New design of a PEFC cathode separator of for water management

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Abstract. Generally, polymer electrolyte fuel cells (PEFCs) need humidifiers to prevent the drying of the membrane, but this use of humidifiers creates water management issues, such as the flooding/plugging phenomena and decreased system efficiency because of an increase in the electric energy needed for auxiliary equipment. Although most researchers have developed high-temperature membranes that do not need humidifiers, a lot of time is necessary for the development of these membranes, and these membranes drive up costs. Therefore, we propose a new cathode separator design that can recycle water generated by power generation in the same cell and a stack structure that can redistribute water collected in the cathode outlet manifold to drying cells. Because the new cathode separator has a bypass channel from the gas outlet to the gas inlet to transport excess water, a dry part in the gas inlet is supplied with excess water in the gas outlet through the bypass channel even if the PEFC is operated under dry conditions. Excess water in the PEFC stack can be transported from the cell with excess water to the drying cell through the cathode outlet manifold with a porous wall. Therefore, we confirm the influence of the plugging phenomenon in the cathode gas outlet manifold on the cell performance of each cell in the stack. As a result, the cell performance of the new cathode separator design is better than that of the standard separator under the low humidity conditions. We confirm that the plugging phenomenon in the cathode outlet manifold affects the cell performance of each cell in the stack.

1. Introduction

Recently, polymer electrolyte fuel cells (PEFCs) are beginning to spread to a wide range of fields, such as “ENE-FARM”, as a domestic use co-generation systems, and “Mirai” and “Clarity Fuel Cell” as fuel cell vehicles. However, PEFCs are not spreading rapidly because of the cost. Although PEFCs need a humidification system to avoid membrane drying during operation, the use of humidifiers causes a decrease in the system’s efficiency and the flooding/plugging phenomena. Hakenjos A, Litster S, Dunbar Z W, Jao T C et al have studied these issues on the cathode side using various visualization, such as optical images, MRI, and x-ray [1-4]. K. Takaya et al have elucidated the flooding phenomenon in GDL by a computational simulation [5]. Tanuma T, Eller J, Nishida K, Kitahara T, Kinumoto T, Date T et al proposed water repellency processing to GDL [6-11] as a solution to these issues. However, this method lacks durability and is not effective. Although Rastedt M and Wippermann K et al have also studied membranes for a high-temperature PEFC (HT-PEFC) that operates at a temperature where water is not condensed [12,13], and a lot of time is necessary for the practical use of HT-PEFC. Because the gas inlets of both electrodes are located at the top of the...
cell, the flooding/plugging phenomena occur downstream of the cell as shown in figure 1 (100% RH condition). We understand that the flooding/plugging phenomena in the cathode do not occur alone, and these flooding/plugging phenomena affect the anode channel through the membrane. The plugging water transport phenomenon through the membrane is affected by the location of the cause of the flooding/plugging phenomena in each gas channel [14]. Therefore, we have developed a self-water management separator with a water absorption layer (WAL), which absorbs the plugging water and drains to the water drain to eliminate the plugging phenomenon as shown in figure 2. As a result, although several cathode channels of the cell with a WAL of 14 μm of mean pore diameter (MPD) were plugged by excess water, the plugging phenomenon hardly occurred on the cell with a WAL of 0.9 μm MPD under 100% RH condition as shown in figure 3. Although the cell voltage and the plugging ratio were unstable on the cell with a WAL of 14 μm MPD, that of the 0.9 μm MPD was extremely stable as shown in figure 4 [15]. Although the self-water management separator with a WAL was effective under a high humidity operation, the WAL promoted the drying of a membrane under a low humidity operation because the WAL absorbed too much water in the membrane. A recent trend is a decrease in the amount of humidity to reduce the power required for accessory units. Because there is a large difference in the humidity between the gas inlet and the outlet mentioned above, if the amount of humidity of both electrodes decreases, cell performance deteriorates due to drying of the membrane near the gas inlet. Therefore, we propose a new self-water management separator with a bypass channel as shown in figure 5. Scilicet, this self-water management separator (SWMS), has a bypass channel connecting the supersaturation area and the low humidity area to move excess water by the capillary phenomenon and water concentration diffusion for effective use of the produced water. Because this improvement only adds to the present PEFC composition, the development time is negligible compared to HT-PEFC. The SWMS can be introduced to the present PEFC at once if the effectiveness of the SWMS on cell performance can be confirmed.

![Figure 1. Plugging phenomenon in both gas channel under Tc/Tda/Tdc=80°C/80°C/80°C (100% RH)](image1)

![Figure 2. Schematic diagram of the self-water management separator with WAL](image2)

![Figure 3. Comparison of plugging phenomena: (a) 0.9 μm MPD and (b) 14 μm MPD.](image3)

However, PEFC stacks have specific issues, such as the flow rate distribution to each cell and the temperature distribution in the stack. The supply flow rate to each cell of the stack depends on the fluid resistance of each cell; the temperature at the middle of the stack increases due to an exothermic
reaction (cell reaction). Even if only one cell of the stack is deteriorated, the performance of each cell influences the other cells, and finally the performance of the entire stack decreases. In the PEFC stack, the humidity difference also occurs because of the peculiar issues of a stack and originates in the difference of the stacked cell positions [16]. If it is possible to move excess water from the supersaturation cells to the low humidity cells through the cathode outlet manifold, the system efficiency can be improved by reducing energy consumption. Therefore, we also propose a self-water management PEFC with an absorption layer made of porous materials in the cathode outlet manifold inner wall as also shown in figure 5. After applying the SWMS to the PEFC stack and verifying the effect of the SWMS on cell performance, this paper also confirms the influence of the plugging phenomenon of the cathode outlet manifold on the cell performance of each cell in the stack.

Figure 4. Influence of MPD of WAL on cell voltage variation and plugging ratio.

Figure 5. Schematic diagram of self-water management PEFC stack.

2. Effect of self-water management separator on cell performances

2.1. Experimental apparatus and conditions

Figure 6. Photograph of SWMS-V made of acrylic fiber for visualization.

Figure 7. Photograph of SWMS-P made of carbon for verifying cell performances.

The SWMS had a bypass channel that connected the cathode gas outlet to the gas inlet and was installed in a place different from the gas channel as shown in figures 6 and 7. The SWMS-V made of the acrylic fiber for visualization had a 3-serpentine gas channel; the gas channel was 2.2 mm in width, 1 mm in depth, and 50 mm in length. Another SWMS-P made of carbon for verifying cell performance had a single serpentine gas channel; its geometry was 2.2 mm in width, and 1 mm in depth, and the geometry of the bypass drainage channel was 1.8 mm in width, and 1.2 mm in depth. A
standard separator made of carbon without the bypass channel was prepared to compare with the effect of the SWMS. This standard separator was the same as the SWMS excluding the bypass channel. The gas channel area of all separators was 50 mm × 50 mm, which was the same as the effective electrode area of 25 cm². In the visualization experiment, MEA was placed between the SWMS-A and the acrylic fiber plate, and water transportation in the bypass channel was measured from the front of SWMS-A by the 12-bit CCD camera. Cotton containing red ink was packed into the bypass channel, and the water transport phenomenon in the cell was confirmed by the change in the color of the cotton in the bypass channel when the cathode standard gas (oxidant gas utilization: 50%, cathode humidifier temperature: 80°C) was supplied to the SWMS-A.

In the power generation experiment, SWMS-P was installed in as the cathode separator, and the standard separator was installed in as the anode separator. The standard cell in which the standard separator was installed in both poles was evaluated as the reference data of the effect of the SWMS-P. MEA was the Primer® 5580/5580CARBEL by Gore-Tex®. Figure 8 shows the schematic diagram of the experimental apparatus for evaluating power generation characteristics. Hydrogen and air were supplied to PEFC single cell through each humidifier. The cell resistance was measured by a milliohm meter with AC 4 probe, and I-V performance was measured by a programmable electronic load device. The cell voltage and temperature were recorded by the data logger. The Cole-Cole plot was also measured by an impedance meter to evaluate the effect of the SWMS-P on the cell performance.

![Figure 8. Schematic diagram of experimental apparatus.](image)

There were two experimental conditions: the high humidity condition and the low humidity condition. The standard experimental condition was the high humidity condition called the full humidity condition (100% RH) because the cell temperature (Tc) was the same as 80°C of the anode/cathode humidifier temperatures (Tda/Tdc). The low humidity condition was 42% RH in the cathode channel because the cathode gas was supplied to the cell through the humidifier at 60°C, and this condition was used to verify the effect of SWMS-P on cell performance under low humidity conditions. The fuel gas utilization was 60%, the oxidant gas utilization was 35%, the standard current density was 0.24 Acm⁻², and the experimental pressure was atmospheric pressure.

2.2. Results and discussions

2.2.1. Confirmation of the water transportation using the capillary phenomenon. The effectiveness of the water transport phenomenon was confirmed using the SWMS-A with a visualization technique. Before the experiment, the cotton colored by a red ink was installed in the bypass drainage channel of the SWMS-A. Air humidified by passing the humidifier at 80°C was supplied to the mock cell in which the SWMS-A was installed; the flow rate was the standard flow rate of 334 mL/min that
corresponds to an oxidant gas utilization of 40%. Because the concentration difference of water between the gas inlet and the gas outlet does not occur only by supplying the humidified gas to the gas channel, there is no water transport through the bypass-channel, as shown in figure 9(a). Therefore, to imitate the plugging phenomenon, water was injected near the gas outlet with a syringe. The injected water is absorbed by the cotton packed into the bypass channel by the capillary phenomenon at the same time as water is being injected near the gas outlet, and this absorbed water is transported from the gas outlet to the gas inlet through the bypass channel by the concentration diffusion of water. This is proven because the red ink in the cotton moves from the outlet to the inlet, as shown in figure 9(b) [17]. Because the SWMS-A was able to transport the imitated plugging water from the gas outlet to the inlet through the bypass channel by the capillary phenomenon and the concentration difference of water, the SWMS-P can be expected to eliminate the plugging phenomenon in the actual cell.

![Image of water transport phenomenon in the bypass of the SWMS-A: (a) Before imitated plugging and (b) After imitated plugging.](image)

**Figure 9.** Image of water transport phenomenon in the bypass of the SWMS-A: (a) Before imitated plugging and (b) After imitated plugging.

2.2.2. *Effect of the SWMS-P on cell performance*. Because the effect of the SWMS could be confirmed by the experiment without the power generation, this effect was confirmed by installing the SWMS-P in the actual cell. Figure 10 shows the influence of the SWMS-P and humidity conditions on each polarization as the Cole-Cole plots.

![Influence of SWMS-P and humidity condition on each polarization (Cole-Cole plot).](image)

**Figure 10.** Influence of SWMS-P and humidity condition on each polarization (Cole-Cole plot).
The load current was 6 A, the alternative current was 165 mA, and the measurement frequency varied from 10.000 Hz to 0.04 Hz. From this figure, the activation polarizations of the cells under the low humidity condition are slightly larger than that under the standard conditions despite using the same MEA. Generally, a three-phase-interface forms as the reaction field is formed by catalysts, reactant gas and water, which is a proton conductor. Under the low humidity condition of the cathode side, although the three-phase-interface of the anode side is formed by supplying water in the anode gas, the interface on the cathode side is not easily formed because the water content in the cathode gas is small, as shown in figure 11(a). Therefore, the activation polarization under low humidity condition becomes a little larger. The resistance polarization of the cell with a standard separator becomes larger under the low humidity condition than under the standard condition. Because the humidity difference between the membrane (100%) and the cathode gas (19.7%) is 80.3 %, if the membrane is wet enough by feed water from the anode side, water in the membrane of the cathode side evaporates to the cathode gas, and consequently the resistance polarization becomes large. The resistance polarization of the cell with the SWMS-P is almost equal to that of the standard separator under the standard condition despite the low humidity, and that hardly changes even if the humidity varies. Therefore, because the membrane is wet by transporting excess water at the gas outlet to the gas inlet through the bypass channel, the resistance polarization of the cell with the SWMS-P hardly changes despite of low humidity. Although the diffusion polarization of the cell with the standard separator begins growing under the standard condition, that of the SWMS-P hardly occurs. As the humidities of MEA, the anode gas and the cathode gas are the same under the standard condition, the cathode system will be supersaturated by adding water generated by a cell reaction. Therefore, oxygen cannot reach the platinum of the cathode catalyst, which is submerged by the condensate, as shown in figure 11(b); thereafter, the diffusion polarization of the cell with the standard separator increases as the power generation reaction progresses. However, the diffusion polarization of the cell with the SWMS-P hardly grows for the reason mentioned above. From these results, the effectiveness of the SWMS-P on the cell performance is confirmed under various humidity conditions.

Figure 11. Schematic diagram of the comparison of the three-phase-interface state with a difference in humidity: (a) Under the low humidity condition and (b) Under the standard condition.

3. Confirmation of the influence of the plugging phenomenon in the cathode outlet manifold on cell performance of each cell in the stack

Because the effectiveness of the SWMS for the self-water management was clarified, next the influence of the plugging phenomenon in the cathode outlet manifold on cell performance of each cell in a stack was confirmed using the visualization technique for applying this SWMS to a stack. To imitate the plugging phenomenon, plugging was caused by packing cotton into the cathode outlet manifold near each cell exit of a three-cell stack.

3.1. Experimental apparatus and conditions

The visualization PEFC stack was composed of two standard separators and one visualization separator as shown in table 1. The thickness of both separators was thicker than that of a conventional separator because these separators have a cartridge heater to change the temperature of each separator. The visualization separator has seven observation windows of 0.8 cm²; three windows were installed
in the cell to observe the cathode gas channel, and four windows were installed in the side of the visualization separator to observe the inside of the gas manifold of the cell-1, as shown in figure 12. Because this paper focused on only the cathode outlet manifold, other windows were not observed. The plugging phenomenon inside of the manifold was measured by the 12-bit CCD camera. Compulsory plugging was caused by packing cotton into the cathode outlet manifold near each cell exit of a three-cell stack, as shown in figure 13; pseudo-plugging-1 was between cell-1 and cell-2, pseudo-plugging-2 was between cell-2 and cell-3, and pseudo-plugging-3 was the outlet of the stack.

Table 1. Specification of each separator for the visualization of the PEFC stack.

| Description | (a) Non-visualized separator | (b) Visualized separator |
|-------------|------------------------------|--------------------------|
| Separator   | Gold-plated stainless steel | Gold-plated stainless steel |
| Material of the separator | Gold-plated stainless steel | 0.8 cm² × 3 parts (2.4 cm²) inside |
| Visualized area | - | 0.8 cm² × 4 parts (2.4 cm²) outside |
| Thickness | 20 mm | 30 mm |
| Resistance | 1.815 mΩ | 4.22 mΩ |

Figure 12. Photograph of the assembly of the visualization PEFC stack.

Figure 13. Schematic diagram of the plugging position in the cathode outlet manifold and the position of the observation window.

The flow rate of each gas was controlled by a mass flow controller, and each gas was humidified by passing each humidifier. The temperature of the pipe between the humidifier and the stack was maintained at 80°C by a ribbon heater. The cell voltage was measured and recorded by a data logger. A milliohm meter that utilized four-terminal probes measured the cell resistance.

As the experimental conditions, the temperatures of the cell, the anode humidifier and the cathode humidifier were 80°C, the fuel/oxidant gas utilization was 70%/40%, and the standard current density was 0.3 A/cm².

3.2. Results and discussions

Figure 14 shows the initial I-V performance in pseudo-plugging-1. Although all cell voltages are quite
stable until 0.06 Acm\(^2\), the cell voltage of cell-1 decreases drastically, and all cells thereafter are not able to generate because of the deterioration of cell-1. The reason is that the stack shorted by the short of cell-1 because the backpressure of cell-1 rises and cell-1 is not supplied air by the plugging. Therefore, it becomes clear that the deterioration of only one cell influences the entire stack.

**Figure 14.** Influence of pseudo-plugging-1 on each initial cell performance of three-cell stack.

**Figure 15.** Influence of pseudo-plugging-2 on each initial cell performance of three-cell stack.

**Figure 16.** Lapse of time of each cell voltage and images of the inside of the cathode outlet manifold near the cell-1 outlet at the current density of 0.18 mAcm\(^2\).

**Figure 17.** Influence of the pseudo plugging-3 on each initial cell performance of three cells stack.

In pseudo-plugging-2, the initial I-V performance of each cell is shown in figure 15. The cell voltages of the cells-1 and -2 located in the upstream part of the plugging position decrease drastically when the current density exceeds 0.18 Acm\(^2\) because cotton was packed into the cathode outlet manifold between cell-1 and cell-2 in pseudo-plugging. However, cell-3 located in the downstream part of the plugging position is hardly influenced by the plugging, and the performance of cell-3 is improved because most of the cathode gas is supplied to cell-3 by the plugging. Figure 16 shows the changes with the lapse of time of each cell voltage under 0.18 Acm\(^2\) of current density and the images of the inside of the cathode outlet manifold near the cell-1 outlet. The voltage of cell-1 immediately after beginning the measurement is steady; water does not exist in the cathode outlet manifold. However, the voltage of cell-1 decreases drastically after 25 minutes, and water reaches half of the observation window. Because the cathode outlet manifold between cell-1 and cell-2 was completely
occluded by water, cell-1 was not supplied air. However, air was supplied to cell-2 because cotton was moved to the cell-1 side at an early stage when cotton began to absorb water. Although the voltage of cell-2 begins to decrease a little immediately after beginning measuring, it improves gradually after moving the cotton. Cell-3, which is located in the downstream part of the plugging position, is hardly influenced by the plugging.

Figure 17 shows the initial I-V performance of each cell in pseudo-plugging-3. A big voltage drop such as in pseudo-plugging-1 and -2 is not caused until the high current density region because the buffer volume in the cathode outlet manifold is larger than that of pseudo-plugging-1 and -2. However, cell-1 located in a place that is the furthest from the plugging position decreases drastically compared with other cells in the high current density region. If the plugging was caused by cotton absorbing water, the entire gas channel in the stack becomes a dead water region without the gas in the stack flowing. Therefore, we understand that the plugging phenomenon was caused first in cell-1, which was the most active to react because water generated by a cell reaction remained in each cell. Figure 18 shows the lapse of time of each cell voltage under a current density of 0.18 mA cm$^{-2}$ and the image of the cathode outlet manifold near the cell-1 exit. Each cell voltage is stable until 3000 seconds, although there is a difference in performance. The voltage of cell-3 drastically decreases when the operating time exceeds 3000 seconds, and it recovers 1000 seconds later. The reason is that the water absorbed by the cotton drains outside the stack and is easily compared with pseudo-plugging-1 and -2 because pseudo-plugging is located near the outlet of the stack. However, the cathode outlet manifold becomes an obstruction at once because the condensate flows from each cell into the cotton even if water in the cotton drains temporarily [18]. Therefore, because the plugging phenomenon was caused anywhere in the stack, if the plugging phenomenon was caused near the exit of the cathode outlet manifold, it is necessary to avoid the plugging phenomenon in this area.

These results show that plugging in the manifold greatly influences the performance of each cell in the stack. We could suggest that it is also possible to move water between cells in the stack through this manifold if water management in this manifold was done appropriately.

4. Conclusions
The objective of this study was to propose a PEFC with water management functions. The results of this study are summarized as follows:

Figure 18. Lapse of time of each cell voltage and images of the inside of the cathode outlet manifold near the cell-1 outlet at the current density of 0.18 mA cm$^{-2}$. 

• The bypass drainage channel of the SWMS absorbed water by the capillary phenomenon and transported the absorbed water from the gas outlet to the inlet using water concentration diffusion.
• The SWMS contributes to the increase of the three-phase interface by managing water inside the cell and improves cell performance despite low humidity.
• The plugging phenomenon in the cathode outlet manifold affects the cell performance of each cell in the stack.
• We suggest that it could also be possible to move water between cells in the stack through this manifold if the water management in this manifold is done appropriately.

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