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Hydrophilic and hydrophobic double microporous layer coated gas diffusion layer for enhancing performance of polymer electrolyte fuel cells under no-humidification at the cathode

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ABSTRACT

Gas diffusion layers (GDLs) coated with a hydrophobic microporous layer (MPL) have been commonly used to improve the water management properties of polymer electrolyte fuel cells (PEFCs). In the present study, a new hydrophilic and hydrophobic double MPL coated GDL was developed to achieve further enhancement of PEFC performance under no-humidification at the cathode. The hydrophobic MPL, which consists of carbon black and polytetrafluoroethylene (PTFE), was coated on the carbon paper substrate. The hydrophilic layer, which consists of carbon black and polyvinyl alcohol (PVA), was also coated on the hydrophobic MPL. The hydrophilic layer is effective for conserving humidity at the catalyst layer, while the hydrophobic intermediate layer between the hydrophilic layer and the substrate prevents the removal of water in the hydrophilic layer via dry air in the substrate. Both decrease in the hydrophilic layer thickness to 5 μm and appropriate enhancement of hydrophilicity by increasing the PVA content to 5 mass% are effective for enhancing PEFC performance. Reducing the maximum pore diameter of hydrophobic intermediate layer to 20 μm is also effective for enhancing PEFC performance. However, when the pore diameter of the hydrophobic layer becomes too small, concentration overpotential tends to increase, thereby lowering PEFC performance.

1. Introduction

Polymer electrolyte fuel cells (PEFCs) generally have external humidifiers to supply humidified fuel and oxidant gases, preventing dehydration of the membrane electrode assembly (MEA). However, the humidification requirements of the anode and cathode are different. At the anode, it is possible to introduce humidified hydrogen gas using the water generated during the reforming process in fuel production. Furthermore, humidified pure hydrogen gas that is not used for the electrochemical reaction can be recirculated in the PEFC system. It is therefore possible to remove external humidification at the anode. At the cathode, air, from which oxygen is consumed in the electrochemical reaction, is usually exhausted without recirculation. Therefore, the humidified air is generally supplied using external humidification. If a PEFC could be operated without cathode humidification, then external humidification may be removed, resulting in a very simplified overall PEFC system with increased total efficiency and reduced cost [1–3].

The design parameters for the gas diffusion layer (GDL), such as thickness, pore size, and hydrophobic and hydrophilic properties play an important role in determining the water management characteristics during PEFC operations. It is essential to clarify the optimum GDL design parameters when the PEFC is operated without cathode humidification [2,3]. Several investigations have demonstrated that a hydrophobic microporous layer (MPL) coated on the GDL substrate is effective for improvement of the water management characteristics, thereby enhancing PEFC performance [4–8]. The authors have also reported that an optimum MPL coated GDL significantly reduces flooding on the electrode under high humidity conditions and also prevents dehydration of the MEA under low humidity conditions [9]. Both reducing pore diameter and lowering the hydrophobicity of the MPL are effective for the prevention of MEA dehydration, thereby enhancing PEFC performance under low humidification. A conventional MPL contains carbon black and polytetrafluoroethylene (PTFE). Decreasing the content of PTFE in the MPL is effective for lowering its hydrophobicity. However, this has a negative aspect in that the binder force between the MPL and the substrate is reduced.

In the present study, polyvinyl alcohol (PVA) was used instead of PTFE as the MPL binder. This enables hydrophilicity to be imparted without a reduction in the binder force between the MPL and the substrate. A new hydrophilic and hydrophobic double MPL was developed to achieve further enhancement of PEFC performance under no-humidification at the cathode. For the double MPL, it

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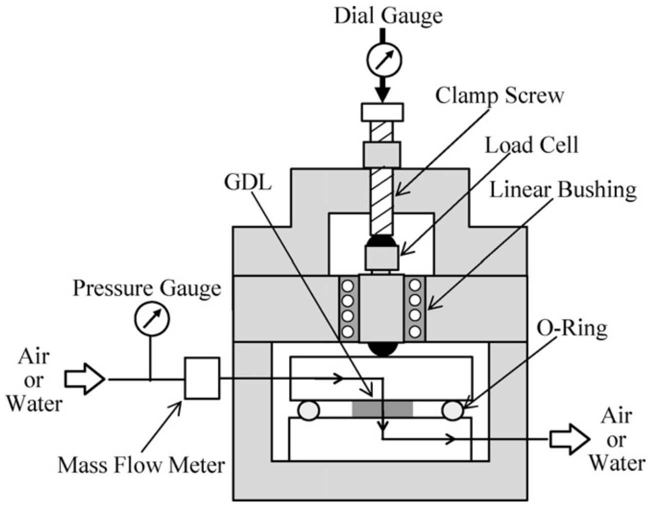


Fig. 1. Schematic diagram of air and water permeability test apparatus.

is expected that a thin hydrophilic layer is effective for conserving membrane humidity, and that a hydrophobic intermediate layer between the hydrophilic layer and the substrate prevents the removal of water in the hydrophilic layer via dry air in the substrate. The influences of thickness, pore diameter, and hydrophilic and hydrophobic properties for the double MPL on PEFC performance were evaluated.

2. Experimental

2.1. Air permeability, pore diameter and contact angle measurements

Fig. 1 shows a schematic diagram of the test apparatus used to evaluate the air and water permeability of the GDL [9]. A 13 mm-diameter GDL was placed between two cylindrical plates. The compression pressure acting on the GDL was set at 1 MPa, which is similar to that measured in a typical PEFC [10]. For the air permeance measurements, the supplied air pressure was set to 1.23 kPa, which is the same as that used in the Gurley method [11]. The permeance q_a was defined as the air flow rate divided by the supplied pressure and permeable cross-sectional area.

The maximum pore diameter of the GDL was measured using through-plane permeability tests according to the ASTM standard test method for pore size characteristics [12]. A low surface tension (=0.0157 N m^{-1}) wetting liquid, Galwick [13], was used to wet the GDL and fill its pores. The contact angle of the wetting liquid is close to 0°. The minimum pressure at which air begins to flow by clearing of the first pore is the maximum pore (bubble point) pressure, which is used to calculate the maximum pore diameter d_{max}.

The contact angle inside the GDL pore was measured using the air and water permeability test results [9]. The water permeability test in the through-plane direction of the GDL was conducted using the same apparatus shown in Fig. 1. The water flow rate Q_w increases with the increase in the supply pressure p_w, as shown in Fig. 2. The minimum pressure at which water begins to flow through the largest pore of the GDL is the maximum pore pressure P_w. For the water permeability tests, the small pores of the hydrophobic MPL are not easily wetted with water. It is not clear whether water can permeate into a mean flow pore of the GDL, which is much smaller than the maximum pore of GDL. Therefore, in the water permeability tests, the relationship between the water flow rate and supply pressure is not stable, which leads to a noticeable error in the mean flow pore pressure obtained from the water permeability test. However, because water must always pass through the maximum pore of the GDL, the maximum pore pressure can be accurately measured. The maximum pore diameter is calculated using Eq. (1):

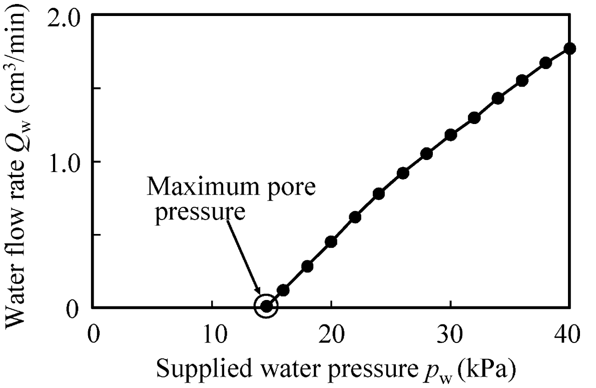


Fig. 2. Typical changes in water flow rate with supply pressure.

$$d_{max} = -\frac{4\gamma \cos\theta}{P_w} \tag{1}$$

where d_{max} is the maximum pore diameter, and γ and θ are the surface tension and contact angle of the water, as shown in Fig. 3. The surface tension of water at 25 °C was set at 0.0720 N m^{-1} [14]. When the maximum pore diameter measured using the water permeability test was assumed to be the same as that measured using the air permeability test, the contact angle inside the GDL pore could be accurately estimated.

2.2. PEFC performance test

PEFC performance tests were carried out as follows. The cell temperature was set at 75 °C. The utilization of hydrogen and air at the anode and cathode was set to 70% and 60%, respectively. The relative humidity of the gas supplied at the cathode was set to 0%, while maintaining a relative humidity of 60% at the anode. The back pressure of the supplied gases was set to zero. The active area of the MEA (PRIMEA® 5580) was 4.2 cm^2. The separator has a triple serpentine flow channel configuration.

IR (ohmic loss), activation and concentration overpotentials were measured separately [15]. The differences in activation overpotentials obtained for all MPL coated GDLs were not significant. Therefore, the influences of the MPL design parameters on IR and concentration overpotentials were discussed with the following test results.

2.3. Gas diffusion layers

The GDL used at the anode was a commercial carbon paper without the MPL (SGL SIGRACET® 24BA), which was treated by 5 mass% PTFE loading to impart hydrophobicity [5]. The 24BA GDL had a thickness of 190 μm, areal weight of 54 g m^{-2}, porosity of 84% and

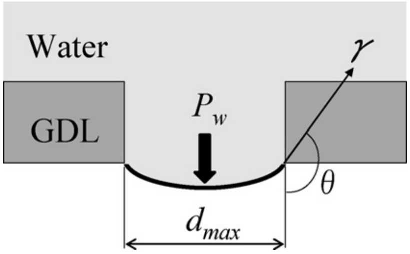
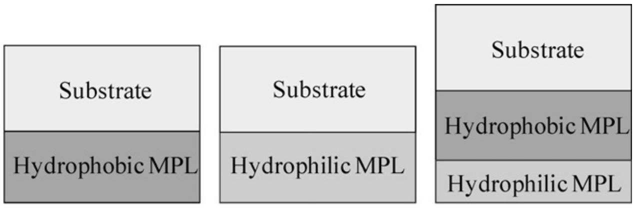


Fig. 3. Equilibrium relationship between surface tension and supply pressure of water.



(a) Hydrophobic MPL (b) Hydrophilic MPL (c) Hydrophilic and hydrophobic MPL

Fig. 4. Hydrophobic MPL, hydrophilic MPL, and hydrophilic and hydrophobic double MPL coated GDLs.

maximum pore diameter of 110 μm. Fig. 4 shows the GDLs used at the cathode; hydrophobic MPL, hydrophilic MPL, and hydrophilic and hydrophobic double MPL coated GDLs. The hydrophobic MPL, which consists of 20 mass% PTFE and 80 mass% carbon black, was coated on the 24BA GDL [9]. The hydrophilic MPL, which consists of 5 mass% PVA and 95 mass% carbon black, was also coated on the 24BA GDL. For the hydrophilic and hydrophobic double MPL, the hydrophilic layer was coated on the hydrophobic MPL coated GDL. The PVA content in the hydrophilic layer was varied between 2 and 10 mass%. The PTFE content in the hydrophobic intermediate layer was varied between 10 and 40 mass%.

Fig. 5 shows the contact angles obtained with the hydrophobic MPL and the hydrophilic MPL coated GDLs. When the PTFE content in the hydrophobic MPL increases from 10 to 40 mass%, the contact angle is extended from 123° to 142°, which enhances the hydrophobic properties of the MPL. The contact angles of the hydrophilic MPLs are less than 60°. When the PVA content in the hydrophilic MPL increases from 2 to 10 mass%, the contact angle is reduced from 56° to 42°, which enhances the hydrophilic properties of the MPL.

Figs. 6 and 7 show SEM micrographs of the surface and cross-section of GDLs with and without MPLs, respectively. For both the hydrophobic and hydrophilic single MPLs, the maximum pore diameter $d_{\max}$ was set at same value of 20 μm. For the hydrophilic and hydrophobic double MPL, $d_{\max}$ of the hydrophobic layer was varied between 1 and 40 μm, and $d_{\max}$ of the hydrophilic layer was varied between 1 and 20 μm. The cross-sectional view of the hydrophobic single MPL coated GDL demonstrates that the MPL is not simply coated on the substrate surface, but penetrates into the porous substrate. The average MPL thickness h_{MPL} considering the penetration into the substrate [9] for both the hydrophobic and hydrophilic single MPLs was set at same value of 80 μm. For the double MPL, the hydrophilic layer was simply coated on the dense hydrophobic MPL surface at thicknesses h_{PVA} varied between 5 and 40 μm.

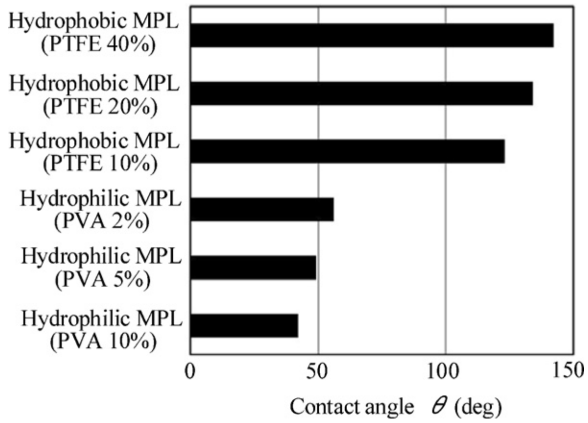
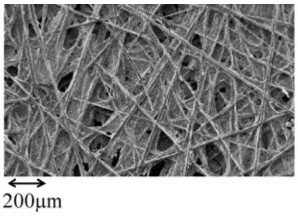
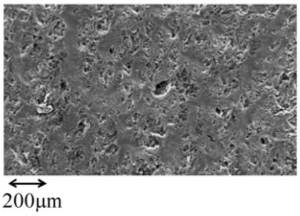


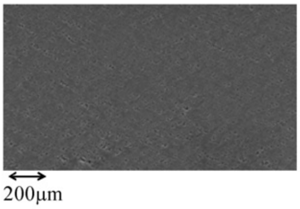
Fig. 5. Contact angles obtained with hydrophobic MPL and hydrophilic MPL.



(a) GDL substrate (24BA, $d_{\max}$=110μm)



(b) Hydrophobic MPL coated GDL($d_{\max}$=40μm)



(c) Hydrophilic and hydrophobic double MPL coated GDL($d_{\max}$=1μm)

Fig. 6. Surface views of GDLs with and without MPL.

Fig. 8 shows the air permeance of the GDLs with and without MPLs. The permeance obtained with all MPL coated GDLs are significantly lower than that with the 24BA GDL without the MPL. The difference in the permeance is not significant between the hydrophilic and hydrophobic single MPL coated GDLs, because the pore diameter and MPL thickness for both GDLs were set to the same values. The permeance of the double MPL is lower than that of the hydrophobic single MPL. The permeance of the double MPL changes depending on the pore diameter and thickness of the hydrophilic layer, and is reduced by both decreasing the maximum pore diameter $d_{\max}$ and increasing the thickness h_{PVA} of the hydrophilic layer.

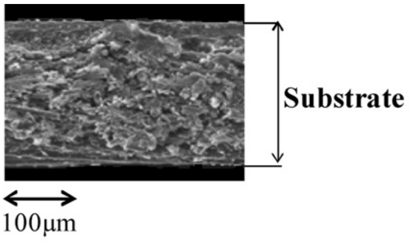
3. Results and discussion

3.1. Influence of the hydrophobic and hydrophilic single MPLs

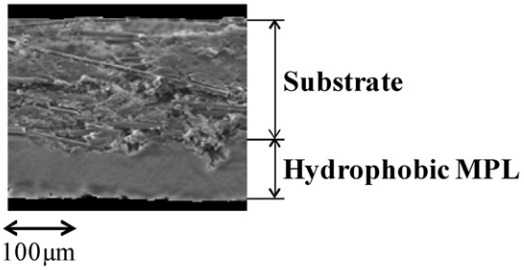
Fig. 9 shows the influence of the cathode GDL on PEFC performance under no-humidification at the cathode, while maintaining a relative humidity of 60% at the anode. The 24BA GDL without the MPL, the hydrophobic MPL and the hydrophilic MPL coated GDLs were used at the cathode. The GDL without the MPL is effective for promoting water transport from the anode gas to the MEA [2,3]. Therefore, the 24BA GDL without the MPL was used at the anode.

For the polarization curve measurements, the current density was increased at a rate of 0.05 mA cm^{-2} per 5 min. The measured polarization curves were stable without any influence of flooding phenomena. The same test was conducted five times to obtain the polarization curve. The averaged polarization curve of the five curves was presented.

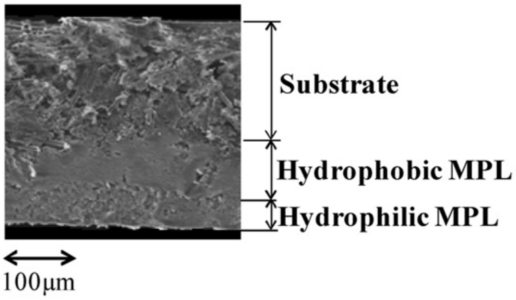
IR and concentration overpotentials obtained with the MPL coated GDLs were lower than those with the 24BA GDL without



(a) GDL substrate (24BA, h=190μm)

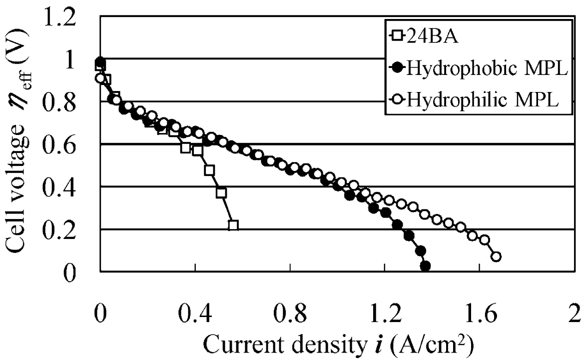


(b) Hydrophobic MPL coated GDL (h=240μm, h_{MPL}=80μm)

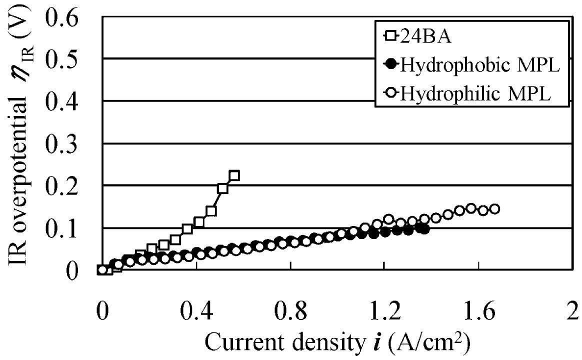


(c) Hydrophilic and hydrophobic double MPL coated GDL (h=280μm, h_{PVA}=40μm)

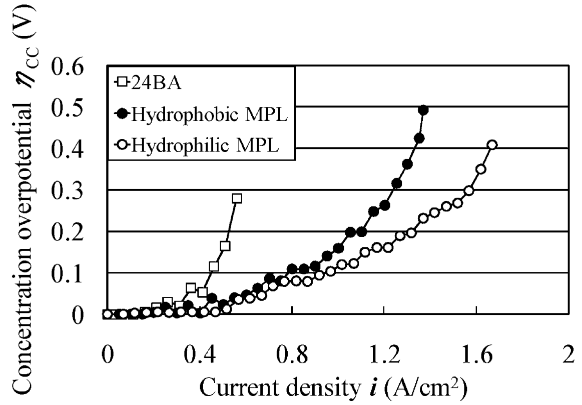
Fig. 7. Cross-sectional views of GDLs with and without MPL.



(a) Output voltage



(b) IR overpotential



(c) Concentration overpotential

Fig. 9. Influence of hydrophobic MPL and hydrophilic MPL on PEFC performance.

the MPL, which indicates that the MPL coated GDL is effective for the enhancement of PEFC performance under no-humidification at the cathode. In the case of the cathode GDL without the MPL, dehydration of the membrane caused by dry air at the cathode cannot be avoided. When the water content in the membrane becomes low, the ionic conductivity is reduced, which results in an increased IR overpotential. Moreover, a decrease in the water content of the cathode catalyst layer reduces the effective reaction area at the triple-phase boundaries, which results in an increased concentration overpotential [9].

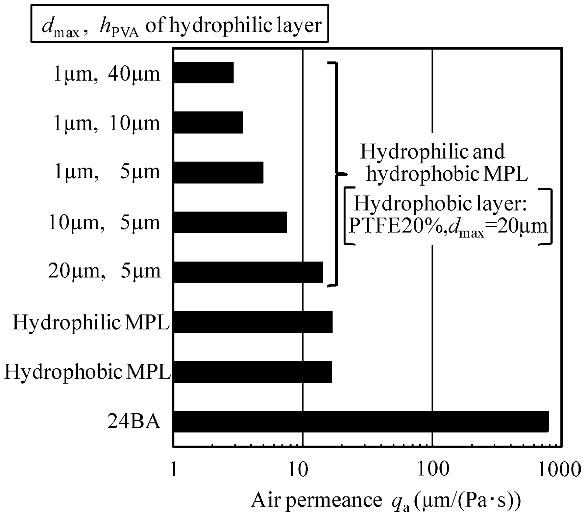
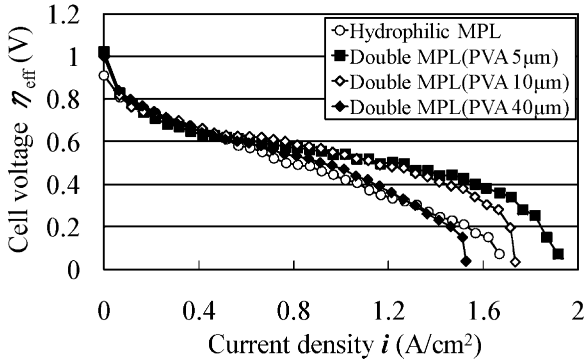
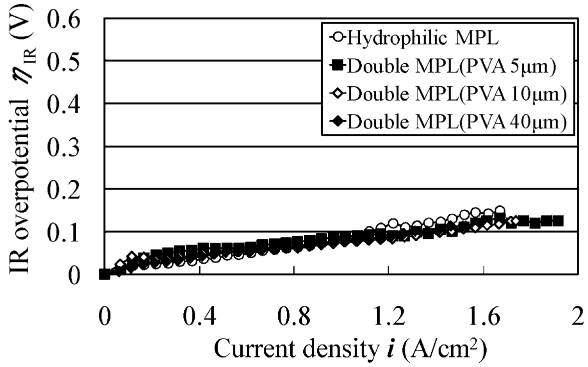


Fig. 8. Air permeance of GDLs with and without MPL.

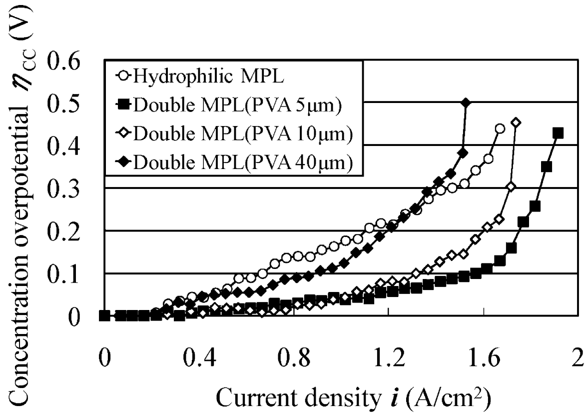
When the dense MPL coated GDLs with low permeability are used at the cathode, it is difficult for the water in the MEA to be expelled to the GDL substrate. This enhances PEFC performance without cathode humidification. The ability of the MPL to prevent dehydration at the catalyst layer varies significantly depending on the hydrophobic and hydrophilic properties of the MPL. PEFC performance obtained with the hydrophilic MPL is higher than that with the hydrophobic MPL. The values of electrical resistivity between the hydrophobic and hydrophilic MPLs are different. However, the difference in IR overpotential obtained for both MPL coated GDLs is not significant, which demonstrates that the influence of the GDL electrical resistivity on IR overpotential is negligible, but the influence of the membrane ionic conductivity on IR overpotential is significant [10]. When the hydrophilic MPL



(a) Output voltage

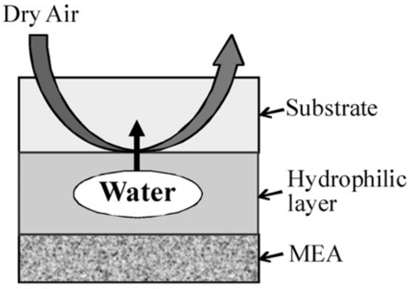


(b) IR overpotential

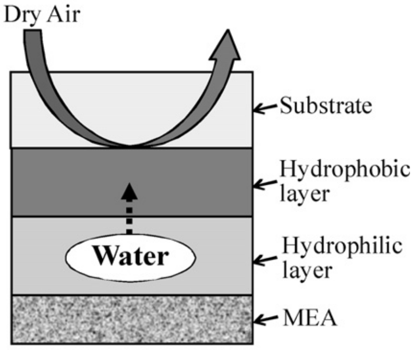


(c) Concentration overpotential

Fig. 10. Influence of hydrophilic layer thickness for hydrophilic and hydrophobic double MPL coated GDL on PEFC performance.



(a) Hydrophilic MPL



(b) Hydrophilic and hydrophobic double MPL

Fig. 11. Enhanced ability of double MPL to prevent drying-up of MEA.

is used, concentration overpotential is reduced, which enhances PEFC performance. Because the pore diameter and the thickness of both the hydrophilic MPL and hydrophobic MPL coated GDLs were set to the same values, the difference in permeability is negligible. When the hydrophilic MPL is used, the ability of the MPL to conserve humidity at the catalyst layer is enhanced, which is effective for enhancing PEFC performance without cathode humidification.

3.2. Influence of the hydrophilic and hydrophobic double MPL

In order to achieve further enhancement of PEFC performance, a new hydrophilic and hydrophobic double MPL coated GDL was developed. Fig. 10 shows the influence of the double MPL on PEFC performance when the thickness h_{PVA} of the hydrophilic layer (d_{max} = 20 μm, PVA 5 mass%) was varied between 5 and 40 μm. For all double MPLs, the hydrophilic layers were coated on the hydrophobic MPL with a PTFE content of 20 mass% and a maximum pore diameter of 20 μm. Later in this paper (see Figs. 14 and 15), the reason for selecting this hydrophobic intermediate layer for the double MPL will be evident. PEFC performance varied significantly with the change in the hydrophilic layer thickness. A hydrophilic layer thickness of 5 μm resulted in a significant enhancement of PEFC performance, compared with the hydrophilic single MPL coated GDL.

Although the ability of the hydrophilic single MPL coated GDL to conserve the humidity at the catalyst layer is enhanced, it is relatively easy for the water in the hydrophilic layer to be expelled to dry air in the substrate, as shown in Fig. 11(a). Because the hydrophilic single MPL is not sufficient to prevent dehydration of the cathode catalyst layer, no significant enhancement in PEFC performance can be expected. On the other hand, for the double MPL coated GDL, the hydrophilic layer is effective for conserving humidity at the catalyst layer, while the dense hydrophobic intermediate layer between the hydrophilic layer and the substrate prevents the removal of water in the hydrophilic layer via dry air in the substrate, as shown in Fig. 11(b). This results in a significant enhancement of PEFC performance.

Fig. 10 also demonstrates that PEFC performance is significantly lowered when the hydrophilic layer thickness is increased from 5 to 40 μm. We have previously reported that increasing the MPL thickness for the hydrophobic single MPL coated GDL is effective for reducing permeability, and thereby enhances PEFC performance without cathode humidification [9]. Increasing the hydrophilic layer thickness for the double MPL also reduces permeability, which seems to be advantageous for maintaining the humidity of the cathode catalyst layer. However, when the hydrophilic layer thickness is increased, accumulated liquid water in the hydrophilic layer reduces oxygen transport to the electrode through the MPL, which increases concentration overpotential and thereby lowers

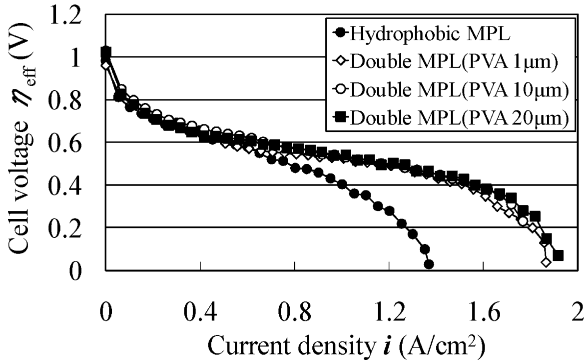


Fig. 12. Influence of maximum pore diameter of hydrophilic layer for double MPL coated GDL on PEFC performance.

PEFC performance. This result clearly indicates that the hydrophilic layer thickness of the double MPL must be as small as possible for the enhancement of PEFC performance.

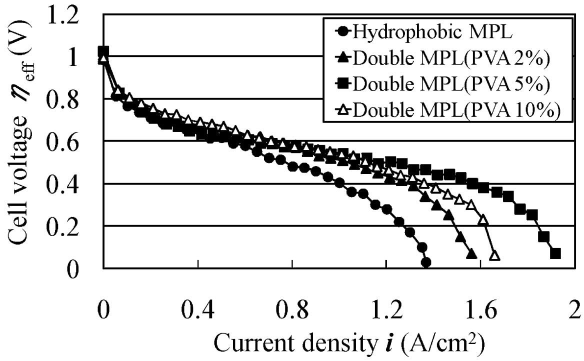
3.3. Influence of pore diameter and PVA content of the hydrophilic layer for the double MPL

Fig. 12 shows the influence of the double MPL on PEFC performance when the maximum pore diameter $d_{\max}$ of the hydrophilic layer ($h_{\mathrm{PVA}} = 5$ μm, PVA 5 mass%) was varied between 1 and 20 μm. We have previously reported that PEFC performance obtained with the hydrophobic single MPL coated GDL varied significantly depending on the MPL pore diameter [9]. Decreasing the MPL pore diameter reduces permeability, which is effective for enhancing the ability of the MPL to prevent dehydration of the MEA caused by dry air. For the double MPL coated GDL, however, the difference in PEFC performance is not significant, even when the maximum pore diameter of the hydrophilic layer is reduced from 20 μm to 1 μm. The dense hydrophobic intermediate layer between the hydrophilic layer and the GDL substrate prevents the removal of water in the hydrophilic layer via dry air in the substrate. As a result, the influence of the hydrophilic layer pore diameter on PEFC performance is not significant.

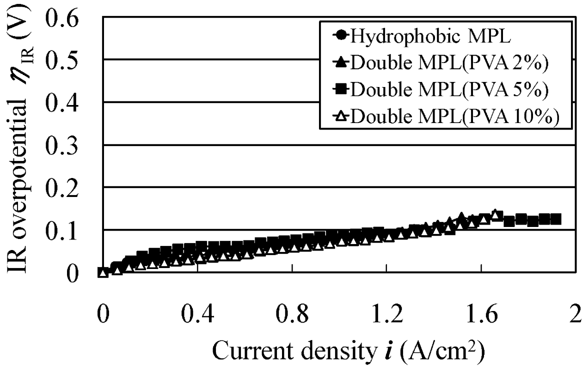
Fig. 13 shows the influence of the double MPL on PEFC performance when the PVA content in the hydrophilic layer ($d_{\max} = 20$ μm, $h_{\mathrm{PVA}} = 5$ μm) was varied between 2 and 10 mass%. PEFC performance obtained with the double MPL coated GDL varies significantly depending on the PVA content in the hydrophilic layer. When the PVA content in the hydrophilic layer is low at 2 mass%, the hydrophilic properties of the MPL are insufficient to conserve humidity at the catalyst layer, so that no significant enhancement in PEFC performance can be expected. Appropriate enhancement of hydrophilicity by increasing the PVA content to 5 mass% is effective for conserving humidity at the cathode catalyst layer, and thereby enhancing PEFC performance significantly. However, when the PVA content is much higher, at 10 mass%, the PEFC performance tends to decrease. Excessive enhancement of the hydrophilic properties results in an increase in accumulated liquid water at the cathode catalyst layer, which reduces the diffusion of oxygen to the electrode. This increases concentration overpotential, thereby lowering PEFC performance.

3.4. Influence of pore diameter and PTFE content of the hydrophobic intermediate layer for the double MPL

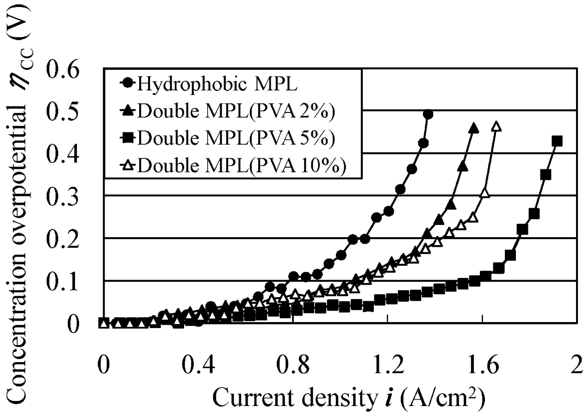
Fig. 14 shows the influence of the double MPL on PEFC performance when the PTFE content in the hydrophobic intermediate layer ($d_{\max} = 20$ μm) was varied between 10 and 40 mass%. For all double MPL coated GDLs, the thickness of 5 μm, the maximum pore



(a) Output voltage



(b) IR overpotential



(c) Concentration overpotential

Fig. 13. Influence of PVA content in hydrophilic layer for double MPL coated GDL on PEFC performance.

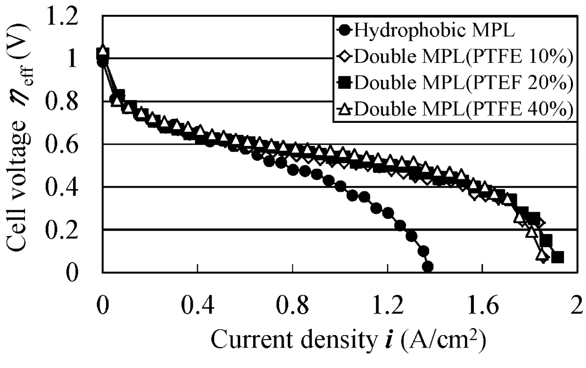
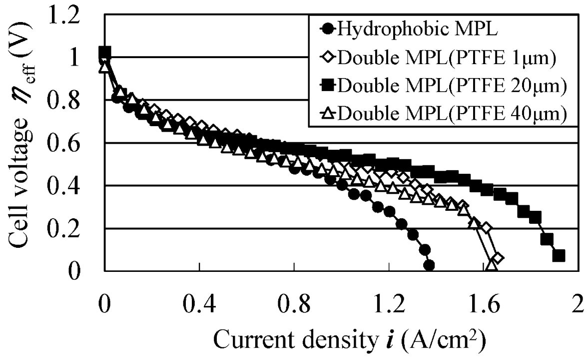
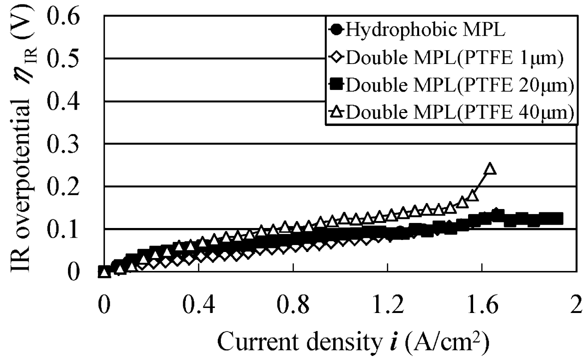


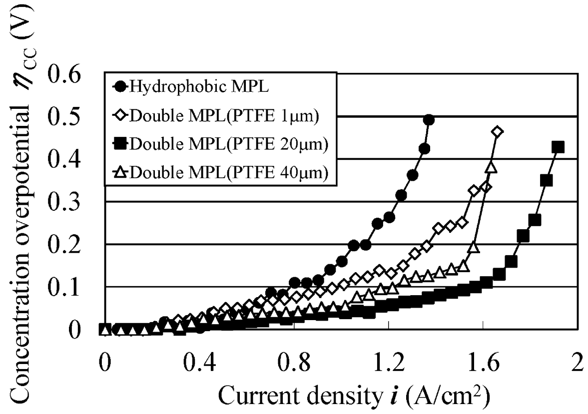
Fig. 14. Influence of PTFE content in hydrophobic intermediate layer for double MPL coated GDL on PEFC performance.



(a) Output voltage



(b) IR overpotential



(c) Concentration overpotential

Fig. 15. Influence of maximum pore diameter of hydrophobic intermediate layer for double MPL coated GDL on PEFC performance.

diameter of 20 μm and the PVA content of 5 mass%, which demonstrated excellent performance as shown in Figs. 10, 12 and 13, were applied to the hydrophilic layer. We have previously reported that PEFC performance obtained with the hydrophobic single MPL coated GDL varied significantly depending on the PTFE content in the MPL [9]. Decreasing the PTFE content is effective for conserving humidity at the cathode catalyst layer, thereby enhancing PEFC performance. For the double MPL coated GDL, however, the difference in PEFC performance is not significant, even when the PTFE content in the hydrophobic intermediate layer is reduced from 40 to 10 mass%.

Fig. 15 shows the influence of the double MPL on PEFC performance when the maximum pore diameter of the hydrophobic intermediate layer (PTFE 20 mass%) was varied between 1 and 40 μm. PEFC performance varies significantly according to the pore diameter of the hydrophobic layer. Decreasing the pore diameter from 40 to 20 μm reduces permeability, so that it is difficult for the water in the hydrophilic layer to be expelled to the GDL substrate. This enhances the ability of the double MPL to prevent dehydration of the MEA, enhancing PEFC performance. However, when the pore diameter becomes too small, at 1 μm, the transport of accumulated liquid water in the hydrophilic layer to the substrate is inhibited, and the diffusion of oxygen gas to the electrode is also reduced. This increases concentration overpotential and thereby lowers PEFC performance.

In the present study, we have demonstrated that the appropriate double MPL coated GDL is effective for enhancing PEFC performance without cathode humidification. When the PEFC operates under high humidity, the flooding phenomenon has a significant influence on PEFC performance. Therefore, we will evaluate the appropriate design parameters of the double MPL to reduce flooding and thereby enhance PEFC performance under high humidity in our future work.

4. Conclusions

A new hydrophilic and hydrophobic double MPL coated GDL was developed to enhance PEFC performance under no-humidification at the cathode. The influences of the thickness, pore diameter, and hydrophilic and hydrophobic properties for the double MPL on PEFC performance were investigated and the following conclusions were obtained.

(1) Further enhancement of PEFC performance was achieved using the hydrophilic and hydrophobic double MPL coated GDL, compared with the hydrophobic single MPL coated GDL. The hydrophilic layer is effective for conserving the humidity of the catalyst layer, while the hydrophobic intermediate layer between the hydrophilic layer and the substrate prevents the removal of water in the hydrophilic layer via dry air in the substrate.
(2) Reducing the hydrophilic layer thickness to 5 μm is effective for enhancing PEFC performance. When the hydrophilic layer thickness is increased, oxygen transport to the electrode through the MPL is decreased, which increases concentration overpotential and thereby lowers PEFC performance. Appropriate enhancement of hydrophilicity by increasing the PVA content to 5 mass% is effective for enhancing PEFC performance. However, when the PVA content is too high, PEFC performance tends to decrease.
(3) When the PTFE content in the hydrophobic intermediate layer is varied between 10 and 40 mass%, the difference in PEFC performance is not significant. Reducing the maximum pore diameter of the hydrophobic layer to 20 μm is effective for preventing the removal of water in the hydrophilic layer to the substrate, thereby enhancing PEFC performance. However, when the pore diameter becomes too small, the transport of accumulated liquid water in the hydrophilic layer to the substrate is inhibited. This increases concentration overpotential, thereby lowering PEFC performance.

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