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Visualization of water accumulation process in polymer electrolyte fuel cell using neutron radiography

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Abstract

In order to clarify the water-accumulation phenomena in an operating polymer electrolyte fuel cell (PEFC), the water distribution in a small fuel cell was measured in the through-plane direction by using neutron radiography. The fuel cell had nine parallel channels for classifying the water-accumulation process in the gas diffusion layer (GDL) under the lands and channels. The experimental results were compared with numerical results. The water accumulation in the GDL under the lands was larger than that under the channels during the period of early PEFC operation. The difference of the water accumulation in the GDL under the land and channel was related to the water vapor. Because of the land, the vapor fraction in the GDL under the land was also higher than that under the channel. As a result, condensation was easy to occur in the GDL under the land.

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Keywords: Water distribution; Neutron radiography; In-situ measurement; Vapor fraction

1. Introduction

Fuel gas (hydrogen gas) and oxidant gas (air) are supplied to a polymer electrolyte fuel cell (PEFC). Condensation may occur on the cathode side, because the water vapor may be super saturated by the fuel cell reactions. If condensed water exists in the gas diffusion layer (GDL), it depresses the gas diffusion as flooding. The generated water must be appropriately supplied to the proton exchange membrane (PEM) for proton conduction.

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Hence, water management is significantly important for the PEFC performances, and clarifying the water-transport mechanisms between the PEM, GDL, and gas channel is of great concern. Neutron radiography is a powerful tool for observing the water distributions in the PEFC, and many investigations have been carried out [1-5].

Purpose of this study is to investigate the water accumulation process in the GDL under the lands and under the channels. The water distributions in a PEFC with nine parallel channels was employed to distinguish the GDLs under the lands and under the channels. The water distributions in the through-plane direction were obtained every 60 s by using neutron radiography. The results were compared with numerical results.

2. Experimental and analysis methods

2.1. Experimental method

The fuel cell used in this research for visualization using neutron radiography is shown in Fig. 1. The separators have nine parallel gas channels with equal widths and depths of 1 mm. It was expected that the water distributions in the GDL between the lands and the channels would be different, which might affect gas diffusion. Therefore, the authors selected parallel channels to visualize the difference in the water-accumulation processes in the GDL by the lands and channels.

A PEM was sandwiched between the GDLs and separators at the anode and cathode. The PEM was Nafion® NR-212 with a thickness of approximately 90 μm having a catalyst layer (CL) on both the anode and cathode sides. The electrode area was 10×19 mm^2. The GDL was carbon paper (Toray Ind.), with thicknesses of 190 μm at the cathode and anode. The porosity of the GDL was 78%. The experimental conditions included the following: current densities i = 158 mA/cm^2, hydrogen flow rate = 28 cc/min, and air flow rate = 66 cc/min without humidification. The hydrogen and oxygen utilization was 7.5%. The experiments were carried out at room temperature, and the temperature of the PEFC varied within the range of 30–35 °C.

The neutron radiography facility at the B4 port in the Kyoto University Reactor (KUR) was used in this study. Neutrons emanatd from the side-view direction and attenuated by the PEFC including the accumulated water. The transmitted neutrons were converted to visible rays using a scintillator. 16-bit gray-scale images were taken using a cooled CCD camera (PIXIS 1024, Princeton Instruments) with a resolution of 1024×1024 pixels. The field of view was 9×9 mm^2, and the pixel size of the image was 8.8 μm. Image processing was applied to the images to obtain the two-dimensional distributions of water thickness along the neutron beam (Murakawa et al., 2011). The thickness represents the increase in the water accumulation referenced to the thickness at 0 min. The measurement area was around the 3rd to 6th channel, as shown in Fig. 1. In this experiment, we focus on the transient changes in the water distributions. Hence, the exposure time was set to 60 s to obtain the water distribution in the through-plane direction of the PEFC every 60 s during operation. L/D was set to 278 using a collimator with diameter of 5 mm.

2.2. Analysis method

A model proposed by Natarajan et al. (2001) was used for the analysis of liquid and gas distributions. Fig. 2 shows the schematic of the model region. The analysis region is the GDL under a land and a channel. The analytical conditions are the same as the experimental conditions. All phases are assumed to be continuous, and the property of the GDL is assumed to be uniform. Water generates at the boundary between the catalyst layer and the GDL as water vapor depending on the local current density. Water saturation in the channel is set to zero. When water locally accumulates in the channel, the boundary condition between the GDL and the channel also changes. However, the change of the boundary condition due to the water accumulation in the channel was neglected. Hence, it is difficult to compare the experimental and analytical results after the time at which this occurs. A periodic boundary condition is applied at the top and bottom in the GDL.

The governing equations for liquid water and gas phase are expressed as:

$$\varepsilon_0 \frac{\partial s}{\partial t} + \nabla \cdot q_{H_2O} = -R_{H_2O} \tag{1}$$

$$\varepsilon_0 \frac{\partial \{(1-s)x_i\}}{\partial t} + \nabla \cdot q_i = R_i \tag{2}$$

where q is the mass flux across the cross-sectional area, s is the liquid water saturation, R is the source term due to the phase change, ε_0 is the porosity in the GDL, x is the mole fraction, and the subscription of H_2O represents the liquid water, i represents the gas species. The gas phases of oxygen, water vapor and nitrogen are solved by using the Stephan-Maxwell multicomponent diffusion equation. Details of the analysis model are shown in Natarajan et al., 2001.

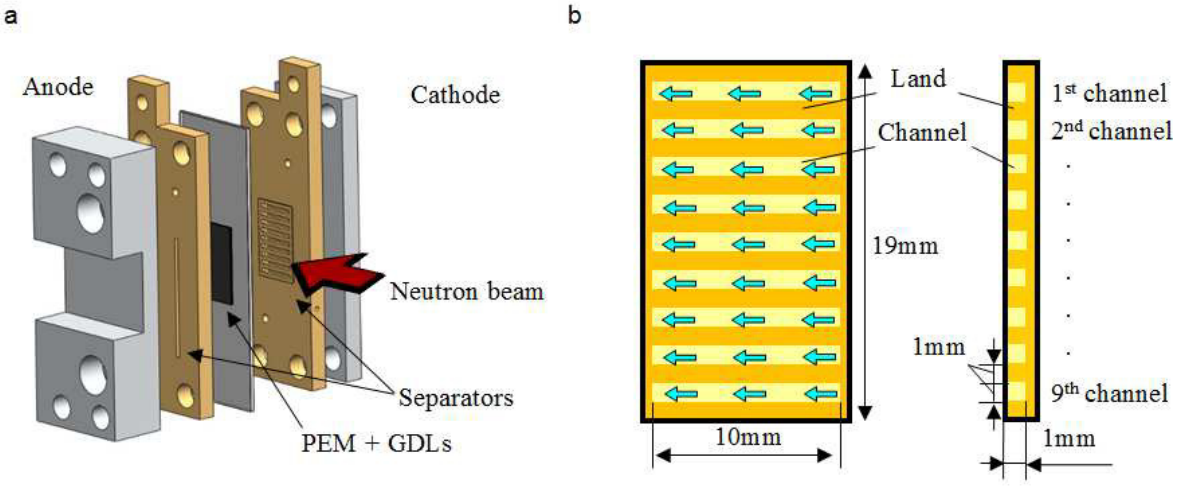

Fig . 1 (a) diagram of the PEFC; (b) geometry of the gas channel.

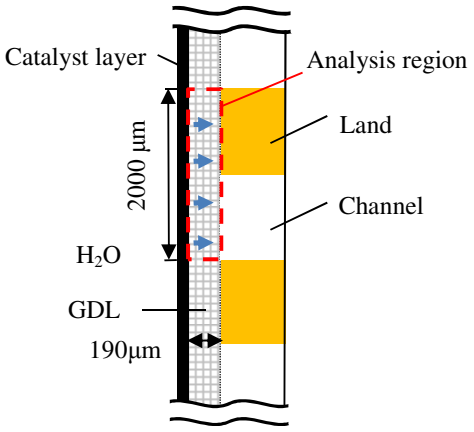

Fig. 2. Schematic of the model region.

3. Results and discussion

3.1. Two-dimensional water distributions

Fig. 3 presents two-dimensional water distributions. It can be observed that water accumulation occurred as the operation time increased, as indicated by the blue color. The difference in the amount of water in the GDLs under the lands and under the channels apparently appears at 8 min. The water accumulation in the GDL under the land was larger than that under the channel during the period of early PEFC operation. Water accumulation started in the GDL, and a large amount of water existed in the GDL under the lands. The water-accumulation area extended to the GDL under the channels. Furthermore, with an increase in the operation time, the water evacuation from the GDL to the channels is confirmed to occur mainly around the land corners. Liquid droplets formed in the channels, and these grew in the channels as the operation time increased. No water evacuation in the downstream channel direction was observed during the experiments. This is because the channels were parallel, and water evacuation to the outside of the channels was difficult.

3.2. Analytical results

Fig. 4 shows results of water saturation and vapor mole fraction distributions in the cathode GDL at 2, 4 and 8 min. The horizontal and vertical scales are not identical for understanding the distributions easily. The liquid water accumulation is confirmed mainly at the boundary between the catalyst layer and the GDL. At the early time of the PEFC operation, the water accumulates almost uniformly along the catalyst layer. However, the difference of the water saturation appears with an increase of the operation time. The tendency of the water accumulation in the GDL is good agreement with the experimental results. Water saturation in the GDL near the channel is lower than that in the other region. The results are strongly affected by the boundary condition between the channel and GDL.

It can be seen that the vapor mole fraction in the GDL under the land is also higher than that under the channel. The difference of the water accumulation in the GDL under the land and under the channel was related with the water vapor. Therefore, it can be thought that the higher vapor mole fraction causes the water accumulation in the GDL under the land.

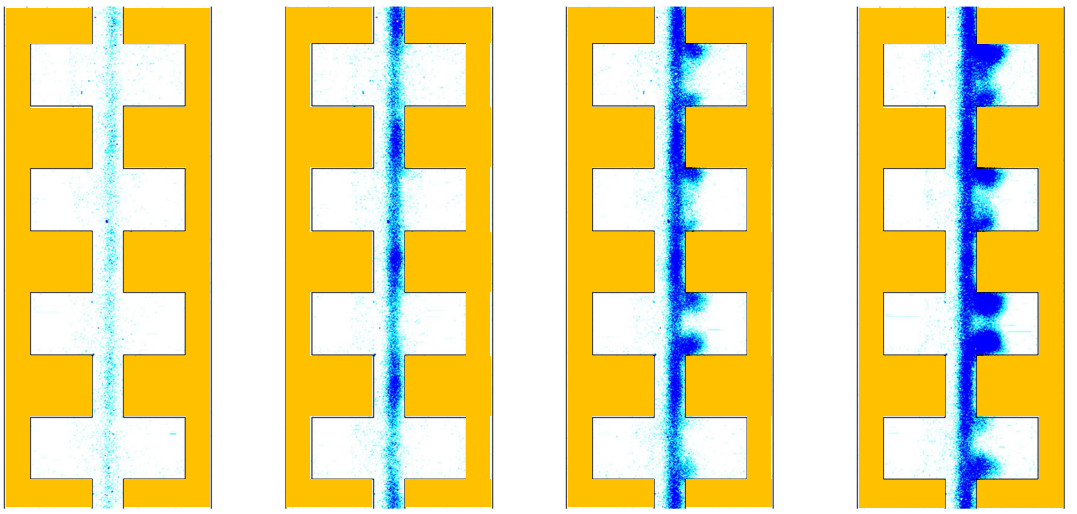

Fig. 3. Time-change of the two-dimensional water distribution around the 3rd to 6th channel obtained by using neutron radiography.

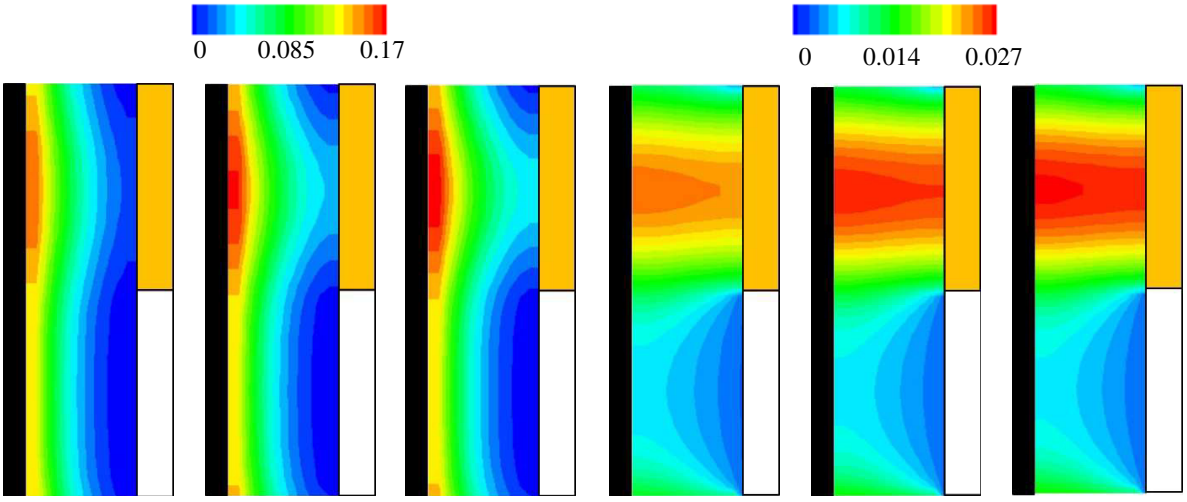

Fig. 4. Analytical results of water saturation and vapor fraction distributions in the cathode GDL under a channel and a land.

4. Conclusions

To investigate the difference of water accumulation in the GDL under the land and under the channel, in-situ measurements of the water distributions in the through-plane direction were carried out by using neutron radiography. A PEFC with nine parallel gas channels was used to distinguish the water accumulation in the GDL under the lands and channels at room temperature. The measurements results were compared with the analytical results. The water accumulation in the GDL under the land was larger than that under the channel during the period of early PEFC operation. The difference of the water accumulation in the GDL under the land and under the channel was related to the water vapor. Because of the land, the vapor mole fraction in the GDL under the land was higher than that under the channel. As a results, condensation was easy to occur in the GDL under the land.

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