

Optimization of Fuel Cell Generation Environment by Water Observation using Neutron Radiography

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ABSTRACT

Electric generation by a polymer electrolyte fuel cell requires an appropriate quantity of water to help ensure proton conductivity of the membrane electrode assembly. However, excess water in the gas channels and the gas diffusion layer is thought to be a factor impeding gas supply, so it is necessary to appropriately control the water inside the fuel cell during electric generation.

This study used neutron radiography to observe the state of the water accumulated in a fuel cell during electric generation in order to clarify the mechanism by which water accumulates and its effect on voltage behavior, and set the optimum electric generation conditions.

Observation using neutron radiography was performed from the two directions of the through-plane direction and the in-plane direction of the fuel cell. Observation in the through-plane direction of an actual cell clarified the correlation between the accumulated water and the voltage behavior. On the other hand, observation in the in-plane direction of a test cell determined the gas channels in which accumulated water exists and the reason for that water accumulation, and suggested directions for countermeasures. Based on these results, the optimum electric generation environment was set in terms of water and electric generation performance, and applied to the control of an actual vehicle.

1. Introduction

1.1. Fuel Cells

Fuel cell electric vehicles, which are the ultimate form of electric vehicle that does not emit CO₂ or hazardous substances, are being developed to address environmental issues such as global warming and air pollution, and energy issues due to depleting fossil energy such as petroleum resources. Honda is also promoting the development of fuel cell electric vehicles using polymer electrolyte fuel cells (PEFC) that can start from low temperatures and also be made smaller.

Fuel cells are power generating devices that produce electricity by the electrochemical reaction of oxygen and hydrogen. The actual reaction comprises the oxidation reaction of hydrogen at the anode shown in Eq. (1), and the oxygen reduction reaction at the cathode shown in Eq. (2).

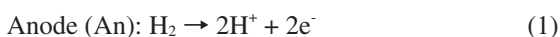


Figure 1 shows a schematic image of a fuel cell. The fuel cell structure consists of a membrane electrode assembly (MEA) with catalyst layers and gas diffusion layers (GDL)

positioned around a polymer electrolyte membrane (PEM), and separators that form gas and coolant channels.

1.2. Importance of Water Management in a Fuel Cell

To obtain high power output during electric generation by a fuel cell, the MEA needs to be moistened with an appropriate quantity of water to help ensure proton conductivity. However, excess water accumulates in the GDL and gas channels, which impedes gas supply, lowers electric generation performance, and results in unstable voltage behavior. In addition, impedance of the gas supply increases the electric potential, which can cause corrosion of

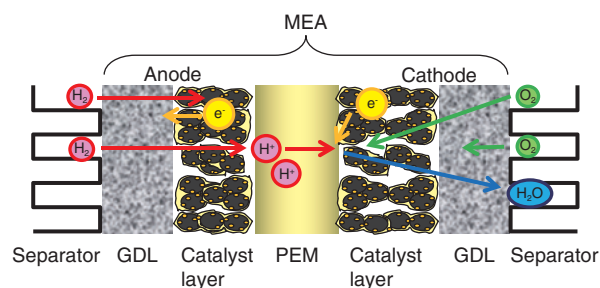


Fig. 1 Schematic image of fuel cell

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the carbon and platinum, and lower durability performance. Therefore, water management to maintain an appropriate MEA water content and to discharge the water from the gas channels is important for the electric generation of fuel cells. Recently, various methods are being investigated to visualize accumulated water due to the importance of this water management. For example, there are numerous reports of methods such as making an acrylic window and directly observing the inside of a fuel cell, or using X-ray radiography measurement and magnetic resonance imaging (MRI) to observe the water⁽¹⁾⁻⁽⁴⁾.

Table 1 shows the measurement speed, spatial resolution and observation range of various visualization technologies. Direct observation via an acrylic window is advantageous for continuously understanding changes in the accumulated water, but this method is unable to observe the water inside the MEA. The other methods can understand the water in the MEA, but neutron radiography and X-ray radiography have an advantage over MRI in terms of measurement speed, and can easily understand time-base changes in the water.

Figure 2 shows the mass attenuation coefficients of various atoms when irradiated by neutron rays and X-rays⁽⁵⁾. The mass attenuation coefficients of water and the atoms that constitute water are higher than those of Al, Fe and other fuel cell component materials, which is effective for observing small quantities of water inside a fuel cell. X-rays have the characteristic that the mass attenuation coefficient increases gradually as the atomic number increases, which makes it a challenge to isolate small quantities of water. On the other hand, neutron rays exhibit higher mass attenuation characteristics for elements with smaller atomic numbers such as hydrogen, and the mass attenuation coefficient of water in particular is an order of magnitude or more higher than that of Fe and Al. This characteristic means that small quantities of water can be observed more accurately using neutron radiography measurement than with X-rays. This study used neutron radiography as the water measurement technology in consideration of the overall balance between the water detection accuracy, measurement speed, and observation range. Neutron radiography measurement

Table 1 Comparison of visualization methods

Method	Time resolution	Spatial resolution	Observation region
Neutron radiography	5-10 sec	10-200 μm	Below 300 mm
X-ray radiography	2-10 sec	1-10 μm	As required
MRI	5 minutes	10 μm	100 mm
Direct visualization	—	—	As required

technology that was formerly limited to small cells was applied to a cell of the size mounted in the Honda FCX Clarity, and the accumulated water state was observed in an actual cell size.

1.3. Research Goals

The goal of this study was to use neutron radiography to clarify the relationship between accumulated water and voltage behavior by checking the state of water accumulated in a cell of the size mounted in the Honda FCX Clarity, and apply the optimum stack electric generation environment to actual vehicle control.

2. Principle of Water Measurement Using Neutron Radiography

Neutron radiography is a method for irradiating the observation subject with neutron rays, converting the transmitted neutron rays to light with a scintillator, and visualizing this light with a CCD camera or other detector. The attenuation coefficient of the neutron rays differs according to the atoms through which the neutron rays are transmitted, so locations consisting of atoms with large attenuation coefficients appear dark, and locations consisting of atoms with small attenuation coefficients appear bright.

The water is quantified by first extracting an image of the water by dividing an image of the state without water (background image) from the neutron radiography image during electric generation. Next, the water can be quantified by using the neutron attenuation characteristics given in Eq. (3) to derive the thickness t [cm] of the measurement subject⁽¹⁾.

$$I_{\text{out}} = I_{\text{in}} \exp(-\alpha t) \quad (3)$$

Here, I_{in} is the incident neutron intensity, I_{out} is the transmitted neutron intensity, and α indicates the neutron mass attenuation coefficient [cm^2/g] of the measurement subject. This study quantified the liquid water inside the fuel cell by multiplying the thickness t [cm] of the liquid water inside the fuel cell under various electric generation conditions by the area to be quantified [cm^2], and expressing the liquid water as a volume [cm^3]. Note that the neutron radiography images in this report indicate regions without water as yellow, and regions with water as blue.

Water measurement using neutron radiography was performed at the NEUTRA beam line of the Paul Scherrer

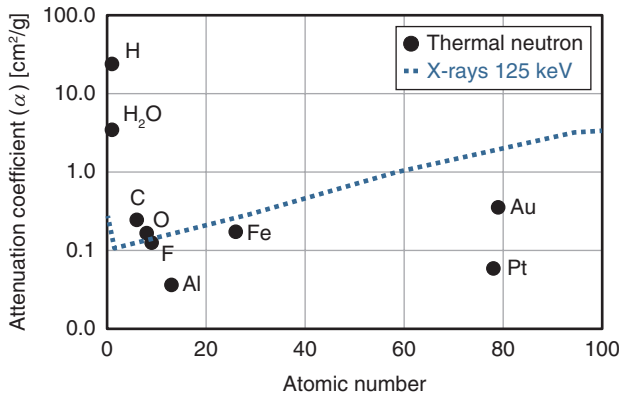


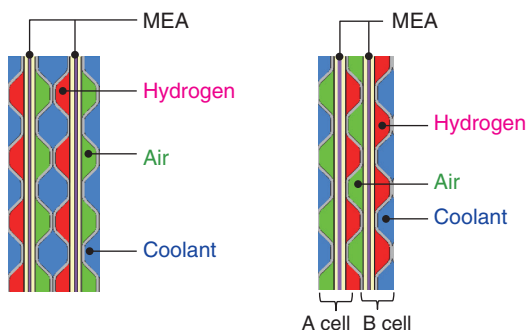
Fig. 2 Attenuation coefficient of elements

Institute (PSI) in Switzerland, in collaboration with the PSI Electrochemistry Laboratory (ECL) and Neutron Imaging and Activation Group (NIAG).

3. Tests

3.1. Directions for Observing Accumulated Water Using Neutron Radiography

Typical fuel cell stacks have a structure in which cooling is performed separately for each cell [Fig. 3(a)]. In contrast, the fuel cell stack mounted in the FCX Clarity uses a structure that performs cooling in two-cell units as shown in Fig. 3(b) in order to reduce size and weight⁽⁶⁾. For this reason, there are two types of cell: A-cells in which the cathode surface is cooled, and B-cells in which the anode surface is cooled. Observation was performed from both the through-plane and in-plane directions to accurately observe the distribution and locations of accumulated water in this fuel cell stack. Figure 4 shows a schematic image of the neutron radiography observation directions. Observation from the through-plane direction enables understanding of the water distribution over the entire electric generation surface, and observation from the in-plane direction enables accurately isolating the positions of the accumulated water in the MEA and each gas channel.



(a) Two cooling layers per two cells (b) One cooling layer per two cells

Fig. 3 Schematic image of two kinds of stacking

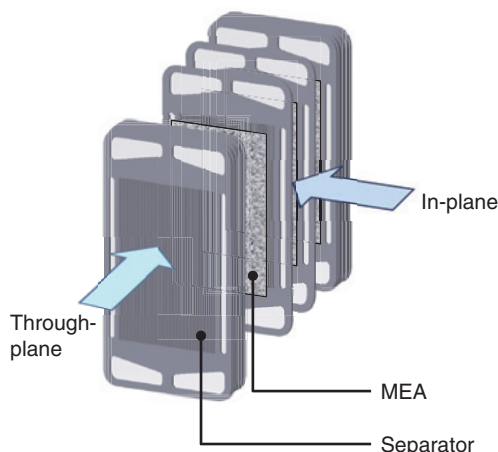


Fig. 4 Schematic image of neutron irradiation direction

3.2. Introduction of Neutron Radiography Cells

3.2.1. Actual cell used for observation in the through-plane direction

Observation in the through-plane direction was performed using an actual cell in order to observe the accumulated water distribution conditions in the cell mounted in the FCX Clarity. Figure 5 shows the cell holder used for observation and the neutron radiography image. The imaging range is wide, so the cell holder is made of materials that take into account both neutron transmittance and the strength to prevent structural warping. This enabled clear observation of accumulated water while maintaining structural integrity. In addition, two-cell observation is possible using a one cooling layer per two cells structure (Fig. 5), and each cell can also be observed separately to check the difference between the A-cell and B-cell.

3.2.2. Cell used for observation in the in-plane direction

The actual cell has a long neutron transmission distance with low transmittance in the in-plane direction, which makes it difficult to measure the water. Therefore, an in-plane observation cell was designed that simulates a one cooling layer per two cells structure (Fig. 6). The MEA

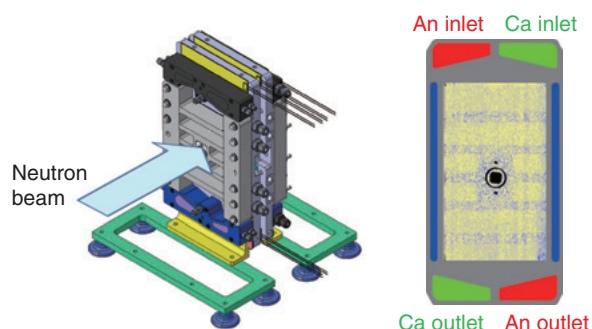


Fig. 5 Schematic and neutron images of through-plane cell

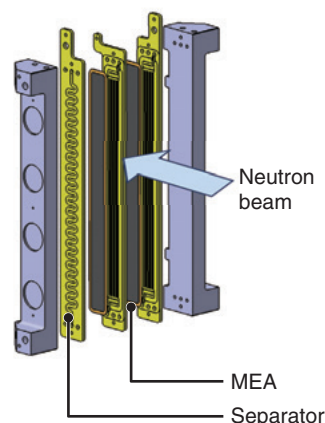


Fig. 6 Schematic image of in-plane cell

seal structure and sealer material of this cell were modified and the number of gas channels was reduced to increase the neutron transmittance in the in-plane direction. All A-cell and B-cell gas channels have the same structure. In addition, the temperature of each separator can be measured using thermocouples.

Figure 7 shows an image of the in-plane observation cell visualized using neutron radiography. The structure simulates the one cooling layer per two cells structure, and both sides of two adjacent cells are cooled by coolant. The red frames in Fig. 7 indicate the anode channels, and the green frames indicate the cathode channels. The region between the anode and cathode channels is the MEA.

3.3. Test Conditions

3.3.1. Neutron measurement conditions

Table 2 shows the neutron radiography measurement conditions. The conditions for the in-plane observation cell were set to increase the resolution in the sideways direction, and the conditions for the actual cell were set to obtain a wide measurement field of view. In addition, a long exposure time was set for the in-plane observation cell to increase the image resolution.

3.3.2. Power generation conditions

The electric generation performed during neutron radiography measurement was low-load electric generation in which the accumulated water has a large effect on the voltage behavior. The standard electric generation conditions in the development stage were set as the base electric generation conditions.

The accumulated water quantity in the actual cell was observed at different cathode stoichiometry values and coolant temperatures as parameters. In addition, tests were also conducted that simulated anode purge, which is performed as control when mounted in a vehicle. A compound test that combined the above conditions was also added as a proving test for application to a vehicle. The electric generation apparatus at the PSI facility was used^{(7), (8)}.

In this paper, the gas flow rate is expressed by the stoichiometry. The stoichiometry is the value calculated

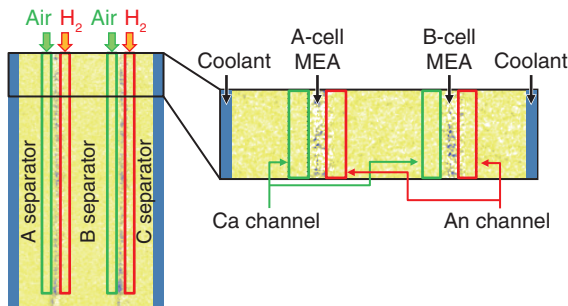


Fig. 7 Neutron radiography image of in-plane cell

Table 2 Neutron imaging condition of each cell

Imaging condition	Through-plane	In-plane
Pixel size (H × W) [μm]	100 × 100	75 × 15
Field of view (H × W) [cm]	25 × 35	25 × 5
Exposure time [sec]	5	10

according to Eq. (4), and is expressed as a relative value, with 1 being the case when the exact quantity of gas required for the fuel cell reaction is supplied.

$$\text{Stoichiometry} = \text{Feed-gas quantity} / \text{Gas quantity required for reaction} \quad (4)$$

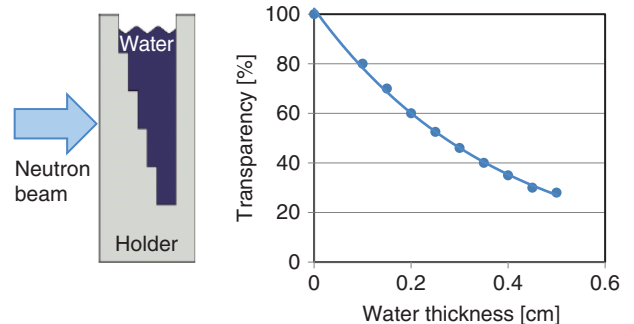
3.3.3. Anode purge

Anode purge refers to periodically opening the anode discharge side valve during electric generation to discharge the nitrogen and other gases not required for electric generation. This operation increases the purity of the hydrogen in the anode gas, and is also thought to be effective from the viewpoint of discharging water. Therefore, a test that simulated anode purge was conducted using the actual cell to check the effects of anode purge on discharging accumulated water.

4. Neutron Radiography Test Results

4.1. Quantitativity Using Neutron Radiography

Different water thicknesses were measured and the neutron transmittance was checked in order to check the quantitativity of water using neutron radiography. Figure 8 shows the measurement results. The transmittance was observed to decrease exponentially as the water thickness increased, which confirmed the validity of Eq. (3). The quantitative values in this study indicate the water using volume [cm³] conversion by first obtaining the water thickness using the quantitative constant obtained through tests, and then multiplying the results by the quantification area.



(a) Schematic representation of water scale (b) Transparency of water filled with different thicknesses

Fig. 8 Validation for water quantification

4.2. Observation of Current Accumulated Water Status and Factor Analysis

4.2.1. Observation of accumulated water during electric generation by an actual cell

Neutron radiography measurement was performed during electric generation using an actual cell to check the accumulated water status in a fuel cell of the size mounted in the FCX Clarity. Figure 9 shows the voltage behavior when electric generation was performed at the standard conditions. The voltage behavior instability was confirmed to be greater for the B-cell than for the A-cell.

Neutron radiography measurement was performed to analyze the difference in the A-cell and B-cell voltage behavior from the viewpoint of accumulated water. Figure 10 shows the images after 30 minutes of electric generation, at which point the accumulated water quantity is constant. More water was observed to have accumulated in the B-cell than in the A-cell. In addition, the results also clearly showed that more accumulated water was present in the lower part of the channels.

Figure 11 shows the change in the accumulated water when electric generation is performed at the standard

conditions. In the initial stages of electric generation, the accumulated water in both the A-cell and B-cell increases at a similar rate. However, after around 10 minutes of electric generation, more water tends to accumulate in the B-cell than in the A-cell, and the accumulated water quantity was observed to stabilize in both cells from around the 30-minute mark. This difference in the accumulated water quantity in the A-cell and the B-cell is due to the differences in the amount of condensation and the water discharge capability. In addition, the time period during which the B-cell exhibited unstable voltage behavior in Fig. 9 is the same as the time during which the quantities of water accumulated in the A-cell and B-cell diverge in Fig. 11. Therefore, the unstable voltage behavior of the B-cell is thought to have occurred because the large quantity of accumulated water in the B-cell impeded the gas supply.

Figure 12 shows the relationship between the standard deviation (SD) at 10-minute intervals of the voltage behavior shown in Fig. 9 and the average value of the accumulated water quantity during the same time period shown in Fig. 11. These results verified that the voltage behavior became unstable as the accumulated water increased.

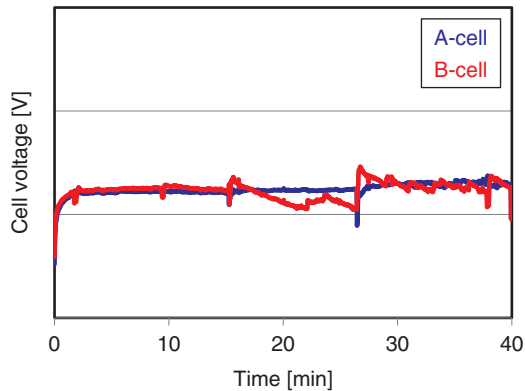


Fig. 9 Cell voltage of each cell under generation

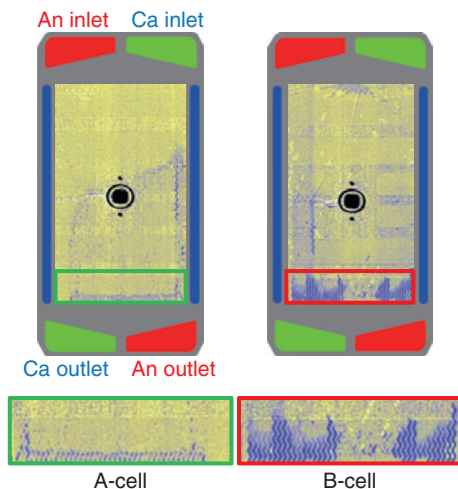


Fig. 10 Neutron radiography image of each cell

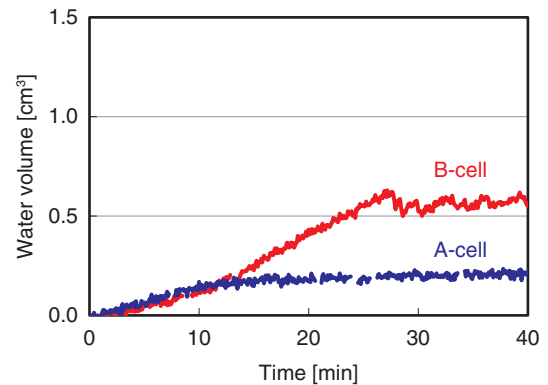


Fig. 11 Water volume transition for each cell under generation

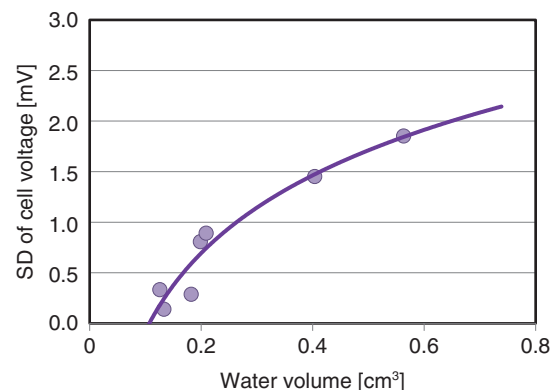


Fig. 12 SD of cell voltage at various water volumes

4.2.2. Results of observation in the in-plane direction of the cell

Figure 13 shows the neutron radiography image when electric generation is continued until the accumulated water quantity becomes constant at the standard conditions. Water was observed to be generated on the cathode side, and also to accumulate on the anode side. This is thought to be due to the effects of back diffusion whereby water moves from the cathode to the anode via the MEA. Figure 14 shows the results of quantifying the accumulated water in each gas channel. This clarified that more accumulated water was present in the B-cell than in the A-cell for both the anode and cathode sides. The separator structure is the same for both the A-cell and the B-cell, so the difference in the accumulated water quantity between the A-cell and the B-cell was determined to be due to the difference in the electric generation environment.

4.2.3. Examination of differences in accumulated water quantities in a one cooling layer per two cells structure

Examination focused on the environment temperature during electric generation in order to investigate the difference in accumulated water quantities between cells in a one cooling layer per two cells structure. Figure 15 shows the temperature of each separator during low-load electric generation. The temperatures of the A and C separators on both sides, which have cooling surfaces, are lower than the temperature of the B separator located in the center. In addition, the C separator

temperature is lower than the A separator temperature. This is thought to be because the C separator is located on the anode side and the A separator is located on the cathode side. That is to say, the C separator is located on the anode side that has lower overvoltage and heat generation, and also has a cooling surface, so this separator is the lowest temperature.

These results confirmed that the B-cell has a lower cell environment temperature than that of the A-cell, and is an environment in which condensation occurs more easily. This is thought to be why more accumulated water is present in the B-cell on both the anode and cathode sides. Accumulated water impedes the supply of gas to the cell, which further reduces water discharge performance, and is thought to result in unevenness in the quantity of gas supplied to each cell.

The cooling surfaces for the anode and the cathode differ in a one cooling layer per two cells structure, which makes it a challenge structurally to maintain a uniform environmental temperature in each cell. Therefore, in order to supply gas evenly to each cell during electric generation, it is necessary to actively discharge the accumulated water and reduce the difference in the quantities of accumulated water. In addition, accumulated water is present not only on the cathode side but also the anode side, which suggests that it is necessary to reduce the accumulated water at both the anode and the cathode.

4.3. Investigation of Application to Control Using the Actual Cell

The results obtained from the actual cell and the in-plane observation cell confirmed that the presence of accumulated water in the gas channels during electric generation and the difference in accumulated water quantities between cells resulted in uneven gas supply, which in turn produced instability in the cell voltage behavior. Therefore, countermeasure methods to reduce the accumulated water quantity were investigated using the actual cell.

4.3.1. Effects of increasing the cathode stoichiometry

The effects of increasing the cathode stoichiometry on reducing the accumulated water were investigated. Tests were conducted using a two-cell structure in

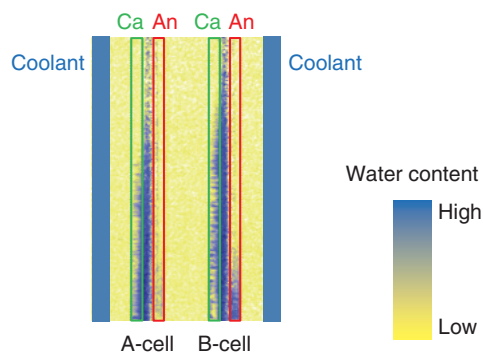


Fig. 13 Neutron radiography image of in-plane cell

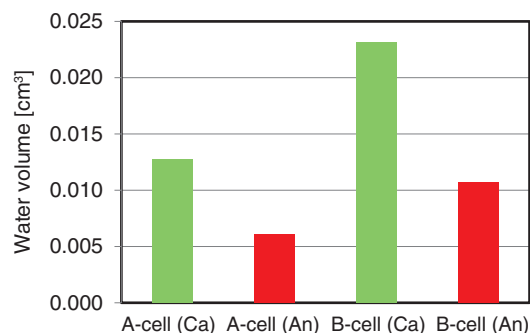


Fig. 14 Water volume of each channel

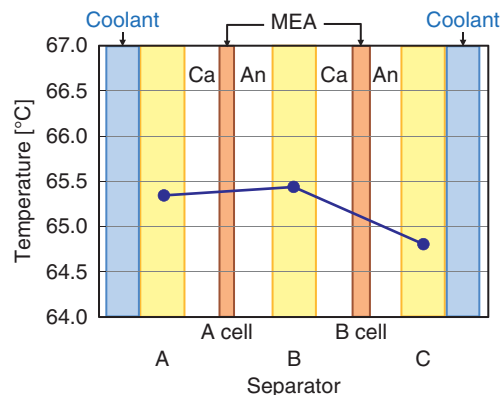


Fig. 15 Separator temperature of each cell

which the A-cell and B-cell perform electric generation simultaneously. Figure 16 shows the relationship between the voltage behavior and the stoichiometry in terms of the standard deviation. It was observed that the voltage behavior tended to stabilize at a stoichiometry of 3.4 or more.

The accumulated water at various stoichiometric quantities was measured to check the relationship between stabilization of the voltage behavior and the accumulated water. Figure 17 shows the neutron radiography image when electric generation is performed at various cathode stoichiometric quantities. The accumulated water in the electric generation area was observed to decrease as the stoichiometry increased.

Figure 18 shows the results of quantifying the accumulated water in the lower part of the gas channels of the electric generation area. This shows that the accumulated water quantity can be reduced by 50% by increasing the stoichiometry from 2.6 to 6.0. This is thought to be because the cathode flow rate increased, which increased the differential pressure and enhanced the water discharge performance.

Figure 19 shows the cell voltage and the resistance overvoltage when electric generation was performed while varying the stoichiometry. An increase in the resistance overvoltage was not observed, but enhanced electric generation performance was confirmed. These results

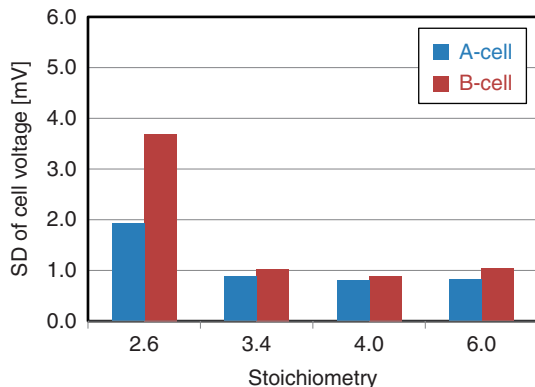


Fig. 16 SD of cell voltage at various stoichiometric quantities

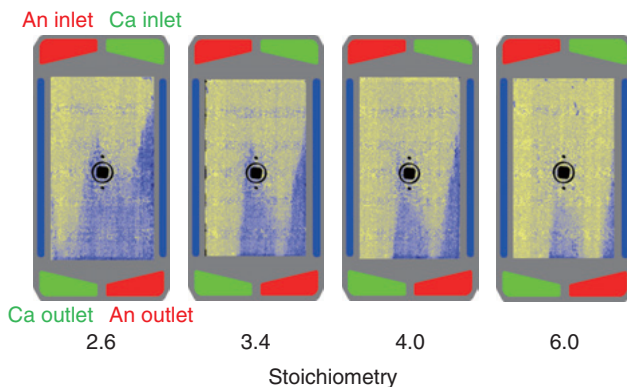


Fig. 17 Neutron radiography image at various stoichiometric quantities

confirmed that increasing the cathode stoichiometry reduces the accumulated water, which stabilizes the voltage behavior and also enhances electric generation performance.

4.3.2. Effects of increasing the coolant temperature

The effects of increasing the coolant temperature on reducing the accumulated water were checked. Figure 20 shows the relationship between the coolant temperature and the voltage behavior as the standard deviation. It was confirmed that the voltage behavior tended to stabilize at a coolant temperature of 72.5°C or more.

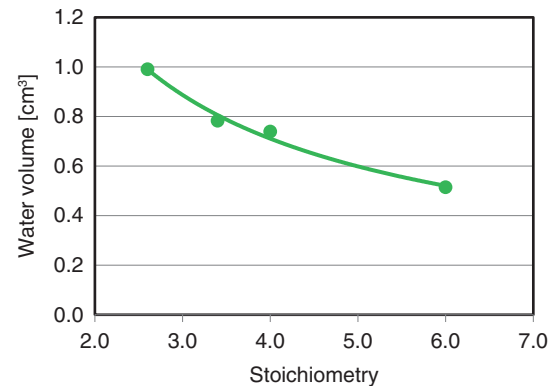


Fig. 18 Water volume at various stoichiometric quantities

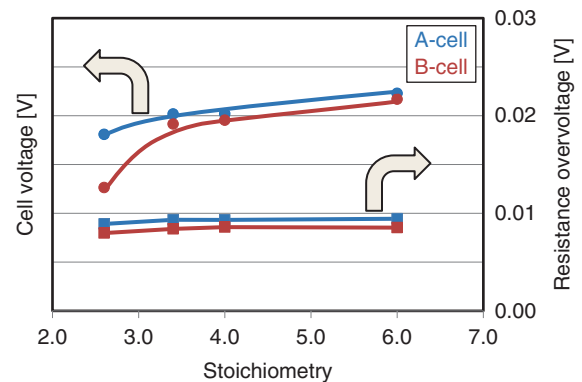


Fig. 19 Cell voltage and resistance overvoltage at various stoichiometric quantities

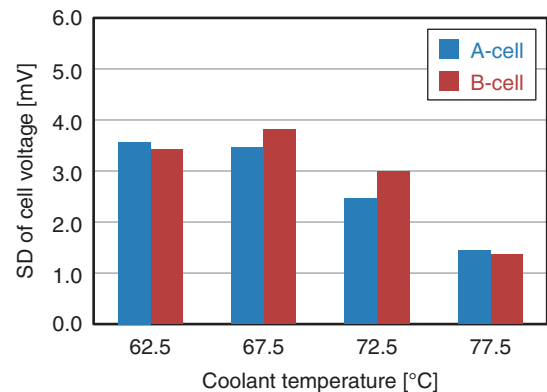


Fig. 20 SD of cell voltage at various temperatures

Figure 21 shows the results of quantifying the accumulated water in the lower part of the gas channels of the electric generation area. Increasing the coolant temperature was confirmed to reduce the accumulated water. This is presumed to be because the cell internal environment temperature rose, so the saturation vapor pressure also rose and the amount of condensation decreased.

Figure 22 shows the relationship between the cell voltage and the resistance overvoltage at various coolant temperatures. Unlike the case when increasing the stoichiometry, increasing the coolant temperature was confirmed to increase the resistance, which reduced the electric generation performance. This is thought to be because while increasing the cell temperature reduced the accumulated water in the gas channels, it also simultaneously reduced the water content of the MEA. This clearly showed that while increasing the coolant temperature effectively reduces the accumulated water, the rise in the resistance overvoltage results in a drop in electric generation performance.

4.3.3. Effects of reducing the accumulated water quantity by anode purge

The effects of anode purge performed in an actual vehicle on the accumulated water quantity were checked. Figure 23 shows the change in the standard deviation of the cell voltage during anode purge control. These results showed

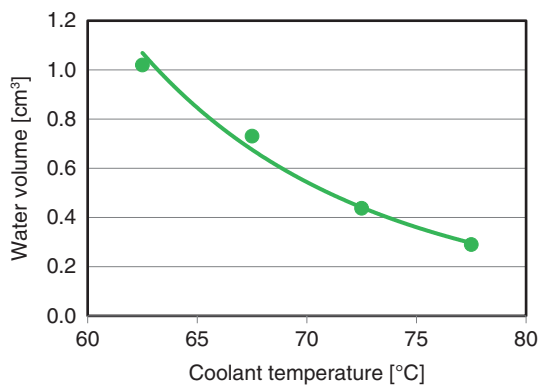


Fig. 21 Water volume at various coolant temperatures

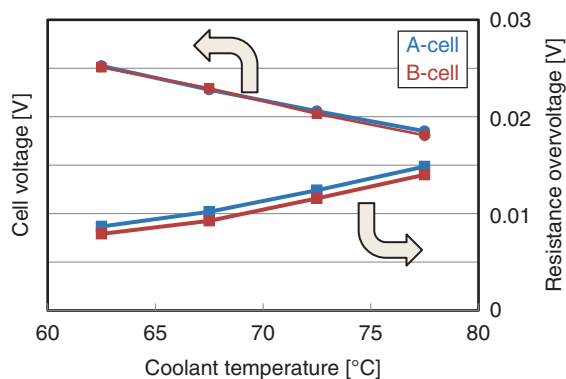


Fig. 22 Cell voltage and resistance overvoltage at various temperatures

that voltage behavior can be enhanced by anode purge.

Figure 24 shows the change in the accumulated water quantity in the lower parts of the A-cell and B-cell gas channels when anode purge control is started. Anode purge was confirmed to reduce the accumulated water in the B-cell to the same level as that in the A-cell.

In addition, Fig. 25 shows the effect of anode purge on the cell voltage. No drop in electric generation performance was observed due to anode purge. Therefore, it is thought that only the accumulated water in the gas channels was discharged, and the MEA water content was maintained.

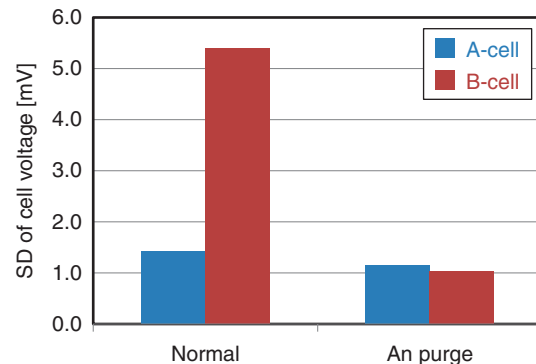


Fig. 23 SD of cell voltage before and after An purge

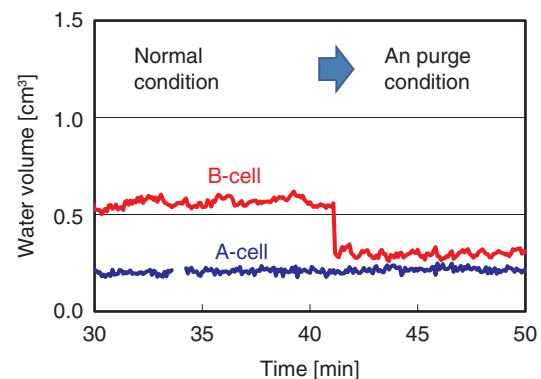


Fig. 24 Transition of water volume before and after An purge

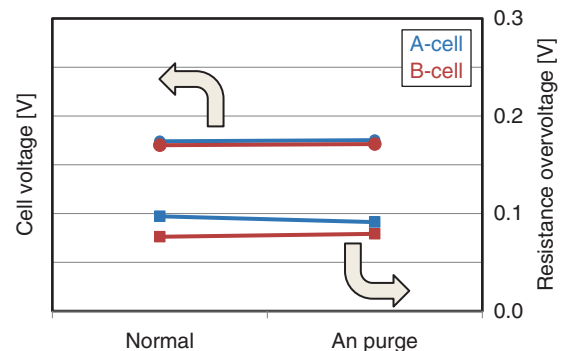


Fig. 25 Cell voltage and resistance overvoltage before and after An purge

These results clarified that anode purge effectively reduces the accumulated water, and can also help stabilize voltage behavior and maintain electric generation performance.

5. Investigation of Methods for Application to an Actual Vehicle System

Application of methods to reduce accumulated water to an actual vehicle system was investigated based on the above results. The results from the actual cell showed that increasing the cathode stoichiometry and performing anode purge are effective methods that can reduce the accumulated water without adversely affecting performance. When applying the above methods as control, the parameters were adjusted within the ranges that do not lower the electric generation efficiency or fuel economy.

Figure 26 shows the neutron radiography images during electric generation before and after the accumulated water countermeasures. It was confirmed that the countermeasures effectively reduce the accumulated water during electric generation.

Figure 27 shows the effects of this countermeasure control in terms of the relationship between the accumulated

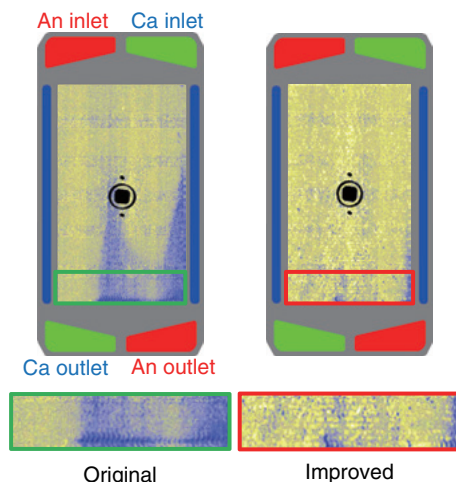


Fig. 26 Neutron radiography image of original and improved generation environment

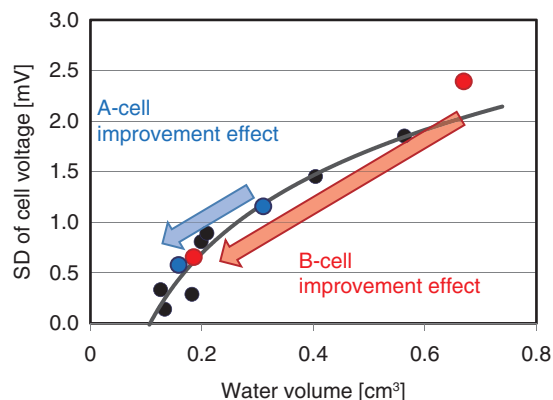


Fig. 27 Relationship of water volume and cell voltage

water and voltage behavior indicated in section 4.2.1. The countermeasure control enabled reduction of the accumulated water quantity in both the A-cell and the B-cell, and reduced the accumulated water quantity in the B-cell to nearly the same level as the A-cell. As the accumulated water quantity decreased, the stability of the voltage behavior also increased.

This countermeasure control controlled the accumulated water quantity and helped to stabilize the voltage behavior, thereby maintaining electric generation performance.

6. Conclusion

The accumulated water was observed by neutron radiography using an actual cell and an in-plane observation cell, the relationship between the accumulated water and the voltage behavior was determined, and methods of reducing the accumulated water were applied to an actual vehicle system. The results are as follows.

- (1) Neutron radiography was used to observe the water behavior during electric generation, and the effects of accumulated water on the cell voltage behavior during electric generation were clarified.
- (2) The water accumulation positions were isolated and analyzed using an in-plane observation cell, and it was proven that the difference in the accumulated water quantity between cells is due to the electric generation environment.
- (3) Methods of reducing accumulated water were investigated while checking the electric generation performance and accumulated water using an actual cell, and increasing the cathode stoichiometry and performing anode purge were found to be effective methods.
- (4) The above methods were applied to the control of an actual vehicle, and enabled stabilizing of voltage behavior while maintaining electric generation performance by simultaneously realizing both an appropriate MEA water content and accumulated water discharge.

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