V.A.6 Neutron Imaging Study of the Water Transport in Operating Fuel Cells

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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
(E) System Thermal and Water Management

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 fuel cells (PEMFCs).
- Data Analysis: Develop accurate data interpretation and quantitative image processing.

Accomplishments

- Developed 25 micrometer resolution as a standard capability for testing with fuel cells.
- Demonstrated a 30% improvement in spatial resolution with prototype 10 micrometer detector system.
- Seven new research groups are involved with using the facility.
- Ten peer-reviewed experimental proposals were awarded beam time.

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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 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 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 PEMFCs 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 year of operation of the NIST neutron imaging facility (NNIF) has seen further improvements in both the facility and the program itself. With both a new freeze testing capability and routine access to high spatial resolution the realm of experiments using neutron imaging to study fuel cells has greatly expanded.

This year there have been 10 experiments proposed and performed at the NNIF and several journal articles have been published or submitted for publication. Using 25 micrometer resolution and imaging the fuel cell edge we are able to unambiguously distinguish anode from cathode. One of these experiments was a collaborative effort with Sandia National Laboratories. A 7 cm by 2.1 cm active area fuel cell was specifically built for this experiment. The cell was operated with fully humidified gases at 40°C, 60°C, and 80°C. Figure 1 shows a set of images at 60°C for increasing current density. The images show that water tended to remain on the cathode side of the cell at low current. At moderate current where both water back-diffusion from the cathode and thermal gradients contribute significantly to the overall water transport, water builds up in the anode gas flow channel. A detailed analysis of the water profile shows that the cathode side of the membrane electrode assembly was moderately more hydrated than the anode.

We have demonstrated the feasibility of even higher spatial resolution with a new 10 micrometer detector. Here a gain of about a factor of 30% was achieved (see Figure 2). A new system is anticipated to be available for general use starting this fall of 2008. This new system will allow users to use beams with 10 times the neutron intensity and will be insensitive to noise due to gamma backgrounds that affect the current 25 micrometer detector.

The facility has seen studies related to freeze/thaw increase throughout the year. From studies of shutdown purging mechanisms to studies of cold start all of these areas can be analyzed using neutron imaging at the fuel cell. Using high resolution neutron imaging to directly measure an increase in water flux from hot to cold in the fuel cell, researchers from Penn State have demonstrated that utilizing the heat pipe effect increases purge efficiency. Researchers from General Motors/Rochester Institute of Technology/Michigan Technological University have demonstrated the importance of and the ability to purge an automotive grade fuel cell without hysteresis at shutdown.

Standard neutron radiography with scintillators is providing valuable water content data in the lands and channels to both qualitatively and quantitatively validate water transport models. In an experiment to simulate non-isothermal startup, researchers from the University of Connecticut were able to show that with inlet gas of 60% relative humidity, the water content reached equilibrium well after the cell reached a stable operating temperature, as expected (Figure 3). Researchers from

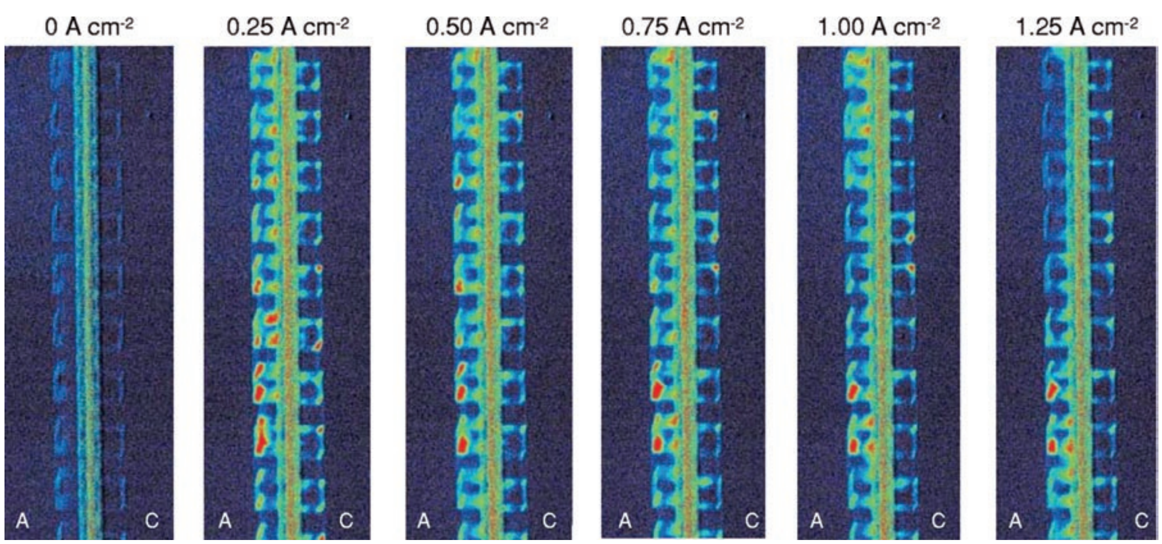

FIGURE 1. Colorized Images of the Water Content of a Fuel Cell Operating at 60°C and Fully Humidified Inlet Gas Streams

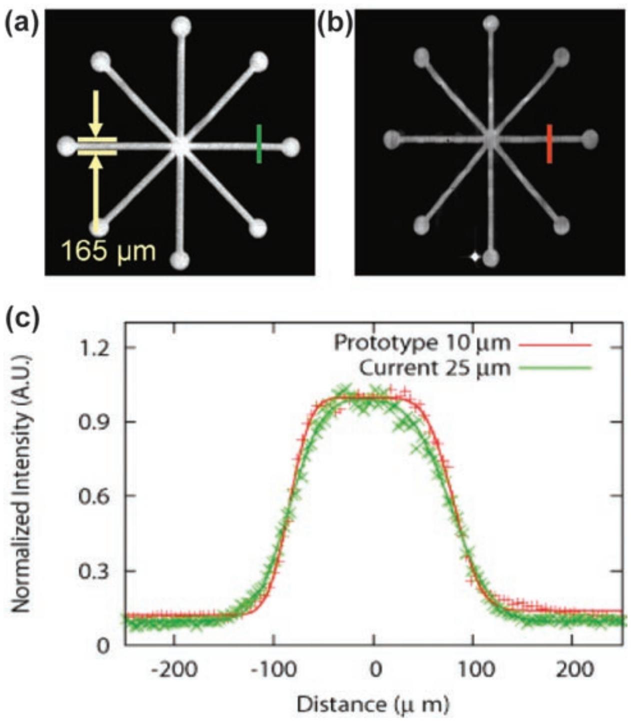

FIGURE 2. Cadimum star pattern imaged by (a) the current 25 micrometer detector and (b) the prototype of the future 10 micrometer detector. (c) The line profiles through the arms demonstrate a 30% improvement in the spatial resolution.

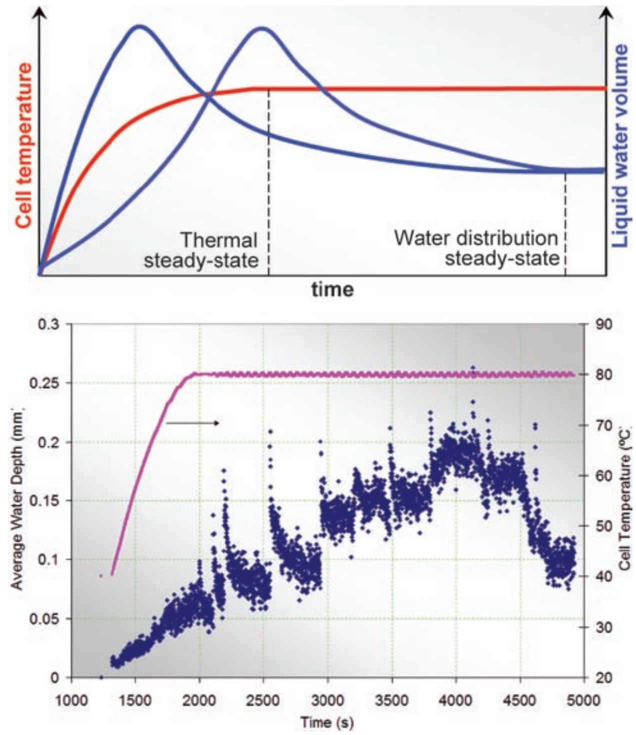

FIGURE 3. Top: Sketch of the fuel cell water content during a non-isothermal start-up. Competition between membrane hydration, water production, evaporation, etc. results in the water content equilibrating at different times than the cell temperature. Bottom: Neutron radiography demonstrates that the water content follows the expected trend for inlet gases with humidity of 60% relative humidity. Cell temperature is plotted in magenta, and the water content is shown in blue.

the University of Michigan compared the water content from a quantitative dead-ended anode model to the neutron radiography data. As Figure 4 demonstrates, there is reasonable agreement between the model results and the measured water content.

Conclusions

- High resolution imaging has provide input into understanding the through-plane water distribution. Results from experiments with Sandia National Laboratories have shown that anode versus cathode water content can be determined and compared to models of water transport.
- In situ neutron radiography studies of fuel cell shutdown procedures are providing key information for meeting DOE cold start targets. Using the new environmental chamber at the NNIF, in situ radiography of fuel cells at temperatures as low as -40°C are possible. This allows the effects of purging cell water content on shutdown and subsequent cold start to be directly studied as was shown by the collaboration General Motors/Rochester Institute of Technology/Michigan Technological University. In addition the new high resolution detector has provided insight into optimized purging mechanisms at shutdown.

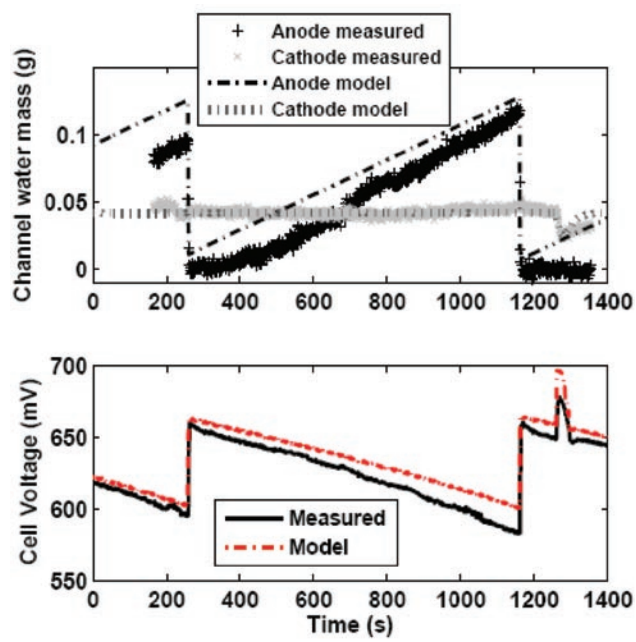

FIGURE 4. Dead-ended anode model predictions of cell voltage as well as channel water in the anode and cathode compared with measured water.

- Neutron radiography data is being used to validate models of water transport. Both experiments using low and high resolution neutron radiography has proven valuable in validation of models of fuel cell water transport as data from Los Alamos, Sandia National Laboratories, University of Michigan and the University of Connecticut have shown.

Future Directions

- Deploy the 10 micrometer resolution detector for routine use. This use will also be extended to freeze/thaw studies of fuel cells. This new direction will allow experimental measurement of the water distribution on anode and cathode under freeze conditions. Test prototype of a sub-micrometer detector.
- Provide electrical impedance spectroscopy for all users for in situ testing. Combined with neutron imaging electrochemical impedance spectroscopy would allow a secondary cross correlation method to better understand water transport. Although separate testing in home laboratories is possible, testing in situ with neutrons is preferred to eliminate a possible source of systematic error.
- Explore tomographic evaluation of hydrogen storage beds. Mapping of the 3-dimensional hydrogen distribution in charged hydrogen storage bed is expected to aid the engineering and development of hydrogen storage beds. Tomography would be sensitive to non-uniform hydrogen concentrations in a storage bed.
- Explore methods for image restoration to improve water profile quantification in the through-plane direction. This will allow us to improve the spatial resolution, which is important to measure the peak water content in the membrane.

FY 2008 Publications/Presentations:

1. A.K. Heller, M. C. Hatzell, M. M. Mench, D. S. Hussey, and D. L. Jacobson, "High Resolution Neutron Imaging of Temperature-Driven Flow in Polymer Electrolyte Fuel Cells", S. Kim, Annual American Nuclear Society Meeting, June 2008.

2. A.K. Heller, M. C. Hatzell, M. M. Mench, D. S. Hussey, and D. L. Jacobson, "High Resolution Neutron Imaging of Temperature-Driven Flow in Polymer Electrolyte Fuel Cells", S. Kim,, In preparation for Journal Submission.

3. M. A. Hickner, N. P. Siegel, K. S. Chen, D. S. Hussey, D. L. Jacobson, and M. Arif, "In Situ High-Resolution Neutron Radiography of Cross-Sectional Liquid Water Profiles in Proton Exchange Membrane Fuel Cells", J. Electrochem. Soc., Volume 155, Issue 4, pp. B427-B434 (2008).

4. M. A. Hickner, N. P. Siegel, K. S. Chen, D. S. Hussey, D. L. Jacobson, and M. Arif, "Understanding Liquid Water Distribution and Removal Phenomena in an Operating PEMFC via Neutron Radiography", J. Electrochem. Soc., Volume 155, Issue 3, pp. B294-B302 (2008).

5. D.S. Hussey, D.L. Jacobson, M. Arif, J.P. Owejan, J.J. Gagliardo, and T.A. Trabold, "Neutron images of the through-plane water distribution of an operating PEM fuel cell", Journal of Power Sources, Volume 172, Issue 1, 11 October 2007, Pages 225-228.

6. 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", International Journal of Hydrogen Energy Volume 32, Issue 17, December 2007, Pages 4489-4502.

7. R. Fu, U. Pasaogullari, D.S. Hussey, D.L. Jacobson, and M. Arif, "Neutron Radiography Imaging of Simulated Non-Isothermal Start-up of a Polymer Electrolyte Fuel Cell", ECS Trans., 11, (1) 395 (2007).

8. J.B. Siegel, D.A. McKay, A.G. Stefanopoulou, D.S. Hussey, D.L. Jacobson, submitted to the Journal of Electrochemistry 2008.

9. R. Mukundan, J.R. Davey, T. Rockward, J.S. Spendelow, B. Pivovar, D.S. Hussey, D.L. Jacobson, M. Arif, and R. Borup, "Imaging of Water Profiles in PEM Fuel Cells Using Neutron Radiography: Effect of Operating Conditions and GDL Composition", ECS Trans. 11, (1) 411 (2007).

10. R. Mukundan, Yu Seung Kim, T. Rockward, J.R. Davey, B. Pivovar, D.S. Hussey, D.L. Jacobson, M. Arif, and R. Borup, "Performance of PEM Fuel Cells at Sub-Freezing Temperatures", ECS Trans., 11, (1) 543 (2007).

11. 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", accepted ASME Journal of Fuel Cell Science and Technology, 2007.

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

13. Park, J., Li, X., Tran, D., Abdel-Baset, T., Hussey, D.S., Jacobson, D.L. and Arif, M., "Neutron imaging investigation of liquid water distribution in and the performance of a PEM fuel cell", International Journal of Hydrogen Energy (in press).

14. D.S. Hussey, D.L. Jacobson, M. Arif, "High Resolution Neutron Imaging", 212^{th} Meeting of the Electrochemical Society, Washington, D.C., October 9, 2007.

15. J.K. Park, K.A. Page, D.S. Hussey, D.L. Jacobson, M. Arif, R.B. Moore, "Morphology and transport considerations in the actuation mechanism of electrically-stimulated artificial muscles", 235^{th} ACS National Meeting, New Orleans, LA, April 2008.

16. J.K. Park, K.A. Page, D.S. Hussey, D.L. Jacobson, M. Arif, R.B. Moore, "Dynamics of water/counterion redistribution in an electrically-stimulated ionic polymer metal composite actuators", 235th ACS National Meeting, New Orleans, LA, April 2008.

17. M. Hickner, K.Chen, N. Seigel, D. McBrayer, D.S. Hussey, D.L. Jacobson, U.S.-Canada Fuel Cell Workshop, NRC Institute for Fuel Cell Innovation, Vancouver, B.C., Canada, March 16, 2008.

18. D.S. Hussey, D.L. Jacobson, M. Arif, "Neutron Radiography of Operating Proton Exchange Membrane Fuel Cells", McClellan RAC Meeting, McClellan, CA, December 13, 2007.

19. D.S. Hussey, D.L. Jacobson, E. Baltic, M. Arif, "Neutron Imaging: The key to understanding water management in hydrogen fuel cells", Washington Capital Science Meeting, Arlington, VA., March 29, 2008.