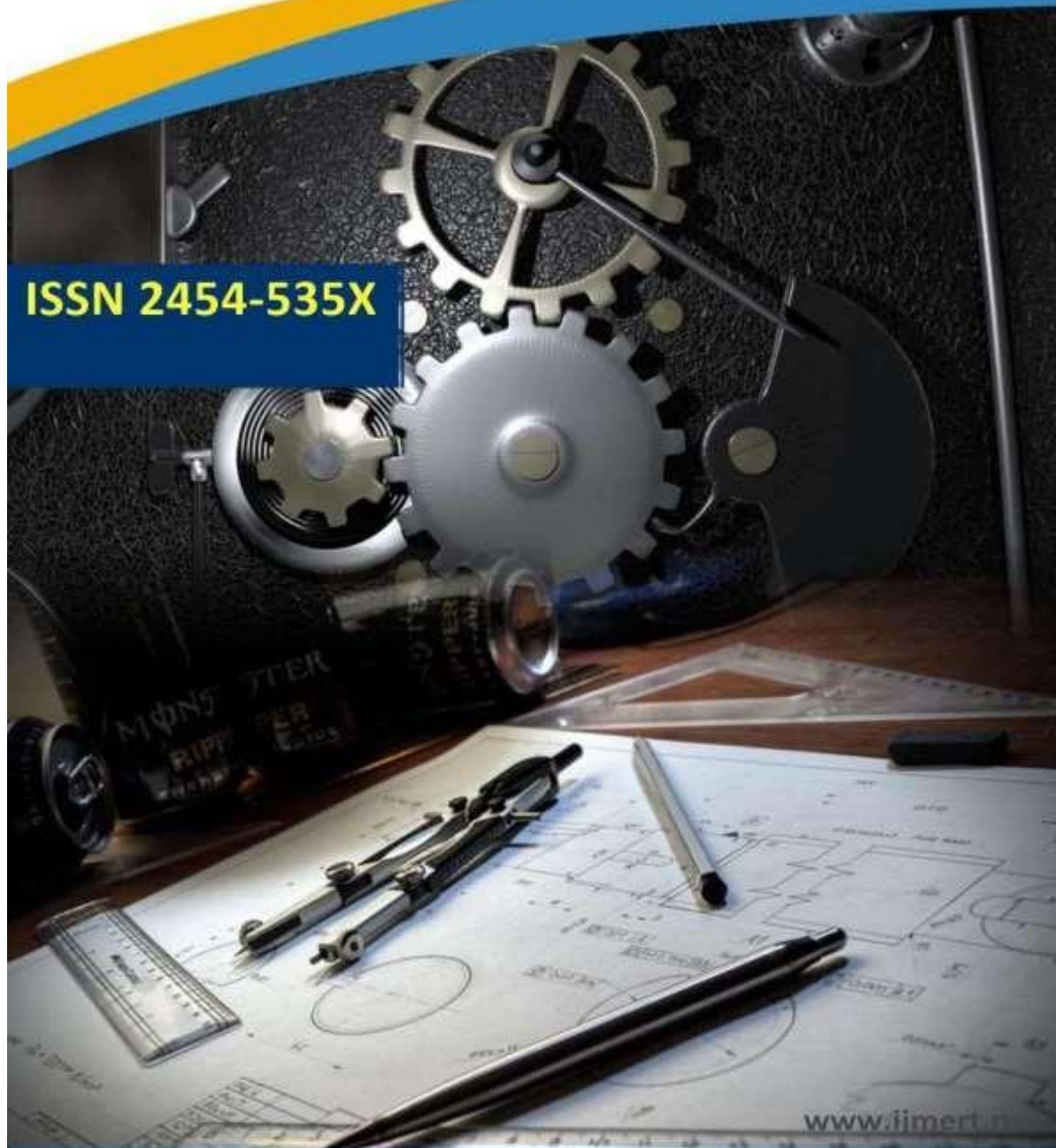




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Creation of a PEFC Self-Management Water Separatory.

Yeshwanth* & K. Suvarna

*

ABSTRACT The flooding phenomena, in which condensate obstructs the pores of the gas-diffusion-layer (GDL), and the plugging phenomenon, in which condensate obstructs the gas channels, are two problems with water management associated with polymer electrolyte fuel cells (PEFCs). The efficiency of the system is increased by lowering the amount of energy needed for humidification if the plugged water can be transferred from the plugged area to the gas intake with low humidity. As a result, since 2006, we have suggested the Self-Management Water Separator (SMWS) as a solution to those problems. Through a bypass channel and a change in water concentration, excess water was absorbed by the capillary action of a water absorption material and transferred from the gas outlet to any point in the gas channel. The earlier research was able to

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 system, “Mirai” and “Clarity Fuel Cell” as fuel cells for vehicles. However, PEFCs are not spreading rapidly because of high cost. Specifically, PEFCs need a humidification system to avoid membranes from drying during operation. However, the use of the humidifier not only depraves the system’s efficiency but also causes performance deterioration by the flooding and plugging of water. Therefore, proper water management in PEFCs is an important parameter to prevent performance losses and improve the system’s efficiency. A lot of researchers have been studying two-phase flow and liquid water transport using various visualization techniques and simulations, and have proposed the water repellency processing to the gas channel and GDL to solve the flooding/plugging phenomena [1-6]. We have also researched the flooding/plugging phenomena of a single PEFC, using the two poles simultaneous imaging technology and that of the PEFC stack using the borescope camera

since 2003 [7]. As a result, a plugging phenomenon was created in the cathode outlet of each separator and the cathode exhaust manifold, and it decreased the cell performance. Since 2017, we have proposed the high performance separator named Self- Management Water Separator (SMWS) [8]. The SMWS has the bypass channel connected from the gas outlet to an arbitrary gas channel. We confirmed the effectiveness of the SMWS by a visualization experiment using an acrylic separator. As a result, excess water was absorbed by the capillary action of the water absorption material, and it moved from the gas outlet to the bypass exit, by a concentration difference of water by which absorption material is packed into the bypass channel of the SMWS. Moreover, in the cell performance test, the voltage of the cell with the bypass channel was more stable than the one without a bypass channel. Additionally, the voltage drop due to the flooding/plugging phenomena is small. Thus, we have verified to improve the water management issues by several methods such as water management of a catalyst layer, a stack and a separator [9]. Furthermore, SMWS can

also be applied to the stack, and excess water can be managed by11

the entire stack by connecting each SWMS and a water absorption layer of an internal manifold in the stack. From these results, we were able to confirm the possibility that the humidifier became unnecessary by effectively using excess water in the

EXPERIMENTAL APPRATUS AND CONDITION

Table 1 shows the specifications of the standard separator and the SMWS. Both separators were made of carbon composite. The standard separator and the SMWS were applied to the anode side and cathode side, respectively. Here, the gas channel area of all separators was 50 mm × 50 mm and the effective electrode area of our single PEFC

® 212 as the electrolyte membrane.

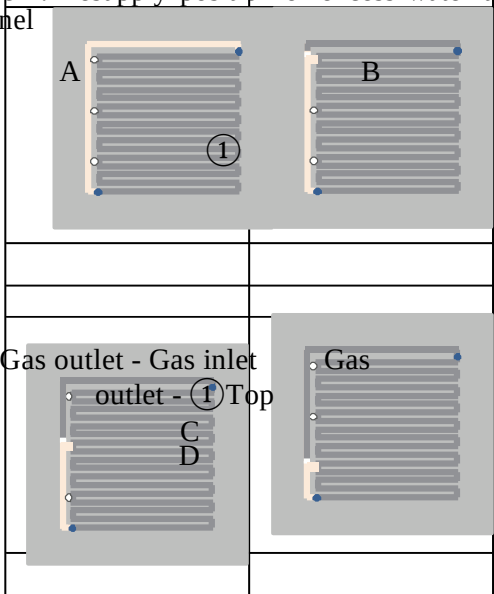
Table 1. Specifications of both Separators

Standard separator and		SMWS made of carbon	
width	50mm	width	50mm
depth	1.2mm	depth	1.2mm
area	2500mm ²	area	2500mm ²
gas channel	1.2mm	gas channel	1.2mm
bypass channel	1.2mm	bypass channel	1.2mm

same cell, and the system’s efficiency and cost were able to be reduced. Therefore, this paper examines the following goals of the practical use of SMWS; (1) Optimization of the position of the bypass exit and confirmation of the effect of the SMWS by a durability test, and (2) the selection of absorption material for the bypass channel [10-13].

was 25cm². The SMWS has a bypass channel that connects from the gas outlet to the inlet, and three exit ports in the place where three aliquots of the gas channel as shown in Table 2. When each port was opened, the other two ports were sealed with a Teflon sheet. All PEFC single cells for this paper used Pt/C as an electrode catalyst, and Nafion

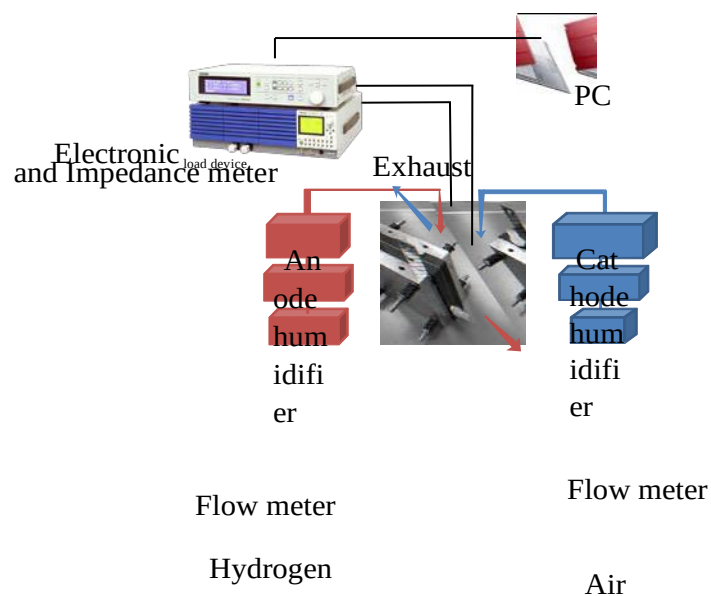
Table 2. Resupply position of excess water through the bypass channel



②③

Gas outlet - ②Middle Gas outlet - ③Bottom

Figure 1 shows the schematic diagram of the cell characteristic evaluation apparatus. Pure hydrogen and air were supplied to the anode and cathode by each mass flow meter, respectively. The cell resistance was measured by a milliohm meter with AC 4 probes. IV characteristics were measured by a programmable electronic load device. The Cole-Cole plot was measured by an impedance meter to evaluate the effect of the SMWS on the cell performance.



PEFC Exhaust

Figure 1. Schematic diagram of experimental apparatus

Table 3 shows the experimental conditions. In the optimization of the bypass exit and the selection of the absorption material, the cell performance was evaluated by IV characteristics and the AC impedance method. Here, the Cole-Cole plot was measured at 0.2 A/cm² and 0.4 A/cm² of the current density, and the fuel/oxidant gas utilization was U_f/U_{ox}=70%/40%. In the optimization of the bypass exit, Gauze was used as the absorption material from knowledge of past studies. In the durability test, the current density was 0.2A/cm², and the fuel/oxidant gas utilization was U_f/U_{ox}=45%/25%. Here, PVA, which had the highest evaluation of all water absorption materials, was used as the absorption material. Additionally, a humidifier was not used in each test. If the cell temperature was not controlled to operate under room temperature, it was greatly changed by each different season. Therefore, the cell temperature was set at 30°C in consideration of room temperature during the summer season.

Table3
Experimental
onditionsOptimization of the
pass exitSelection
ofabsorption material

Durability test

Fuel flow	80ml/min	80ml/min		
Fuel gas utilization	70% @0.3A/cm ²	45% @0.2A/cm ³		
Anode humidifier	N/A			
N/A Relative humidity	(0%RH)			
(0%RH) Air flow	334ml/min			
334ml/min				
Oxide gas utilization	40% @0.3A/cm	25% @0.2A/cm ²		
Cathode humidifier	N/A			
N/A Relative humidity	(0%RH)			
(0%RH) Cell temperature	30°C			
30°C Frequency range	10000~0.04Hz			
10000~0.04Hz Cell pressure				
Atmosphere	Atmosphere			

RESULTS AND

DISCUSSIONS

Optimization of the bypass exit

Figure 2 and 3 show the IV characteristics (IR Free) and the Cole-Cole plots by the AC impedance method, respectively. Here, Gauze was applied to the bypass channels of Pattern-A~D as an absorption material. In Figure 2, the voltage of the cell without the bypass channel is lower than the one with the bypass in a high current density region. As shown in Figure 3(b), the activation polarization, of not using a bypass channel, increases as compared with others. Although the difference of bypass ports hardly affects IV performance, they do have effects on the Cole-Cole plot. From Figure 3 (a), the activation polarization increases in order of Pattern-B, A, D and C, and the resistance polarization of Pattern-D is a little larger than others, respectively. Excess water transported



from near the gas outlet to an upper area of the cell where especially prone to drying in Pattern-B. This phenomenon wets the membrane and decreases the activation polarization, by increasing the three phase interface. Thus, Pattern-B made excess water circulate by making the bypass function in the same cell; consequently, the cell performance was enhanced. The bypass port of Pattern-A is located at the gas inlet, and the length of the bypass is the longest of the other patterns. Therefore, the movement time of water through the bypass is long; though the stored water amount is the largest. Moreover, the three phase interface might increase because a large amount of absorption material absorbs water at the plugging area (gas outlet). The bypass ports of Pattern-C and D are the middle and the bottom of the gas channel area, respectively. Therefore, because the membrane located at the top of these bypass ports dries without water supply, the resistance polarization increases according to the order of the size of these dry regions. Moreover, the cell reaction generally actively occurs from the middle of the cell to the bottom and a lot of steam is generated; there is little concentration difference of water at both ends of the bypass, consequently excess water at the gas outlet was unable to be transported by the concentration difference diffusion, especially in Pattern-C. Therefore, we are considering that the activation polarization of Pattern-C became the largest.

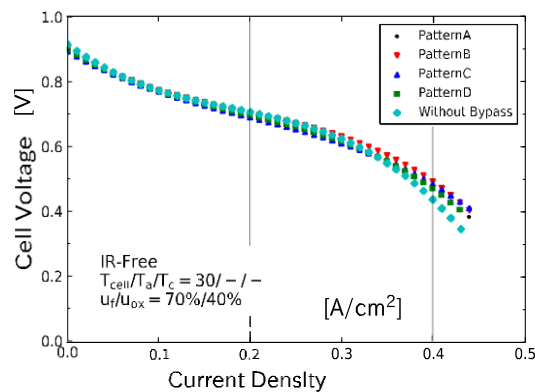
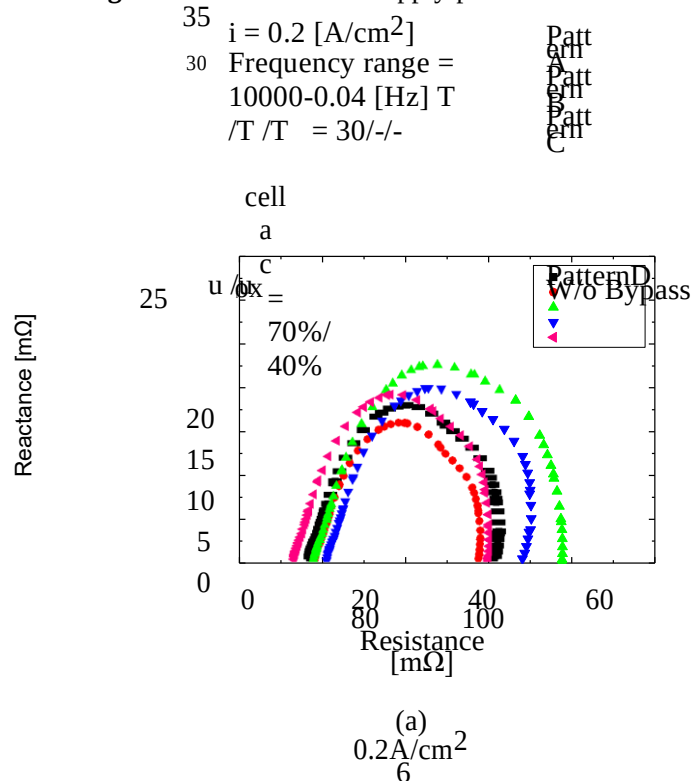


Figure 2. Influence of resupply position on IV characteristics





80 $i = 0.4 \text{ [A/cm}^2\text{]}$
70 Frequency range
= 10000-0.04 [Hz]
 $T_{-}/T_{+}/T_{-} = 30/-/-$
cell a c

Patt
cell
Patt
cell
Patt
cell
PatternD

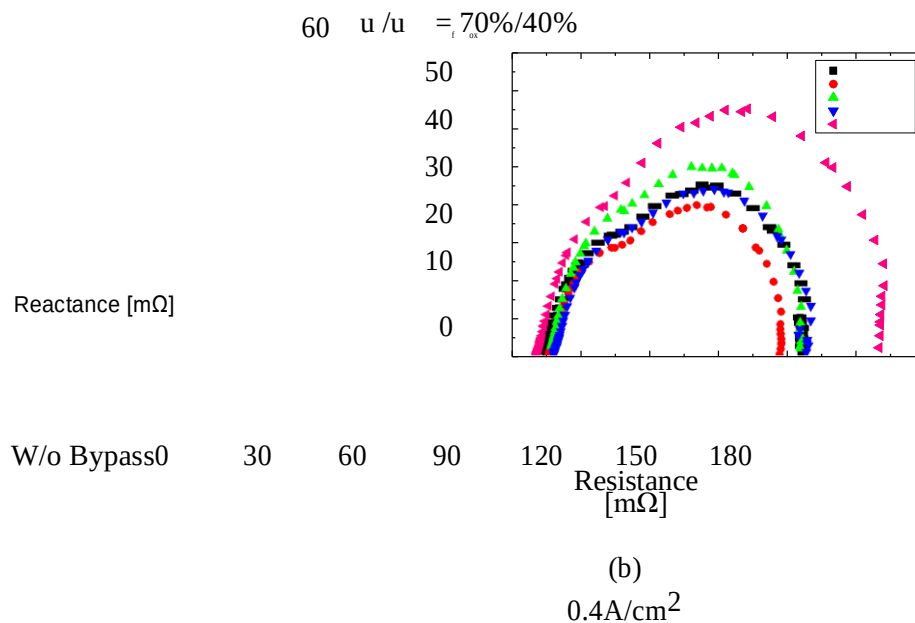


Figure 3. Influence of resupply position on Cole-Cole plot

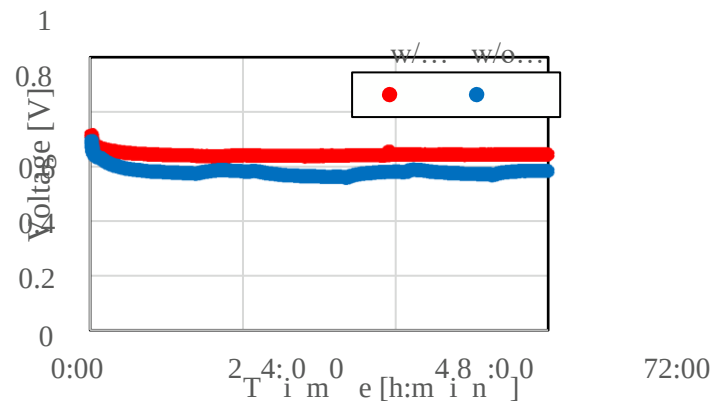
From Figure 3 (b), because the circular arc begins to separate into two, we believe that the anode reaction has dulled compared with the cathode reaction. Water generated, is increased by the amount of increase in the current density; the three phase interface in the cathode electrode increases by supplying water to the ionomer in the catalyst layer and the membrane on the cathode side, using the effect of the bypass. However, because water is hardly transported from the cathode side to the anode side through the membrane, we believe that the anode activation polarization will increase. Because the activation polarizations of the patterns, except for Pattern-B, are almost corresponding and are larger than Pattern-B, Pattern-B is optimal as the bypass exit.

Confirmation of the durability of the bypass effect

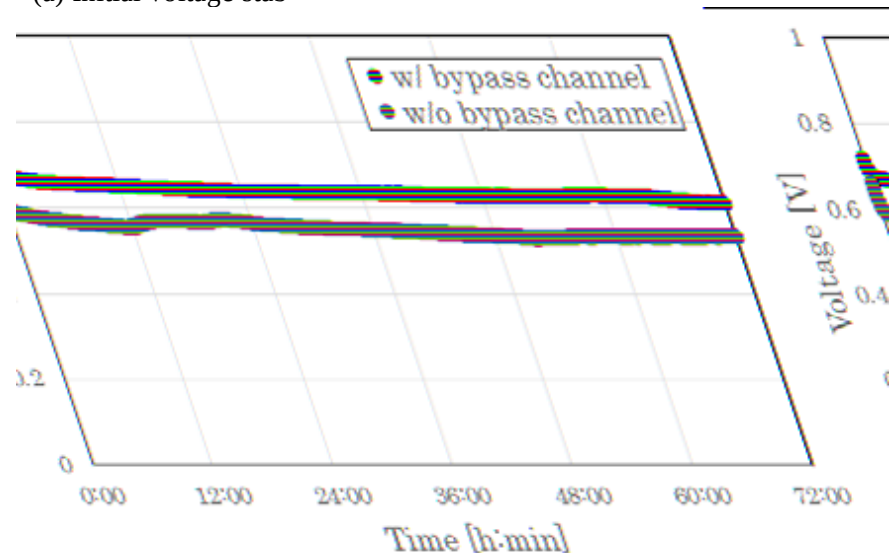
Figure 4 shows the influence of the bypass effect on the voltage stability for 72 hours. Here, the current density was set to a 0.2A/cm² constant, and PVA was utilized as an absorption material packed into the bypass channel of the SMWS. Figure 4(a) shows the initial cell voltage stability, and (b) shows it after 230 hours, respectively. Although the voltage of the cell with the bypass channel is stable and kept at a high voltage on the initial performance, the cell without the bypass channel is unstable. After 230 hours, both cells gradually deteriorated, and the deteriorating rate of the cell with the bypass channel was smaller than the one without the bypass channel. Moreover, the voltage of the cell without the bypass channel is also unstable after 230 hours. Therefore, the IV performance and the Cole-Cole plot were measured every 75 hours to examine the effect of the bypass channel, in more detail. Figure 5 shows the influence of the bypass effect on the IV performance (IR Free) every 75 hours. Although there is little change, with the lapse of time, in the voltage of the cell with the bypass channel, the IV performance of the cell without a bypass channel, varies greatly in time around the middle and high current density region. Although the cell voltage at 150 hours is the highest, we



believe that this swing of the cell voltage in a high current density region was caused due to periodic drainage. Figure 6 shows the influence of the bypass effect on the Cole-Cole plot under $0.4\text{A}/\text{cm}^2$ of current density. The activation polarization of the cell with the bypass channel is smaller than that without the bypass channel. Moreover, because the resistance polarization is almost the same without any relation to the presence of the bypass, the bypass channel influences the activation polarization, especially with the three phase interface in the cathode electrode. The bypass channel functioned at high current density where a lot of water is generated by the active cell reaction, and it removes excess water from the near gas outlet to the top of the cell where moisture is needed. Oppositely, because the standard cell without the bypass channel is not able to control excess water in the cell, the cell performance is influenced by the excess water removal ability of the gas outlet. However, the activation polarization of the cell with the bypass channel increases with the operation time. Because the heat of reaction, by power generation, is accumulated in the cell as the operating hour lapses, water of the three phase interface is evaporated by this accumulation heat; consequently the activation polarization increases. However, we are taking into consideration that the water transport in the cell equilibrates if the operating hour is made longer; thereafter the activation polarization is stable.



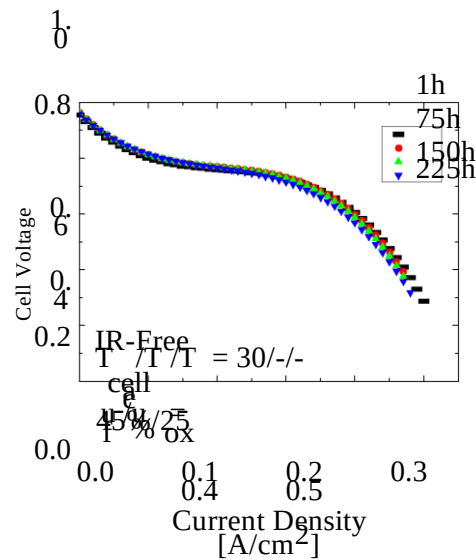
(a) Initial voltage stab



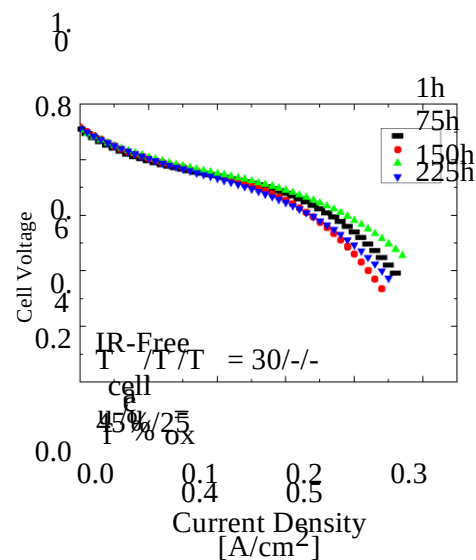
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(b) Voltage stability after 230hours

Figure 4. Influence of bypass effect on voltage stability

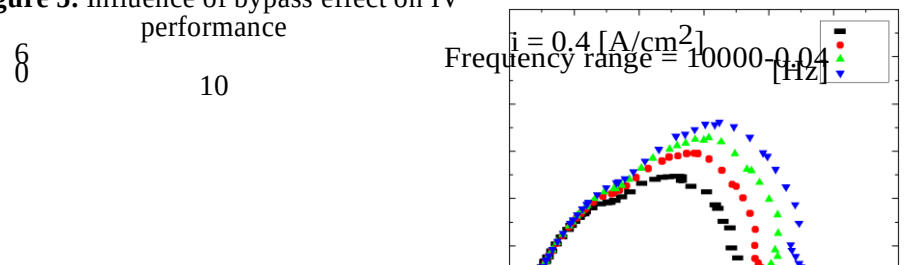


(a) W/ bypass channel



(b) W/o bypass channel

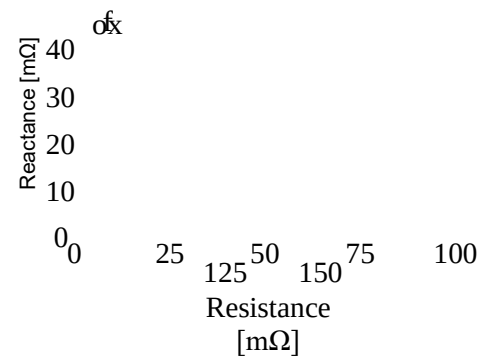
Figure 5. Influence of bypass effect on IV performance



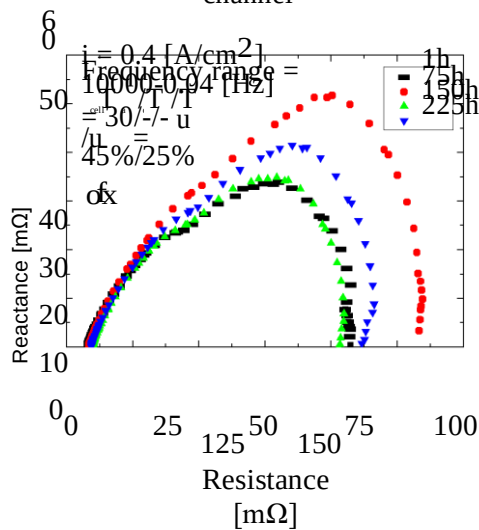


$$50 \frac{T}{u} = \frac{30}{45\%} \frac{T}{u} = \frac{2}{3} \frac{T}{u}$$

1h
75h
150h
225h



(a) W/ bypass channel



(b) W/o bypass channel

Figure 6. Influence of bypass effect on Cole-Cole plot

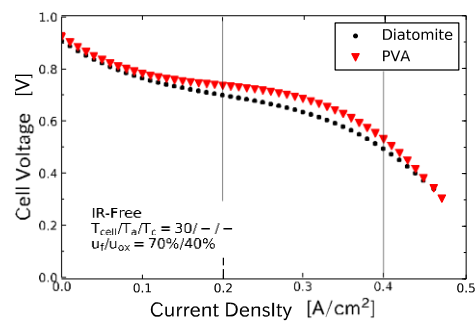


Figure 7. Comparison of water absorption materials on IV characteristics

$i = 0.2 \text{ [A/cm}^2\text{]}$

F
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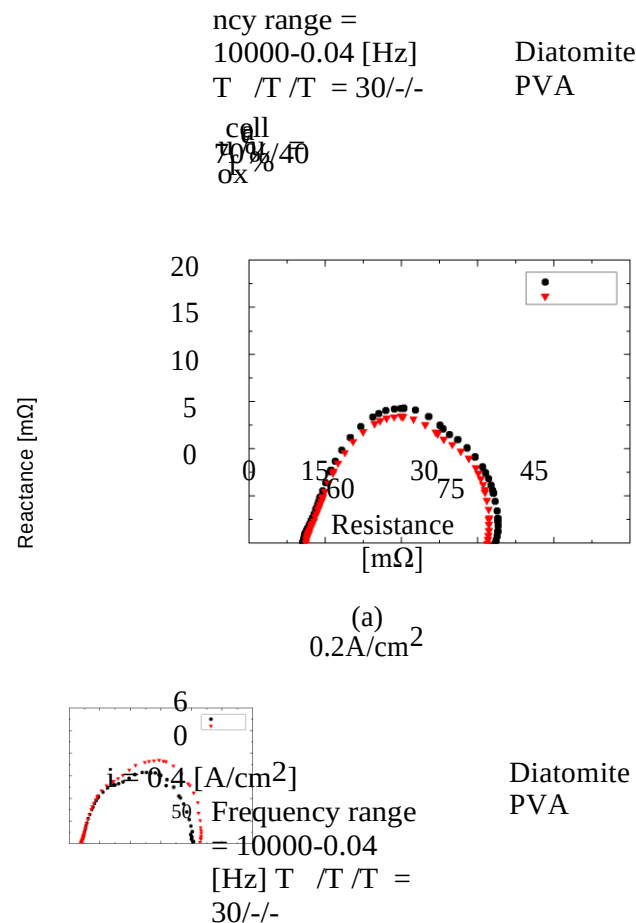


Figure 8. Comparison of water absorption materials on Cole-Cole plot

Selection of absorption material

The previous study proposed that PVA was optimal as an absorption material. However, recently, Diatomite, which has excellent moisture adsorption/emission characteristics, has attracted attention. The price of Diatomite is about 100 times higher than PVA because Diatomite is made to be a reagent. However, if Diatomite was sold for industrial use, the cost would not become a problem. Therefore, the possible application, of Diatomite, as the water absorption material of the bypass channel was verified by an actual cell test. Figures, 7 and 8, show the influence of water absorption material on the IV performance (IR Free) and the Cole-Cole plot under non- humidifier and room temperature conditions, respectively. Here, the Cole-Cole plot was measured at 0.4 A/cm² of the current density. Although the IV performance of PVA is better than that of Diatomite in all current density regions,

this difference decreases with the increase of the current density. Although the activation polarization of PVA is a little smaller than that of Diatomite at 0.2 A/cm², it is reversed at 0.4 A/cm². Moreover, we confirmed by a visualization experiment that the water-absorption-speed/amount of PVA were greater than that of Diatomite. From these results, PVA packed into the bypass channel transports excess water from the gas outlet to the cell upstream part appropriately; thereafter the activation polarization is improved by increasing the three phase interface up to a middle current density region. However, in a high current density region, the voltage of the cell with PVA decreases drastically; this behavior is similar to that of a cell when the gas diffusion polarization enlarges; though the growth of gas diffusion



polarization does not appear in the Cole- Cole plot at 0.4 A/cm^2 . Therefore, we are considering that because the returned water through the bypass channel with PVA is too abundant, the three phase interface decreases. According to a theoretical calculation, the amount of condensate on the cathode side is determined by the differential of saturated vapor pressure at the cell temperature and the water vapor concentration in the cathode exhaust gas; calculated from the vapor quantity generated by the cell reaction and oxygen consumption amount. As a result, at under 30°C of cell temperature and 0.4A/cm^2 of current density, the amount of condensate on the cathode side is about 12.2g/min . On the other hand, the capacity in which GDL could absorb water is about 0.47g if all the pores of GDL would be able to absorb water. Therefore, even if half of the generated condensate was drained from the cell, the possibility that creates the flooding

CONCLUSION

The objective of this study is the development of Self-Management Water Separators for PEFCs in practical use. The results obtained in this study are summarized as follows.

- (1) The optimum bypass exit is located upstream of the cell, under non-humidifying/heating conditions.
- (2) In the durability test, the voltage of the cell with the bypass channel is higher than that without the bypass channel and is stable. Therefore, we have to verify the change of cell voltage with further long term experiments.
- (3) PVA is the best material for water absorption. As our next work, we have to optimize the geometry of abypass channel.

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