# V.F.5 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 (NIST)
100 Bureau Drive
Gaithersburg, MD 20899
Phone: (301) 975-6303
Email: arif@nist.gov

DOE Manager
Nancy Garland
Phone: (202) 586-5673
Email: Nancy.Garland@ee.doe.gov

Contract Number: DE-AI-01-01EE50660

Project Start Date: Fiscal Year (FY) 2001
Project End Date: Project continuation and direction determined annually by DOE

## Overall Objectives

- Provide state-of-the-art research and testing infrastructure to enable the fuel cell industry to design, test, and optimize prototype to commercial grade fuel cells using in situ neutron imaging techniques
- Provide a secure facility for proprietary research by industry; provide beam time at no cost to non-proprietary research through a competitive proposal process; 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

## Fiscal Year (FY) 2015 Objectives

- Collaborate and support groups from the DOE Hydrogen Program performing water transport measurements with neutron imaging at NIST
- Improve fuel cell measurement infrastructure based on needs of the fuel cell community
- Provide support to fuel cell infrastructure to enable testing of automotive-scale test sections
- Explore and develop high resolution neutron imaging methods to enable water transport studies of catalyst and membrane electrode assemblies (MEAs)
- Employ a high resolution imaging method to achieve resolution approaching 1 μm to resolve water concentration in fuel cell electrodes
- Determine and correct systematic effects due to spatial resolution effects

## Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section (3.4) of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan:

(A) Durability

(B) Cost

(C) Performance

## Technical Targets

This project is conducting fundamental studies of water transport in the fuel cell. Insights gained from these studies will be applied toward the design of components and operational strategies of polymer electrolyte membrane fuel cells that meet the following DOE fuel cell targets:

- Durability with cycling at operating temperature of ≤80°C: 5,000 h
- System energy density: 650 W/L
- System specific power: 850 W/kg
- Energy efficiency: 60% at 25% rated power
- Cost: $30/kWe
- Start-up time to 50% power: 30 s from -20°C, 5 s from 20°C
- Assisted start from low temperatures: -40°C
- Durability with cycling: 5,000 h

## FY 2015 Accomplishments

- Performed first user experiments employing the large scale test stand to study water transport and coolant distribution in a five cell stack
- Installed a new, second neutron imaging instrument
- Installed micro-focus X-ray source for simultaneous neutron/X-ray imaging
- Acquired components for new high resolution imaging detector systems

✧ ✧ ✧ ✧ ✧

## INTRODUCTION

At NIST, we maintain the premier fuel cell neutron imaging facility in the world and continually seek to improve its capabilities to meet the changing needs of the fuel cell community. This facility provides researchers with a powerful and effective tool to visualize and quantify water transport inside operating fuel cells. Imaging the water dynamics of a fuel cell 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, fuel cell industry personnel and researchers can obtain in situ, nondestructive, quantitative measurements of the water content of an operating fuel cell. Neutron imaging is the only in situ method for visualizing the water distribution in a "real-world" fuel cell. 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 length scales of interest in a fuel cell are: channels approximately 1-mm wide and 1-mm deep, the diffusion media (DM) is 0.1-mm to 0.3-mm thick, the membrane is 0.01-mm to 0.02-mm thick, and the active area of test sections can range from 2 $cm^2$ to 500 $cm^2$. Though the study of water transport within these length scales are technically very challenging, the unique capabilities of neutron imaging have already successfully addressed many questions. However, as fuel cell research matures, the water transport questions become increasingly more demanding, requiring for instance resolving the water content in catalyst layers. To meet these demands, based on fuel cell community feedback and need, we continue to develop new facilities and improve existing capabilities for obtaining higher spatial and temporal resolution neutron images. These improvements will enable users to perform even more detailed, nondestructive, and in situ studies of the water and hydrogen transport in fuel cells to meet DOE goals. In addition, employing mathematical models of neutron scattering, we will develop a software suite that enables users to obtain reliable, accurate, quantitative measurements of the water content in an operating fuel cell. Due to the complexity of fuel cells and the large number of remaining open questions regarding water transport, we will develop partnerships with industry, academia, and national laboratories to train them in the use of the facility, seek their feedback, and collaborate with them on research projects, to seek measurement breakthroughs that will facilitate the rapid, efficient, and robust development of fuel cells.

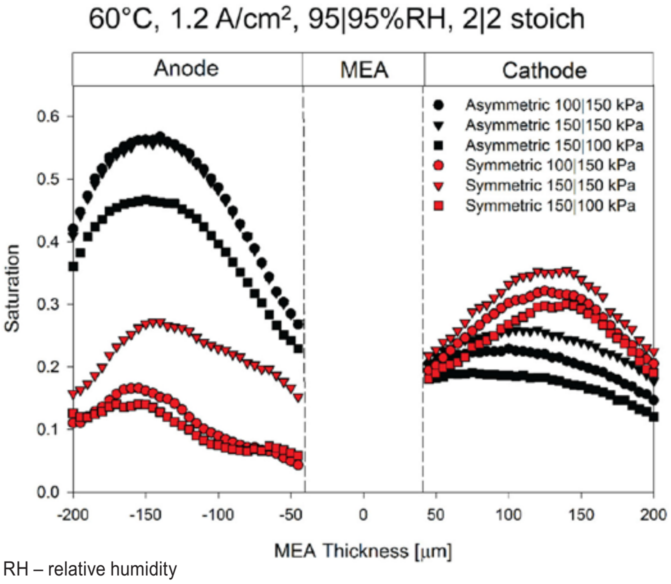
## RESULTS

The NIST Neutron Imaging Facility provides year-to-year support for the DOE Hydrogen and Fuel Cell Program projects through beam time and by collaboration with users on a variety of related neutron imaging projects that support the DOE mission. For FY 2015 General Motors (GM), Nissan, Los Alamos National Laboratory (LANL), University of Connecticut, University of California, Merced, Commissariat à l'énergie atomique et aux énergies alternatives (CEA), and University of Tennessee, Knoxville have received project support for experiments at the facility accounting for more than 70 days of beam time. Published results during FY 2015 from these experiments are reflected in the publication list attached to this report.

NIST now provides full support to full sized commercial and automotive grade fuel cell testing at the facility with a large scale fuel cell test stand. This stand was developed through the NIST partnership with GM. The facility technical staff has received extensive onsite training on calibration, operation and validation testing from our testing partners at GM. This test stand is capable of operating fuel cells and small stacks at 800 W, 6–1,000 A at 0.2 V, 0–50 V, hydrogen: 0.065–11.31 SLPM, air: 0.239–26.92 SLPM. The first user experiments employing the test stand were conducted in March, 2015 by researchers from CEA. CEA studied the water content and coolant distribution in a five cell stack.

The University of Tennessee in collaboration with GM have performed an extensive study of how changes to flow channels and diffusion media properties effect thermal transport of water through the cell [1]. One example from this study showed how reducing the number of serpentine flow channels on the anode bipolar plate in order to increase anode flow rates to better facilitate water transport out of the anode could result in more retained water instead of less (see Figure 1). The reason for the anode water content increasing is the fact that decreasing the number of serpentine flows increases the land surface area and thereby increases the thermal transport of heat away from the anode (see Figure 2). This can then result in an increase in the liquid water content on the anode. The study further showed that as the diffusion media saturates the thermal conductivity increases overall and results in a mitigation of the thermally driven transport. Using the diffusion media water saturation profile measured with neutron imaging the study was able to determine that the temperature driven effects are strongest at the inlets (see Figure 3) where the diffusion media is driest and becomes less pronounced near the outlets where the water saturation is the highest due to the flow of product water and humidified water out of the cell.

Normally groups come to NIST with their fuel cells and MEAs and perform testing in person at the facility. This can in some cases be difficult to schedule as travel is



RH – relative humidity

**FIGURE 1.** Water saturation data measured with neutrons for both symmetric and asymmetric flow field design. More water is retained in the asymmetric design even though the flow velocity is increased.

costly and user schedules must be commensurate with the facility schedule. NIST has been working with LANL to develop a protocol to facilitate users sending MEAs that can be run using standardized fuel cell hardware based on the LANL small scale cell design. This would allow users to reduce travel costs and have more timely results. To test the feasibility of this new capability LANL sent NIST three MEAs with non-precious metal cathode catalysts, each with a different catalyst thicknesses in an effort to determine if flooding of the catalyst support played a significant role in the reduced performance of the MEAs relative to MEAs made purely from platinum catalysts. The neutron imaging results from running the MEAs in the standard small scale hardware were consistent with flooding forming a significant contribution to performance loss. However, due to the lack of in situ impedance testing during the imaging it was not possible to cross validate catalyst flooding with impedance measurements. To address this issue at the facility our testing partners at GM have identified an impedance measurement apparatus that is fully supported by the existing stand software that would allow in situ impedance measurements

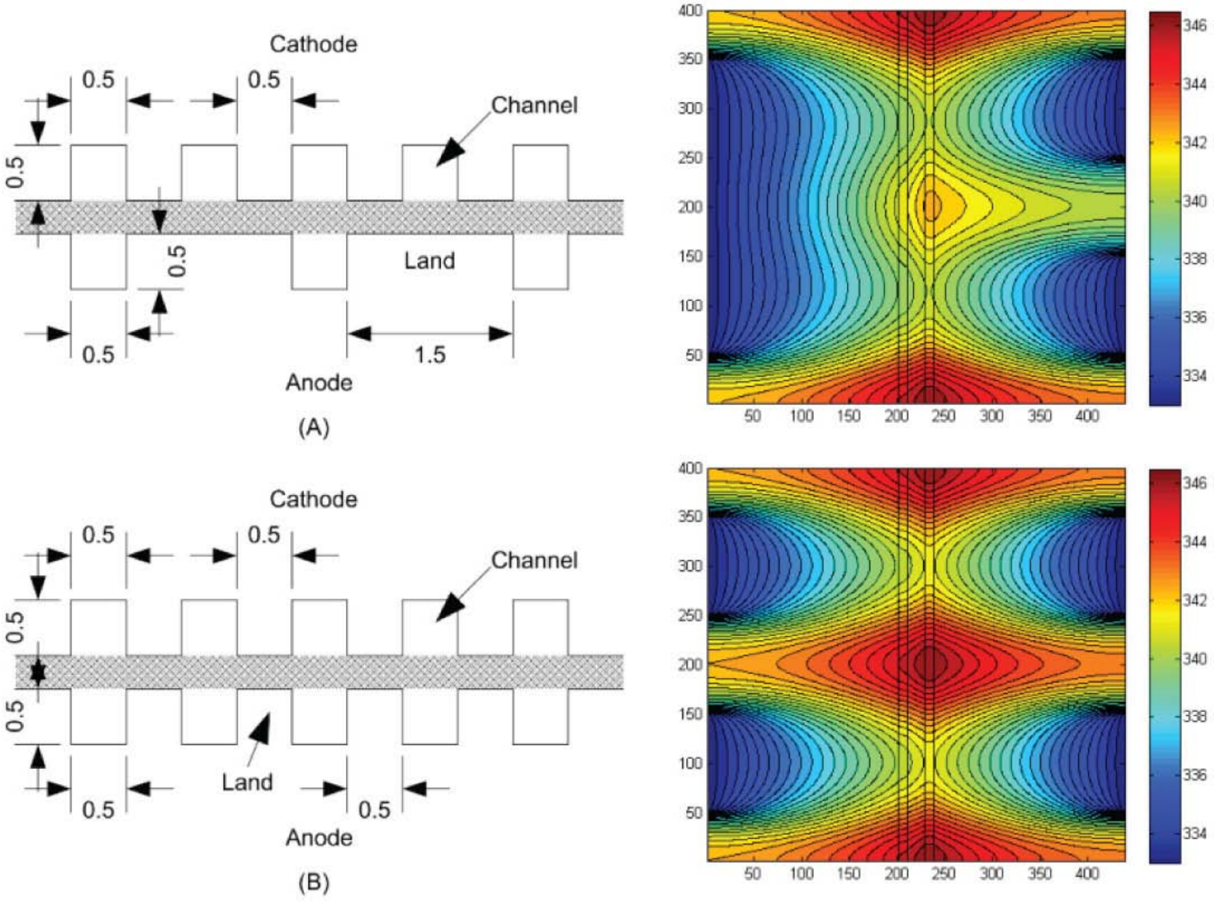


**FIGURE 2.** Asymmetric flow channels shown in (A) increase contact area on the anode thereby raising the thermal conductivity and decreasing the anode temperature demonstrated by the COMSOL generated temperature model shown on right. In (B) a symmetric flow field design is symmetric in temperature. The net effect if reducing the flow fields in (A) to increase flow rate and reduce water saturation is nullified by the increase in thermal conductivity. In fact the opposite of the desire effect occurs as water saturation increases.

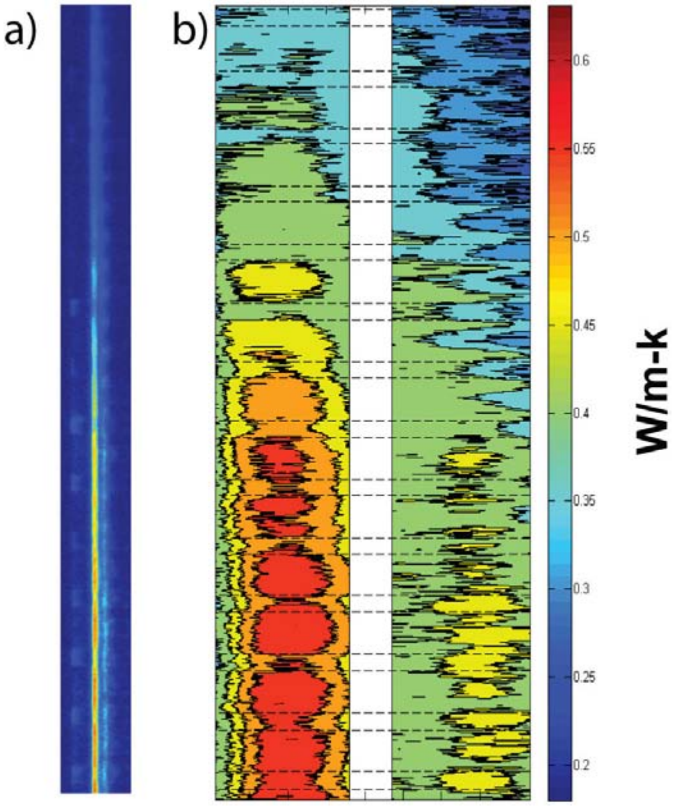


**FIGURE 3.** Water saturation of the diffusion media increases thermal conductivity and will serve to balance the temperature profile in the cell. The water saturation in a cell measured with neutrons is shown in a) and the calculated thermal conductance based on the measured water saturation profile is shown in b). Here neutrons allow one to directly determine the spatially distributed contributions to temperature driven water transport effects.

at the facility. An impedance measurement apparatus that is fully integrated with the fuel cell test stands at the facility, will be purchased and made available to facility users during FY 2016.

The facility has added a new X-ray modality to the facility capabilities with an X-ray microfocus tube oriented 90 degrees to the neutron beam. This allows one to concurrently image with neutrons and X-rays. This new X-ray imaging capability has spatial resolution approaching 10 µm.

Understanding flooding and degradation issues due to liquid water in the catalyst layers is a critical step towards improving durability and cycling of fuel cells. To study commercial grade platinum based catalysts requires at least a factor of 10 improvement in spatial resolution over current state-of-the-art (about 15 µm). The limiting factor in spatial resolution for current detector systems stems from the range of charged particles (3.5–150 µm) that are used to detect neutrons. To overcome this limit we are exploring several methods. The first method uses nanofabricated neutron absorbing gratings with an opening of ~2 µm or less in width to define the neutron illuminated area of the fuel cell with high spatial resolution in one dimension. By translating the grating across the through-plane direction of the fuel cell, one obtains a high resolution image of the water content from anode to cathode, overcoming the resolution limit of the detector. New gratings with thicker deposits of GadOx particles were obtained, and an image intensifier was purchased, but delivery has been delayed as the vendor addresses problems with the device to meet specifications. It is anticipated that users will have access to this new method by the end of calendar year 2015.

The data from the high resolution fuel cell images show that improvements to the signal to noise ratio are necessary to improve the quality of the data. This can be achieved with longer integration times or more efficient detectors. Longer integration times will limit the number of fuel cell operating conditions that can be measured. Therefore emphasis has been placed on improving the detector efficiency in order to improve the signal quality. Gadox scintillators have similar spatial resolution and four times the stopping power of the microchannel plates currently used for high resolution imaging. However, Gadox produces little light for each stopped neutron and has not been used due to small signal to noise ratios. Modern image intensifiers have been developed that enable high image resolution with stable long-term performance and initial tests showed that an intensifier improved the signal to noise ratio by over a factor of 30. A new detector system based on an intensifier has been designed, and an image intensifier is being procured for fuel cell users. This new system is expected to be available at the facility by January 2015 and is expected to increase the time resolution for high resolution neutron imaging by a factor of four due to the increased neutron detection efficiency. In addition, by magnifying the GadOx scintillation light and taking images at high frame rate, the intensifier may make it possible to obtain higher resolution by calculating the centroid of the emitted light.

By using a neutron lens, it could be possible to increase the neutron intensity by 50 to 100 times than currently available. Previously, practical lenses for neutrons have not been available due to the low neutron refractive power of all materials. However, a new X-ray telescope lens technology using thin nickel foil mirrors developed by the National Aeronautics and Space Administration has shown great promise to provide a practical lens for neutron imaging. By nesting several mirrors, the flux can be increased up to a factor of 100 over BT2. An engineering demonstration lens with 20 µm spatial resolution is expected at the end of calendar year 2016. If successful, a complete optic with 10 nested shells will be produced in 2017 for 20 µm resolution imaging, and in 2018 a magnifying optic to reach ~1 µm resolution. Such "Wolter optics" will be installed at the new cold neutron imaging instrument. This new imaging instrument is being completed in August 2015 and will begin commissioning measurements in September 2015. The new instrument will also serve as a test bed for new high spatial resolution detector systems and methods and with the lower energy neutron spectrum enable discrimination of ice and water in the fuel cell during freeze operation.

## CONCLUSIONS AND FUTURE DIRECTIONS

- NIST Neutron Imaging Facility continues to maintain a robust fuel cell user program with:
  - Eight publications and 10 presentations in FY 2015.
  - Over 50 days of dedicated fuel cell beam time.
- Fuel cell infrastructure supports automotive-scale testing.
  - NIST staff were trained at General Motors to support calibration and use of the test stand.
  - The first user tests were performed by CEA.
- An MEA mail-in program is currently being developed.
  - The program makes use of the standard high resolution fuel cell that is available to users.
  - The first tests with MEAs provided by LANL show it could be a valuable new capability.
  - The program will be fully optimized by new fuel cell test stand supported in situ impedance testing capability.
- With the goal to study catalysts, NIST continues to improve the image spatial resolution. New avenues toward resolving the MEA water content include:
  - Employing a grating method to achieve resolution approaching 1 µm with 12 hour acquisition time (end of 2015).
  - Developing a magnifying neutron lens to reach 1 µm with 20 min acquisition time (2018).
- NIST will improve fuel cell high resolution image time.
  - Time resolution for through-plane water content measurements improved by a factor of four with 20 µm scintillator detector.
  - Future neutron lens will increase time resolution by 50x to achieve image times of 10 s with 10 µm resolution.
- Additional general improvements will be pursued.
  - A second new cold neutron imaging facility will begin operation by Fall of 2015.
  - Improvements to achieve 1 µm imaging will be continued.
    - Develop neutron lens.
    - Improve grating method.

## FY 2015 PUBLICATIONS/PRESENTATIONS

**1.** Rupak Banerjee, Satish G. Kandlikar, “Liquid water quantification in the cathode side gas channels of a proton exchange membrane fuel cell through two-phase flow visualization,” Journal of Power Sources, **247**, (2014), Pages 9–19.

**2.** Jon P. Owejan, Thomas A. Trabold, and Matthew M. Mench, “Oxygen transport resistance correlated to liquid water saturation in the gas diffusion layer of PEM fuel cells,” International Journal of Heat and Mass Transfer, **71**, (2014), Pages 585–592.

**3.** Adam Z. Weber, et al., “A Critical Review of Modeling Transport Phenomena in Polymer-Electrolyte Fuel Cells,” Journal of the Electrochemical Society, **161** (12), (2014), Pages F1254–F1299.

**4.** Jacob M. LaManna, Subhadeep Chakraborty, Jeffrey J. Gagliardo, Matthew M. Mench, “Isolation of transport mechanisms in PEFCs using high resolution neutron imaging,” International Journal of Hydrogen Energy, **39**, (2014), Pages 3387–3396.

**5.** Toshikazu Kotaka, Yuichiro Tabuchi, Ugur Pasaogullari, Chao-Yang Wang, “Impact of Interfacial Water Transport in PEMFCs on Cell Performance,” Electrochimica Acta, **146**, (2014), Pages 618–629.

**6.** Toshikazu Kotaka, Yuichiro Tabuchi, Ugur Pasaogullari, Chao-Yang Wang, “Investigation of Interfacial Water Transport at the Gas Diffusion Media by Neutron Radiography,” ECS Transactions, **64**, (2014), Pages 839–851.

**7.** Thiago Lopes, Jose-Maria Sansiñena, Rangachary Mukundan, Daniel S. Hussey, David L. Jacobson, Fernando H. Garzon, “Diagnosing the Effects of Ammonia Exposure on PEFC Cathodes,” Journal of The Electrochemical Society, **161**, (2014) Pages F703–F709.

**8.** Bradley M. Wood, Kyungmin Ham, Daniel S. Hussey, David L. Jacobson, Adel Faridani, Anders Kaestner, John J. Vajo, Ping Liu, Tabbetha A Dobbins, Leslie G Butler, “Real-time observation of hydrogen absorption by $LaNi_5$ with quasi-dynamic neutron tomography,” NIM-A, **324**, (2014), Pages 95–101.

**9.** D.J. Fenton, J.J. Gagliardo, and T.A. Trabold, “Analysis of Water Morphology in the Active Area and Channel-to-Manifold Transitions of a PEM Fuel Cell,” ASME 2014 12th International Conference on Fuel Cell Science, Engineering and Technology, Boston, Massachusetts, USA, June 30–July 2, 2014.

**10.** J.M LaManna, M.M. Mench, “Comparison of Water Transport Mechanisms Between Commercial and Experimental Virgin and Aged Diffusion Media Using High Resolution Neutron Radiography,” The Electrochemical Society Meeting Abstracts, Issue 14 (2014), Pages 646–646.

**11.** R. Mukundan, R.L Borup, D. Spernjak, G. Wu, A. Steinbach, P. Zelenay, D. Hussey, D. Jacobson, “Neutron Imaging to Optimize PEM Fuel Cell Performance,” World Conference on Neutron Radiography, Grindelwald, Switzerland, October 2014.

**12.** R. Mukundan, D. Spernjak, G. Wu, D.S. Hussey, D.L. Jacobson, A.J. Steinbach, R.L. Borup, P. Zelenay, “Visualizing water in non-precious metal catalyst-based polymer electrolyte fuel cells using neutron imaging,” Abstracts of Papers of the American Chemical Society, **248**, August 2014.

**13.** M. Arif, D.S. Hussey, E. Baltic, and D.L. Jacobson, “Recent Developments at the NIST Neutron Imaging Facility,” World Conference on Neutron Radiography, Grindelwald, Switzerland, October 2014.

**14.** D.S. Hussey, C. Brocker, D.L. Jacobson, E. Baltic, and M. Arif, “A New Cold Neutron Imaging Instrument at NIST,” World Conference on Neutron Radiography, Grindelwald, Switzerland, October 2014.

**15.** D.S. Hussey, D.L. Jacobson, D. Liu, B. Khaykovich, M. Gubarev, M. Arif, “Design of Neutron Microscope Objectives,” World Conference on Neutron Radiography, Grindelwald, Switzerland, October 2014.

**16.** J.M. LaManna, D.S. Hussey, D.L. Jacobson, and M.M. Mench, “Role of Land Drainage on Water Storage in Polymer Electrolyte Fuel Cells Using High Resolution Neutron Imaging,” World Conference on Neutron Radiography, Grindelwald, Switzerland, October 2014.

**17.** Md. A. Uddin, T. Kotaka, U. Pasaogullari, Y. Tabuchi, D.S. Hussey, D.L. Jacobson, “High Resolution Neutron Radiography of the Behavior of Liquid Water at the Interfaces in Polymer Electrolyte Fuel Cells,” World Conference on Neutron Radiography, Grindelwald, Switzerland, October 2014.

**18.** P. Saggu, D.L. Jacobson, D.S. Hussey, D. Pushin, E. Baltic, and M. Arif, “Using Micro-Channel Plates to Achieve Both High Spatial and Temporal Resolution in Neutron Radiography,” World Conference on Neutron Radiography, Grindelwald, Switzerland, October 2014.

## REFERENCES

**1.** LaManna, Jacob Michael, “The Role of Channel-Land Architecture, Diffusion Media Transport Properties, and Aging Effects on Water Transport and Storage in Polymer Electrolyte Fuel Cells.” PhD diss., University of Tennessee, 2014. (http://trace.tennessee.edu/utk_graddiss/3145).