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Water flow in the gas diffusion layer of PEM fuel cells

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Abstract

Water flow through carbon cloth and carbon paper gas diffusion media used for polymer electrolyte membrane fuel cells treated with Teflon was measured. The gas diffusion layer (GDL) media are hydrophobic and it is necessary to apply pressure to force water to penetrate into the pores of these materials. Teflon treatments made the carbon cloth and paper more hydrophobic, no water flowed through the media until pressures of 5–10 kPa were applied to overcome the surface energy of the water/Teflon interface in the largest pores. The largest pores were ~250 μm in the carbon cloth and ~40 μm in the Toray carbon paper. The largest pores in a catalyst layer applied to the woven carbon cloth were ~20 μm. Increasing the applied hydrostatic pressure permitted water to flow through smaller pores in the GDL. Water flows through less than 1% of the void volume in the GDL; the small pores remain free of water and permit gas to get to the catalyst layer.

Keywords: PEM fuel cells; Gas diffusion layer; Water transport

1. Introduction

The gas diffusion layer (GDL) plays the essential role in fuel cells of permitting gas to be transported from the supply channel to the electrode/electrolyte interface. In polymer electrolyte membrane (PEM) fuel cells the GDL must also permit water to be transported from the electrode/electrolyte interface into the gas flow channels where it is convected out of the fuel cell. The gas diffusion layer is a porous membrane that must permit transport of reactant gases to the electrode/electrolyte interface and provide a path for the product water to be removed from the catalyst layer. The transport of gas and liquid in a PEM fuel cell is shown schematically in Fig. 1. When liquid water accumulates in the GDL the gas transport from the gas flow channel to the catalyst layer is hindered and it reduces the power output from the fuel cell layer (the catalyst layer is at the electrode/electrolyte interface). The problem of liquid water is especially acute at the cathode where water is formed in a catalyst layer at the electrolyte/electrode interface. At high current densities water accumulation at the cathode hinders oxygen from getting to the electrode/electrolyte interface causing "mass transport" limitations that limit the maximum current density. The ideal GDL would permit water to be removed from the electrode/electrolyte interface without hindering gas transport.

In addition to permitting gas flow, the gas diffusion layer must be electrically conductive to shuttle electrons between the catalyst layer and the conductors that make up the flow channel. Gas diffusion media are almost always made of a porous carbon cloth or paper. These materials are both porous to permit the gas flow to the electrolyte/electrode interface and have sufficient electrical conductivity to carry the current [1–3]. GDL materials for liquid based fuel cells (e.g. phosphoric acid fuel cells and alkaline fuel cells) had to be designed to hold the liquid electrolyte in between the electrodes [4]. The carbon GDLs in these systems were treated to make them hydrophobic so the liquid would not enter the pores of the GDL. Liquid water had to be prohibited from flowing through the GDL in fuel cells with liquid electrolytes to avoid flooding the gas flow channels. Liquid water in phosphoric acid and alkaline fuel cells is removed by two routes: (1) water vapor diffusion through the GDL and convection out the gas flow channels, and (2) circulation of the electrolyte and vaporizing the water.

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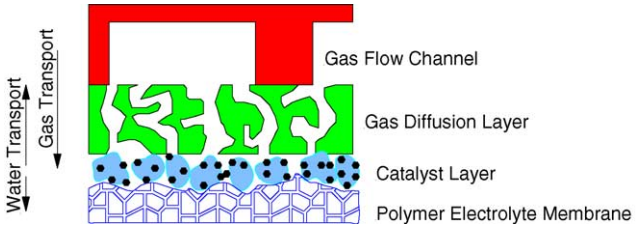

Fig. 1. Schematic of the cathode side of the membrane–electrode assembly. Oxygen or air flows through the gas flow channel, and must diffuse through the gas diffusion layer to the catalyst layer where it reacts with protons coming through the polymer electrolyte membrane to make water. The water formed must go from the catalyst layer through the gas diffusion layer into the gas flow channel where it can be convected out of the fuel cell. Some water may also enter the polymer electrolyte membrane.

In the PEM fuel cell all the water must be removed through the GDL. If the GDL is made of a hydrophilic material liquid water will condense in the GDL and hinder the diffusion of oxygen to the cathode/electrolyte interface. However, if the membrane is hydrophobic it inhibits liquid water from entering the GDL. This can cause water to build up at the catalyst membrane interface. Water formed at the cathode can be absorbed into the membrane which will cause the membrane to swell. The membrane swelling can compress the GDL and eventually the pressure from the membrane will be sufficient to push the water through the hydrophobic pores of the GDL.

This paper reports measurements of the pressure required to push water through different layers of gas diffusion media. These measurements provide details about the pore sizes of different gas diffusion media, and elucidate the role of surface treatments of the GDL in affecting liquid water transport.

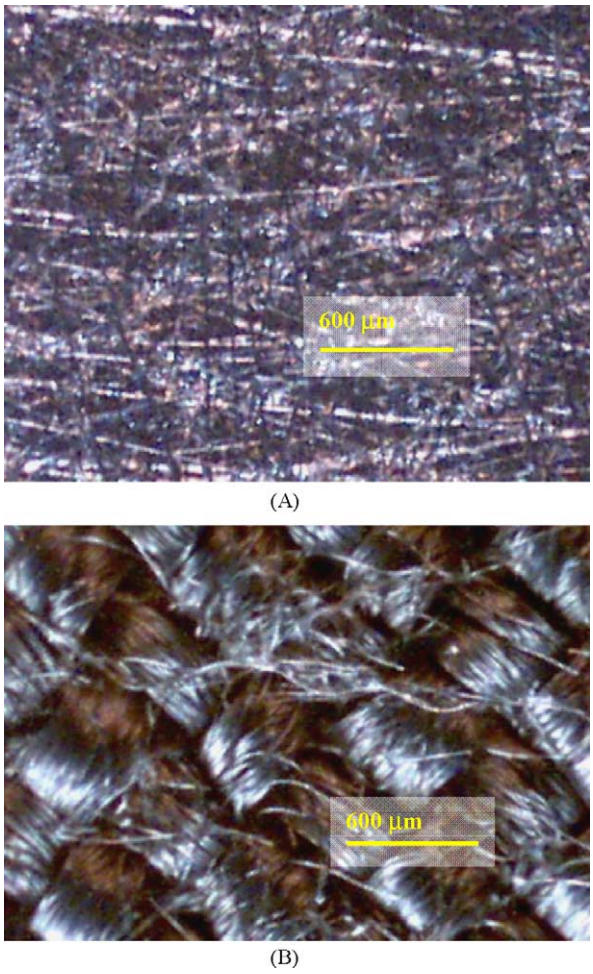

Fig. 2. (A) Micrograph of the Toray carbon paper gas diffusion media. (B) Micrograph of the woven carbon cloth gas diffusion media.

2. Experimental

2.1. Materials

The gas diffusion media tested were all obtained from E-TEK (E-TEK Div. of De Nora N.A., Inc., 39 Veronica Ave Somerset, NJ 08873-6800, USA). Carbon paper GDL material was obtained with different Teflon loadings. TGPH-120 carbon paper (nominal 0.35 mm thick) with 0, 20, 40 and 60% Teflon loading was supplied by E-TEK. Woven carbon cloth (nominal 0.35 mm thick) was also obtained with the same Teflon loadings. Lastly, a single sided ELAT electrode based on the carbon cloth was tested. Shown in Fig. 2 are micrographs of the two different materials. The individual carbon fibers are approximately 12 μm in diameter. The fiber bundles in the woven cloth are ∼400 μm.

The mass per unit area and bulk density of the different media were measured. The void fraction of the different media materials was determined from their uptake of kerosene in an Archimedes displacement measurement. Kerosene completely wetted the carbon cloth and paper and could be seen to wick into both materials. Lastly, the advancing and receding contact angle with water was measured using a Whilemy Plate technique [5]. These physical characterizations are summarized in Table 1.

2.2. Water flow measurements

A 5.0 cm diameter piece of the diffusion media was placed on a porous plate in a pressurized membrane filtration cell (Cole Parmer). The cell was 12 cm high. Clear tygon tubes (1.2 cm diameter) were attached to the top of the cell; one was used to fill the cell and the second tube allowed air to escape. A schematic of the apparatus is shown in Fig. 3. Water was slowly added to the filtration cell, waiting about 5 min after every 1 cm increase in water height in the cell. The hydrostatic head when water flow commenced was recorded. The amount of water drained was recorded as a function of time after water flow commenced. After the water drained out the diffusion media was removed from the cell and was weighed to determine the amount of water retention. The samples were dried in an oven at 75 °C for >2 h, reweighed and the tests were repeated. The drying temperature was kept below 80 °C to minimize the possibility of the Teflon flowing

Table 1
Physical characterization of diffusion media

Media	Dry areal mass (kg/m^2)	Areal mass after liquid water contact (kg/m^2)	Void fraction	Advancing/receding contact angle
Carbon paper (Toray)	0.259 ± 0.007	0.469 ± 0.052	0.72 ± 0.05	115°/30°
Carbon paper + 20% Teflon	0.374 ± 0.007	0.423 ± 0.035	0.69 ± 0.05	170°/120°
Carbon paper + 40% Teflon	0.456 ± 0.010	0.525 ± 0.047	0.59 ± 0.05	170°/120°
Carbon paper + 60% Teflon	0.476 ± 0.009	0.526 ± 0.044	0.50 ± 0.05	170°/120°
Carbon cloth	0.355 ± 0.009	0.484 ± 0.027	0.75 ± 0.05	95°/30°
Carbon cloth + 20% Teflon	0.476 ± 0.012	0.551 ± 0.047	0.73 ± 0.05	170°/120°
Carbon cloth + 40% Teflon	0.595 ± 0.011	0.648 ± 0.041	0.68 ± 0.05	170°/120°
Carbon cloth + 60% Teflon	0.697 ± 0.017	0.839 ± 0.055	0.52 ± 0.05	170°/120°
E-TEK/ELAT electrode	0.435 ± 0.011	0.620 ± 0.036	0.74 ± 0.05	170°/120°

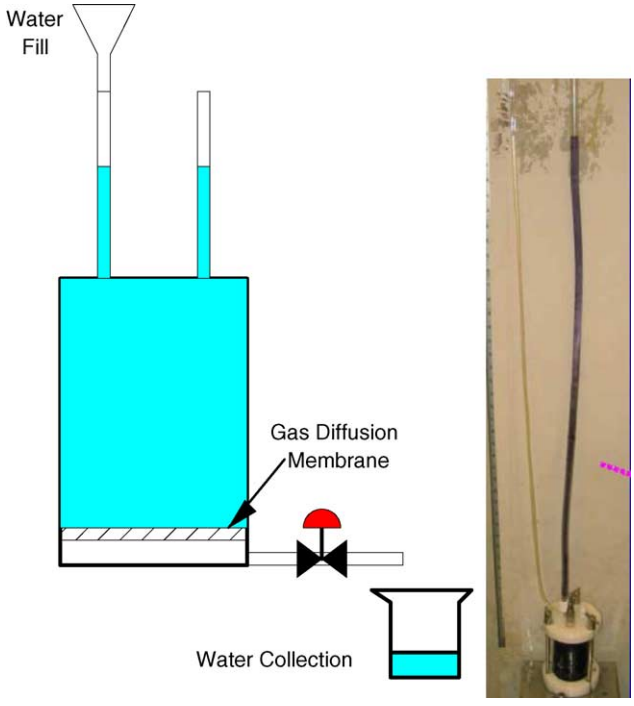

Fig. 3. Schematic of the flow measurement. The hydrostatic pressure for flow is measured from the height of the water above the gas diffusion media. A clamp on the outlet can stop the flow to allow greater hydrostatic pressure heads to be applied. The photo at the right shows the apparatus. Dye was added to the water in the photo to make it easier to identify the height of water in the tube. (No dye was added when making measurements.)

and redistributing in the carbon cloth and paper. The tests were also repeated where the water was permitted to drain and then water was added without removing the diffusion media and drying it.

Additional flow experiments were carried out where a clamp on the outlet from the cell blocked the flow and permitted a larger hydrostatic head of water to be added. The valve was opened and the mass of the water flowing out of the cell was measured as a function of time.

3. Results

The determination of the minimum pressure required for flow was done by slowly adding water to the cell and then

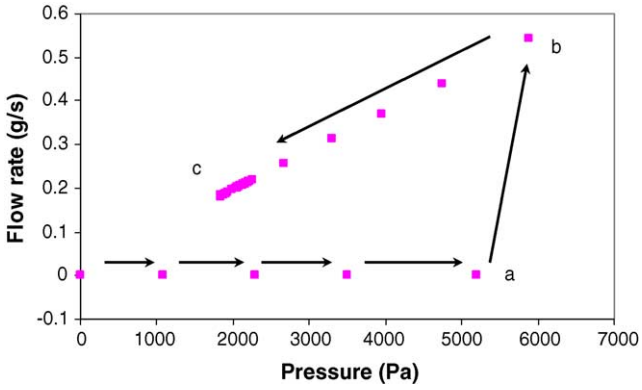

Fig. 4. Flow rate as a function of applied pressure through a carbon paper with 20 wt.% Teflon treatment. No water flow was observed through the carbon cloth until the hydrostatic head exceed 5200 Pa (point a). Once the minimum head was exceeded water flow began (point b) and the water flow decreased over time as the hydrostatic pressure head decreased (points b → c).

measuring the flow rate out of the cell. The flow rate was determined by collecting the water effluent from the cell on a balance and recording the weight every 5 s. A typical data set for these experiments is shown in Fig. 4. Once the minimum pressure was exceeded the water would drain freely nearly emptying the cell. Typically a pool of water approximately 1–3 mm deep was retained on the top of the membrane. Table 2 summarizes the minimum pressure for flow. The flow rates for draining at the minimum pressure were variable. Small variations in the amount of water added beyond the minimum caused variations in the water flux through the GDL media. Each measurement was repeated after removing the membrane from the cell and dried in an oven for 2–4 h at 75 °C. The minimum pressure for flow decreased from the first to the second trial. Additional trials were clustered about the minimum pressure determined in the second trial. The minimum pressure for flow and the flow rates each had variations of ±20%.

After allowing the water to drain and flow had stopped the cell was refilled with water without drying the GDL media. The minimum water pressure to cause flow to recommence was recorded, along with the flow rate. The results for flow through an initially wet GDL show there is a small decrease in the necessary pressure head to cause water to enter the

Table 2
Minimum Pressure Head to Induce Water Flow

Media	Minimum pressure for flow in an initially dry gdl first trial/second trial (Pa)	Pore radius based on minimum pressure (μm)	Minimum pressure for flow in an initially wet GDL (Pa)
Carbon paper (Toray)	5300/3300	25	1200
Carbon paper + 20% Teflon	7100/3800	21	3200
Carbon paper + 40% Teflon	7200/4800	21	4200
Carbon paper + 60% Teflon	7400/4700	21	4000
Carbon cloth	400/200	140	200
Carbon cloth + 20% Teflon	1800/850	140	500
Carbon cloth + 40% Teflon	2200/1500	130	800
Carbon cloth + 60% Teflon	2400/1200	130	850
E-TEK/ELAT electrode	14,000	10	11,000

pores of the GDL and flow compared to an initially dry GDL (see Table 2). The results also show there is a small decrease in the necessary pressure head to cause flow between a virgin GDL and a previously used GDL.

A second set of experiments was carried out where the flow through the GDL media was blocked by closing the effluent valve from the cell. Different initial hydrostatic heads were applied and the valve opened and the effluent flow rates were measured. Fig. 5 shows the data obtained for the carbon cloth with a 20% Teflon treatment. The data was plotted in two ways, in Fig. 5A is the integrated flux (mass of water collected) as a function of time. In Fig. 5B the flux is plotted as a function of time beginning when the water level was at the top of the filtration cell, corresponding to a hydrostatic pressure of 1200 Pa. The mass balance for the water effluent is given by Eq. (1). Neglecting any pressure drop through the effluent tube and valve the slope of $\ln(h/h_o)$ versus time gives the permeability of the GDL membrane (Eq. (2)).

$$\frac{dV}{dt} = A_{cell}\frac{dh_{water}}{dt} = K\,\Delta P = K\rho_{water}gh_{water} \quad (1)$$

$$\ln\left(\frac{h}{h_o}\right) = \left(\frac{K\rho_{water}g}{A_{cell}}\right)t \quad (2)$$

As expected the water effluent flow rate increased with increased hydrostatic head. A more interesting result was that the permeability depended on the initial hydrostatic head applied. Table 3 provides a comparison of the permeability of the different GDL membranes as a function of initial applied hydrostatic head. The larger hydrostatic head permitted water to enter into smaller pores and increased the water flow through the GDL membrane.

4. Discussion

The gas diffusion layer plays an essential role in regulating the flow of water away from the catalyst layer in PEM fuel cells. For water to move from the electrolyte/electrode interface across the GDL there must be a driving force. Gaseous water transport can be driven by a gradient in the partial pressure of water between the electrode/electrolyte interface and the gas flow channel. However, for current densities greater than 200 mA/cm^2 in a PEM fuel cell below 90 °C liquid water must be removed in the effluent from the gas flow channel. When liquid water is present in the gas flow channel there must be a pressure gradient to force the liquid to flow from the catalyst layer to the gas flow channel. The pressure at the electrolyte/electrode interface must be greater that the pressure in the gas flow channel.

In order for the GDL to effectively transport gas from the gas flow channel to the electrolyte/electrode interface the GDL must maintain free void volume for diffusive flow of the gases. To keep water from condensing in the pores of the GDL and blocking gas diffusion the GDL material must be hydrophobic. The pressure gradient to drive water flow must overcome surface energy of the hydrophobic pores of the GDL and the pressure to drive the flow. The pressure gradient to overcome the surface energy is much greater than the pressure to drive the flow, so once water penetrates the

Table 3
Water flow through gas diffusion media

Media	Initial hydrostatic pressure applied (kPa)	Water permeability at applied hydrostatic head of 1000 Pa, K (m^4 s/kg)
Carbon paper + 20% Teflon	7.5	0.5×10^{-10}
Carbon paper + 20% Teflon	10	2.8×10^{-10}
Carbon Paper + 20% Teflon	12.5	5.2×10^{-10}
Carbon paper + 60% Teflon	10	1.7×10^{-10}
Carbon paper + 60% Teflon	12.5	2.6×10^{-10}
Carbon cloth + 60% Teflon	3.6	3.5×10^{-10}
Carbon cloth + 60% Teflon	5.0	7.2×10^{-10}
Carbon cloth + 60% Teflon	10.0	11.1×10^{-10}

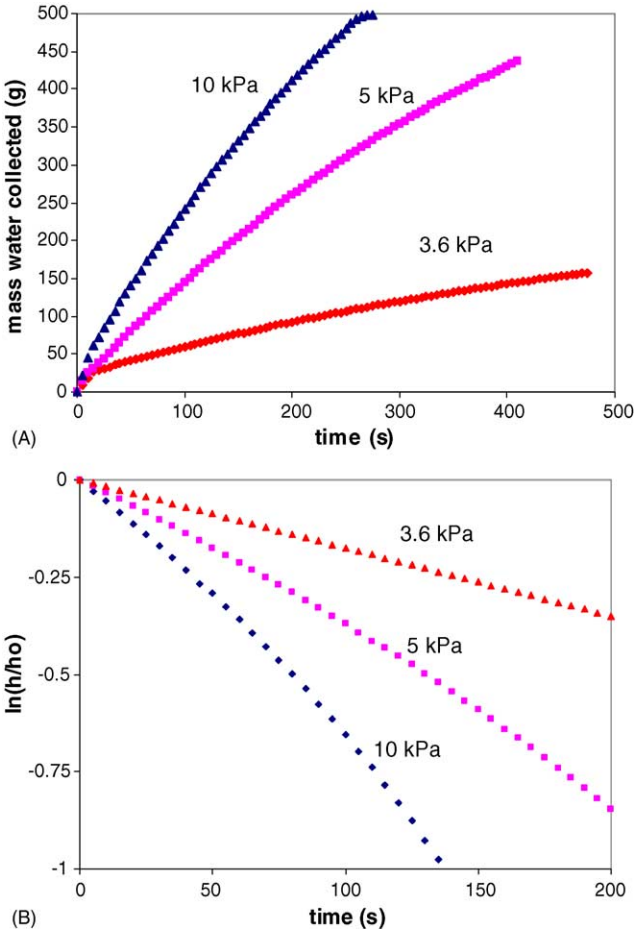

Fig. 5. (A) Integrated mass flux through a 20 wt.% Teflon treated carbon cloth at different initial applied hydrostatic heads. (B) Water flux through a 20 wt.% Teflon treated carbon cloth from a water head of 1200 Pa after draining from different initial applied hydrostatic heads.

GDL the water is easily carried from the electrolyte/electrode interface to the gas flow channel.

There are numerous models of transport in PEM fuel cells which make various assumptions about the flow through the GDL. There are two recent excellent reviews of modeling of PEM fuel cells, which summarize the state of modeling in the GDL [6,7]. Gas transport through the GDL from the gas flow channel to the electrode/electrolyte interface is handled with diffusion models that account for molecular and Knudsen diffusion with some tortuosity factor to account for irregular pore structures [8–11]. Some models include pressure driven gas flow [8,12–14], but it is not clear that there is a significant pressure gradient between the gas flow channel and the electrode/electrolyte interface [7]. There are more significant differences between models in the treatment of water transport, and the effect of liquid water on gas phase transport. Liquid water will occupy some fraction of the void volume of the GDL and hinder gas phase diffusion. Some models treat the liquid water as a solid that simply hinders gas phase diffusion [15–18]. Liquid water flow is most simply handled as single-phase flow, assuming that water and gas move through separate pores [9,10,19,20]. More complete models introduce two phase mixtures in the pores of the GDL [13,21–23].

There are several reports of the importance of hydrophobic surface treatments to assist with flooding control [24–26]. Recently Mathias et al. at GM have published a very thorough characterization of diffusion media materials for PEM fuel cells [2]. Mathias et al. reported water flux measurements at applied pressures of 2.5 kPa and higher and pointed out that the measured fluxes correspond to water flows that greatly exceeded the water production at current densities of 1 A/cm^2. They also reported measurements of pore size distributions in GDL media, but they did not consider the pressure requirement to cause flow through GDL media.

We have found no reports in the literature that discuss the pressure requirement to initiate liquid flow through the gas diffusion layer. There have been efforts to model the liquid flow through a porous media using Darcy's law; but this approach does not recognize that the liquid flow only goes through the largest pores and is not directly related to the void fraction of the GDL. The results reported here indicate that the commercially available GDL media has a unique structure that facilitates counter current flow of liquid water and gaseous reactants.

4.1. Pore dimensions for liquid flow in the gas diffusion layer

The simplified picture of the GDL from the experiments presented here is that it consists of pores with different sizes that run between the catalyst layer and the gas flow channel. There is a distribution of pore sizes as suggested in Fig. 6. For simplicity we assume that the pores are cylindrical and run transverse across the GDL. When liquid water flows through a cylindrical pore it must wet the walls of the pore. If the pore walls are hydrophilic water will be drawn into the pores

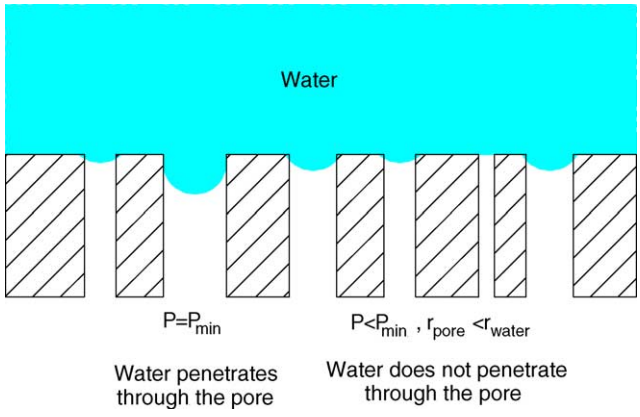

Fig. 6. Idealized model of gas diffusion layer, consisting of parallel cylindrical pores of different diameters. By applying pressure the water is forced into the hydrophobic pores. The largest pores are penetrated first. The more hydrophobic the surface (larger contact angle), the more pressure is required to force the water into the pores.

and liquid condensation will occur in the GDL before liquid water accumulates at the electrode/electrolyte interface. For a hydrophilic GDL there would be no restriction to liquid flow and liquid flow would begin with any applied pressure. On the other hand, if the pore walls are hydrophobic liquid water is excluded from the pores until sufficient work is done to overcome the surface energy. The carbon fibers of both the cloth and paper are slightly hydrophobic; work must be done to push the water into the hydrophobic pores. The larger the pores the less work is required to overcome the unfavorable surface energy. Coating the carbon fibers with Teflon makes the pores highly hydrophobic and requires a higher pressure to push the water into the pores. The pressure, ΔP, that must be applied to force water into the pores of radius r_{pore} is given by the Young and Laplace Equation (Eq. (3)),

$$\Delta P = \frac{2\gamma_{\text{water}} \cos\theta}{r_{\text{pore}}} \tag{3}$$

where γ_{water} is the surface energy of water and θ is the contact angle of water with the surface of the pore [5]. If the applied pressure is less than the right hand side of Eq. (3) the water is excluded from the pore. The experimental results for minimum pressure for the flow of water through the different GDL media gives a measurement of the largest pores in the GDL material. It is the largest pores that have smallest total surface energy and require the smallest pressure for liquid water to penetrate. (This experiment is similar to a mercury porosimetry experiment!) The calculated values for the largest pore size are summarized in Table 2. For water entering the pores the contact angle used in the calculation is the advancing contact angle from the Whilemy plate measurement.

The results in Table 2 show that the woven cloth has the largest pores, those pores being ~250 μm in diameter. The carbon paper has pores about five times smaller and the ELAT catalyst layer has pores that are only 20 μm in diameter. The pores in the woven carbon cloth can be imaged and the model of the parallel pores is close to reality. Fig. 7 shows a micrograph of the carbon cloth taken with back lighting (this corresponds to the image in Fig. 2B, but with lighting from the back). The micrograph shows clearly the pores in the cloth with dimensions of ~200–300 μm. Backlit images of the carbon paper did not show anything; the pores in the Toray paper are more tortuous and irregular.

4.2. Liquid water in the gas diffusion layer

Knowing the size of the pores and the total water flux through the GDL media the number of pores required and the volume of pores required for the water flow can be determined. Assuming laminar flow of liquid water through cylindrical pores with length L_{pore} = GDL thickness, the volumetric flow of water through each pore is given by Eq. (4). Dividing the measured water flux by the flux through the penetrated pore gives the number of pores (Eq. (5)). The total pore volume of the GDL media associated with liquid water flow is found by multiplying the number of pores by the volume of each pore. The volume of the GDL media filled with water, $\varepsilon_{\text{water}}$, is found from the mass gain after the water flow experiment (Eq. (6)).

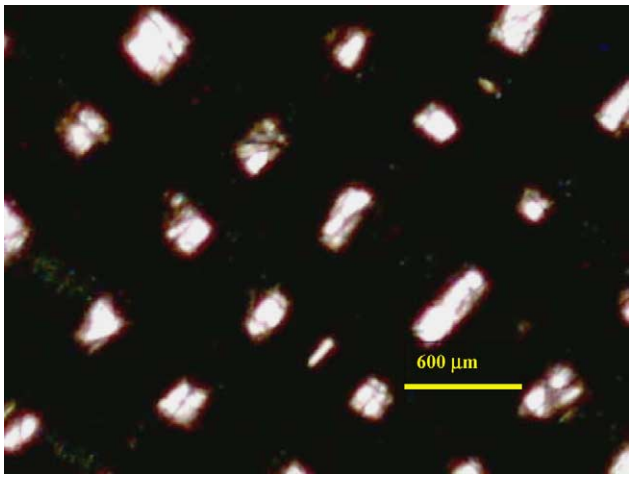

Fig. 7. Backlit image of carbon cloth GDL material. The pores are seen at the intersections between the carbon fibers.

$$Q_{\text{water}}^{\text{pore}} = \frac{\pi r_{\text{pore}}^4 \Delta P}{8\mu_{\text{water}} L_{\text{pore}}} \tag{4}$$

$$N_{\text{pore}} = \frac{\text{water flux}}{Q_{\text{water}}^{\text{pore}}} \tag{5}$$

$$\varepsilon_{\text{water}} = \frac{m_{\text{wet}} - m_{\text{dry}}}{\rho_{\text{water}} V_{\text{GDL}}} \tag{6}$$

Table 4 summarizes the volumes needed for liquid–water flow in the different GDL media. The volumes correspond to a liquid water flow of 4×10^{-6} m^3/s/m^2 for a constant applied pressure of 1 kPa. (This water flux corresponds to water production for a current density of 1.1 A/cm^2 in the fuel cell). The number of pores necessary to conduct the water flow is small, and the volume needed to carry the liquid flow is a small fraction of the volume of the GDL. The large pores of the carbon cloth easily carry the water flow and less volume is needed to carry the water flow than in the carbon paper. The volume for liquid–water flow is compared to total void volume fraction of the GDL, $f_{\text{flow,water}}(\varepsilon)$ = fraction of GDL void volume needed to carry water flux. This can be compared to the volume fraction of the GDL filled with water, $f_{\text{total,water}}(\varepsilon) = \varepsilon_{\text{water}}/\varepsilon$. Water occupies a much greater volume in the GDL membranes than is accounted for by the water flow through the GDL membrane; $f_{\text{flow,water}}(\varepsilon)/f_{\text{total,water}}(\varepsilon) \ll 1$. Even at the highest liquid flow rates imaginable in a fuel cell (1 A/cm^2) the fraction of the void volume carrying the liquid water flow will be less than 0.1%.

The membrane holds much more water than is necessary for the liquid–water flow because the pore structure is not simple cylindrical pores. Pot-bellied pores, that have larger cavities than exits, will trap liquid in the cavities. A simplified picture of water trapped in the cavities of the GDL is illus-

Table 4
Water Filling of GDL Media

Media	GDL volume fraction for liquid–water flow	GDL volume fraction filled by liquid–water ε_{water}	$f_{flow}(\varepsilon)$	$f_{water}(\varepsilon)$
Carbon paper (Toray)	1.1×10^{-4}	0.60 ± 0.05	1.6×10^{-4}	0.83 ± 0.11
Carbon paper + 20% Teflon	2.9×10^{-5}	0.14 ± 0.05	4.1×10^{-5}	0.20 ± 0.11
Carbon paper + 40% Teflon	3.2×10^{-5}	0.18 ± 0.05	5.4×10^{-5}	0.31 ± 0.11
Carbon paper + 60% Teflon	2.5×10^{-5}	0.11 ± 0.05	5.1×10^{-5}	0.22 ± 0.11
Carbon cloth	2.1×10^{-6}	0.43 ± 0.05	2.9×10^{-6}	0.57 ± 0.11
Carbon cloth + 20% Teflon	1.7×10^{-6}	0.21 ± 0.05	2.4×10^{-5}	0.29 ± 0.11
Carbon cloth + 40% Teflon	1.8×10^{-6}	0.15 ± 0.05	2.7×10^{-5}	0.22 ± 0.11
Carbon cloth + 60% Teflon	2.0×10^{-6}	0.16 ± 0.05	3.8×10^{-5}	0.31 ± 0.11
E-TEK/ELAT electrode	7.0×10^{-4}	0.52 ± 0.05	9.5×10^{-4}	0.71 ± 0.11

trated in Fig. 8. Water gets pushed into these cavities, but these do not play a role in the flow of liquid–water through the GDL membrane. The liquid flow will go through a percolation path of the largest pores. Once the percolation path is established there is no longer any work required to overcome the surface energy and the liquid–water flow becomes rapid.

Teflon treatments of the GDL material make the pores more hydrophobic, thus limiting water penetration into the GDL. More work is needed to force water into the hydrophobic pores, so the Teflon treatment keeps the smaller pores liquid–water free. The values of $f_{water}(\varepsilon)$ in Table 4 show the Teflon treatments greatly reduce the fraction of the void volume into which water penetrates. The addition of the Teflon reduces the total void volume (see Table 2), but it has a much greater effect on the amount of water that can penetrate into the GDL membrane. The results from this study also suggest that not much is gained in terms of altering the water flux by increasing the Teflon loading from 20–60 wt.%. The ELAT catalyst layer from E-TEK is also very hydrophobic. The preparation of the ELAT catalyst layer is proprietary, but E-TEK has acknowledged that Teflon is used as a binder to hold the carbon particles together. The pore size in the catalyst layer is smaller than in the GDL. This indicates that the catalyst layer may pose the largest resistance to water flow in a typical membrane–electrode assembly. However, the catalyst layer generally has Nafion added when making the membrane–electrode assembly [27]. The Nafion would make the catalyst layer more hydrophilic and could reduce the pressure needed to promote liquid water flow.

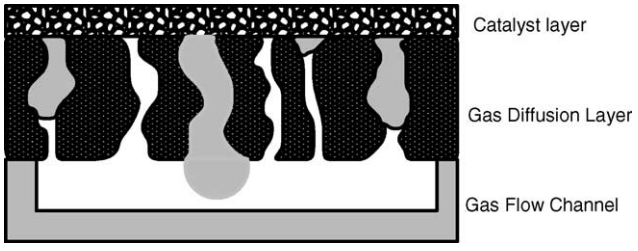

Fig. 8. Water in the gas diffusion layer. The water enters pores where the radius of the entrance is greater than the critical radius for penetration, but if the pore narrows down the water is trapped, preventing either water or reactant gases from moving through the pore. In the smaller pores no water enters. Pores with entrances larger than the critical radius and with exits larger than the entrances permit the water to flow into the gas flow channel (the center pore).

4.3. Implications for fuel cell engineering

The results presented here suggest that a distribution of pore sizes is a key feature in the functioning of the gas diffusion layer. Liquid water is pushed out of a small number of large pores from the electrode/electrolyte interface, while the small pores are kept free of liquid condensation. Gas is able to move through the small pores from the gas flow channel to the electrode/electrolyte interface. Only a small fraction of the pores are necessary to carry the liquid water from the electrode/electrolyte interface. Some pores get filled with water, but do not contribute to the water flow from the electrode/electrolyte interface to the gas flow channel. Water trapped in the GDL can hinder the gas diffusion from the gas flow channel to the electrode/electrolyte interface. Teflon treatments of the GDL reduce the amount of water that can be trapped, and help maintain good gas transport through the GDL.

Another essential finding from this work is that a pressure must build up at the electrolyte/electrode interface to push the water through the pores of the gas diffusion layer. Either the water will accumulate as a separate phase, building up a pressure because it is physically constrained, or water will be absorbed into the electrolyte membrane causing the polymer to swell. The swelling will generate the pressure to push the water into the gas diffusion layer. The mechanical properties of the polymer electrolyte and MEA construction will both play crucial roles in affecting the water transport through the GDL.

The results presented here suggest that the ideal GDL material should have a bimodal pore size distribution. There should be a few large pores to carry water and lots of small pores to carry gas. The small pores should be very hydrophobic. The large pores for carrying the water would perform best is they were mildly hydrophobic (contact angle near 90°) to minimize the pressure needed to push the water through the pores, but also to keep the water from accumulating in the pores. It would also be best if the pores were not interconnected. The GDL media tested all retained some water after water flow experiments. If the pores were straight or had hour-

glass figures the water would freely drain out once flow had been initiated. But in the real materials there were potbellied pores that retained water in the cavities after the pressure was reduced. The ideal GDL would eliminate these cavities where liquid was retained hindering the reactant transport.

5. Conclusions

Liquid water is transported across the gas diffusion layer through the largest pores. Only a few percent of the void fraction of the GDL is necessary for liquid water transport and the smaller pores remain free for gas diffusion of reactants from the gas flow channel to the electrolyte/electrode interface. Water formed at the electrolyte/electrode interface must build up sufficient pressure to force the water to penetrate into the largest pores of the GDL. Once the water penetrates the largest pores the liquid can freely drain from the applied pressure. Commercially available GDL media made from carbon fibers treated with Teflon possess the desirable bimodal pore structure necessary to carry liquid water and gas in counter current flow. Improvement of commercial GDL media would reduce the amount of water that is trapped in pores that do not carry the liquid water flow from the electrode/electrolyte interface to the gas flow channel.

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Nomenclature

A_{cell}	cross-sectional area of ultrafiltration cell
$f_{flow}(\varepsilon)$	fraction of GDL void volume required for liquid water transport
$f_{water}(\varepsilon)$	fraction of GDL void volume filled with water
g	gravitational constant
h, h_{water}	height of water head
h_o	initial height of water head
L_{pore}	length of pore in GDL
N_{pore}	number of pores for water flow
ΔP	pressure differential across the GDL
ΔP_{min}	minimum pressure across the GDL for water to penetrate
Q	volumetric flow rate
r_{pore}	radius of pore in GDL for liquid transport
γ_{water}	surface tension of water
ρ_{water}	density of water
ε	void fraction of the GDL
ε_{water}	volume fraction of the GDL occupied by water
ε_{flow}	volume fraction of the GDL required for 1.86×10^{-6} $m^3/s/m^2$ liquid–water flow
θ	water/GDL contact angle
μ_{water}	viscosity of water

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