V.A.2 Neutron Imaging Study of the Water Transport in Operating Fuel Cells

Muhammad Arif (Primary Contact),
David L. Jacobson, Daniel S. Hussey
National Institute of Standards and Technology
100 Bureau Drive
Gaithersburg, MD 20899
Phone: (301) 975-6303; Fax: (301) 926-1604
E-mail: arif@nist.gov

DOE Technology Development Manager:
Ingrid M. Milton
Phone: (202) 586-9583; Fax: (202) 586-9811
E-mail: Ingrid.Milton@ee.doe.gov

Project Start Date: 2003
Project End Date: Project continuation and direction determined annually by DOE

Objectives:

- Provide neutron imaging-based research and testing infrastructure to enable the fuel cell industry to design, test, and optimize prototype to commercial grade fuel cells.
- Provide secure facility for proprietary research by industry. Make open research data available for beneficial use by the general fuel cell community.
- Continually improve and develop methods and technology to accommodate rapidly changing industry/academia needs.

Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section (3.4.4.2) of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

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

Technical Targets

- Instrument: Develop and operate fuel cell neutron imaging testing facility.
- Research: Nondestructive, *in situ* study of dynamic water transport in operating proton exchange membrane (PEM) fuel cells.
- Data Analysis: Develop accurate data interpretation and quantitative image processing.

Accomplishments

- Developed 25-micrometer resolution as a standard capability for testing with fuel cells.
- Incorporated freeze chamber into facility for *in situ* neutron radiography of cold startups and freeze-thaw testing.
- Demonstrated stack stimulation with a single cell.
- Increased neutron intensity by a factor of two.
- Nearly doubled the use of the facility by fuel cell research groups.

✧ ✧ ✧ ✧ ✧

Introduction

At the National Institute of Standards and Technology (NIST), we maintain the premier fuel cell neutron imaging facility in the world and continually seek to improve its capabilities. This facility provides researchers a powerful and effective tool to visualize and quantify water transport inside operating fuel cells. Imaging the water dynamics of a proton exchange membrane fuel cell (PEMFC) is carried out in real time with the required spatial resolution needed for fuel cells that are being developed today. From these images, with freely available NIST-developed image analysis routines, PEMFC industry personnel and researchers can obtain *in situ,* non-destructive quantitative measurements of the water content of an operating PEMFC. Neutron imaging is the only *in situ* method for visualizing the water distribution in a “real world” PEMFC. Unlike X-rays, whose interaction with materials increases with the number density of electrons, neutrons interact via the nuclear force, which varies somewhat randomly across the periodic table, and is isotopically sensitive. For instance, a neutron’s interaction with hydrogen is approximately 100 times greater than that with aluminum, and 10 times greater than that with deuterium. It is this sensitivity to hydrogen (and insensitivity to many other materials) that is exploited in neutron imaging studies of water transport in operating fuel cells.

Approach

The typical scales of interest in a PEMFC are: channels approximately 1 mm wide and 1 mm deep, the gas diffusion media (GDM) is 0.1 mm to 0.3 mm thick, the membrane is 0.01 mm to 0.05 mm thick, and the active area is 50 cm^2 to 500 cm^2. Thus, to study *in situ*, nondestructive water/hydrogen transport in PEM fuel cells while in operation and hydrogen transport/distribution in hydrogen storage media we will develop new facilities and improve existing capability for obtaining high spatial and temporal resolution neutron imaging. Employing the mathematical models of neutron scattering, we will develop a software suite that enables users to obtain quantitative measurements of the water content in an operating PEMFC. Due to the complexity of PEMFCs and the large number of open questions regarding water transport in PEMFCs, we will develop partnerships with industry and academia to train them in the use of the facility, collaborate with them on research projects, and seek their feedback to pursue future technical breakthroughs.

Results

The first year of operation of the new NIST neutron imaging facility (NNIF) has seen a continual series of improvements in both the facility and the program itself. We have realized two major achievements in our neutron imaging capabilities. We have brought into routine service a neutron-imaging detector with 25-micrometer resolution, which is a 10-fold improvement. With this new detector, the water content in commercial gas diffusion layer (GDL) material can be unambiguously measured and fundamental measurements of the water content of thick membranes can be performed. In addition to the improved spatial resolution, we have increased the neutron intensity by a factor of two by optimizing the thickness of upstream radiation filter material. The intensity increase results in a 40% decrease in the time required to achieve a desired measurement uncertainty. A major fuel cell infrastructure addition was the purchase and installation of a sample environment chamber to allow fuel cell testing at temperatures as low as -40°C (see Figure 1). The custom-built chamber enables *in situ* neutron imaging of a fuel cell during cold startups and freeze-thaw cycles. The freeze chamber was first used with our fuel cell testing partners from Los Alamos National Laboratory in May of 2007 and is freely available to all users of the NNIF. Additionally, as a member of the NIST Center for Neutron Research proposal and beam time allocation program, the facility use by academic and national laboratory research groups more than doubled and interest continues to grow.

We have performed a series of experiments aimed at understanding the water transport in the GDL under differential conditions. The first experiment measured the water distribution in three dimensions using neutron tomography (see Figure 2).

FIGURE 1. Photo of the freeze chamber installed inside the Neutron Imaging Facility (top). The sample chamber with 100 cm^2 cell inside is also shown (bottom). Windows on front and back are designed for neutron transmission and thermal insulation.

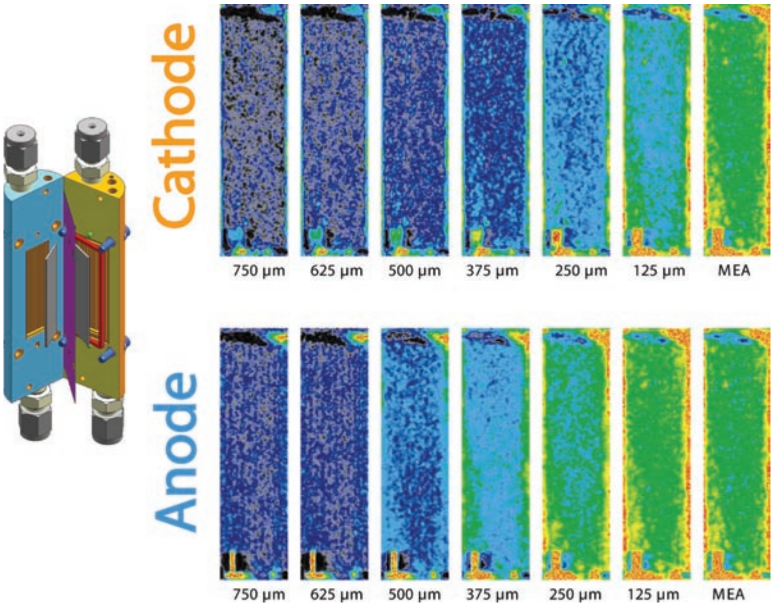

FIGURE 2. Tomography of a single cell using 800-micrometer thick gas diffusion layers and Nafion-117. Each slice shows the water content as a function of the distance from the center of the membrane electrode assembly (MEA).

The results showed that the in-plane water content was very uniform, while the through-plane water content peaked in the anode GDL. This seeming surprising result was due to the use of a thick GDL, which significantly increased the time required to reach an equilibrium state. Since the in-plane water content was uniform, we performed a follow-on experiment viewing the cell "edge on" to measure the through-plane water content. The cell was run for two hours to ensure stable equilibrium and in this case, the through-plane water content was maximum in the cathode GDL as expected. Further, we explored the relative impact of capillary pressure versus evaporation and advection on the water transport by measuring the change in the water content after ceasing the load. In our differential cell the preliminary results show that capillary pressure plays a minimal role as a mechanism for water removal in the fuel cell.

We have extended the edge-on radiography capability by developing a 25-micrometer spatial resolution detector that is now in routine use at the NNIF. We have also added low flow testing capabilities to our fuel cell test stand to operate smaller scale fuel cells at low stoichiometric ratios. Initial tests showed that membrane swelling can be significant and could cause dimensional changes that make normalizing images to the dry state difficult. However, using a hydrated state as a reference enables one to measure the change in water content from open circuit voltage, for instance. Further studies of how membranes swell during operation will be feasible. Cells using relatively thick membranes such as Nafion®-117 have shown a pronounced water gradient from anode to cathode with the water distribution peaking on the cathode side (see Figure 3) and we are in the process of comparing the measured water content with previously reported correlations.

We simulated the operation of a fuel cell stack using a single cell connected to a by-pass manifold (see Figure 4). Full-scale fuel cell stacks have additional water management challenges that are not present with single cells, for instance the design of a stack manifold. As a single cell develops water in the cell, the flow streams begin to develop a pressure

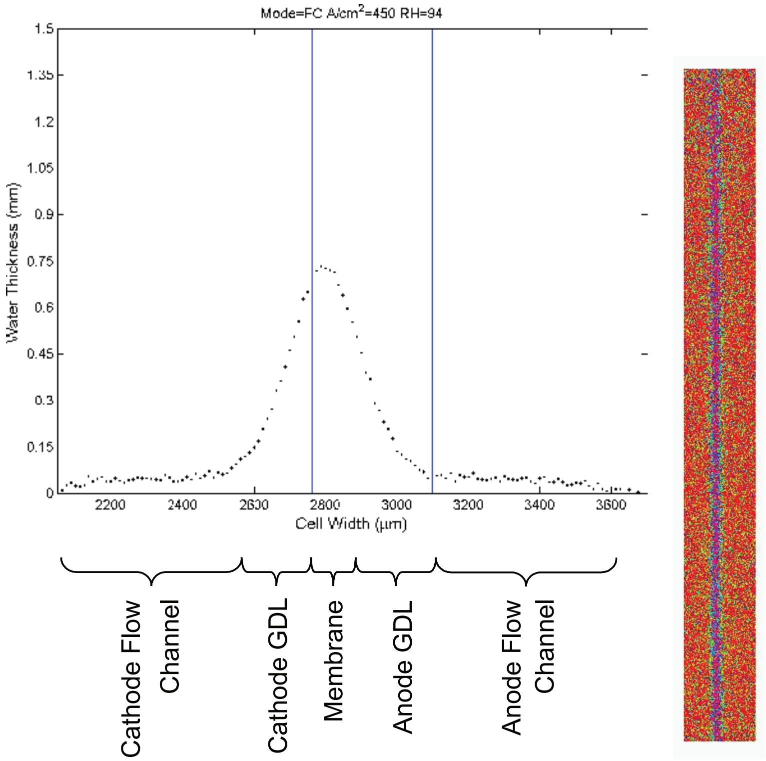

FIGURE 3. Image and water profile of Nafion®-117 using 25-micrometer spatial resolution (Ludlow, et al). Here a water gradient between anode and cathode can be seen that peaks at the cathode.

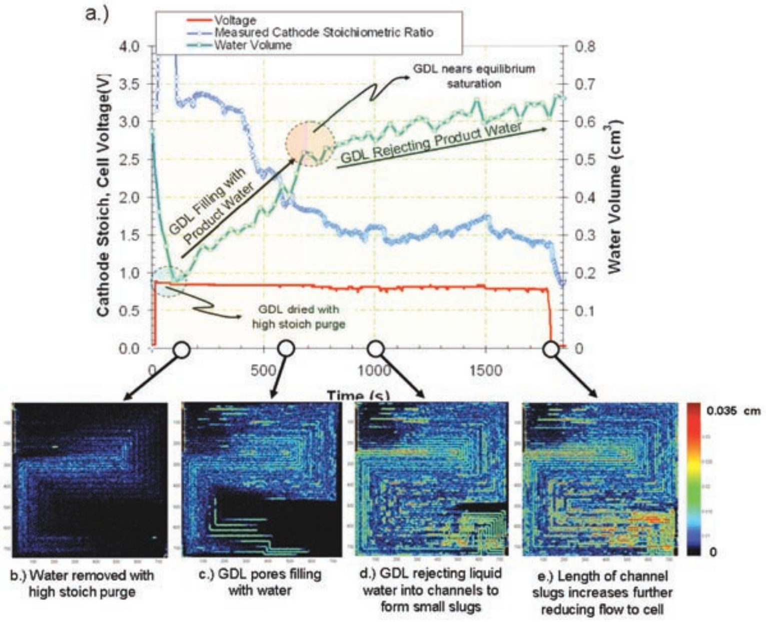

FIGURE 4. Simulating a fuel cell stack using a bypass manifold. Here the total water content measured with neutrons is plotted along with the corresponding stoichiometry on the cathode side from which a correlation of water content to stoichiometery on the cathode could be determined. The images represent the corresponding neutron images at the various times.

differential that assists in water removal. However, when connected in parallel in a stack the input and output manifolds of the cells are tied together, so instead of developing a pressure gradient the flow simply bypasses the cell. The lack of gas flow leads to further flooding which eventually leads to a catastrophic failure of the cell. From the results of the stack simulation (reported by Owejan et al [7]) it was possible to formulate a correlation of water content to cathode stoichiometry providing valuable input for stack models.

Conclusions

1. Initial cold-start experiment with the freeze chamber showed direct one-to-one correlation between integrated current and cell water content.
2. Simulating aspects of stack operation with a single cell enabled simultaneous measurement of the water content via neutron imaging and cathode stoichiometry to obtain a correlation between water content and cathode stoichiometry for incorporation into stack models.
3. Nafion®-117 swelling and water content, as well as GDL water content have been directly observed and measured with the new 25-micrometer detector.
4. With a factor of two increase in neutron intensity and a factor of two increase in user base, the NNIF continues to improve and be applied to an ever wider array of water management and fundamental water transport questions in PEM fuel cells.

Future Directions

- Perform transient studies of fuel cell start-up at various initial operating temperatures and compare measured water content and production with model predictions.
- Develop and deploy neutron imaging detectors with ever increasing higher spatial (5 micrometers or better is the eventual goal) and temporal resolution.
- Accelerate technology transfer to industry.
- Expand non-proprietary collaborations.

FY 2007 Publications/Presentations

1. T.J. Kim, Y.M. Jung, M.H. Kim, M. Arif, D.L. Jacobson, D. Hussey, C.M. Sim, S.W. Lee, and J.S, Jeon, "The Visualization of water distribution at the PEMFC using the Neutron Imaging Technique: Feasibility Test". 2005 IEEE Nuclear Science Symposium Conference Record, 639, 2006.

2. T. V. Nguyen, G. Lin, H. Ohn, D.S. Hussey, D.L. Jacobson, and M. Arif, "Capillary Pressure Measurements of the Gas Diffusion and Catalyst Layers in PEMFCs", ECS Transactions, 1 (6) 481-489 (2006).

3. D.J. Ludlow, C.M. Calebrese, S. Yu, C. Dannehy, D.S. Hussey, D.L. Jacobson, M. Arif, M.K. Jensen, G.A. Eisman, "PEM fuel cell membrane hydration measurement by neutron imaging", Journal of Power Sources, Volume 162, Issue 1 , 8 November 2006, Pages 271-278.

4. T. Trabold, J. Owejan, D. Jacobson, M. Arif and P. Huffman, "In-Situ Investigation of Water Transport in an Operating PEM Fuel Cell Using Neutron Radiography: Part 1 - Experimental Method and Serpentine Flow Field Results", International Journal of Heat and Mass Transfer 49 (25-26), pp. 4712-4720, 2006.

5. J. Owejan, T. Trabold, D. Jacobson, D. Baker, D. S. Hussey, M. Arif, "In-Situ Investigation of Water Transport in an Operating PEM Fuel Cell Using Neutron Radiography: Part 2 - Transient Water Accumulation in an Interdigitated Cathode Flow Field", International Journal of Heat and Mass Transfer 49 (25-26), pp. 4721-4731, 2006.

6. O.H.W. Siegmund, J.V. Vallerga, A. Martin, B. Feller, M. Arif, D.S. Hussey, D.L. Jacobson, "A high spatial resolution event counting neutron detector using microchannel plates and cross delay line readout", Accepted Nuclear Instruments and Methods, Section A (in press).

7. J. P. Owejan, T. A. Trabold, J. Gagliardo, D. L. Jacobson, R. N. Carter, D. S. Hussey and M. Arif, "Voltage Instability in a Simulated Fuel Cell Stack Correlated to Cathode Water Accumulation", accepted Journal of Power Sources.

8. J. P. Owejan, T. A. Trabold, D. L. Jacobson, M. Arif and S.G. Kandlikar, "Effects of Flow Field and Diffusion Layer Properties on Water Accumulation in a PEM Fuel Cell", accepted Journal of Power Sources.

9. D.S. Hussey, J.P. Owejan, D.L. Jacobson, T.A. Trabold, J. Gagliardo, D.R. Baker, D.A. Caulk, M. Arif, "Tomographikc Imaging of an Operating Proton Exchange Membrane Fuel Cell", Proceedings of the 8th World Conference on Neutron Radiography, (2006).

10. D. L. Jacobson, D. S. Hussey, E. Baltic, J. LaRock, M. Arif, J. Gagliardo, J. Owejan, T. Trabold, "Neutron Radiography and Tomography Facilities at NIST to analyze in-situ PEM Fuel Cell Performance", Proceedings of the 8th World Conference on Neutron Radiography, (2006).

11. Y-S Chen, H. Peng,D. S. Hussey, D. L Jacobson, D. T. Tran, T. Abdel-Baset, M. Biernacki, "Water distribution measurement for a PEMFC through neutron radiography", Journal of Power Sources 170, pp. 376-386 (2007).

12. D.S. Hussey, D.L. Jacobson, M. Arif, J.P. Owejan, J.J. Gagliardo, T.A. Trabold, "Nuetron images of the through-plane water distribution of an operating PEM fuel cell.", accepted Journal of Power Sources.

13. J. Park, X. Li, D. Tran, T. Abdel-Baset, D.S. Hussey, D.L. Jacobson and M. Arif, "Dynamic liquid water distribution in an operating PEM fuel cell with a long serpentine flow channel", submitted Electrochimica Acta.

14. J. Park, X. Li, D. Tran, T. Abdel-Baset, D.S. Hussey, D.L. Jacobson and M. Arif, "Neutron imaging investigation of liquid water distribution in and the performance of a PEM fuel cell", submitted Journal of Power Source.

15. D.S. Hussey, D.L. Jacobson, M. Arif, K.J. Coakley, D.F. Vecchia, "In situ fuel cell water metrology at the NIST neutron imaging facility", submitted ASME Fuel Cells 2007 conference proceedings.

Presentations

1. D. Jacobson, et al, "Neutron Radiography and Tomography Facilities at NIST to Analyze In-Situ PEM Fuel Cell Performance." 8th World Conference on Neutron Radiography, October 16–19, 2006, National Institute of Standards and Technology, October 16, 2006.

2. D.S. Hussey, et al, "Tomographic imaging of an operating proton exchange membrane fuel cell" 8th World Conference on Neutron Radiography, October 16-19, 2006, National Institute of Standards and Technology, October 18, 2006.

3. D. Jacobson, et al, "Neutron Radiography and Tomography Facilities and Experiments to Analyze *In Situ* PEM Fuel Cell Performance," plenary session for the 2006 Joint International Meeting, 210th Meeting of The Electrochemical Society, Cancun, Mexico, October 29 – November 3, 2006, 10/31/2006.

4. D. Jacobson, et al, "Using Neutron Radiography to Study Hydrogen Fuel Cells," colloquium for the Hopkins Applied Physics Laboratory, February 2, 2007.