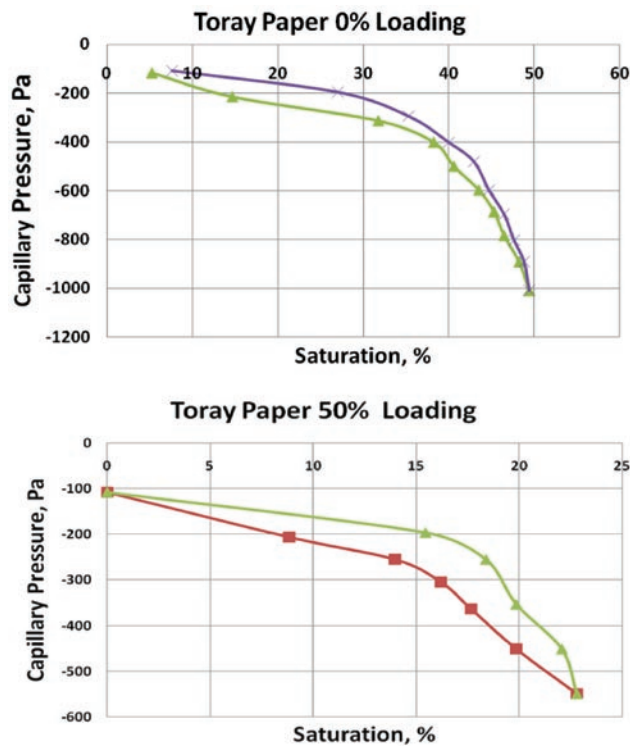


FIGURE 2. Measured Capillary Pressure for Toray Paper GDL Media During Imbibing and Drainage

transport in porous media and is typically assumed to vary with water saturation cubed. The measurements indicate that a linear variation is more appropriate, although there is still uncertainty due to the challenges in specifying and controlling the water saturation while measuring air flow through the sample.

Additional water transport studies were performed to provide quantitative information on water transport in GDLs and across the GDL-channel interface. The

GDL transport studies use a system for simultaneous monitoring of the capillary flow, pressure, and injected water volume as water percolates through GDL samples. These experiments complement the relative permeability studies, and are providing improved understanding of the governing physics as water begins to percolate through and exit the GDL material. Key findings include history-dependent invasion, namely that the observed pressure required to drive a fixed volumetric flow rate increases for a previously wet sample. Image analysis is also being performed during these percolation studies, to quantify the growth and clustering of water droplets on the surface while simultaneously measuring the injected water volume and pressure. At the GDL-channel interface, detachment of water droplets to allow rapid water transport down the channel is a key process governing effective water removal. We are studying this process on controlled surfaces, with quantitative imaging, Figure 3, to determine dynamic water/substrate contact angles and correlate droplet removal frequency with flow conditions.

Model development efforts have focused primarily on the CFD-based cell-scale models. The key physical sub-models for water transport in GDLs and catalyst layers, capillary pressure and Darcy’s Law resistance to flow in porous media, have been incorporated into the two-phase fluid flow solver. The resulting models have been integrated with the chemical species and electric current conservation equations to allow simulation of operational fuel cells, and integration with energy conservation is underway. These models have been evaluated for a variety of relevant test problems, primarily simplified representations of the water transport characterization experiments described above. The tests have demonstrated that the sub-models are correctly implemented and experimental trends can be reproduced, but there are still numerical stability issues and transient simulations are typically required to obtain satisfactory convergence. Ongoing efforts include evaluation of alternate models for fluid-fluid drag in channels, inclusion of surface tension effects to allow simulation of droplet formation at GDL-channel interfaces, and development of a physically based interface matching condition at the GDL-channel interface.

We have developed a fixture for measuring pressure drops and pressure profiles during two-phase flows in single fuel cell channels with the option to inject water at specific locations or distributed by a GDL. A test matrix for a range of air flow rates and water injection rates, corresponding to stoichiometric operation at current densities of 0.3, 1, 1.5, and 2 A/cm², were characterized for two channel designs. For two-phase flows in channel design 1 with water injection at a single upstream point, significant pressure transients were observed at the lowest water and air flow rates and run-to-run repeatability was also worse. The steady-state models predicted pressure drop trends with water

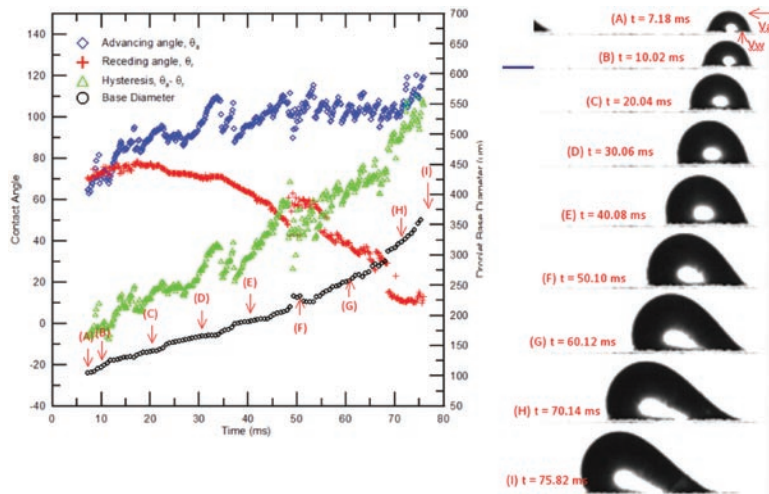


FIGURE 3. Apparent Contact Angles and Droplet Base Diameter Time Evolution during Single Hole Water Injection Experiment (left) and Droplet Shape Evolution (right)

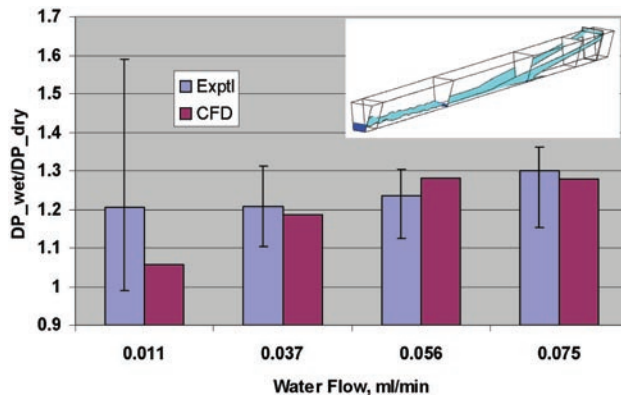


FIGURE 4. Measured and Predicted Two-Phase Pressure Drop in Single-Channel Fixture at 61 sccm Air Flow with Varying Water Flow; Predicted Water Distribution for 0.011 ml/min Water Injection (inset)

flow rate variation for a given air flow relatively well, Figure 4, despite the transient nature of the system at low water flows. However, the model predictions for wet-to-dry pressure drop ratio versus overall flow rate did not produce the observed variation. For channel design 2, with a wider but shorter cross-section, the predicted wet/dry pressure drop ratio variation with total flow had worse agreement with experiment than design 1. The models are qualitatively predicting the correct water distribution in the channel, based on experimentally derived flow field regime maps, namely a horizontal stratified film as shown in Figure 4. Incorporation of gravity effects improved the pressure drop agreement in the design 1 simulations, and caused the water film to form at the lower surface of the channel.

Characterization of water management related effects in operational cells has continued, with

diagnostics based on both single cell polarization and internal resistance measurements. The impact of a variety of GDL materials on performance was assessed in small-scale fuel cells, 25 cm² active area, under self-humidified operating conditions. Performance sensitivity with PTFE treatment of the GDL was demonstrated, and a custom membrane electrode assembly (MEA) was developed with 20% PTFE in the microporous layer of carbon cloth type GDL that demonstrated better performance than both standard commercial and 5% PTFE content MEAs operated at 60°C and 1.75 air stoichiometry. The custom GDL modifications allow effective operation at higher temperatures, and consequently increased power density, without external humidification.

Conclusions and Future Directions

In the past year, we have characterized key material properties and processes governing water transport in individual fuel cell components and across material interfaces. Initial improvements of cell performance via materials modifications have been demonstrated. Modeling and simulation has been applied at the microscale to improve understanding of water transport within GDL materials, and to guide formulation of fuel cell scale multiphysics models for integrated prediction of cell performance and water management. Specific accomplishments include:

- Completed characterization of key physical and transport properties for SGL, BMP and Toray GDL materials, including variation of these properties with PTFE loading;
- Analyzed impact of GDL microstructure and PTFE loading on GDL permeability, wetting characteristics and water breakthrough, and capillary pressure behavior with experimental characterization and validated micro-scale multiphase LBM transport models;
- Measured water droplet growth rate, dynamic contact angles, and break-away frequency during water emergence at model GDL-channel interfaces;
- Measured wet pressure drop and transient pressure signatures of two-phase flows in channels and cells to evaluate design sensitivity and support CFD model development and validation; and
- Demonstrated sensitivity of cell operation to water management, and performance improvement, through materials and design modifications.

Key activities planned for the coming year include:

- Complete experimental characterization of water transport in cell components and non-operational cells to screen water management improvement concepts and provide validation data for cell-scale models;
- Complete integration of heat transfer and electrochemistry with two-phase CFD-based cell-scale models and evaluate against operational cell experimental data, improve fluid-fluid drag models and surface tension implementation for better prediction of wet channel pressure drop; and
- Devise and screen concepts for improved water management by applying the developed experimental diagnostic tools and numerical models, with initial emphasis on channel and GDL material designs for effective water removal with low pressure drop and minimized pressure transients.

Future work will consist primarily of testing the models against diagnostic data and improving them as necessary, while demonstrating an improved understanding of the key factors controlling water transport by developing improved water management approaches.

FY 2009 Publications/Presentations

1. Kunal Jain, James Vernon Cole, Sanjiv Kumar, Ashok Gidwani, and N. Vaidya, "A Multiphase, Two-Fluid Model for Water Transport in a PEM Fuel Cell", ECS Trans. 16, (2) 45 (2008). Presented at 214th Meeting of the Electrochemical Society.
2. Shiladitya Mukherjee, James Vernon Cole, Kunal Jain, and Ashok Gidwani, "Lattice-Boltzmann Simulations of Multiphase Flows in PEM Fuel Cell GDLs and Micro-channels", ECS Trans. 16, (2) 67 (2008). Presented at 214th Meeting of the Electrochemical Society.
3. H.P. Dhar and S.K. Chaudhuri, "Detections Of Electrode Flooding in Single Cell Experiments Through Measurements of Cell Internal Resistances," Poster presentation in Fuel Cell Seminar, 2008. Extended Abstracts, published by Courtesy Associates.
4. H.P. Dhar and S.K. Chaudhuri, "Measurements of Fuel Cell Internal Resistances for the Detection of Electrode Flooding," J. Solid State Electrochemistry. In Press.
5. H.P. Dhar and S.K. Chaudhuri, "Detection and Verification of Electrode Flooding in Single Cell Studies," Accepted for publication in ECS Transactions, Vol. 17 (2009).
6. Kunal Jain, James Vernon Cole, Sanjiv Kumar, N. Vaidya, Ashok Gidwani, "Formulation, Implementation and Validation of a Two-Fluid Model in a Fuel Cell CFD Code," Presented at 2008 Annual Meeting of the AIChE.
7. Shiladitya Mukherjee, J. Vernon Cole, Kunal Jain, Ashok Gidwani, "Lattice-Boltzmann Simulations of Multiphase Flows in Gas-Diffusion-Layer (GDL) of a PEM Fuel Cell," Presented at 2008 Annual Meeting of the AIChE.
8. Ashok S. Damle, J. Vernon Cole, "Water Transport Characteristics of Gas Diffusion Layer in a PEM Fuel Cell," Presented at 2008 Annual Meeting of the AIChE.
9. Shiladitya Mukherjee, James Vernon Cole, Kunal Jain, and Ashok Gidwani, "Water Management in PEM Fuel Cell - A Lattice-Boltzmann Modeling Approach," Presented at 2009 ASME 7th International Fuel Cell Science, Engineering & Technology Conference.