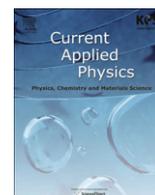




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## Breakthrough/drainage pressures and X-ray water visualization in gas diffusion layer of PEMFC

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### ABSTRACT

The primary role of the gas diffusion layers (GDLs) in polymer electrolyte membrane fuel cells (PEMFC) is to maintain the delicate balance between water retention and removal in GDLs. Water management in the fuel cell is related to the breakthrough pressure at which water starts to pass through GDL, and the drainage pressure, which is maintained after the breakthrough. These pressures are both related to water management in fuel cells. Here we measured these pressures for two different GDLs and used X-ray tomography to visualize the water distributions within them. We then relate the variations in liquid pressures to the visualization and discuss water management in PEMFC.

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### 1. Introduction

A fuel cell is an electrochemical device that converts the bond energy of reactants directly to electricity. PEMFC, which uses a soft-matter proton-conducting polymer membrane as the electrolyte, produces water and this water either remains in the pores of the gas diffusion layer (GDL) and membrane or leaves the fuel cell. Management of the pore water is required to maintain appropriate water content in the electrolyte phase and to ensure proton transport through it, while avoiding flooding, which hinders the transport of reactants by filling the pores in the GDL and by covering catalytic reaction sites. Failure to do this results in a significant reduction in cell performance and durability [1,2]. Because pore water content is directly influenced by the GDL structure, understanding water distribution in the GDL is important.

Many studies have examined two-phase transport in PEM fuel cells using constitutive relationships for liquid water flow in the porous layers. Two approaches exist for modeling the liquid water behavior in PEM fuel cells. The first is the macroscopic PEM fuel cell model, based on the two-phase Darcy law under assumptions of a homogeneous pore structure and uniform wetting characteristics [3–6]. The second approach is pore-scale modeling and simulation, which is based on a more precise investigation of the liquid water transport mechanism through a fibrous diffusion media (DM) structure [7,8]. These approaches use the capillary pressure in a GDL as an important parameter in analyzing water transport. However, the capillary pressure from analytic equations has limitations in modeling the characteristics of real-life GDLs, because GDL carbon fibers have heterogeneous surfaces and structures. The breakthrough pressure [9,10] and the drainage pressure can be used to represent GDL characteristics. These pressures are strongly related to the water distribution in a GDL. In this study, the water distributions in GDLs with and without the microporous layers (MPLs) were visualized with the X-ray tomography, which was employed to visualize the water transport in the PEM fuel cells. Previous studies [11–15] have also used the X-ray imaging, here we used the X-ray microscope at the Pohang Accelerator Laboratory in Korea to

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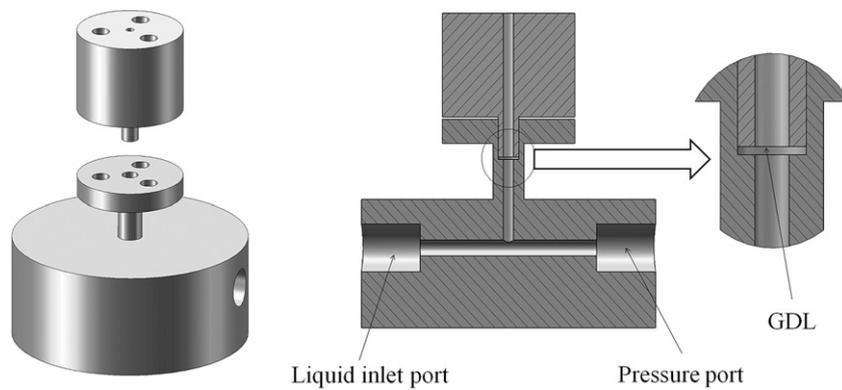


Fig. 1. GDL housing for measurement of capillary pressure.

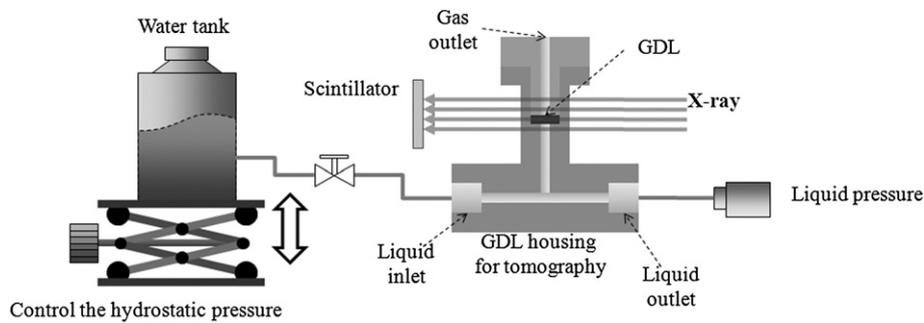


Fig. 2. Schematic diagram of experimental apparatus. The liquid inlet port is connected to the pressurized water. The liquid outlet, air inlet, and outlet ports were exposed to ambient air.

transient observations and the accompanying measurement of the breakthrough and the drainage pressures.

## 2. Experiment and discussion

An experimental system was designed to visualize GDLs with X-rays while measuring the liquid pressures. As shown in Fig. 1, a GDL housing consisting of two cylindrical blocks was used to enclose a GDL. Pressurized water was supplied to bottom of the GDL and the water pressure was measured with a pressure transmitter as shown in Fig. 2. The experiments were performed at the 7B2 X-ray Microscopy Beamline of the Pohang Accelerator Laboratory. The X-ray beam from a bending magnet was created with dimensions  $120$  (horizontal)  $\times$   $45$  (vertical)  $\mu\text{m}^2$  with the microscopy system located at a distance of  $34.8$  m. The beam angular divergences were  $8.3$   $\mu\text{rad}$  (vertical) and  $20$   $\mu\text{rad}$  (horizontal), and the beam strengths were  $2.8$  keV at  $2$  GeV/ $5.5$  keV at  $2.5$  GeV. The X-ray beam was directed through the sample to a  $150$   $\mu\text{m}$   $\text{CdWO}_4$  scintillator that converted the X-rays to visible beam. This visible beam reflected off a mirror into a  $4008 \times 2672$ -pixel charge-coupled-device camera through a microscope. The field of view was  $5.05 \times 3.36$   $\text{mm}^2$ .

The typical experimental process was as follows:

- 1) Supply pressurized water through liquid inlet.
- 2) Close the liquid inlet valve when the liquid pressure reaches the hydrostatic pressure.
- 3) Record X-ray images of the test section while the liquid pressure is at steady state.
- 4) Repeat steps 1–3.

The first experiment was done with an untreated Toray TGPB-090 GDL. In step 3 of the experimental process, the liquid pressure decreased until it reached steady state (Fig. 3). The reason for this behavior is that the supplied water needed time to diffuse fully in the GDL. As the experimental processes were repeated, the liquid pressure increased to the breakthrough pressure  $P_1$  (Fig. 3). Breakthrough occurred at pressures above  $P_1$ , and the liquid pressure decreased dramatically to almost zero. The liquid pressure did not drop completely to zero after breakthrough because the water that passed through the GDL into the gas outlet hole increased the

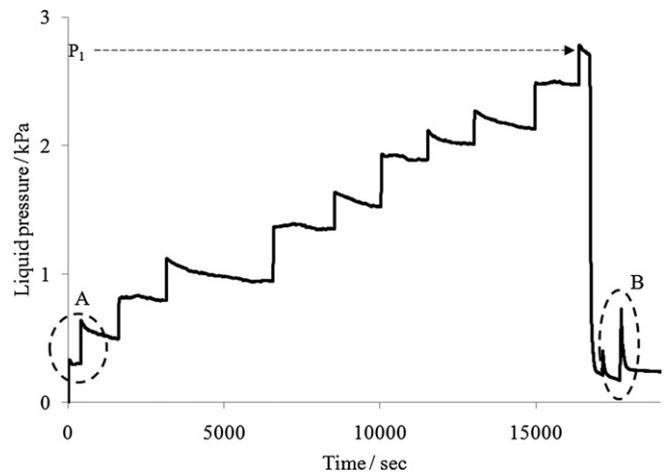
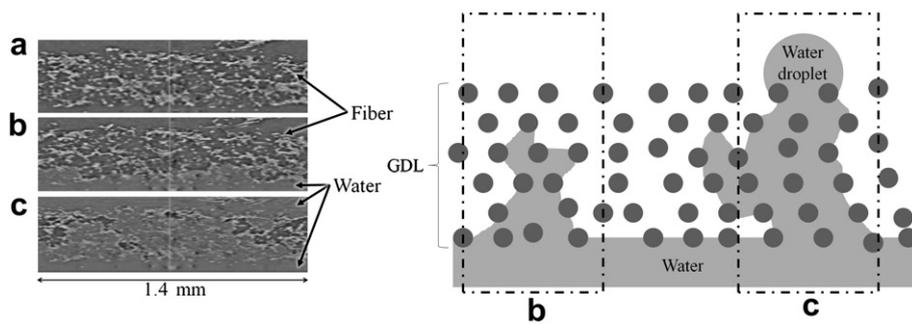


Fig. 3. Liquid pressure for the Toray TGPB-090 GDL.



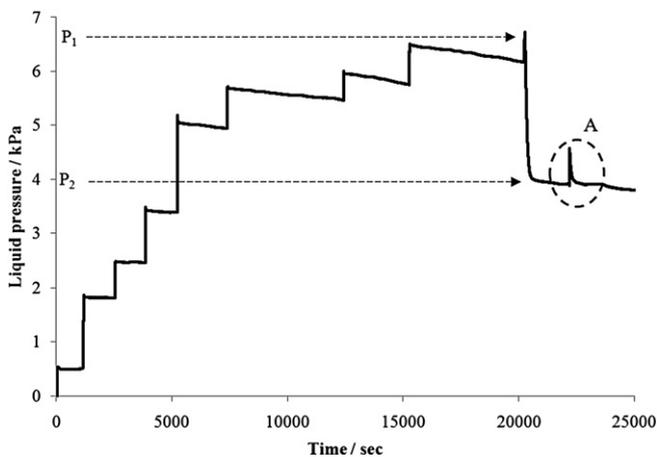
**Fig. 4.** The X-ray tomography and the simplified model results for GDL water distribution of the untreated TGPH-090, (a) before water injection, (b) at less than 2.13 kPa, and (c) after the breakthrough.

hydrostatic pressure. The liquid inlet valve was then opened to increase the liquid pressure again. In this case, the liquid pressure also decreased dramatically (region B in Fig. 3) to near zero, in contrast to region A (Fig. 3) where the liquid pressure remained constant.

This difference between regions A and B can be explained by X-ray tomography water visualization and the simplified model (Fig. 4). Before the liquid pressure reached  $P_1$ , the percolated water could not form a complete water column from the liquid side to the other side of the GDL. Thus, capillary force, which prevented water movement, affected the head of the water column. Otherwise, after the liquid pressure reached  $P_1$ , the water column connected both sides of the GDL and the capillary force at the head of the water column vanished. For this reason, after the liquid pressure reached  $P_1$ , water could readily move up through the water column under

low pressure conditions, whereas it could not move up before the liquid pressure reached  $P_1$ . This indicates that this GDL easily vented water after breakthrough.

The experiment was repeated with an SGL 10BB GDL, which was made by applying a MPL coating to a 10BA GDL (Fig. 5). In this case, the breakthrough pressure was greater than that of the TGPH-090 GDL, even though previous research reported that 10BA has a lower capillary pressure than the TGPH-090 according to the pressure–saturation relationship [16]. Above  $P_1$ , the liquid pressure also decreased, but the decrease stabilized at a certain pressure ( $P_2$ , the drainage pressure) while the liquid pressure of the TGPH-090 after breakthrough decreased to almost zero. In the visualization of the water distribution in 10BB, no water was observed in the fiber structure of the GDL before breakthrough and percolated water existed in the fiber structure of GDL after breakthrough (Fig. 6). This result contrasts with the case of the TGPH-090 in that some water percolated into the GDL before breakthrough. After water passed through the MPL, its pressure was higher than the breakthrough pressure of the GDL fiber structure because the MPL had a higher capillary pressure than that of the GDL. Thus, the breakthrough in the whole GDL occurred after breakthrough in the MPL, and water was observed in the fiber structure. To maintain the liquid pressure after breakthrough ( $P_2$ ), we suggest that the water column formed was broken in the MPL below the drainage pressure, because the water could not attach to the surface of the hydrophobic particles of the MPL. These behaviors of pressure indicate that the MPL is the dominant factor in the water behavior in GDLs and is a useful tool for maintaining the hydrated condition of the membranes in operating fuel cells where flooding often occurs.



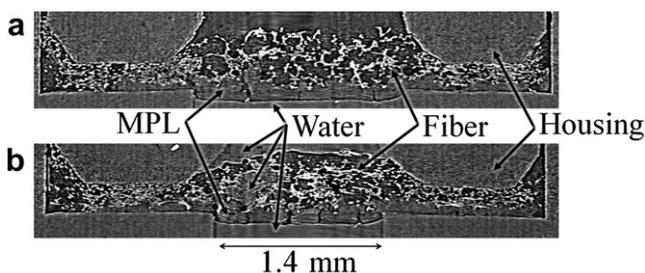
**Fig. 5.** Liquid pressure for the SGL 10BB GDL.

### 3. Conclusions

In this study, the liquid water pressures in two different GDLs are measured and analyzed using X-ray tomography of the water distribution. The liquid pressure in TGPH-090 GDL dramatically vanishes after the breakthrough, since a complete water column is formed when the breakthrough occurs. In the case of the 10BB, the breakthrough pressure is larger, even though 10BA has a lower capillary pressure. Moreover, the decrease in liquid pressure after the breakthrough stabilizes at a nonvanishing drainage pressure. These results demonstrate that the MPL controls the water behavior in GDLs and is a useful tool for controlling the humidity in PEMFC.

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**Fig. 6.** Water distribution in the GDL of the bare SGL 10BB (a) at less than 6.2 kPa and (b) after breakthrough.

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