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

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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 and academia needs.

Fiscal Year (FY) 2016 Objectives

- Collaborate and support groups from the DOE Hydrogen and Fuel Cells Program performing water transport measurements with neutron imaging at NIST.
- Deploy new cold neutron fuel cell imaging facility for high resolution imaging of fuel cells.
- Install fuel cell and support infrastructure at the new cold imaging facility.
- 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.
- Deploy and develop in situ X-ray imaging for fuel cells at the neutron imaging facility.

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

(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 operation strategies of proton exchange membrane fuel cells that meet the following 2020 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: \$40/kW$_e$
- Start-up time to 50% power: 30 s from -20°C, 5 s from 20°C
- Assisted start from low temperatures: -40°C

FY 2016 Accomplishments

- First 4 µm resolution fuel cell images using slits
- Installation of a new, second neutron imaging instrument
- Installation of micro-focus X-ray source for simultaneous neutron/X-ray imaging
- Components for new high resolution imaging detector systems acquired

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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, non-destructive, 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) are 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 is 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, national laboratories, and the DOE Fuel Cell program consortia 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 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 2016 General Motors, Los Alamos National Laboratory, University of California, Merced, University of Toronto, South African Nuclear Energy Corporation, and University of Tennessee, Knoxville have received project support for experiments at the facility. The results published during FY 2016 from these experiments are reflected in the publication list attached to this report.

Researchers from the Thermal and Electrochemical Energy Laboratory, at the University of California, Merced performed a study of two different diffusion media looking at the onset liquid water condensation using a fuel cell based on the Los Alamos National Laboratory high resolution cell. Two cells were built: the first labeled here as Cell 4, with a Nafion® XL (~30 µm) membrane, and Toray (~178 µm) DM and the second labeled Cell 5, with Nafion XL (~30 µm) membrane and Freudenberg (~203 µm) DM. The test conditions were: 50°C, 77% relative humidity; 0.3V; 300 kPa abs, high flow conditions (stoichiometry ratios > 30/30 anode/cathode), 100% hydrogen concentration with 2%, 8%, and 16% oxygen concentration. Starting under dry conditions (2% O_2), the water saturation in the DM of both cells is similar (see Figure 1). Under wet condition (8% O_2), liquid water is saturated throughout the diffusion media thickness for Toray DM. In contrast, liquid water is only saturated away from the MEA near the land for Freudenberg DM.

The same trend is observed for DM under the channel area. It can be clearly observed that Freudenberg DM provides much more open path for oxygen diffusion compared to Toray DM.

In consultation with the fuel cell community, one of the leading issues this project has been asked to address is fuel cell flooding and degradation due to liquid water in the catalyst layers. 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 µm to 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

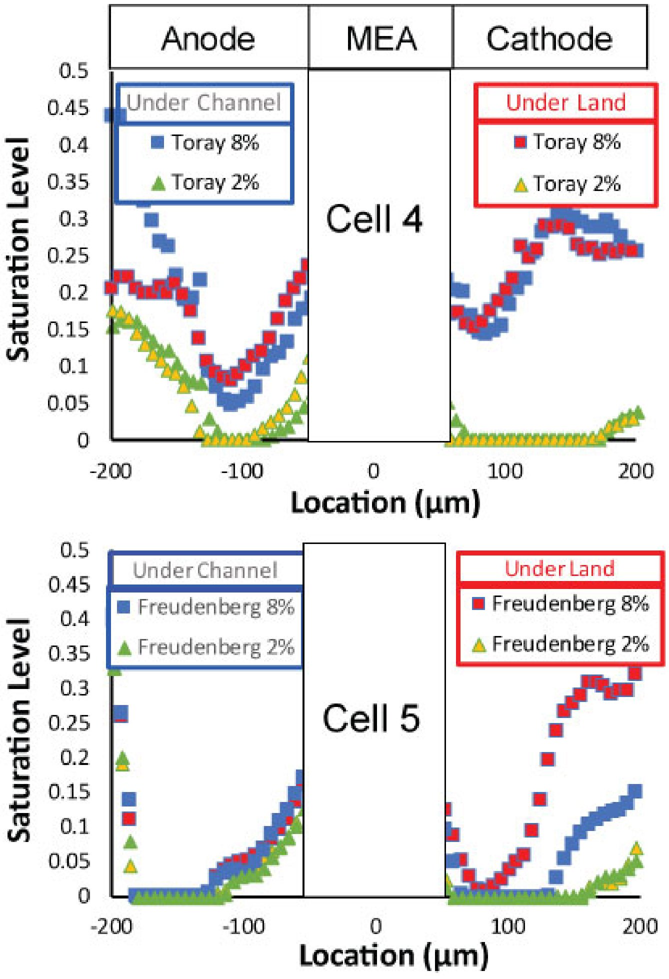

FIGURE 1. Measurements as the onset of liquid water saturation measured with neutrons for both Toray and Freudenburg DM. Under (8% O_2) liquid water saturation, the Freudenburg DM shows less liquid water saturation allowing for greater pathways of O_2 diffusion than Toray DM.

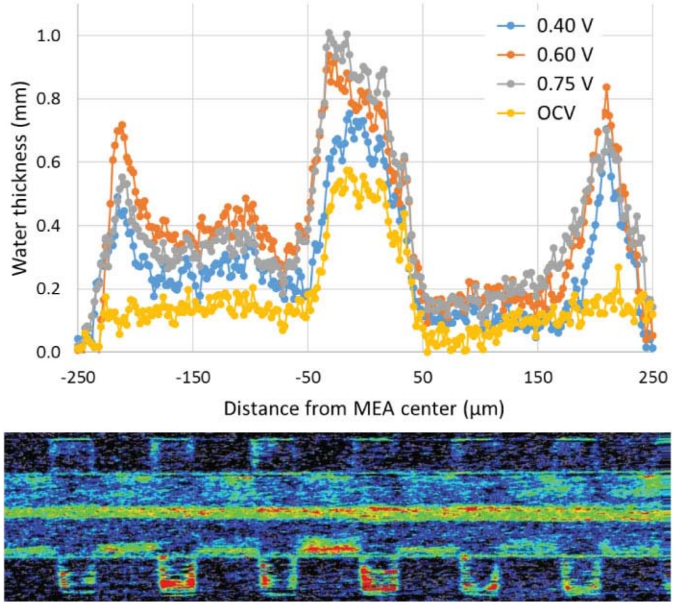

FIGURE 2. Resolution of 4 µm was achieved to measure through-plane water saturations of a fuel cell running at several different current densities. The images were taken using 2 µm gratings translated across the fuel cell during a 17 h acquisition.

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 silicon gratings with thicker deposits of neutron absorbing gadolinium oxysulfide particles were developed by collaborators from Pusan University, Korea, and a new image intensifier required to improve the detector signal-to-noise ratio was finally accepted in January. This allowed for the first time to image a fuel cell with 4 µm spatial resolution (double the sampling resolution as required by the Shannon-Nyquist theorem) (see Figure 2).

With the new intensified camera, it was also possible to test a centroiding method with the gadolinium oxysulfide scintillators. By capturing images quickly (0.005 s exposures), it was possible to see individual neutron events in the camera and find the center of mass of each event. This method has shown improved spatial resolution of about 5 µm and may be able to be used to further improve spatial resolution with other methods. As this first test involved saving many images and post processing them, future work will look toward a hardware based real time centroiding method.

Ultimately resolution of 1 µm is expected to be efficiently and practically achieved using a neutron magnifying lens. 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 NASA 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 that achievable at BT2. An engineering demonstration of the new lens with 20 µm spatial resolution was performed in July 2016 and has shown good results that are currently being analyzed. A picture of the test lens is shown in Figure 3. 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 that was commissioned August of 2015 and is shown in Figure 3. The cold neutron imaging 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.

FIGURE 3. Top showing the inside the new cold neutron imaging instrument that was commissioned in August of 2015. Bottom showing the engineering test optic mounted on the beam line to test the NASA fabrication methods. Based on the analysis of the results a final 1:1 optic will be delivered to NIST for the user program in 2017.

CONCLUSIONS AND FUTURE DIRECTIONS

- NIST Neutron Imaging Facility continues to maintain a robust fuel cell user program.
- New cold imaging facility will allow more rapid development of high resolution methods to measure MEA water content.
- Good progress has been made towards developing the method to measure liquid saturation values in the catalyst and membrane.
  - Slit scanning
    - 4 µm spatial resolution was demonstrated.
    - Acquisition time is 17 h, but could be improved to less than 8 h with a smaller grating period.
  - Centroiding has shown that 5 µm is possible
    - Method needs further refinement.
    - Future: develop hardware based centroiding to allow high throughput.
    - Future: method could be combined with the lens to improve resolution beyond the targets of this project.
  - Wolter optics
    - Flagship method to achieve spatial resolution of 1 µm
    - Validation of NASA fabrication techniques during July 2016
    - Future 2017: high speed 20 µm optics, 2018: 1 µm optics
- User program
  - New cold imaging facility is currently being upgraded to include full support.
  - Including electrochemical impedance spectroscopy into the scripting of the test stand would be a great benefit to the users.
  - It was observed from fuel cell testing that Freudenberg DM shows improved performance under wet and cold operating condition due to improved oxygen diffusion over Toray DM.

FY 2016 PUBLICATIONS/PRESENTATIONS

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3. F. Nandjou, J.-P. Poirot-Couvezier, M. Chandesris, S. Rosini, D.S. Hussey, D.L. Jacobson, J.M. LaManna, A. Morin, Y. Bultel, “A pseudo-3D model to investigate heat and water transport in large area PEM fuel cells – Part 2: Application on an automotive driving cycle,” submitted to Int. J. Hydrogen Energy.

4. Jacob LaManna, Matthew Mench, “Channel-land configuration control of temperature driven water transport in polymer electrolyte fuel cells,” in preparation (2016).

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14. LaManna, Jacob M; Hussey, Daniel S; Jacobson, David L; Mench, Matthew M; "Influence of Thermal Conductivity and 2-D Temperature Distribution of Liquid Water Saturation," The Electrochemical Society Meeting Abstracts 37 (2015) 1539–1539.

15. Sperjnak, Dusan; Chung, Hoon T; Mukundan, Rangachary; Borup, Rod L; Hussey, Daniel S; Jacobson, David L; Wu, Gang; Zelenay, Piotr; "Water Management in PEM Fuel Cells with Non-Precious Metal Catalyst Electrodes," The Electrochemical Society Meeting Abstracts 37 (2015) 1540–1540.