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In situ Liquid Water Visualization in Polymer Electrolyte Membrane Fuel Cells with High Resolution Synchrotron X-ray Radiography

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Abstract. In this work, we investigated the dominating properties of the porous materials that impact water dynamics in a polymer electrolyte membrane fuel cell (PEMFC). Visualizations of liquid water in an operating PEMFC were performed at the Canadian Light Source. A miniature fuel cell was specifically designed for X-ray imaging investigations, and an in-house image processing algorithm based on the Beer-Lambert law was developed to extract quantities of liquid water thicknesses (cm) from raw X-ray radiographs. The X-ray attenuation coefficient of water at 24 keV was measured with a calibration device to ensure accurate measurements of the liquid water thicknesses. From this experiment, the through plane distribution of the liquid water in the fuel cell was obtained.

INTRODUCTION

Polymer electrolyte membrane fuel cells (PEMFCs) are electrochemical conversion devices, which use hydrogen and oxygen to produce electricity, with heat and water as the only by-products. Although this technology is considered as a promising alternative to the internal combustion engine for automotive applications, increasing the fuel cell performance, reliability and durability are necessary for reaching commercial viability. In particular, fuel cell performance is hindered by the non-optimal management of the liquid water produced by the electrochemical reaction [1]. Liquid water is necessary for ensuring ionic conductivity of the membrane, but excess liquid water also reduces pathways for the reactants (air and hydrogen) to reach the catalyst layers. This pathway reduction leads to the decrease in fuel cell power output.

Gas diffusion layers (GDLs) are porous media inserted between the fuel cell flow field plates and the catalyst layers (see Fig 1). GDLs are typically composed of carbon fibers (either woven or non-woven) and are 80% porous with a mean pore size of 50 μm. The GDL is a critical component in fuel cells, because it moderates the transport of liquid water out of the fuel cell.

Synchrotron X-ray radiography has been recognized as a powerful method for studying liquid water transport behavior in an operating fuel cell. This technique provides high spatial resolutions, between 1–10 μm, and high temporal resolutions with minimal alteration to the original hardware [2], offering advantages over other in situ visualization techniques [3]. However, Kim et al. [4] recently pointed out that the accurate measurement of in situ liquid water quantities requires the calibrating of the X-ray attenuation coefficient of liquid water.

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In this work, we used synchrotron X-ray radiography to visualize the through-plane liquid water at the micron scale in the GDLs of an operating fuel cell. The water attenuation coefficient was also measured to ensure an accurate quantification of the liquid water; a calibration device was specifically designed to achieve this goal. This enabled us to obtain the through-plane liquid water profile in the fuel cell.

METHODOLOGY

The fuel cell used in this study was specifically designed for in-plane liquid water visualizations using X-ray radiography. The membrane electrode assembly (MEA) was composed of a thin membrane (thickness of 15 µm) coated with two 10 µm-thick catalyst layers. A TGP-H-060 (Toray Industry©) carbon paper was used as anode and cathode GDLs. A schematic of a fuel cell with the MEA, the GDLs and flow field plates is depicted in Fig. 1. The in situ X-ray visualizations were performed at the Biomedical Imaging and Therapy (BMIT) bending magnet beamline (05B1-1) at the Canadian Light Source Inc. in Saskatoon, Canada [5]. An energy level of 24 keV was selected using a double-crystal Bragg monochromator placed upstream of the cell. The intensity of the X-ray beam that passed through the cell was measured by a Hamamatsu C11440-22CU charge-coupled device (CCD) camera with a Hamamatsu M11427-42 scintillator. The pixel resolution was 6.5 µm and the temporal resolution was 3 s.

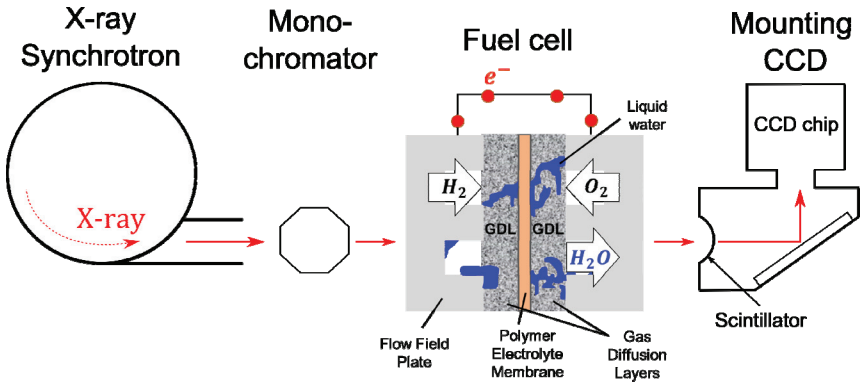

FIGURE 1. A schematic of the experimental setup used to measure the in situ liquid water distribution in an operating fuel cell.

The Beer-Lambert law was used to translate the grayscale values of the radiographs recorded by the CCD camera into water thicknesses X_w, given by

$$X_w = \frac{1}{\mu_w}.\ln\left(\frac{I_d}{I_w}\right) \quad (1)$$

This image processing was based on determining the difference in pixel values between a dry image (I_d) obtained during open circuit voltage operation and a wet image (I_w) obtained once liquid water had accumulated in the fuel cell, during normal cell operation. The difference between the wet image and the dry image corresponded to liquid water thickness values, calculated from the liquid water attenuation coefficient μ_w [6].

A calibration device was designed to measure the liquid water attenuation coefficient (see Fig. 2b). This device was composed of Polydimethylsiloxane (PDMS), chosen specifically to simulate similar scattering and attenuation effects due to the higher harmonics that would occur during in operando fuel cell imaging. This material has similar X-ray properties to the PEN sheet gaskets used in our fuel cell (attenuation coefficient of 1 cm^{-1} vs. 0.54 cm^{-1} at 24 keV for the PDMS and the PEN gasket, respectively). PDMS was also chosen for the high microscale precision and ultra-fast fabrication that is afforded through soft-lithography. Eleven reservoirs of thicknesses ranging from 1 to 12 mm were fabricated with high precision. These reservoirs were used to measure the X-ray attenuation of known liquid water thicknesses, from which the liquid water attenuation coefficient was calculated at the energy level of 24 keV.

EXPERIMENTAL RESULTS

Measurement of the Water Absorption Coefficient

The ratio of the radiographs of the reservoir with and without liquid water was presented versus the reservoir thickness, see Fig. 2a. These experimental data were least squares fitted, and a water attenuation coefficient of 0.4936 cm^{-1} was obtained at the energy level of 24 keV. The water attenuation coefficient from the NIST database, which is usually used to convert the raw radiograph into liquid water thickness [3], was 10% higher (0.5426 cm^{-1}) at the same energy level. This difference is caused by higher-than-expected X-ray intensities in the wet image that resulted from the phase contrast effect, the scattering of the photons, and higher harmonics. Specifically the portion of scattered photons that traveled towards the scintillator led to a higher intensity measured by the scintillator. The higher harmonics are contaminating high energy photons from the double-crystal monochromator. As a result, the higher harmonics were attenuated less by the imaging sample which led to undesirably high measured intensities, and this effect was amplified by the relatively thick sample used in this study (31 mm). Similar mechanisms takes place in fuel cells where the X-ray beam need to pass through an equivalent thickness of PEN gaskets before they reach the liquid water. Thus, the calibration of the X-ray absorption coefficient of liquid water is critical to ensure an accurate measurement of the liquid water quantities. This conclusion is in agreement with Kim et al. [4].

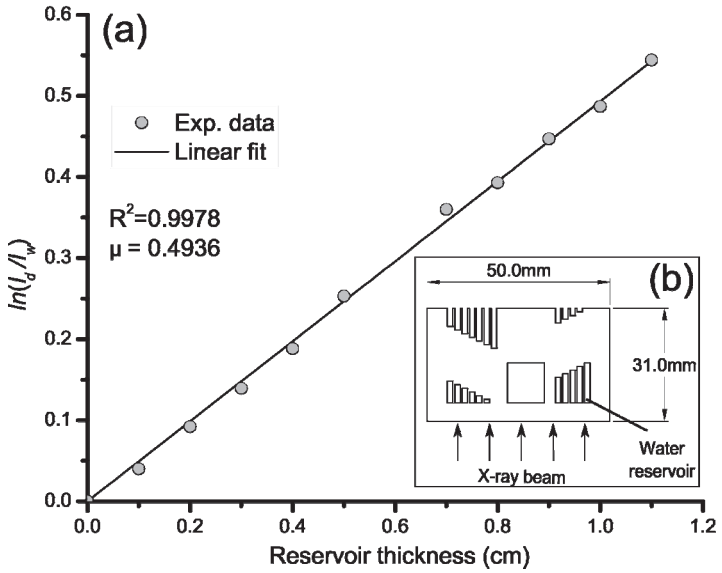

FIGURE 2. (a) Measurement of the attenuation coeficient of liquid water at 24 keV. (b) Design of the calibration device used to measure the attenuation coefficient.

Measurement of Liquid Water in the Fuel Cell

Figure 3a presents a raw radiograph of the fuel cell; the MEA, the flow field, and the GDL of the anode and the cathode are visible in greyscale values. This radiograph was processed using equation (1), with the water attenuation coefficient previously measured, and our in-house algorithm to obtain the water thicknesses. In Fig. 3b, the color of each pixel indicates the water thickness between -0.1 and 0.6 cm. The image processing assumed that the cell did not move between the dry and wet images, but micro-metric movements cannot be avoided during the experiment. These movements resulted in artifacts in the measurement of the water thicknesses which produced negative values.

The profile of liquid water (Fig. 3c) was extracted by averaging each pixel in Fig. 3b over the vertical direction in the rectangle with white dashed lines. Both anode and cathode GDLs exhibit a significant amount of liquid water, particularly close to the MEA where two local maxima of thickness at 0.38 cm and 0.54 cm were measured in the anode and the cathode, respectively. A significant decrease of the water thicknesses toward the flow field was observed and almost no liquid water is present close to the GDL-flow field interface. This kind of water distribution can be associated with a capillary dominated transport regime in the GDL [7], where the liquid water percolates in the MEA-GDL interfaces and invades the GDL toward the flow field.

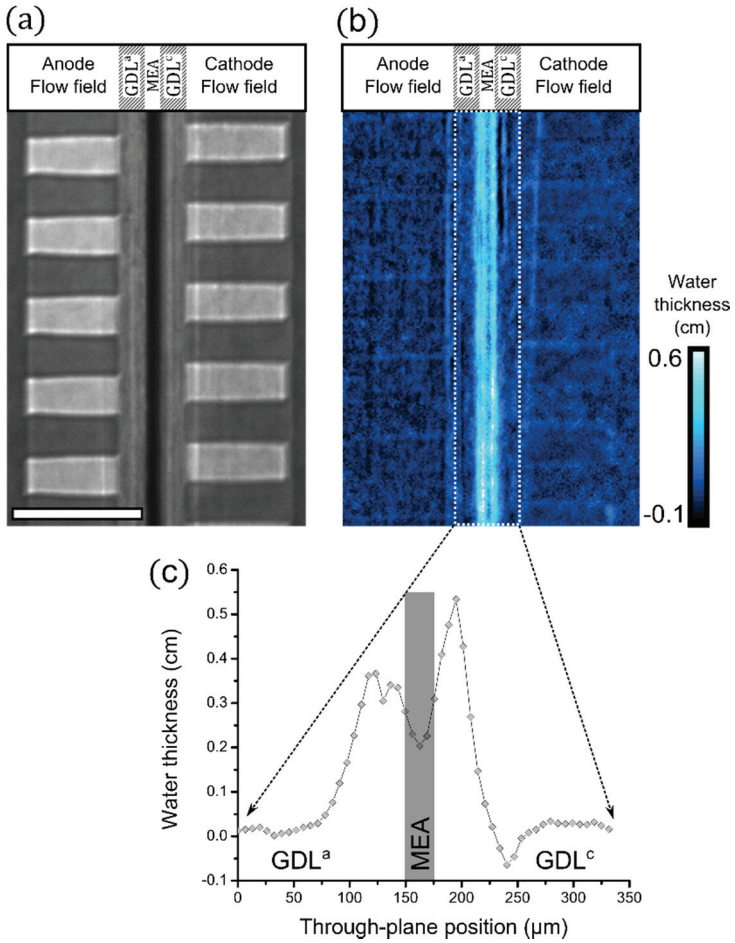

FIGURE 3. Synchrotron X-ray radiographs, (a) raw image (b) processed image (c) water thickness profile in the GDLs and the MEA of the processed image. The length bar in (a) is 1 mm.

CONCLUSIONS

The calibration of the water attenuation coefficient was performed to ensure the accurate in situ measurement of the liquid water thicknesses in an operating fuel cell. The in situ profile of water was obtained which provides new insights in the transport of liquid water in the GDL. In particular, the transport was identified to be capillary dominated. As a result, the structure of the GDL, such as the pore size and the porosity distribution, can be tailored to decrease the liquid water buildup and improve the reactants diffusion toward the catalyst layers.

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