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Characteristics of Membrane Humidifiers for Polymer Electrolyte Membrane Fuel Cells

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Abstract−Water management plays an important role in obtaining high performance from a polymer electrolyte membrane fuel cell (PEMFC). To reduce the volume and energy consumption of widely-used bubble humidifiers, membrane humidifiers were fabricated by using an ultrafiltration (UF) membrane and Nafion membranes. The performance of the membrane humidifiers was examined as a function of gas flow rate and operating temperature. A single cell was operated using the UF membrane humidifiers exhibiting almost the same performance with that employing bubble humidifiers.

Key words: PEMFC, Membrane Humidifier, Bubble Humidifier, Ultrafiltration Membrane, Nafion®

INTRODUCTION

The polymer electrolyte membrane fuel cell (PEMFC) has various desirable features as a power source for transportation and portable equipment and as a residential power generation system: high power density, relatively low operating temperature, simple design of the whole system, no electrolyte loss, etc. In the operation of a PEMFC, water management is very important since proton conductivity of the polymer electrolyte membrane, which plays a critical role in determining the cell performance, is proportional to the hydration level of the membrane. However, excessive water in the membrane suppresses diffusion of reactant gases towards catalytic layers and, hence, lowers the cell performance accompanied by concentration polarization [David et al., 1998; Fuller and Newman, 1992; Xie and Okada, 1995]. Therefore, to obtain high-performance for a PEMFC, it is essential to maintain optimal water content in the membrane, which is achieved by feeding humidified reactant gases through a humidifier. Without humidification of the reactant gases, a PEMFC exhibited lower performance by 20-40% than that operating on the humidified reactant gases [Büchi and Srinivasan, 1997].

Humidification methods for PEMFC are classified mainly into two types: external and internal humidification. Most of the external humidifiers, representatively bubble humidifiers, supply water to PEMFC by injecting water vapor into the reactant gases. However, the external humidifiers need additional equipment such as a bubbler and a heater and make the whole system complex as well as increase parasitic energy consumption [Reid, 1998; Nguyen and White, 1993]. To make the system compact by integrating the humidifiers into the PEMFC stack, internal humidifiers have been developed employing membranes [Chow and Wozniczka, 1995; Watanabe et al., 1993], porous bipolar plates [Staschewski, 1996], and hydrophilic channels [Wilson et al., 1998] as water transportation media. Among the humidification methods described above [Büchi and Srinivasan, 1997; Reid, 1998; Nguyen and White, 1993; Chow and Wozniczka, 1995; Watanabe et al., 1993; Staschewski, 1996; Wilson et al., 1998], internal humidification, particularly the membrane-based humidification, is considered to be effective.

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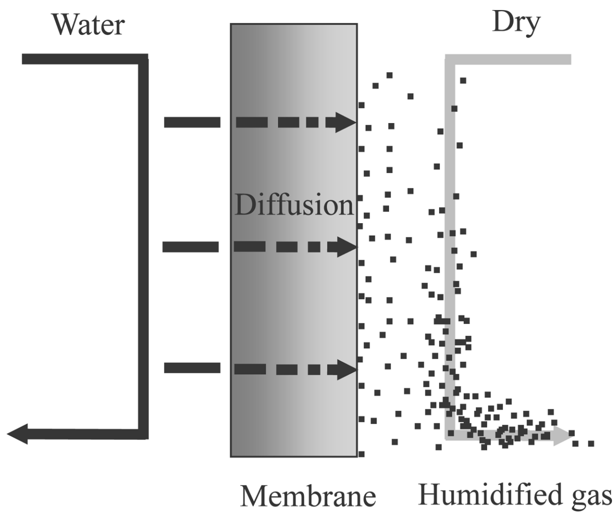

Fig. 1. Principles of membrane-based humidification.

The principle of membrane-based humidification is depicted in Fig. 1. Liquid water and a dry reactant gas are fed to each side of a membrane, respectively. Then, due to the difference in water content across the membrane, water diffuses through the membrane from the water side towards the gas side and evaporates at the membrane/gas interface to humidify the reactant gas [Shin-Hsiung et al., 2001]. As the humidifying water, cooling water of the PEMFC stack is used without additional water supply system and heating equipment required for most external humidification, which reduces the system size and parasitic energy consumption.

In this study, membrane humidifiers were fabricated by using ultrafiltration (UF) membrane and Nafion membranes, and their performance was evaluated as a function of gas flow rate and operating temperature. Then, a single cell was operated on the reactant gases humidified by the UF membrane humidifiers, in comparison with the commonly-used external bubble humidifiers.

EXPERIMENTAL

1. Membrane Humidifier

A membrane humidifier was assembled with a membrane, two flow field plates, two gaskets, and two end plates, as shown in Fig. 2. As the membrane, ultrafiltration (UF) membrane, Nafion® 112, Nafion® 115, and Nafion® 117 were used. Properties of the membranes are summarized in Table 1 [Choi et al., 1998; Sivashinsky et al., 1981]. Flow field plates of the PEMFC were employed as they were. Water and dry gas were supplied through the flow channels on both sides of the membrane, respectively. Fig. 3 shows a schematic experimental apparatus used for characterizing the membrane humidifiers. Water whose temperature was controlled to be the same as the operating temperature was circulated via the humidifier. The active area of the membrane humidifier was 25 cm^2.

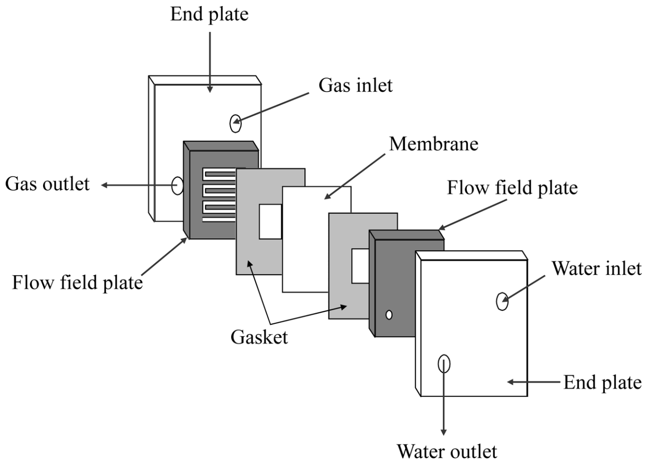

Fig. 2. Structure of membrane humidifiers.

Table 1. Properties of UF membrane and Nafion® 112, 115, and 117 membranes

	UF membrane	Nafion® 112/115/117
Material	Polysulfone	Perfluorosulfonic acid
Thickness (μm)	160	50/125/175
Pore size (Å)	70-80	<12
Tensile strength (kpsi)	2.9-4.4	9.4-36
Cost (US $/$m^2$)	12	616/950/1,065

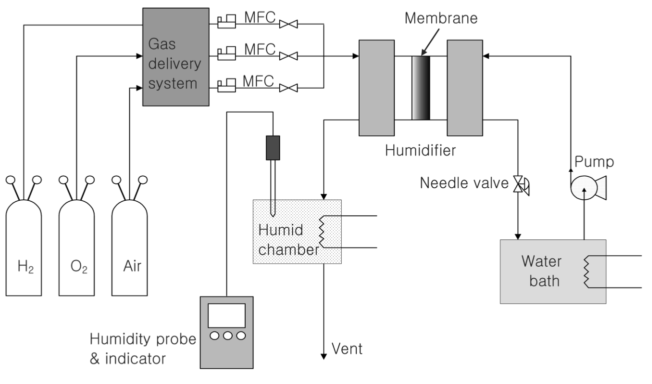

Fig. 3. A schematic diagram of the experimental apparatus used for characterization of the membrane humidifiers.

The performance of the membrane humidifiers was evaluated by measuring the relative humidity (RH) of the reactant gas passing out the humidifiers by using a humidity indicator (Vaisala, HMP 46). Effects of gas flow rate and operating temperature on the humidifier performance were investigated at gas flow rates from 1 to 10 L/min and at operating temperatures from 25 to 75 °C. The pressure of the water and gas was 1 atm.

2. Single Cell

To test the membrane humidifiers in connection with a PEMFC, a single cell was fabricated and operated on the gases humidified through the membrane humidifiers. The ink for the catalytic layers was prepared by mixing Pt/C powders, Nafion solution, and isopropyl alcohol. The prepared ink was screen-printed on the carbon papers to make electrodes. The Pt loading was 0.4 mg/cm^2 for both the anode and cathode. MEA was prepared by placing the prepared electrodes on both sides of a pre-treated Nafion 115 membrane, followed by hot pressing at 140 °C and 200 atm for 90 seconds. The electrode area was 25 cm^2, which was the same as the active membrane area of the membrane humidifiers.

The performance of the single cell employing the membrane humidifiers and the conventional bubble humidifiers was evaluated by measuring the current-voltage characteristics by using an electronic load (Daegil electronics, EL 500P) at cell temperatures of 60 and 75 °C. Oxygen and air were used as the oxidant. The gas stoichiometry was 1.5 for hydrogen and 2 for oxygen and air. All the reactant gases passed through the humidifiers before being fed to the single cell. The humidifier temperature was the same as that of the single cell.

RESULTS AND DISCUSSION

1. Effects of Flow Rate on the Humidification Characteristics

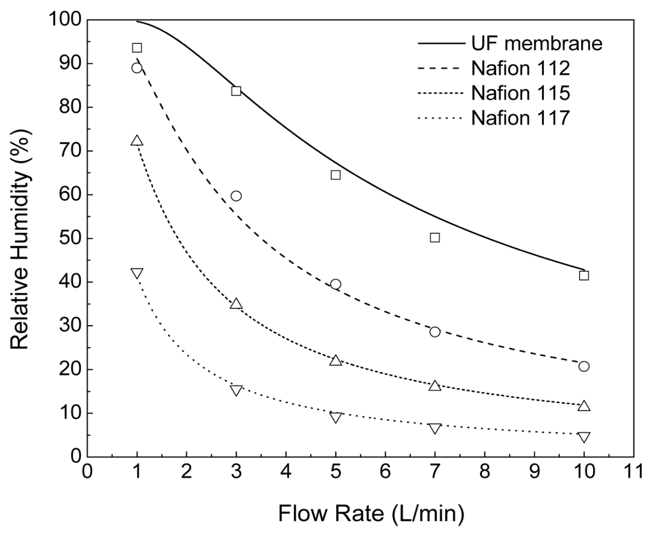

Fig. 4. Effects of flow rate on relative humidity of outlet gas of the membrane humidifiers using UF membrane and Nafion 112, 115, and 117 membranes at a humidifier temperature of 60 °C.

In designing a humidifier for polymer electrolyte membrane fuel cells (PEMFCs), the gas flow rate is one of the most important parameters to consider since humidification characteristics of the humidifier are strongly affected by the gas flow rate that is determined by power of the PEMFCs. To examine the effects of flow rate on the performance of the membrane humidifiers, the relative humidity of the outlet gas passing through the membrane humidifiers employing UF membrane and Nafion membranes was measured at gas flow rates from 1 to 10 L/min. For all the membrane humidifiers, as presented in Fig. 4, the relative humidity of the outlet gas decreased with increasing flow rate, which could be attributed to the limited rate of water transport through the membrane and of water evaporation in the membrane/gas interface. As the gas flow rate increases, the volume of the gas to be humidified by the humidifier also increases. On the other hand, the amount of water transporting and evaporating into the gas is mainly restricted by the membrane active area at a given temperature. Therefore, with increasing gas flow rate and volume of the gas passing the humidifier per unit time, the relative humidity of the outlet gas decreases, since the amount of water vapor included in the gas passing out of the humidifier per unit time remains almost constant. If stagnant in the flow field, the gas would be saturated with water vapor after certain time goes by.

Among the membrane humidifiers used in this study, the UF membrane humidifier exhibited the highest relative humidity of the outlet gas, implying the best humidifying performance probably due to the highest porosity as revealed in Table 1. Compared with Nafion 117 with similar thickness, the UF membrane showed 2-10 times higher performance, depending on the flow rate. At a higher flow rate, the UF membrane was more preferred. For Nafion membranes, the relative humidity decreased with membrane thickness, which could be associated with the reduced gradient of water concentration across the membrane. It can be supposed that the concentration difference of water between both sides of the membrane is the same for all the membrane. Thus, the water gradient across the membrane and, hence, driving force for the water transport would be reduced with increasing the membrane thickness.

From data in Fig. 4, the number of single cells that one unit of the membrane humidifier can afford was calculated under the following assumptions and summarized in Table 2: active area of the membrane humidifiers and single cells is 25 cm^2; current density of the single cells is 1 A/cm^2; stoichiometry of hydrogen and oxidant is 1.5 and 2, respectively; relative humidity of humidifier outlet gases (reactant gases of the single cells) is 80%; and temperature of the membrane humidifier and the single cells is 60 °C. Under these conditions, the flow rate of hydrogen, oxygen, and air is about 0.29, 0.19, and 0.97 L/min, respectively. For the UF membrane humidifier, the relative humidity of the outlet gas is about 80% at a flow rate of 3.2 L/min. Therefore, one UF membrane humidifier can humidify the hydrogen required for feeding a maximum of 10 single cells. If a 30-cell PEMFC stack operates on hydrogen and oxygen, 5 units of the UF membrane humidifier are necessary; 3 units for hydrogen and 2 units for oxygen. In contrast, the 30-cell PEMFC stack needs 50 units of the membrane humidifier using Nafion 117; 30 units for hydrogen and 20 units for oxygen. Those results show that the humidifying characteristics of the membrane play an important role in determining size of the humidifier for a PEMFC stack.

Table 2. Number of PEMFC cells that can be humidified by one unit of the membrane humidifier; active areas of the single cells and the membrane humidifiers are 25 cm^2; current density of the single cells is 1 A/cm^2; stoichiometry of hydrogen and oxidant is 1.5 and 2, respectively; and relative humidity of humidifier outlet gases (reactant gases of the single cells) is 80%; temperature of the membrane humidifier and the single cells is 60 °C

	Anode (H_2)	Cathode (O_2)	Cathode (Air)
UF membrane	10.8	16.2	3.4
Nafion® 112	4.9	7.4	1.6
Nafion® 115	2.5	3.7	0.8
Nafion® 117	1.1	1.6	0.3

2. Effects of Temperature on Humidification Characteristics

Fig. 5 shows the relative humidity of the outlet gas passing through the membrane humidifiers at humidifier temperatures of 25, 40, 60, and 75 °C at flow rate of 1 L/min. With increasing the temperature, the relative humidity of the outlet gas decreased for all the membrane humidifiers but at different rates. For the UF membrane humidifier, the relative humidity of the outlet gas decreased from 96 to 82%, whereas from 91 to 29% for the Nafion 117 membrane humidifier. For membrane humidifiers, except Nafion 117 membrane humidifiers, the relative humidity of the outlet gas markedly decreased at 75 °C. Those results are associated with the rapid increase in saturated water vapor pressure with temperature, as shown in Fig. 6. Compared with the saturated water vapor pressure, the water vapor pressure of the outlet gas increased gradually, implying that the diffusion rate of water through the membrane did not increase with temperature as rapidly as the saturated water vapor pressure. Figs. 5 and 6 also show that the UF membrane humidifier exhibited the best humidifying performance, and with increasing membrane thickness, the humidifying performance of the Nafion mem-

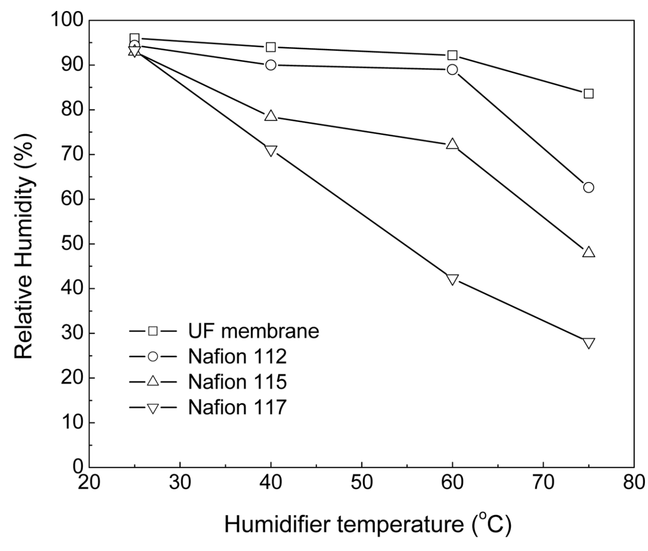

Fig. 5. Effects of humidifier temperature on relative humidity of outlet gas of the membrane humidifiers using UF membrane and Nafion 112, 115, and 117 membranes at a flow rate of 1 L/min.

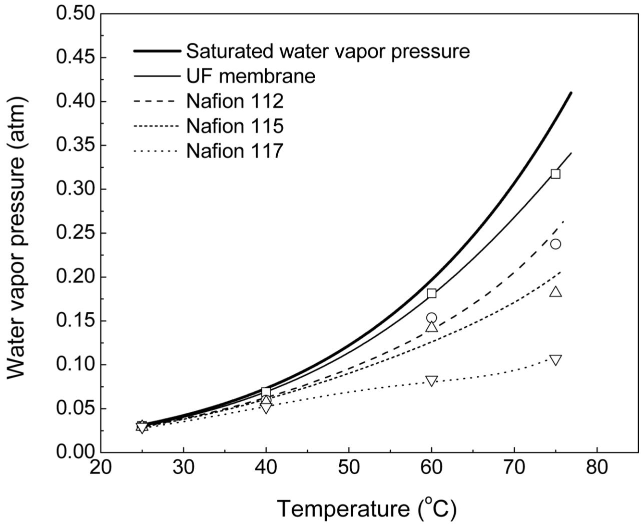

Fig. 6. Effect of temperature on saturated water vapor pressure and experimentally measured water vapor pressure of the outlet gas of the membrane humidifiers using UF membrane and Nafion 112, 115, and 117 membranes at a flow rate of 1 L/min.

brane was lowered.

3. Cell Performance of Bubble Humidifier and UF Membrane Humidifier

To evaluate the membrane humidifiers connected with a single cell in comparison with the commonly-used bubble humidifier, a single cell was operated on the reactant gases humidified through the UF membrane humidifier and the bubble humidifier. Fig. 7 shows performance of the single cells operating on H_2/O_2 and H_2/air at cell temperatures of 60 and 75 °C. The temperature of the humidifiers was controlled to be the same as the cell temperature. At a cell temperature of 60 °C, the single cell exhibited almost the same performance as the bubble humidifier and the UF membrane humidifier as shown in Fig. 7(a); on H_2 and O_2, the open circuit voltage (OCV) was 1.01 V for both humidifiers, and the current density at a cell voltage of 0.6 V was 624 and 632 mA/cm^2 for bubble and UF membrane humidifier, respectively. At a cell temperature of 75 °C, the single cell showed slightly lower performance with the UF membrane humidifier than with the bubble humidifier; on H_2 and O_2, the open circuit voltage (OCV) was 1.01 V for both humidifiers and the current density at a cell voltage of 0.6 V was 776 and 756 mA/cm^2 for bubble and UF membrane humidifier, respectively. Those results could be associated with the sudden decrease in relative humidity of the outlet gas of the UF membrane at 75 °C, as demonstrated in Fig. 5. In contrast, the single cell operating on H_2/air with bubble humidifier exhibited lower performance than with UF membrane humidifier at high current densities, since the bubble humidifier produced more highly humidified air leading to flooding at the cathode. In the case of H_2/O_2 operation, the flow rate of oxygen and, hence the absolute amount of water supplied to the single cell, is only 1/5th of those of air operation. Therefore, flooding at the cathode less readily occurs under O_2 operation. The cell performance presented in Fig. 7 clearly shows that the humidifying performance of the UF membrane humidifier developed in this study can replace the bubble humidifier without loss in cell performance.

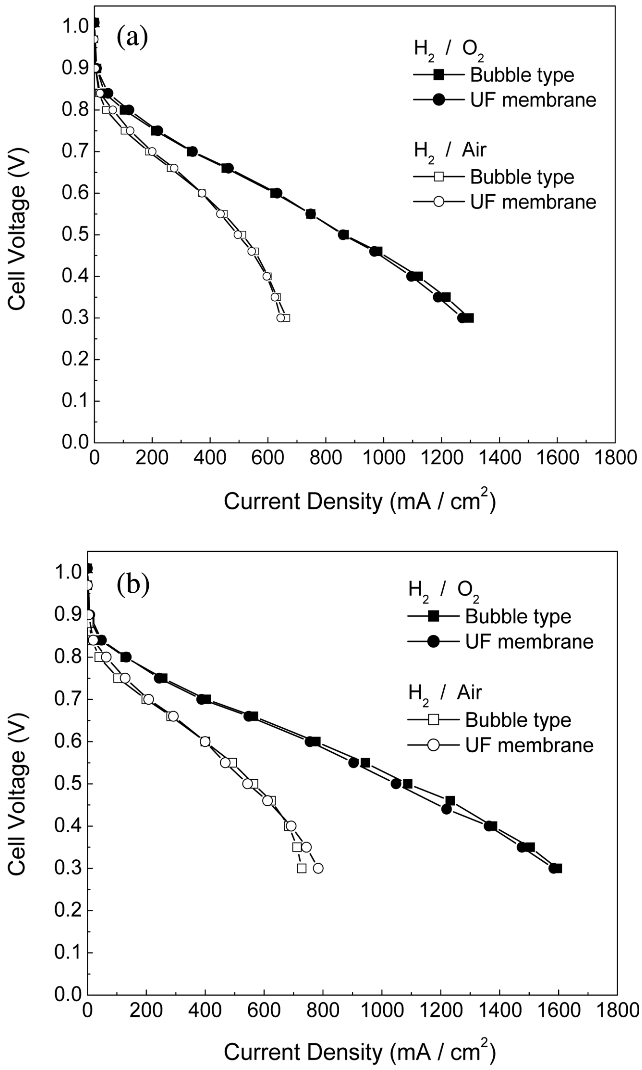

Fig. 7. Performance of the single cell employing the UF membrane humidifiers and bubble humidifiers at cell temperatures of (a) 60 and (b) 75 °C.

CONCLUSIONS

As an alternative humidifier to replace bubble humidifiers requiring additional space and energy consumption, membrane humidifiers for PEMFCs were developed by using an ultrafiltration (UF) membrane and Nafion membranes. Among the membrane humidifiers, the UF membrane humidifier exhibited the best humidifying performance owing to the highest porosity and facilitating water transport. The humidifying performance of the Nafion membranes decreased with increasing membrane thickness. As the gas flow rate increased from 1 to 10 L/min, the relative humidity of the outlet gas decreased due to the limited transfer of water through the membrane. With increasing the humidifier temperature from 25 to 75 °C, the performance of the humidifier was lowered. A single cell was operated on the reactant gases humidified by the UF membrane humidifier and the bubble humidifier and exhibited almost same performance, reflecting that the UF membrane humidifier can be applied to a PEMFC system.

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