Isolation of Transport Mechanisms in PEFCs with High Resolution Neutron Imaging

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Cell water balance and anode to cathode water transport were investigated using high resolution neutron radiography in the through-plane direction. Induced pressure and relative humidity gradients were imposed across the membrane electrode assembly to determine how these gradients alter saturation profiles in the diffusion media. The asymmetric anode/cathode channel pitch used in this study resulted in large anode water saturation changes with applied gradients while cathode saturation remained more constant. A new low-cost core-shell diffusion media is introduced as a potential replacement of current diffusion media. This new material was found to reduce overall cell water content, but suffers poor overall performance due to excessive ohmic losses, and more work is needed to develop this concept.

Introduction

Proton exchange fuel cells (PEFC) are a top candidate for the replacement of fossil fuel based internal combustion engines. Proper water management has been identified as critical to fuel cell performance (1). To determine the liquid water content within an operating fuel cell, neutron radiography has been used for nearly a decade. The in-plane water distribution has been extensively researched to determine water distribution (2-6), flow-field influences (7, 8), diffusion media effects on saturation (9), and differential pressure effects on saturation (10). With the advent of the microchannel plate technology with spatial resolution now approaching 10 μm, through-plane water distribution studies have been conducted to determine liquid water saturation profiles through the diffusion media (11-13).

The work presented in this paper is motivated by the need to understand the impacts of imposed gradients of concentration, temperature, and pressure on water redistribution in an operating cell. When complete this data and analysis will be used to provide detailed computational model validation and develop simplified transport relationships. The neutron imaging in this study was accomplished with a specially designed fuel cell at the high-resolution neutron facilities at the NIST Center for Neutron Research. The channel geometry used in the fuel cell is designed to simulate full cell automotive fuel cell hardware and appropriately reflect industrial conditions including generic manifold design and channel features.

Experimental
Fuel Cell Design

In this work, a 4.8 cm² fuel cell was utilized that was specifically designed for simulation of full cell geometry and high resolution neutron imaging. The total cell width was minimized to reduce geometric blurring effects in the images. Liquid coolant channels are located on the outside faces of the current collectors to provide precise thermal boundary conditions, as shown in Figure 1. The temperature of the coolant is controlled with circulator baths with accuracies of ±0.2°C. The flow field design utilizes a triple serpentine channel with dimensions of 0.5 mm depth and width. Land widths are asymmetrical between the anode with lands of 1.5 mm and the cathode with lands of 0.5 mm. The asymmetrical design is considered the baseline geometry for this project as defined in (14) to build on results of a previous US Department of Energy supported project (15).

Figure 1. (a) Schematic of fuel cell hardware, reactant flows from top to bottom in the direction of gravity, (b) Coolant channel placement on outside faces of current collectors to obtain precise thermal boundary conditions, and (c) Schematic of asymmetrical flow field design from ref (15).

Neutron Imaging and Processing

At each of the operating conditions, five current densities were tested, 0.1, 0.4, 0.8, 1.2, and 1.5 A/cm² to obtain a wide range of characteristic working conditions. At each current density, the fuel cell was allowed to attain and maintain steady state for 30 minutes. A 13 μm resolution microchannel plate (MCP) imaging system at NIST was utilized for these tests (16). Images were collected with an exposure time of 5 seconds, which yielded approximately 300 images for each test condition. A single compiled image was extracted by averaging these 300 images. After a
gamma correction, the images were normalized with the flat field to remove the detector structure, since the detector field response is not generally uniform.

The resulting image was further normalized with respect to an invariant part of the image to compensate for small changes in neutron fluence and then a ratio of intensities for the wet and dry image was calculated. For the saturation profile plots, a ratio of the sum of intensities along the axis (Transmission ratio \( T_r \)) was calculated for each pixel location across the thickness of the DM and MEA. Saturation values were calculated based on a compressed porosity calculated to be 68% for the baseline material and 66% for the core-shell set.

For the density images, the intensities were not summed in any directions, rather the transmission ratio was calculated for each pixel. The final images were filtered with a \( 6 \times 6 \) median filter, to de-noise the images.

Test Cases and Materials

Materials tested consist of Mitsubishi Rayon Corporation Grafil U-105 diffusion media, W.L. Gore 18 μm membrane, and catalyst loadings of 3.5 mg/cm\(^2\) on the cathode and 0.05 mg/cm\(^2\) on the anode. More details on material composition are available in ref. (14).

Results and Discussion

Influences of Pressure Gradient

Pressure gradients were applied across the cell by adjusting the cell exhaust pressure which ranged from 100 to 150 kPa. Three exhaust pressure pairs were used as follows to produce the gradients 100/150, 150/150, 150/100 kPa to produce a 50 kPa gradient towards the anode, no gradient, and a 50 kPa gradient towards the cathode. With the cell configuration used in this study, it was found that the anode saturation was significantly more sensitive to pressure gradients than the cathode. As seen in Figure 2, cathode saturation quantities remain relatively constant for all three pressure gradients at 0.8 and 1.5 A/cm\(^2\) current densities whereas the anode varies significantly. These results will be used to validate ongoing modeling work (14, 17).

![Comparison of anode/cathode saturation sensitivity as a function of backpressure at 0.8 A/cm\(^2\) (left) and 1.5 A/cm\(^2\) (right). Pressure gradient of -50 kPa corresponds to 100/150 kPa.](image-url)
Influences of Relative Humidity Gradient

Four cell inlet relative humidity (RH) pairs were applied to the cell to study the influence of RH gradients on saturation profiles. Gradients of 45% inlet RH were achieved with 50/95% and 95/50% anode/cathode cell inlet RH. Zero percent gradients were produced with 50/50% and 95/95% inlet RH. As shown in Figure 3, the relative humidity was found to have a strong influence on the anode water content. While the cathode saturation remained relatively stable.

![Saturation profiles as a function of relative humidity across the diffusion media and MEA with anode on left and cathode on right. Anode shows greater sensitivity to relative humidity gradients than does the cathode at constant operating conditions. Operating conditions: 60°C, 150/150 kPa exhaust pressure, 1.5 A/cm².](image)

The larger cooling area of the anode lands increases liquid water content at high anode inlet humidification. This increase results in approximately double the saturation in the anode diffusion media compared to the cathode diffusion media. Cathode saturation increases for the 95/50% RH case but not for the 50/95% RH case due to the large accumulation of liquid water in the anode for the 95/50% RH case. Water is diffused to the cathode side by the concentration gradient present across the membrane. The results suggest the net water balance in the through plane direction of the cell can be manipulated with an asymmetric channel/land design. This can be useful to optimize performance at extreme operating conditions where anode or cathode dry-out, for example, limits performance.

Influences of Current Density
Current density was varied from 0.1 A/cm² to 1.5 A/cm² to determine the influence on diffusion media saturation levels. Cathode saturation levels increase with increasing current density due to the increase in water production and retention. Interestingly, for this asymmetric design the anode maintains higher average saturation levels compared to the cathode, even without the presence of imposed gradients in Figure 4. This is a result of the additional area located under lands.

![Figure 4. Influence of current density on saturation at baseline conditions. The low channel area of the anode results in increased water accumulation with current. The sensitivity of the anode is higher than the cathode at moist conditions. Operating conditions: 60°C, 150/150 kPa exhaust pressure, 95/95%](image)

**Experimental Core-Shell Diffusion Media**

A candidate for next generation diffusion media was developed by General Motors (18) that consists of a core-shell construction. A low-cost fiber core with conductive metallic shell make up the structure of the material. The goal of this design is to custom tailor thermal and electrical properties of the diffusion media while reducing cost by up to 40% over graphite based diffusion media. The largest cost associated with graphite based diffusion media is the carbonization process that requires large amounts of energy. By switching to a non-graphite based diffusion media, the graphitization process can be removed, thus significantly reducing the manufacturing costs. Fiber structure and cross-sectional images are given in Figure 5.
The core-shell material was found to produce reduced saturation profiles for both the anode and cathode diffusion media, as shown in Figure 6. Large accumulations of liquid water did not build up in the diffusion media due to the small compressed thickness of approximately 95-100 μm and increased heat load associated with a 2x increase in real resistance. The high ohmic losses is mainly due to uneven plating through the thickness of the substrate. However, with further development this material may enable more optimization parameters for reducing water content when compared with carbonized polymer materials.

Figure 6. Saturation profiles as a function of current density of the core-shell diffusion media. Profiles for anode and cathode remain similar through all current densities tested with no indication of increased sensitivity on the anode. Test conditions: 60°C, 95/95% RH, 150/150 kPa backpressure.

Conclusions

High resolution through-plane neutron radiography was used in this work to elucidate the influences of pressure and relative humidity gradients on cell water balance. Additional analysis of the results is ongoing at the time of publication and will be presented at a later date.
innovative, low cost core-shell diffusion media material was introduced that resulted in low relative performance but reduced liquid water content.

- The asymmetrical flow field design with large anode land areas used in this study increases the steady state retention of liquid in the diffusion media, resulting in saturation levels greater than that in the cathode diffusion media. The results suggest the net water balance in the through plane direction of the cell can be manipulated with an asymmetric channel/land design. This can be useful to optimize performance at extreme operating conditions where anode or cathode dry-out, for example, limits performance.
- The liquid saturation level in the cathode diffusion media was shown to be less sensitive to pressure and humidity gradients than the anode diffusion media, which was extremely sensitive for the asymmetric fuel cell design tested.
- Core-shell diffusion media construction shows promise of reducing cell water content and reducing diffusion media costs by up to 40%, but requires further work to reduce ohmic losses.

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