Effects of a Microporous Layer on Water Transport Behaviour in PEM Fuel Cell Gas Diffusion Layers

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Abstract
This paper studies water transport behaviours in proton exchange membrane (PEM) fuel cell gas diffusion layers (GDLs) under the effects of a microporous layer (MPL) and some GDL structure parameters, namely thickness and pore size. Different paper GDL samples with and without MPL treatment were used in this study. The breakthrough pressure of liquid water and the amount of water retention in the GDL were measured. The results indicate that applying MPL on the GDL substrate has a greater impact on water transport behaviours in the GDL than changing the structure parameters of the GDL substrate. Compared to the GDL without MPL, the results show that applying MPL on the GDL surface considerably increases breakthrough pressure up to 4.3 times, while it greatly decreases water retention in the GDL by up to 13.7 times. For the GDL thickness, the results indicate that thicker GDL of the same structure requires up to 60% higher pressure to break through the GDL, while it can retain up to 4.8 times more water in its structure than thinner versions. Besides, the results indicate that twice-larger mean pore size GDL requires about 1.4 times lower breakthrough pressure, while it retains approximately twice more water in the GDL.

Keywords: Gas diffusion layer, Microporous layer, Water transport behaviour, Breakthrough pressure, Water retention

1. Introduction
The gas diffusion layer (GDL) plays an important role in the performance of a proton exchange membrane fuel cell (PEMFC) in the form of facilitating gas transport from a gas channel to a catalyst layer (CL), and product water removal in the opposite direction. An excessive presence of liquid water in the GDL limits the amounts of gaseous reactants accessing the reaction areas of the CL, causing a decrease in the cell performance of the PEMFC. Therefore, GDLs are usually wetproofed with a hydrophobic substance, e.g. PTFE (polytetrafluoroethylene), in order to alter their fibres towards becoming more hydrophobic, which eases gas and water transport across the GDL. Several studies [1–8] have demonstrated experimentally and numerically a decrease in water saturation level in the GDL with an increasing degree of hydrophobicity by applying PTFE. Benziger et al. [1, 2] illustrated that PTFE treatment made the pores more hydrophobic limiting the penetration of water into the GDL, thus providing free void space for gas transport. Also, a fine microporous layer (MPL), generally consisting of carbon black and PTFE particles is usually added on one side of the GDL between the GDL and the
CL in order to improve water management and cell performance. In recent studies, Lu et al. [9, 10] demonstrated the saturation reduction in GDL with MPL, compared to GDL without MPL. They attributed this reduction to the role of MPL in limiting the number of locations where liquid water can penetrate to the GDL. In the presence of MPL, Gostick et al. [11] also reported a considerable decrease in water saturation in the GDL, which might benefit gas transport in the PEMFC.

Similarly, in [12], the present author studied the impact of MPL on water transport in paper and cloth GDLs, and its effect was found to be much greater than applying PTFE, both in terms of increasing breakthrough pressure and limiting the amounts of water retention in the GDL. The samples used in this work, however, are limited to a pair of paper and cloth GDLs with and without MPL, which might not be enough to comprehend the role of MPL on water transport in the GDL. In the present work, different commercially available paper GDLs, which are the most commonly used GDLs, with and without MPL, was investigated in order to evaluate the effect of MPL on water transport behaviour in the GDL. By varying the GDL substrate, the effects of its structure on water transport in the GDLs can also be examined. This could contribute to the understanding of liquid water transport characteristics in the GDLs, which is crucial for the design and development of PEMFC.

2. Experimental

2.1. Materials

The GDL samples tested are listed in table 1. They are commercially available carbon paper GDLs with and without MPL. The selected GDL samples cover a range of GDLs commonly used in various PEM fuel cell applications. The thickness and pore diameter of these GDL samples range from about 103 μm to 355 μm, and from less than 1 μm to about 60 μm respectively. The micrographs of the tested samples are presented in figure 1. In this study, similar GDLs with different thicknesses were also examined to evaluate the effect of thickness on water transport in the GDL. In addition, different GDLs with almost similar pore sizes were investigated.

| GDL sample      | MPL | Thickness (μm) | Mean pore diameter (μm) |
|-----------------|-----|----------------|------------------------|
| Sigracet SGL29AA| No  | 180            | 59.5                   |
| Sigracet SGL29BC| Yes | 220            | < 1                    |
| Sigracet SGL39AA| No  | 277            | 47.9                   |
| Sigracet SGL39BC| Yes | 322            | < 1                    |
| AvCarb P75      | No  | 209            | 54.6                   |
| AvCarb GDS2230  | Yes | 235            | < 1                    |
| Toray TGP030    | No  | 103            | 30.5                   |
| Toray TGP060    | No  | 184            | 27.7                   |
| Toray TGP090    | No  | 277            | 30.4                   |
| Toray TGP120    | No  | 355            | 27.6                   |

Figure 1. Micrographs of (a) Sigracet SGL29AA, (b) Sigracet SGL29BC (MPL side), (c) Sigracet SGL39AA, (d) Sigracet SGL39BC (MPL side), (e) AvCarb P75, (f) AvCarb
GDS2230 (MPL side), (g) Toray TGPH030, (h) Toray TGPH060, (i) Toray TGPH090, and (j) Toray TGPH120

2.2. Experiments
In this work, a test apparatus, as shown schematically in figure 2, was built to examine the breakthrough pressure and water retention in the GDL samples. This apparatus is composed of a 2-m high clear acrylic tube of 10 mm in diameter, which is located on top of the two acrylic plates where the GDL sample is placed. A water column is allowed to build up in the acrylic tube creating hydrostatic pressure according to the height of the water above the sample.

For the breakthrough experiment, the required minimum pressure to force liquid water through the GDL was measured. Liquid water was added into the acrylic tube by a peristaltic pump. As the water started to pass through the sample thickness, the height of the built-up water column above the tested sample corresponding to hydrostatic pressure was measured. Also, each sample was weighed before and after the experiment in order to determine the amount of water retained in it.

3. Results and discussion

3.1. Effects of MPL on breakthrough pressure
In order to examine the effect of MPL on the pressure required to force liquid water through the GDL, different GDL samples with and without MPL were used in this study. These include the Sigracet series, i.e. SGL29AA/BC, SGL39AA/BC, and the AvCarb series, i.e. P75/GDS2230. As shown in figure 3, a striped bar represents GDL without MPL, while the adjacent solid bar represents the same GDL substrate but with MPL. The figure shows that MPL has a great impact on the pressure required to force water through the GDL samples. By applying MPL, the minimum pressure required has increased by about 3.7, 4.3 and 2.0 times for the SGL29, SGL39 and AvCarb series respectively. This is because the MPL has diminutive pore sizes (less than 1 μm), which creates large capillary resistance to water flow. Consequently, much greater pressure is needed to push liquid water through the GDL with MPL. This agrees with the results in [9, 10], which report a considerable increase in the required breakthrough pressure for the GDL with MPL.
3.2. Effects of MPL on water retention

Figure 4 compares the amount of water retention in the GDL samples with and without MPL. A striped bar represents the GDL without MPL, while a solid bar represents the GDL with MPL. It is observed that applying MPL greatly decreases the amount of water retention in the GDL samples, amounting to over 5.4, 12.8 and 13.7 times reduction in water retention for the SGL29BC, SGL39BC and GDS2230 respectively, compared to their corresponding GDL without MPL, namely SGL29AA, SGL39AA and P75 respectively. This indicates that the MPL can limit liquid water from entering the GDL through its diminutive pore sizes, as seen in figure 1. These small pore sizes create much larger resistance to water flow than those of the GDL without MPL and, thus, lower water saturation in the GDL, offering more available pores for gas transport. This, again, corresponds to the results in [9, 10].

3.3. Effects of thickness on breakthrough pressure

Figure 5 shows the minimum pressure required to push liquid water through GDL samples with different thicknesses. Only the GDL samples without MPL were used in this study, which includes the Toray TGPH series, represented by the solid bar, with a thickness ranging from about 103 μm to 355 μm, and the Sigracet AA series, represented by the striped bar, with a thickness of 180 μm to 277 μm. For both the Toray and Sigracet series, it is observed that the required breakthrough pressure increases alongside the thickness. For example, the Toray TGPH030 requires around 2,449 Pa to push liquid water through its structure, while the thick TGPH120, the thickest in this series, requires about 60% greater pressure.
at around 3,911 Pa. The increase in breakthrough pressure with increasing GDL thickness agrees with the results reported by Tamayol and Bahrami [13], and Mortazavi and Tajiri [14], who compared similar carbon paper GDLs with different thicknesses. Likewise, in this study, a comparison can be made only among GDLs in the same series, which have a similar structure, and not the entire range of GDLs. For example, the Toray TGPH090 and Sigracet SGL39AA have a similar thickness at around 277 μm, but the Toray requires about 1.3 times larger pressure to push water through it than the Sigracet.

![Graph showing breakthrough pressure for different GDLs](image)

**Figure 5.** Effect of thickness on the breakthrough pressure

### 3.4. Effects of thickness on water retention

In order to examine the effect of thickness on water retention in the GDL, the GDL samples without MPL with different thicknesses were used. Figure 6 shows the amount of water retention in the GDLs, which include the Toray TGPH series with a thickness ranging from about 103 μm to 355 μm, represented by a solid bar, and the Sigracet AA series with a thickness of about 180 μm to 277 μm, represented by a striped bar. For both the Toray and Sigracet, it is observed that water retention in the GDLs of the same series increases as the GDL thickness increases. The Toray TGPH120, the thickest GDL in this series, retains about 4.8 times more water than the TGPH030, the thinnest one. This indicates the higher water retention capability of a thick GDL over its thinner counterpart.

![Graph showing water retention for different GDLs](image)

**Figure 6.** Effect of thickness on water retention in the GDL

### 3.5. Effects of pore size on breakthrough pressure

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Figure 7 shows the required breakthrough pressure to push water through the GDL as a function of mean pore size. In this study, only the GDL samples without MPL were used. The solid line represents the breakthrough pressure required by the GDLs with a thickness of about 180 \( \mu \text{m} \), while the dashed line corresponds to GDLs with a thickness of about 277 \( \mu \text{m} \). It is observed that the breakthrough pressure decreases as the mean pore size increases. For example, the 180-\( \mu \text{m} \)-thick GDL with a mean pore size of about 27.7 \( \mu \text{m} \) requires about 1.4 times larger pressure than that of the 59.5-\( \mu \text{m} \)-pore-size GDL to break through the other side. According to the Young-Laplace equation, this is due to the decrease in capillary resistance at the increased pore size of the GDL. As a result, smaller pressure is required to overcome the decreased resistance of large pore sizes.

![Figure 7. Breakthrough pressure as a function of mean pore diameter at different GDL thicknesses](image)

3.6. Effects of pore size on water retention

Figure 8 shows the amount of water retention in the GDL samples as a function of mean pore size. The solid line represents the amount of water retention in the 180-\( \mu \text{m} \)-thick GDLs, whilst the dashed line represents that in the 277-\( \mu \text{m} \)-thick GDLs. For both thicknesses, it is observed that the amount of water retention increases with increasing mean pore size. For example, the amount of water retention in the thin GDL increases about twofold from 48.2 \( \text{g m}^{-2} \) to 149.3 \( \text{g m}^{-2} \), as the mean pore size increases from 27.7 \( \mu \text{m} \) to 59.5 \( \mu \text{m} \). This is because the GDL with larger pore sizes creates smaller capillary resistance to water flow according to the Young-Laplace equation, which allows more water entering the GDL and being retained by it. The results also show that the thick GDL absorbs more water than the thin one of about similar pore size. For example, the thick GDL with a pore size of about 30.4 \( \mu \text{m} \) can hold about twice more water than the thin one of about a similar pore size of 27.7 \( \mu \text{m} \).

![Figure 8. Water retention as a function of mean pore diameter at different GDL thicknesses](image)
4. Conclusion

The effects of MPL and some GDL structure parameters on breakthrough pressure and water retention in GDLs were investigated. In applying MPL on GDL substrates, it was found that larger pressure is needed to force liquid water through the GDL due to its diminutive pore sizes creating greater resistance to water flow, while water retention in GDLs with MPL was greatly decreased compared to GDLs without MPL. Applying MPL on the GDL substrate, therefore, could effectively control water saturation levels in the GDL of a PEMFC operating at high current densities, where product water is rapidly produced. This could help in providing more available pores for gas transport during high water production rates. For the GDL structure parameters, which are thickness and pore size, it was shown that they have significant effects on both breakthrough pressure and water retention in the GDL, although their influence is much less than the MPL. It was observed that thicker GDLs require larger pressure for water to travel through the GDL, and they can hold more of it in their structure than thinner ones. As regards pore size, it was found that GDLs with larger mean pore size require smaller pressure to break through the GDL, while they allow more water accumulation within their structure than GDLs with smaller mean pore size.

References

[1] Benziger J Nehlsen J Blackwell D Brennan T Itescu J 2005 J. Membrane Sci. 261 98-106
[2] Gauthier E Duan Q Hellstern T Benziger J 2012 Fuel Cells 12 835-847
[3] Bevers D Rogers R von Bradke M 1996 J. Power Sources 63 193-201
[4] Chapuis O Prat M Quintard M Chane-Kane E Guillot O and Mayer N 2008 J. Power Sources 178 258-268
[5] Hao L Cheng P 2010 J. Power Sources 195 3870-3881
[6] Rama P Liu Y Chen R Ostadi H Jiang K Gao Y Zhang X X Fisher R Jeschke M 2012 Aiche J. 58 646-655
[7] Gao Y Zhang X X Rama P Chen R Ostadi H and Jiang K 2013 Comput. Math Appl. 65 891-900
[8] Jinuntuya F Whiteley M Chen R Fly A 2018 J. Power Sources 378 53-65
[9] Lu Z J Daino M M Rath C Kandlikar S G 2010 Int. J. Hydrogen Energ. 35 4222-4233
[10] Kandlikar S G Garofalo M L Lu Z 2011 Fuel Cells 11 814-823
[11] Gostick J T Ioannidis M A Fowler M W Pritzker M D 2009 Electrochem Commun. 11 576-579
[12] Jinuntuya F Kamsanam W 2019 IOP Conf. Ser., Mater. Sci. Eng. 501 012051
[13] Tamayol A Bahrami M 2011 J. Power Sources 196 6356-6361
[14] Mortazavi M Tajiri K 2014 Int. J. Hydrogen Energ. 39 9409-9419