

# Effect of cathode gas diffusion layer on water transport and cell performance in direct methanol fuel cells

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## Abstract

The optimal design of the cathode gas diffusion layer (GDL) for direct methanol fuel cells (DMFCs) is not only to attain better cell performance, but also to achieve better water management for the DMFC system. In this work, the effects of both the PTFE loading in the cathode backing layer (BL) as well as in the micro-porous layer (MPL) and the carbon loading in the MPL on both water transport and cell performance were investigated experimentally. The experimental data showed that with the presence of a hydrophobic MPL in the GDL, the water-crossover flux through the membrane decreased slightly with increasing the PTFE loading in the BL. However, a higher PTFE loading in the BL not only lowered cell performance, but also resulted in an unstable discharging process. It was also found that the PTFE loading in the MPL had little effect on the water-crossover flux, but its effect on cell performance was substantial: the 40-wt% PTFE loading in the MPL was found to be the optimal value to achieve the best performance. The experimental results further showed that increasing the carbon loading in the MPL significantly lowered the water-crossover flux, but a too high carbon loading would decrease the cell performance as the result of the increased oxygen transport resistance; the 2.0-mg C cm<sup>-2</sup> carbon loading was found to exhibit the best performance.

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**Keywords:** Direct methanol fuel cells; Water crossover; Water management; Micro-porous layer; Back convection

## 1. Introduction

The liquid-fed direct methanol fuel cell (DMFC) has recently received much attention as it has been identified as a leading candidate to compete with the conventional batteries for powering portable electronic devices. However, the commercialization of the DMFC technology is still hindered by several technological problems, among which the water management is one of the key issues [1–5]. Unlike in gas-hydrogen-fed polymer electrolyte fuel cells (PEFCs), in the DMFC, liquid methanol solution is fed to its anode. As a result, liquid water can transport, along with methanol, through the membrane from the anode to the cathode. Water crossover through the membrane may cause two problems for the DMFC technology [6–13]. Firstly, it results in a water loss from the anode, and thus make-up water is needed,

especially for passive DMFCs [6,7]. Secondly, a high rate of water crossover increases the difficulty in avoiding the cathode-flooding problem, limiting the DMFC performance seriously. Therefore, suppressing water crossover through the membrane is of significant importance for simplifying the DMFC system and improving cell performance. Over the past decade, the problem of water crossover through Nafion membranes in DMFCs has been extensively studied [6–20].

It has been understood that the cathode gas diffusion layer (GDL), which typically consists of a backing layer (BL) made of carbon papers or carbon cloths and a micro-porous layer (MPL), plays an important role in both water crossover through the membrane and water ejection from the cathode. Both the BL and MPL are usually waterproof treated with polytetrafluoroethylene (PTFE) for better water management and cell performance. Recently, the effect of cathode GDL on water transport in DMFCs has been investigated extensively [6–13,18–21]. Peled et al. [6] developed a novel approach to recycle water and reduce the rate of water crossover through the membrane for DMFCs

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by building up a high hydraulic pressure at the cathode with the use of a hydrophobic liquid water-barrier layer. They showed that the rate of water crossover through the membrane varied from negative, through zero, to positive values as a function of the thickness and properties of the water-barrier layer. With the optimized water-barrier layer, the water loss from the cathode could be minimized and water-neutral operation conditions could be achieved in a passive DMFC. Lu et al. [10] reported a novel MEA design consisting of a thin membrane (Nafion 112) and a GDL coated with a MPL. The hydraulic liquid pressure built up in the MPL resulting from the large MPL contact angle and small pore size created the back flow of water from the cathode to the anode, which substantially reduced the rate of water crossover through the membrane. To reduce rates of both methanol and water crossover, a similar design of the MEA was also reported by Liu et al. [12], in which, a highly hydrophobic cathode MPL was employed to build up the hydraulic pressure at the cathode that drove the liquid water from the cathode to the anode to offset the water flow dragged by electro-osmosis.

Most recently, we proposed a measurement method that enabled an in situ determination of the water-crossover flux through the membrane in a DMFC [13]. With this method we investigated the effects of various design and geometric parameters as well as operating conditions, such as properties of cathode gas diffusion layer (GDL), membrane thickness, cell current density, cell temperature, methanol solution concentration, and oxygen flow rate, etc., on water crossover through the membrane in a DMFC. Three different cathode GDLs were tested in the experiments, including untreated BL (Toray-090 carbon paper), 30 wt% PTFE treated BL with and without a hydrophobic MPL. It was found the addition of a hydrophobic MPL to the highly PTFE-treated BL not only markedly lowered the water-crossover flux as the result of the enhanced back convection, but also significantly improved the cell performance. However, the PTFE-treated BL without a MPL showed a significantly lower mass-transport limitation than did the untreated BL, because during the PTFE treatment process, thin Teflon films were prone to form in the carbon paper, which covered the gas pores and thus increased the mass transfer resistance of gases. The decreased mass transport limitation as the result of PTFE treatment was also found in a PEMFC [28]. Since this adverse effect of PTFE treatment for the BL can hardly be avoided and it is the highly hydrophobic MPL mainly controls the water and oxygen transport, whether the BL should be highly PTFE-treated with the presence of a hydrophobic MPL should be re-examined.

In addition to the BL, the design and material properties of the cathode MPL for DMFCs also affect the transport behavior of both oxygen and water. Thus, the optimal design of this key cathode component is essential to reduce the anode water loss and to upgrade cell performance. Over the past decade, extensive work has been performed to optimize the cathode MPL for hydrogen-fed PEMFCs, mainly focusing on the effect of the PTFE loading, carbon loading, and carbon types [18,22–34]. Giorgi and co-workers [22,23] investigated the influence of the PTFE loading in the MPL on the cell performance of a PEMFC. They found that a high PTFE loading led to a decrease in the MPL porosity, whereas a low PTFE loading (i.e., <10 wt%)

caused a low hydrophobicity of the MPL, resulting in an ineffective removal of the water that generated in the cell. They found that the best cell performance could be achieved with a MPL consisting of 20 wt% PTFE. Jordan et al. [24] reported the conditions leading to an optimal MPL of a PEMFC; an intermediate loading about  $1.9 \text{ mg C cm}^{-2}$  was found to show the best cell performance. They found that a low loading could not give a non-permeable support for coating with catalyst layer (CL), whereas a high loading resulted in poor oxygen transport through the thicker diffusion layer. Qi and Kaufman [26] investigated different carbon-paper BLs, as well as the effect of MPL with different carbon loadings and different PTFE loadings for a PEMFC. They found that the MPL was the predominated component that controlled mass transport of water and oxygen. Pasaogullary and Wang [29] developed a two-phase numerical model to analyze the multi-layer GDL in PEMFCs. They particularly investigated the effects of porosity, thickness and wettability of a MPL on the two-phase transport. The results showed that placing a MPL could enhance liquid water removal and reduce the liquid saturation in the catalyst layer.

Our literature review indicates that most of previous studies were focused on the effects of the cathode BLs and MPLs on the cell performance of PEMFCs, but ignored their effects on water crossover through the membrane [34], which is another critical factor for the fuel cell system. Moreover, few papers have been reported on the study of the effect of the cathode MPL for DMFCs [35]. Since the water transport and the degree of cathode flooding in DMFCs appears to be significantly different from those in PEMFCs, it is critical to optimize the cathode MPL for DMFCs such that the anode water loss can be reduced and the cell performance can be upgraded. This experimental study was undertaken to elucidate the effects of the PTFE loading in the cathode BL and in the MPL, as well as the carbon loading in the MPL, on both cell performance and water crossover for a DMFC.

## 2. Experimental

### 2.1. The DMFC

The in-house fabricated DMFC consisted of a MEA, with an active area of  $2.0 \text{ cm} \times 2.0 \text{ cm}$ , sandwiched between two bipolar plates, which were fixed by two fixture plates. The MEA consisted of a Nafion 112 membrane and two electrodes. The anode electrode was a single-side ELAT electrode from E-TEK, which used carbon cloth (E-TEK, type A) with 30 wt% PTFE wet-proofing treatment as the backing layer.  $4.0 \text{ mg cm}^{-2}$  unsupported Pt/Ru (1:1 at.%) was used as the catalyst on the anode. On the cathode, the CL was fabricated in-house by the decal method [36]. The catalyst ink was prepared by the method reported elsewhere [36] and sprayed onto the Teflon blank. The CL was then transferred onto the membrane by hot pressing the catalyst coated Teflon blank and the anode electrode on the two sides of the membrane at  $135^\circ \text{C}$  and  $4.0 \text{ MPa}$  for 3 min. The cathode catalyst loading was about  $2.0 \text{ mg cm}^{-2}$  using 80% Pt on Vulcan XC-72 from E-TEK. The content of Nafion ionomer in the cathode CL was maintained to be about 15 wt%. The

decal method for preparing the cathode ensured that the effect of cathode GDL be investigated using different cathode GDLs for the same cathode CL, the same membrane and the same anode. To study the effect of cathode BL, Toray-090 carbon papers treated with different PTFE loadings were tested, and the same hydrophobic MPL was applied on all the BLs. The MPL consisted of Vulcan XC-72 carbon powder, with a carbon loading of about  $2.0 \text{ mg cm}^{-2}$ , and 30 wt% PTFE. Then, to study the effect of cathode MPL, different MPLs with different PTFE loadings and different carbon loadings were tested, which were all applied onto the BL with the optimal PTFE loading.

For convenience of temperature control, both the anode and cathode fixture plates were made of stainless-steel blocks. Single serpentine flow fields, having 0.8 mm channel width, 1.2 mm rib width, and 0.8 mm depth, were formed in the fixture plates for both the anode and cathode sides.

## 2.2. Measurement instrumentation and test conditions

The experiments were carried out in the test rig detailed elsewhere [3]. On the anode, 1.0 M aqueous methanol solution at the flow rate of  $1.0 \text{ ml min}^{-1}$  was fed by a digital HPLC micro-pump (Series III). Before entering the cell, methanol solution was pre-heated to a desired temperature by a heater connected to a temperature controller. On the cathode, synthetic air at the flow rate of 40.0 sccm was supplied without humidification. A mass-flow meter (Omega FMA-7105E), along with a multiple-channel indicator (Omega FMA-5876A), was used to control and measure the flow rate of air. The experiments were performed at the temperature of  $60.0^\circ\text{C}$ . The Arbin BT2000 (Arbin Instrument) electro-load interfaced with a computer was employed to control the cell operation and measure voltage–current (polarization) curves.

## 2.3. Determinations of water-crossover flux

The water-crossover flux through the membrane  $J_{\text{wc}}$  and the corresponding net water-transport coefficient  $\alpha$  are determined, respectively, from [13]:

$$\begin{aligned} J_{\text{wc}} &= \frac{N_{\text{H}_2\text{O}}}{A} - \frac{i}{2F} - \frac{i_c}{3F} \\ &= \frac{N_{\text{H}_2\text{O}}}{A} - i \left( \frac{1}{2F} + \frac{1}{3F} \frac{1 + (ai/Fk_m)}{1 + (ai/F(k_m + k_d))} \right) \\ &\quad \times \left( \frac{i_{c,\text{ocv}}}{i} - \frac{i_{c,\text{ocv}}}{i_{\text{lim}}} \right) \end{aligned} \quad (1)$$

and

$$\begin{aligned} \alpha &= \frac{J_{\text{wc}}F}{i} \\ &= \frac{N_{\text{H}_2\text{O}}F}{Ai} - \left( \frac{1}{2} + \frac{1}{3} \frac{1 + (ai/Fk_m)}{1 + (ai/F(k_m + k_d))} \right) \left( \frac{i_{c,\text{ocv}}}{i} - \frac{i_{c,\text{ocv}}}{i_{\text{lim}}} \right) \end{aligned} \quad (2)$$

where  $N_{\text{H}_2\text{O}}$  is the water flow rate collected at the cathode channel exit,  $A$  the electrode area,  $i$  the cell current density,  $i_c$  the

equivalent methanol-crossover current density,  $F$  the Faraday's constant,  $i_{c,\text{ocv}}$  represents the equivalent methanol-crossover current density at the open-circuit voltage (OCV),  $i_{\text{lim}}$  the anode limiting current density associated with the methanol transport limitation,  $a$  the constant related to the electro-osmotic drag coefficient,  $k_d$  the effective mass-transport coefficient from the channel to the anode CL and  $k_m$  is the effective mass-transport coefficient through the membrane. To ensure that the measured limiting current density of the DMFC ( $i_{\text{lim}}$ ) was caused only by the mass-transport limitation of methanol at the anode, measurements were made under the conditions of feeding rather low methanol solution (0.25 and 0.5 M) and feeding pure oxygen at a high flow rate [4]. The equivalent limiting current density corresponding to the rate of methanol crossover  $i_{\text{lim},c}$  was measured by the voltammetric method [13] by applying a voltage of 0.85 V to the cell under the condition that methanol solution at  $1.0 \text{ ml min}^{-1}$  was fed to the anode, while liquid water, at the same flow rate, was fed to the cathode to create an inert atmosphere. A water trap filled with Dryerite® (anhydrous  $\text{CaSO}_4$ ) was connected to the exit of the cathode channel to collect the water. The water was collected by maintaining a constant current density for about 0.5–3 h. Fifteen minutes were usually needed to make the operating point to be stabilized at every water-collecting point. To eliminate the influence of back pressures on the water transport through the membrane, the respective back pressures on the anode and cathode compartments were kept to the atmosphere pressure. More details about the determination of  $J_{\text{wc}}$  can be found elsewhere [13].

## 3. Results and discussion

### 3.1. Effect of PTFE loading in the BL

Fig. 1 shows the variation in the water-crossover flux with current density for the same MPL but different cathode BLs consisting of Toray-090 carbon papers that were treated with different PTFE loadings (i.e., 0, 5, 10 and 15 wt%). It can be seen that the water-crossover flux, as well as the corresponding

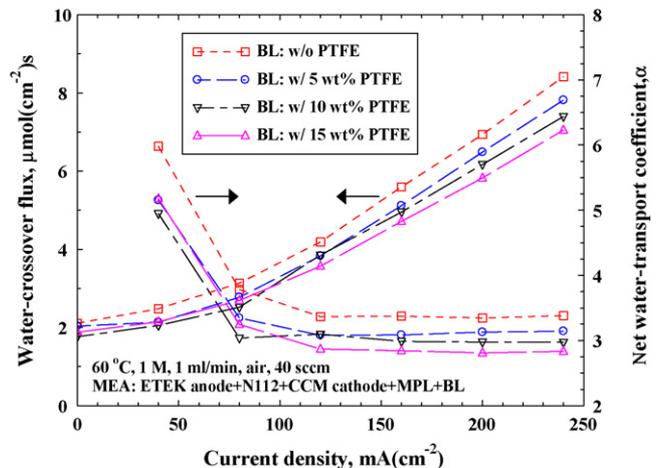


Fig. 1. Effect of the PTFE loading in the cathode BL on the water-crossover flux and the net water-transport coefficient.

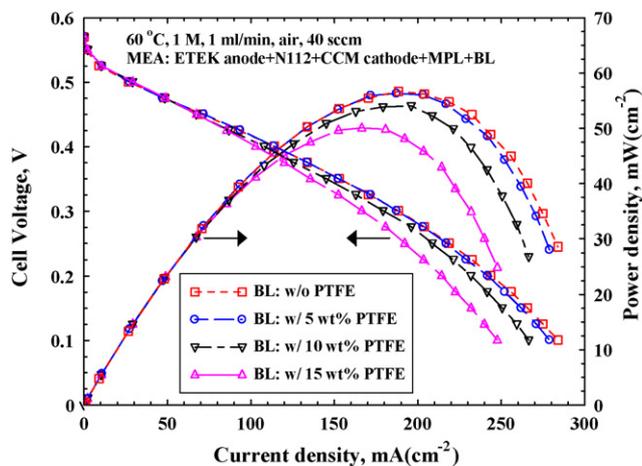


Fig. 2. Effect of the PTFE loading in the cathode BL on the cell performance.

net water-transport coefficient, decreased with the increase in the PTFE loading. For instance, the water-crossover flux was lowered less than about 15% when the PTFE loading was increased from 0 to 15 wt%. The reduction in water crossover as the result of the increased PTFE loading in the BL can be attributed to the enhanced back flow (from the cathode to anode) due to the increased hydrophobic level and decreased permeability in the BL [13].

The effect of the PTFE loading in the BL on the cell performance is shown in Fig. 2. It should be mentioned first that the polarization curves for different cathode GDLs nearly coincided at low current densities, implying that the performance differences at high current densities were mainly caused by the change in cathode GDLs. From Fig. 2 it is seen that the BL without PTFE treatment exhibited the best performance but the cell performance was degraded with increasing the PTFE loading. For instance, when the PTFE loading was increased from 0 to 15 wt%, the cell performance was degraded about 15%. This performance reduction with increasing the PTFE loading in the BL was primarily caused by the increased oxygen transport resistance through the BL, as during the PTFE treatment process, PTFE films might form and block gas pores in the BL [5,13,28]. The pore blockage by PTFE films can not only induce high oxygen transport resistance, but also hinder liquid water removal through the highly restricted BL. Even for low PTFE loadings, the pore blockage effect could be very serious: the SEM images, reported elsewhere [5], showed that over 60% of pores were blocked by PTFE films, although the PTFE loading was only about 14.1 wt%. In the meantime, it is seen from Fig. 2 that the non-PTFE treated BL exhibited a significantly higher limiting current density, indicating that the non-PTFE treated BL could also provide enough gas passages. This is because non-uniform pore-size distributions in carbon papers themselves can provide distinct passages for liquids and gases. Our experimental results indicate that with the presence of a hydrophobic MPL, using non-PTFE treated BLs can not only achieve a sufficient hydrophobic level but also avoid the pore blockages as the result of PTFE treatment.

Fig. 3 presents the effect of the PTFE loading in the BL on the constant-voltage discharging behavior. It is seen from this figure

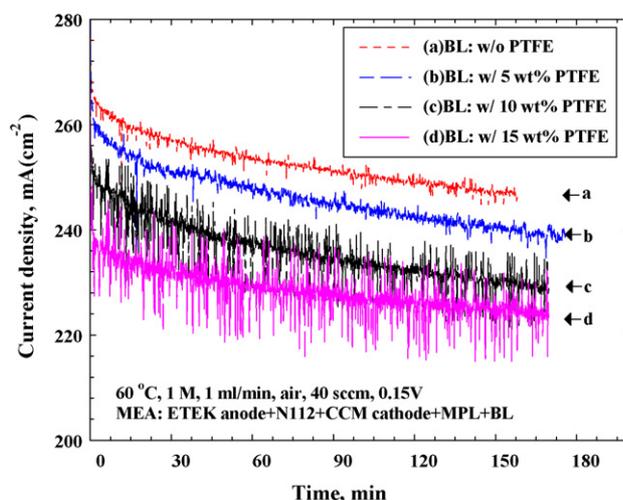


Fig. 3. Transient discharging current at a given cell voltage (0.15 V) with different cathode BLs.

that the increase in the PTFE loading not only led to a decrease in the discharging current density, but also caused serious fluctuations in current density. This unstable discharging behavior may also be attributed to the increased transport resistance as the result of pore blockages by the PTFE films formed during the PTFE treatment process. As a result, the liquid water could not be removed from the cathode unless a sufficient pressure was built up, leading to a periodical removal of water and associated transport process of oxygen.

In summary, the experimental results shown in Figs. 1–3 indicate that although increasing the PTFE loading in the BL can lead to a reduction in water crossover through the membrane, the PTFE treatment will not only yield lower cell performance but also result in significant fluctuations in current density as the result of the increased transport resistance associated with PTFE film blockages. These results suggest that there is no need to treat the cathode BL with PTFE if a hydrophobic MPL is coated on the BL. On the other hand, however, it should also be recognized that the PTFE treatment of carbon papers will increase the mechanical strength of MEAs. Therefore, it is suggested that the PTFE loading in the cathode BL be as low as possible for better oxygen transport and better cell performance.

### 3.2. Effect of PTFE loading in the MPL

The effect of the PTFE loading in the MPL was investigated by testing different MEAs with the same anode and cathode BL (without PTFE treatment) but different cathode MPLs, which consisted of the same carbon loading ( $2.0 \text{ mg cm}^{-2}$ ) but different PTFE loadings (i.e., 20, 30, 40 and 50 wt%). Fig. 4 shows the variation in the water-crossover flux with current density for the different cathode MPLs. It can be seen that the water-crossover flux, as well as the corresponding net water-transport coefficient, were nearly the same for all the tested PTFE loadings in the MPL, indicating that the effect of PTFE loading in the MPL on the water crossover through the membrane is rather small. This finding is somehow different from what it is expected to be: an increase in the PTFE loading will increase the hydrophobic

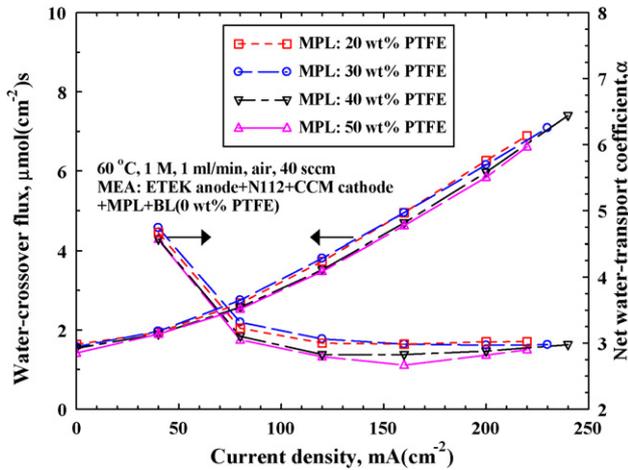


Fig. 4. Effect of the PTFE loading in the cathode MPL on the water-crossover flux and the net water-transport coefficient.

level of the MPL, which is supposed to increase the capillary pressure, thereby enhancing the back flow of water and lowering the rate of water crossover from the anode to cathode. However, it should be recognized that the capillary pressure depends not only on the hydrophobic level but also on the pore size in the porous structure [6,13]. It has already been well understood that during the fabrication process of the MPL, the formation of some macro-pores (between 2 and 20  $\mu\text{m}$ ), or so-called mud cracks [27,31], is hard to be avoided. These relatively larger mud cracks can lead to a reduction in the capillary pressure even though the MPL is highly hydrophobic [6]. It was just based on this understanding, Peled et al. [6] proposed a new method to reduce the formation of large mud cracks in the water-barrier layer such that the water-crossover flux was reduced. In our work, we found that the mud cracks were formed in all the MPLs; the morphology of mud cracks was very similar to that reported elsewhere [22,27,31]. Therefore, it is believed that the presence of these cracks in conventional MPLs significantly reduced the capillary pressure, which mainly account for the negligible effect of PTFE loading in the MPL. Accordingly, reducing the pore size of mud cracks formed during the fabrication process of the MPL is a key issue to reduce water crossover [6].

The effect of the PTFE loading in the MPL on the cell performance is shown in Fig. 5. It can be seen that when the MPL PTFE loading was increased from 20 to 30 and to 40 wt%, the cell performance got significant improvements. However, a further increase in the PTFE loading from 40 to 50 wt% significantly lowered the cell performance. The cell performance turnover at the 40 wt% PTFE in the MPL suggests that this is an optimal PTFE loading to achieve the best cell performance. This value is consistent with that reported elsewhere [37]. This behavior was primarily because that increasing PTFE loading had two effects: increasing the hydrophobic level but lowering the porosity of the MPL, which was also reported by Giorgi and co-workers [22,23]. With the MPL PTFE loading increasing from 20 to 40 wt%, the increased hydrophobic level enabled to lower the water saturation, which led to better oxygen transport and, accordingly, improved cell performance. But when further increasing the

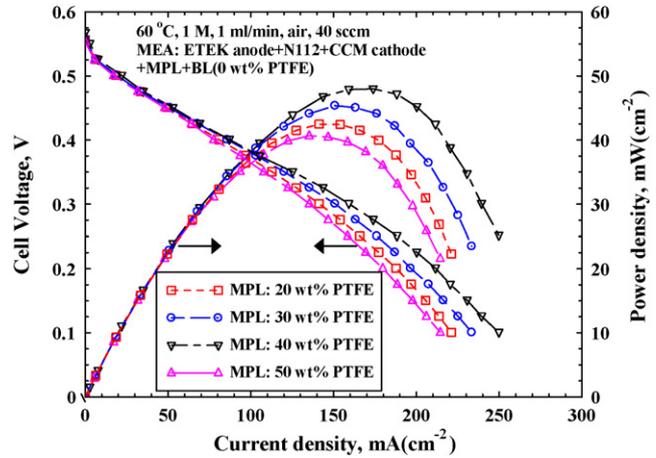


Fig. 5. Effect of the PTFE loading in the cathode MPL on the cell performance.

PTFE loading to 50 wt%, the effect of the decreased porosity of the MPL became critical: the oxygen transport through the MPL became more difficult due to the very small porosity, and thus the cell performance was largely reduced.

### 3.3. Effect of carbon loading in the MPL

With the optimal PTFE loading in the BL (0 wt%) and in the MPL (40 wt%), different MPLs with various carbon loadings (i.e., 1.0, 2.0, 4.0 and 6.0  $\text{mg C cm}^{-2}$ ) were tested. Fig. 6 shows the measured water-crossover flux, as well as the corresponding net water-transport coefficient. It is clear that the water-crossover flux decreased with increasing the carbon loading in the MPL. When the carbon loading was increased from 1.0 to 6.0  $\text{mg C cm}^{-2}$ , the water-crossover flux was reduced from 8.33 to 6.12  $\mu\text{mol cm}^{-2} \text{s}^{-1}$ , a reduction of about 27%. This result indicates that increasing the carbon loading or thickening the MPL can significantly reduce the rate of water crossover.

The effect of the carbon loading in the MPL on the cell performance is shown in Fig. 7. It is seen that increasing the carbon loading from 1.0 to 2.0  $\text{mg C cm}^{-2}$  led to an improved

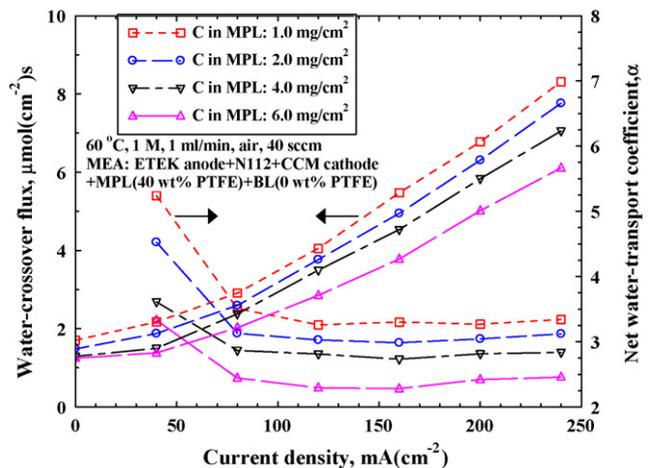


Fig. 6. Effect of the carbon loading in the cathode MPL on the water-crossover flux and the net water-transport coefficient.

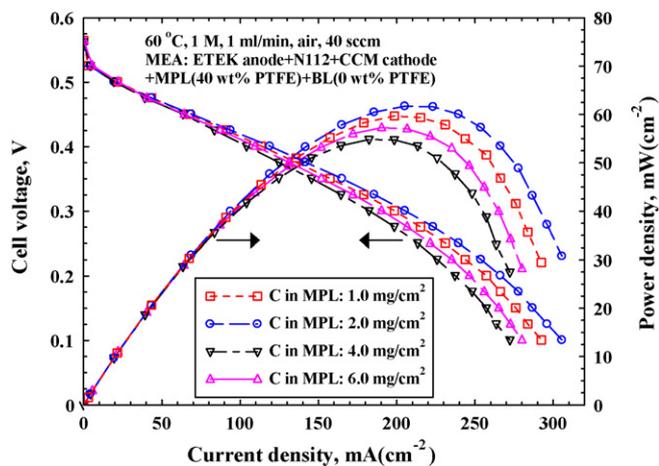


Fig. 7. Effect of the carbon loading in the cathode MPL on the cell performance.

cell performance. However, a further increase in the loading to  $4.0 \text{ mg C cm}^{-2}$  markedly lowered the cell performance. Thus, the intermediate loading of  $2.0 \text{ mg C cm}^{-2}$  was the optimal value for the cell performance. This behavior was also possibly because that increasing the carbon loading led to two effects: lowering the water flooding due to less water crossover (as shown in Fig. 6) but increasing the oxygen transport distance [24]. When the carbon loading was increased from  $1.0$  to  $2.0 \text{ mg C cm}^{-2}$ , the increased thickness in the MPL reduced the water-crossover flux, which led to a reduction in water saturation in the cathode porous layer and better oxygen transport. Hence, the cell performance got improved. However, a further increase in the carbon loading to  $4.0 \text{ mg C cm}^{-2}$  resulted in a too thick MPL, which significantly increased the oxygen transport distance; as a result, the limiting current density was substantially lowered. It is also interesting to notice that the cell performance was slightly increased when the carbon loading was increased from  $4.0$  to  $6.0 \text{ mg C cm}^{-2}$ . One possible reason for this slight change in cell performance is that the cell internal resistance usually decreases with increasing the carbon loading [24]. Our experimental results showed that the cell resistance decreased from  $144$  to  $96 \text{ m}\Omega \text{ cm}^2$  with the carbon loading increased from  $1.0$  to  $6.0 \text{ mg C cm}^{-2}$ . In summary, in general increasing the carbon loading or thickening the hydrophobic MPL can reduce the water-crossover flux, but the carbon loading should not be too high to avoid lowering the cell performance as the result of the increased mass transport polarization.

#### 4. Conclusion

The design and material properties of both the cathode BL and MPL for DMFCs affect the transport behavior of both oxygen and water. Thus, the optimal design of these two key cathode components is not only to upgrade cell performance, but also to reduce the anode water loss and maintain a good water balance in the entire DMFC system. In this work we have experimentally investigated the effects of both the cathode BL and MPL that were treated with different PTFE and carbon loadings on both water crossover and cell performance for a DMFC. Salient

findings and conclusions are summarized as follows:

- (1) With the presence of a hydrophobic MPL, the water-crossover flux through the membrane decreased slightly with increasing the PTFE loading in the cathode BL. However, a higher PTFE loading in the BL not only lowered cell performance but also resulted in significant fluctuations in current when the DMFC discharged at a given voltage, which is attributed to the increased transport resistance associated with PTFE film blockages. Therefore, the PTFE loading in the BL should be as low as possible for better oxygen transport and better cell performance.
- (2) The PTFE loading in the MPL had little effect on water crossover, but its effect on cell performance was substantial. Increasing the PTFE loading in the MPL tends to lower the water saturation, facilitating oxygen transport to the reaction sites and upgrading performance. However, too much PTFE in the MPL will decrease the porosity of the MPL, leading to an increase in the oxygen transport resistance through the MPL and thus lowering the performance. The 40-wt% PTFE loading in the MPL was found to be the optimal value to achieve the best cell performance.
- (3) The experiments showed that increasing the carbon loading in the MPL could significantly lower the water-crossover flux. However, a too large carbon loading will result in a too thick MPL, which increases the oxygen transport resistance and thus lowers the cell performance. The  $2.0\text{-mg C cm}^{-2}$  carbon loading was found to exhibit the best cell performance.

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