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Effects of porosity distribution variation on the liquid water flux through gas diffusion layers of PEM fuel cells

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Abstract

Flooding of the membrane electrode assembly (MEA) and dehydrating of the polymer electrolyte membrane have been the key problems to be solved for polymer electrolyte membrane fuel cells (PEMFCs). So far, almost no papers published have focused on studies of the liquid water flux through differently structured gas diffusion layers (GDLs). For gas diffusion layers including structures of uniform porosity, changes in porosity (GDL with microporous layer (MPL)) and gradient change porosity, using a one-dimensional model, the liquid saturation distribution is analyzed based on the assumption of a fixed liquid water flux through the GDL. And then the liquid water flux through the GDL is calculated based on the assumption of a fixed liquid saturation difference between the interfaces of the catalyst layer/GDL and the GDL/gas channel. Our results show that under steady-state conditions, the liquid water flux through the GDL increases as contact angle and porosity increase and as the GDL thickness decreases. When a MPL is placed between the catalyst layer and the GDL, the liquid saturation is redistributed across the MPL and GDL. This improves the liquid water draining performance. The liquid water flux through the GDL increases as the MPL porosity increases and the MPL thickness decreases. When the total thickness of the GDL and MPL is kept constant and when the MPL is thinned to 3 μm, the liquid water flux increases considerably, i.e. flooding of MEA is difficult. A GDL with a gradient of porosity is more favorable for liquid water discharge from catalyst layer into the gas channel; for the GDLs with the same equivalent porosity, the larger the gradient is, the more easily the liquid water is discharged. Of the computed cases, a GDL with a linear porosity $0.4x + 0.4$ is the best.

Keywords: Polymer electrolyte membrane fuel cell; Gas diffusion layer; Gradient; Porosity; Liquid water flux

1. Introduction

Water management is essential for effective and steady operation of polymer electrolyte membrane fuel cells (PEMFCs), which require the membranes to be hydrated and the membrane electrode assemblies (MEAs) not to be flooded. Therefore on one side, researchers study all kinds of humidification methods to make up the water that the membranes lose; while on the other side workers hope drive the liquid out of the MEAs. Humidification includes liquid water injection, bubbles, membrane humidification, self-humidification and many other methods [1–6]. Flooding of MEA and the liquid water transport in fuel cells were diagnosed or observed by people with transparent fuel cells [7,8], a pressure drop method [9] and neutron radiography [10,11]. As for liquid water discharge studies, Tuber et al. [7], Yang et al. [8] and Liu et al. [12] carried out a gas channel two-phase flow test, while Quan et al. [13] and Jiao et al. [14] carried out a two-phase gas channel flow modeling. Meanwhile much attention has been paid to the gas diffusion layer (GDL). Kong et al. deemed that the pore-size distribution of the GDL should be of bimodal, with large pores for liquid water discharge and small pores for gas diffusion, and so the material and the structure of GDL should be improved [15]. Wang et al. added magnetic particles inside the cathode catalyst layer to attract the paramagnetic oxygen to the catalyst layer while discharging the diamagnetic liquid water out of the MEA [16]. Adding a sub-layer micro porous layer (MPL) between the catalyst layer and the GDL is considered an effective

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Nomenclature

d_f	diameter of fiber (μm)
F	Faraday constant (C mol^{-1})
J	current density (A m^{-2})
K	permeability
k_{rl}	relative permeability
k_K	Kozeny constant
M	Mole molecular weight (g mol^{-1})
p	pressure (Pa)
Q	liquid water volume remaining in GDL (m^3)
q	liquid water flux through GDL (kg s^{-1} m^{-2})
r	radius of pore (μm)
s	saturation
V	volume of liquid or porosity (m^3)
x	dimensionless thickness of GDL and MPL

Greek symbols

α_e	effective diffusion coefficient of water through membrane
δ	thickness of GDL or MPL (μm)
ε	porosity
μ	dynamic viscosity (Pa s)
ν	kinematic viscosity (m^2 s^{-1})
θ	contact angle
ρ	density (kg m^{-3})
σ	surface tension (N m^{-1})
ξ	thickness of GDL + MPL

Subscripts

c	capillary
g	gas
H_2O	water
l	liquid
w	wetting phase
nw	non-wetting phase

method to improve the liquid water drainage and gas diffusion. Jin et al. studied two-phase flow phenomena in the GDL and the MPL [17], Pasaogullari et al. compared the difference of the two-phase transport in the GDL and the MPL between the unsaturated flow theory and the multi-phase mixture formalism [18]. Wilkinson et al. put forward the concept of a gradient, and thought both the operational parameter and the MEA structure should be optimized [19]. Chu et al. suggested a gradient porosity for the GDL, and investigated the effect of average porosity on the oxygen transport and then on the variation of current density, i.e. the influence of porosity on the concentration polarization, the effect of the phase change and liquid water were not considered [20]. Roshandel et al. revealed the effects on PEM fuel cell performance of the porosity distribution variation resulting from the compression pressure corresponding to the assembly process and the presence of liquid water [21].

So far, almost no papers published have especially focused on the studies on the liquid water flux through differently structured GDLs. In this paper, for gas diffusion layers (GDLs) including structures of uniform porosity, a sudden change porosity (GDL with microporous layer (MPL)) and a gradient change in porosity, using a one-dimensional model, the liquid saturation distribution is analyzed based on the assumption of a fixed liquid water flux through the GDL. The liquid water flux through the GDL is calculated based on the assumption of a fixed liquid saturation difference between the interfaces of catalyst layer/GDL and GDL/gas channel. In the following, the computation models and solution procedure are introduced. Then the results are presented and analyzed. Finally, some interesting conclusions are drawn.

2. Model development

2.1. Capillary pressure and saturation distribution

Capillary pressure is the pressure difference between the non-wetting phase and the wetting phase; for the hydrophobic GDL, it is expressed as

$$p_c = p_{nw} - p_w = p_l - p_g \tag{1}$$

$$p_c = \frac{\sigma \cos\theta_c}{(K/\varepsilon)^{0.5}}(1.417s - 2.12s^2 + 1.26s^3), \quad \theta_c > 90^\circ \tag{2}$$

where $s = V_l/V_p$ is liquid saturation, $K = \varepsilon^3 d_f^2/(16k_K(1-\varepsilon)^2)$ the absolute permeability and $k_{rl} = s^3$, is relative permeability.

Liquid water transport in GDL is caused by capillary pressure difference. Suppose under steady condition, the water produced by electrochemical reaction in catalyst layer and the water transferred effectively from the membrane is changed into liquid fully and discharged through GDL to gas channel, hence

$$q_{H_2O} = -\frac{\rho_l K k_{rl}}{\mu_l}\nabla p_c = -\frac{\rho_l K k_{rl}}{\mu_l}\left(\frac{dp_c}{ds}\right)\frac{ds}{dx} \tag{3}$$

or

$$q_{H_2O} = -\frac{\sigma \cos\theta_c}{\nu}\frac{\varepsilon^2 d_f}{4(1-\varepsilon)\sqrt{k_K}}s^3(1.417 - 4.24s + 3.789s^2)\frac{ds}{dx} \tag{4}$$

2.1.1. GDL with uniform porosity

For GDL with uniform porosity, there is no change of the porosity along the GDL thickness. Integrating equation one gets

$$q_{H_2O}x = -\frac{\sigma \cos\theta_c}{\nu}\frac{\varepsilon^2 d_f}{4(1-\varepsilon)\sqrt{k_K}} \times \left(\frac{1.417s^4}{4} - \frac{4.24s^5}{5} + \frac{3.789s^6}{6}\right) + C \tag{5}$$

2.1.2. A GDL with gradient in porosity

Suppose the porosity varies along the thickness following the polynomial rules, a linear type and a parabola type are chosen.

Porosity varying along the thickness follows linear rules, i.e. $\varepsilon = a_1 x + a_0$, then

$$q_{H_2O} = -\frac{(\varepsilon/(1-\varepsilon))s^3(1.417 - 4.24s + 3.789s^2)}{[(\nu/\sigma\cos\theta_c)(4\sqrt{k_K}/d_f) + (a_1\varepsilon/(1-\varepsilon)^2)s^3(1.417s - 2.12s^2 + 1.263s^3)]}\frac{ds}{dx} \quad (6)$$

Porosity varying along the thickness follows parabola rule, i.e. $\varepsilon = a_2x^2 + a_1x + a_0$, then

$$q_{H_2O} = -\frac{(\varepsilon/(1-\varepsilon))s^3(1.417 - 4.24s + 3.789s^2)}{(\nu/\sigma\ \cos\theta_c)(4\sqrt{k_K}/d_f) + (\varepsilon(2a_2x + a_1)/(1-\varepsilon)^2)s^3(1.417s - 2.12s^2 + 1.263s^3)}\frac{ds}{dx} \quad (7)$$

2.2. Liquid water flux through GDL and liquid water volume remaining in the GDL

The GDL is made up of carbon fiber or other porous materials, which act as diffusing gas, draining water, conducting electrons, etc. The electrical conductivity of the GDL has never been an obstacle to the effective and steady operation of PEMFC, but flooding and then gas diffusion decrease are often serious problems. Therefore the GDL structure should be optimally designed to increase liquid water flux through the GDL, and to decrease liquid water volume remaining in the GDL so as to improve oxygen diffusion from gas channel to catalyst layer.

2.2.1. Liquid water flux through the GDL under steady conditions

In the GDL, liquid water is supposed to be discharged by capillary pressure. Under steady conditions, the liquid water flux through the GDL can be obtained from

$$q_{H_2O} = -\frac{\rho_l K k_{rl}}{\mu_l}\nabla p_c = -\frac{\rho_l K k_{rl}}{\mu_l}\left(\frac{dp_c}{ds}\right)\nabla s$$

It is hoped that the liquid water flux through GDL is be large enough when a fixed liquid saturation difference exists between the interfaces of catalyst layer/GDL and GDL/gas channel, in other words, when a fixed liquid flux through GDL presents a liquid saturation difference that is small.

2.2.2. Liquid water volume remaining in the GDL under steady conditions

Considering the saturation and porosity variation along the thickness of the GDL, the liquid water volume remaining in the GDL under steady conditions can be obtained from

$$Q_l = \int_0^\xi v_l(x)\,dx = \int_0^\xi \varepsilon(x)s(x)\,dx \quad (8)$$

This equation presents the volume occupied by the liquid water when a fixed liquid water flux flows through the GDL. As part of the GDL porosity is filled with liquid water, the oxygen diffusion channels will decrease; hence Q_l should be as small as possible.

3. Solution procedure

Software was developed by the authors using Visual Basic to solve the equations. The values of the main variables chosen are shown as in Table 1.

The liquid saturation distribution in GDL was analyzed based on the assumptions that the water produced by the electrochemical reaction with a current density of 1 A cm^{-2} in the catalyst layer and the water transferred from the membrane with effective diffusion coefficient of 0.5 are all changed into liquid and discharged out of GDL, i.e.

$$q_{H_2O} = M_{H_2O}\frac{J}{2F}(1 + 2\alpha_e) \quad (9)$$

The liquid flux through differently structured GDLs is calculated based on an assumption that liquid saturation at the interfaces of catalyst layer/GDL and GDL/gas channel are 0.105 and 0.01, which correspond to the values calculated from Eqs. (6) and (9) for a GDL with 250 μm thickness, 0.5 uniform porosity and 140° contact angle.

In Eq. (5), $(K/\varepsilon)^{0.5}$ is equivalent to half of the pore radius, i.e. $r = 1/2(K/\varepsilon)^{0.5}$, which is an average pore radius of porous medium.

4. Results and discussions

4.1. A GDL with uniform porosity

For a GDL with uniform porosity, the liquid saturation distribution in GDL is calculated from Eq. (5), while the saturation on the interface of GDL/gas channel is chosen as 0.01 to determine the integral constant, the value of which is believed to depend on the channel condition, current density, etc. [17]. Fig. 1 shows the results. For Fig. 1(a, c, e), x-coordinate is the dimensionless thickness of GDL, and y-coordinate is the liquid water saturation. For Fig. 1(b, d, f), x-coordinate is the porosity, the contact angle and the thickness of GDL, respectively, and y-coordinate is the liquid water flux through the GDL. Fig. 1(a and b) shows the liquid water saturation and liquid water flux for different porosities of the GDLs with a fixed thickness of 250 μm and a fixed contact angle of 140°; Fig. 1(c and d) for different contact

Table 1
Values of the variables

Variable	Value
Current density (A cm^{-2})	1
Effective diffusion coefficient of membrane (m^2 s^{-1})	0.5
Kozeny constant	6
Faraday constant (C mol^{-1})	96487
Surface tension (N m^{-1})	0.0625
Viscosity of liquid water (m^2 s^{-1})	3.65×10^{-7}
Mole molecular weight of liquid (g mol^{-1})	18

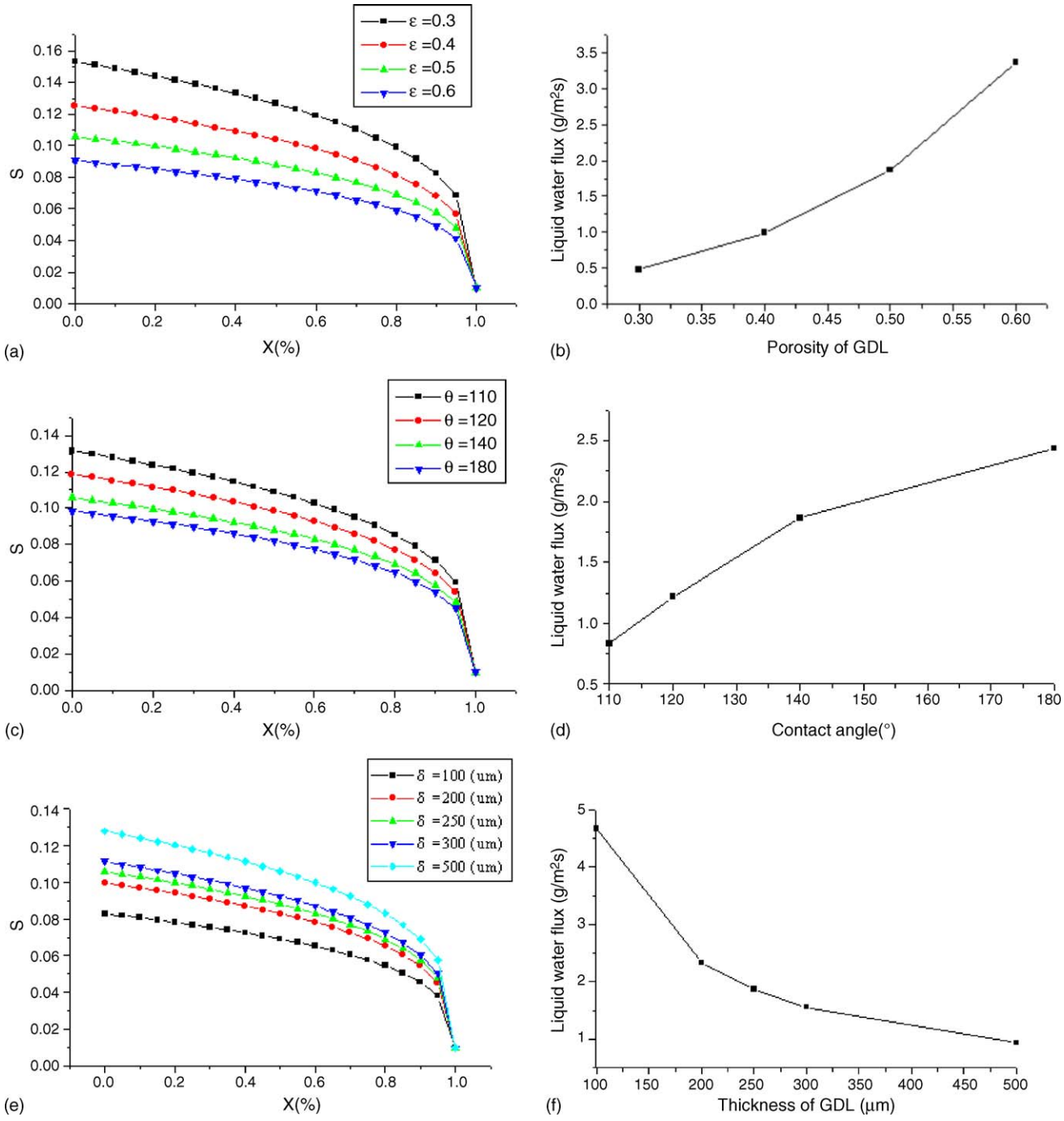

Fig. 1. Liquid saturation and liquid flux through GDL with uniform porosity. (a) Liquid saturation (thickness 250 μm, contact angle 140°), (b) liquid flux through GDL (thickness 250 μm, contact angle 140°), (c) liquid saturation (thickness 250 μm, porosity 0.5), (d) liquid flux through GDL (thickness 250 μm, porosity 0.5), (e) liquid saturation (porosity 0.5, contact angle 140°), (f) liquid flux through GDL (porosity 0.5, contact angle 140°).

angle GDLs with a fixed thickness of 250 μm and porosity of 0.5; Fig. 1(e and f) for different thickness of GDLs with a fixed porosity of 0.5 and fixed contact angle of 140°, respectively. It can be seen from these figures that the local liquid saturation in GDL decreases as the porosity and contact angle increase and the GDL thickness decreases. As the liquid water volume remaining in the GDL is an integral of the saturation in the GDL, its change rule is therefore the same as that of saturation for different GDL. Under a fixed liquid saturation difference between the interfaces of catalyst layer/GDL and the GDL/gas channel, the liquid water flux through the GDL increases with the increase of the porosity (Fig. 1(b)) and the contact angle (Fig. 1(d)), and with the decrease of the thickness, which implies that when the porosity and the contact angle of the GDL increase and its thickness decreases (Fig. 1(f)), fuel cells can produce more current while the MEAs are not be easily flooded.

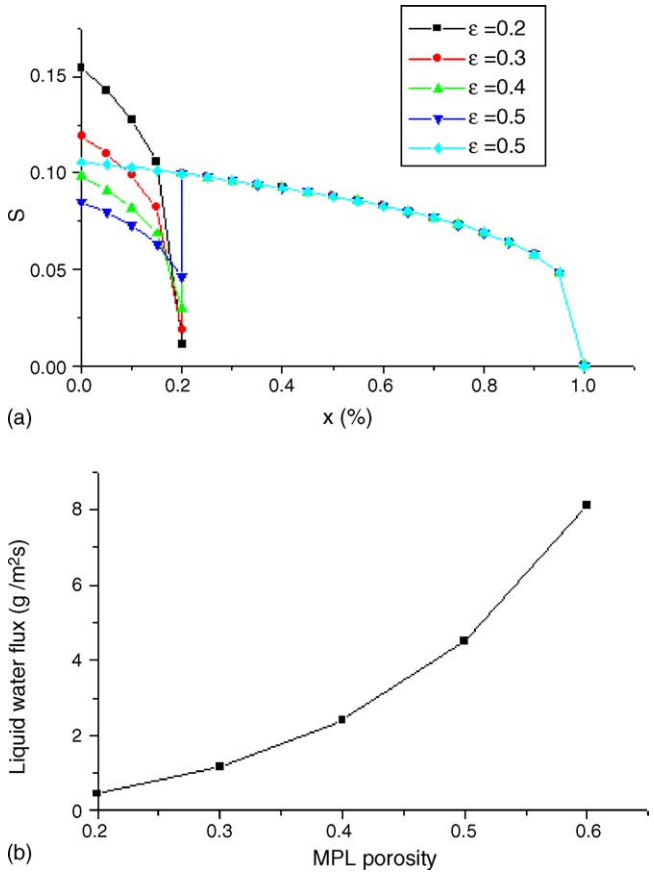

Fig. 2. Liquid saturation and liquid flux through GDL with different porosity of MPL.

4.2. A GDL with an MPL

In order to increase the liquid flux of the GDL and to reduce MEA flooding, many PEMFCs are inserted with MPLs between the catalyst and the normal GDL. The saturation distribution in the MPL is also governed by Eq. (5), but some material or structure properties, such as porosity, particle diameter, etc. are changed. The capillary pressure at the interface of MPL/GDL is continuous while the saturation has a sudden change, thus the saturation distribution in the GDL + MPL is different from a single GDL, and the liquid water volume remaining in the GDL and the flux through the GDL are improved.

4.2.1. A GDL with a different porosity for the MPL

Fig. 2(a and b) shows the liquid saturation and liquid flux through a GDL with a different porosity MPL. For Fig. 2(a), x-coordinate is the dimensionless thickness of GDL + MPL, and y-coordinate is the liquid saturation. For Fig. 2(b), x-coordinate is the porosity of MPL, and y-coordinate is the liquid water flux through GDL. The thickness of the GDL and the MPL are 200 μm and 50 μm while diameters of the fibers or particles are 2 μm and 1 μm, respectively, both contact angles are 140°, porosity of GDL is 0.5 and porosity of MPL is chosen as 0.2, 0.3, 0.4 and 0.5. Therefore $x = 0$, 0.2 and 1 are the interfaces of catalyst layer/MPL, MPL/GDL and GDL/gas channels respectively. The green continuous curve is for a single GDL of porosity 0.5. It can be seen from Fig. 2(a) that the saturation at the interface of catalyst layer/MPL decreases with the increase of the MPL porosity; when porosity of MPL is 0.4 and 0.5, the saturation value at this interface is less than that of a single GDL, which means there is more porosity remaining for oxygen diffusion. Under a fixed liquid saturation difference between the interfaces of catalyst layer/MPL and GDL/gas channel, the liquid water flux through GDL (or through MPL) increases with the increase of MPL porosity (Fig. 2(b)); when porosity of MPL is 0.5 and 0.4, the liquid water flux is 4.505×10^{-3} and 2.405×10^{-3} (kg s^{-1} m^{-2}), which increases by 141.6% and 29% compared with 1.865×10^{-3} kg s^{-1} m^{-2} of a single GDL. Therefore in these two cases, the fuel cells can produce more current while the MEAs are not easily flooded.

4.2.2. A GDL with different thickness of MPL

Fig. 3(a and b) shows the liquid saturation in GDL with different thickness of MPL, while the porosity of the MPL is 0.5 and 0.3, respectively. Fig. 3(c) is the liquid flux through the GDL or MPL. The total thickness of the GDL and MPL is fixed as 250 μm, but divided as 200 μm and 50 μm, 180 μm and 70 μm, 160 μm and 90 μm. The diameters of the fibers or particles are 2 μm and 1 μm, respectively, both contact angles are 140°, and porosity of GDL is 0.5. The Figures show that the saturation at the interface of catalyst layer/MPL decreases with the decrease of the MPL thickness. When the porosity of the MPL is 0.5, under all different MPL thicknesses, the saturation value at this interface is less than that of a single GDL (Fig. 3(b)), which is beneficial to oxygen diffusion. Under a fixed liquid saturation difference between the interfaces of catalyst layer/MPL and GDL/gas channel, the liquid water flux through GDL (or through MPL) increases with the decrease of MPL thickness (Fig. 3(c)); the liquid water flux of three MPL thickness increases by 141.6%, 73.2% and 34.3% when compared with 1.865×10^{-3} kg s^{-1} m^{-2} of a single GDL. When the porosity of MPL is 0.3, the saturation value at the interface of catalyst later/MPL is always larger than that of a single GDL (Fig. 3(b)); meanwhile the liquid water flux is 1.16×10^{-3} (kg s^{-1} m^{-2}), 0.83×10^{-3} (kg s^{-1} m^{-2}) and 0.645×10^{-3} (kg s^{-1} m^{-2}), decreasing by 37.8%, 55.5% and 65.4% compared with that of a single GDL (Fig. 3(b)). It is clear that this kind of MPL is beneficial neither to the oxygen diffusion nor to liquid water drainage.

Therefore when an MPL is placed between the catalyst layer and the GDL, the liquid saturation is redistributed across the MPL and GDL; the saturation at the interface of the catalyst layer/MPL decreases with the increase of MPL porosity and with decrease of MPL thickness; the liquid water flux through the GDL increases with increase of MPL porosity and decrease of MPL thickness. A case of a MPL with a 3 μm thickness was also computed. The result shows that for a 0.5 porosity MPL, the saturation at the interface is quite low, while the liquid flux is almost 10 times larger than that of MPL with a 50 μm thickness (Fig. 3(d)). According to such a rule, it can be seen that the thinner the MPL, the better its performance is. It should be noted, however, that in an extreme condition when the thickness

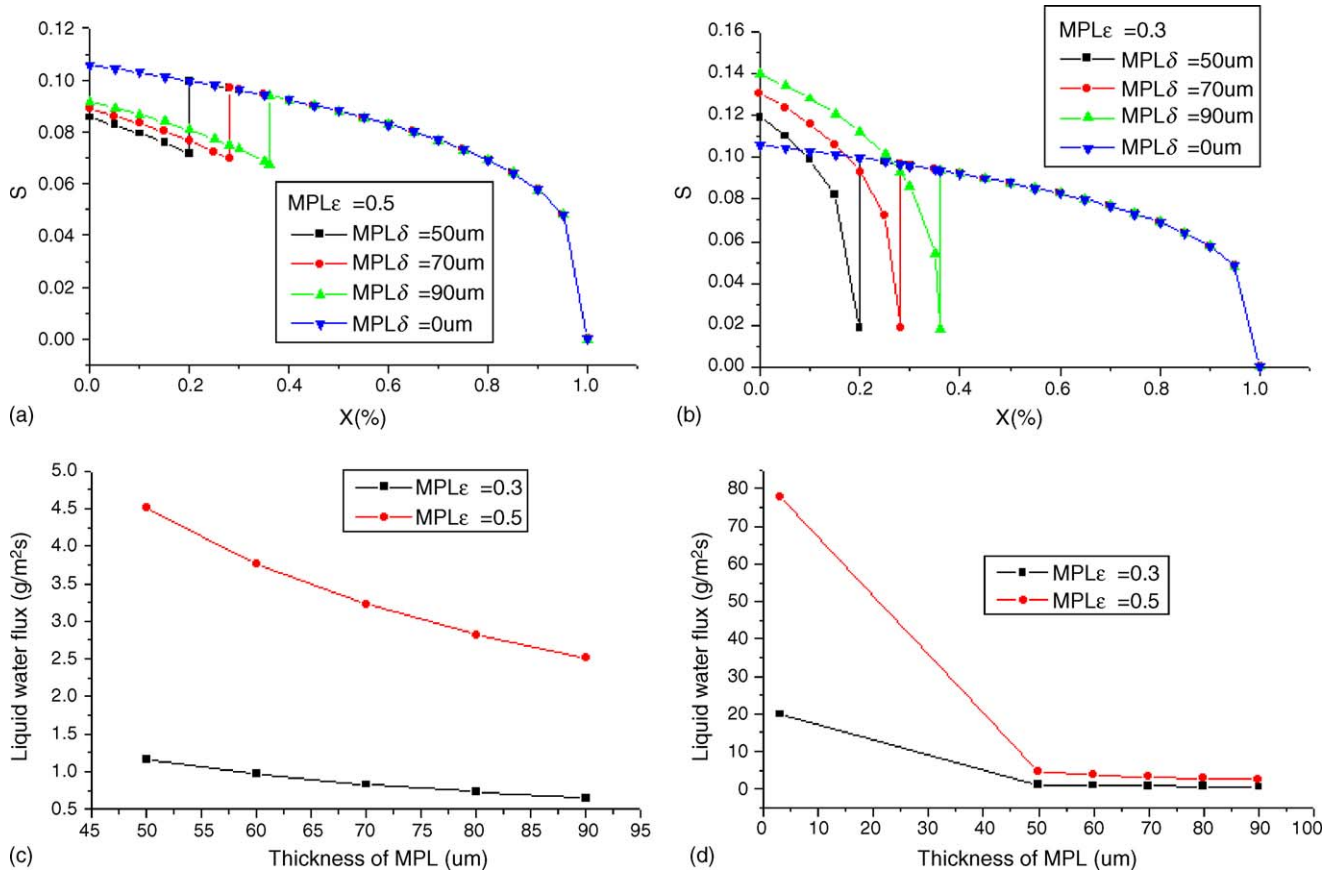

Fig. 3. Liquid saturation (a and b) and liquid flux (c and d) through GDL with different thickness of MPL. (a) $\varepsilon = 0.5$; (b) $\varepsilon = 0.3$.

decreases to zero, the GDL + MPL changes into a single GDL, and both the liquid saturation distribution and the liquid flux are not as good as that of the GDL + MPL. Possibly this is caused by some interfacial action.

4.3. A GDL with a gradient in porosity

When an MPL is inserted between the catalyst layer and the GDL, the saturation distribution is changed, hence the liquid flux through the GDL or MPL is improved and MEA flooding alleviated. The basic reason is that the capillary pressure difference driving the liquid out of the GDL increases. It is speculated that if a GDL with a gradient porosity is used, the performance should be optimized. In the following computed cases, the thickness, contact angle and fiber diameter of GDL are fixed as 250 μm, 140° and 2 μm, respectively.

4.3.1. A GDL porosity varying along the thickness following a linear rule

Suppose the porosity varies along the thickness following the linear rules, i.e. $\varepsilon = a_1 x + a_0$, the governing equation is Eq. (6). Values of a_1 and a_0 should ensure that ε is between 0 and 1. a_1 chosen as 0.2, 0.3 and 0.4, a_0 as 0.2, 0.3, 0.4 and 0.5, totaling 12 cases, the average porosity changes from 0.3 to 0.7. Fig. 4 shows part of the result. Fig. 4(a) is about the liquid volume remaining in GDL versus average porosity; Fig. 4(b) is about the liquid flux through GDL versus average porosity. It can be seen that when the gradient of the porosity a_1 is fixed, the average porosity increases with the increase of a_0, and the liquid volume remaining in the GDL and the liquid flux through GDL increases with increase of average porosity, which is the same as that of a single GDL ($a_1 = 0$); when a_0 is a constant, the average porosity increases with the increase of a_1. With the increase of average porosity the liquid volume remaining in GDL decreases but the liquid flux through GDL increases. Comparing $\varepsilon = 0.2x + 0.4$ and $\varepsilon = 0.4x + 0.3$, whose average porosity is 0.5, with a single GDL with porosity $\varepsilon = 0.5$, the liquid volume remaining of the former type GDLs decreases by 23.73%, 35.42%, while liquid flux increases by 152.82%, 171.85%, respectively. Such a result is caused by the saturation and the porosity variation along the thickness of GDL. If considering the oxygen flux transported to the electrochemical reaction in the catalyst layer, the electric conductivity etc., a GDL with $0.4x + 0.4$ porosity should be optimal. Its average porosity is 0.6, and liquid water remaining in GDL and flux through GDL are 7.39412×10^{-6} kg and 9.365×10^{-3} kg s^{-1} m^{-2}, respectively.

4.3.2. A GDL porosity varying along the thickness following a parabolic rule

Suppose the porosity varies along the thickness following a parabolic rule, i.e. $\varepsilon = a_2 x^2 + a_1 x + a_0$, the governing equation is Eq. (7). Values of a_2, a_1 and a_0 should ensure that ε is between 0

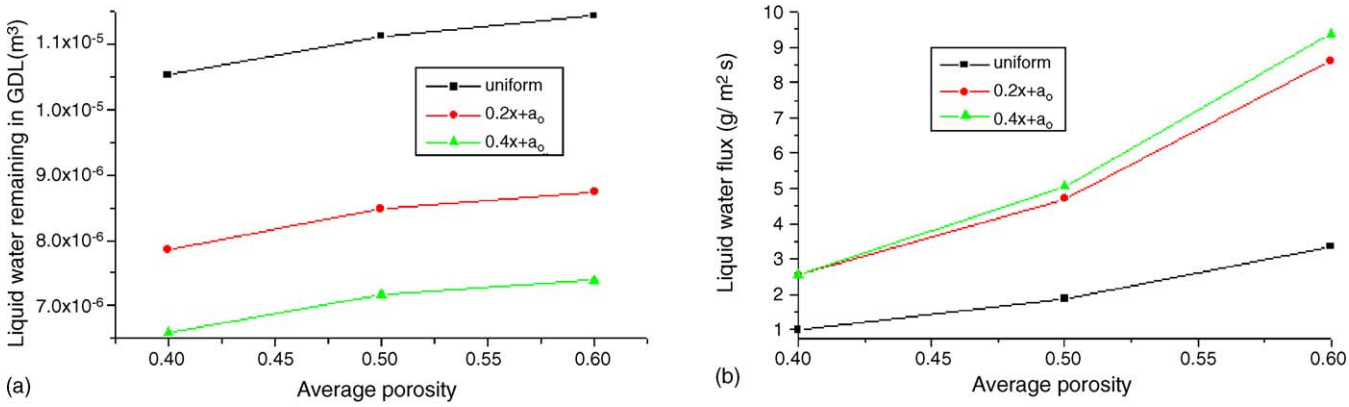

Fig. 4. Linear porosity of GDL. (a) Liquid water remaining in GDL; (b) liquid flux through GDL.

and 1. a_2 chosen as 0.1 and 0.2, a_1 as 0.1, 0.2 and 0.3, a_0 as 0.2, 0.3 and 0.4, totalling 7 cases. The average porosity changes from 0.42 to 0.62. The results show that the liquid volume remaining in GDL and the liquid flux through GDL increase with increase of average porosity, which is the same as that of a single GDL. Considering the difficulties to prepare a GDL with a parabola gradient porosity, it has no advantage over the linear type of GDL porosity.

4.4. Comparison of GDLs with the same average porosity but with a different porosity structure

For GDLs with the same average porosity but different porosity structures, a comparison is made to determine the ones having optimal performance. Fig. 5(a) shows the different porosity structures, including a single GDL, two linear GDLs and a parabola GDL; in each case the average porosity is 0.6. Fig. 5(b

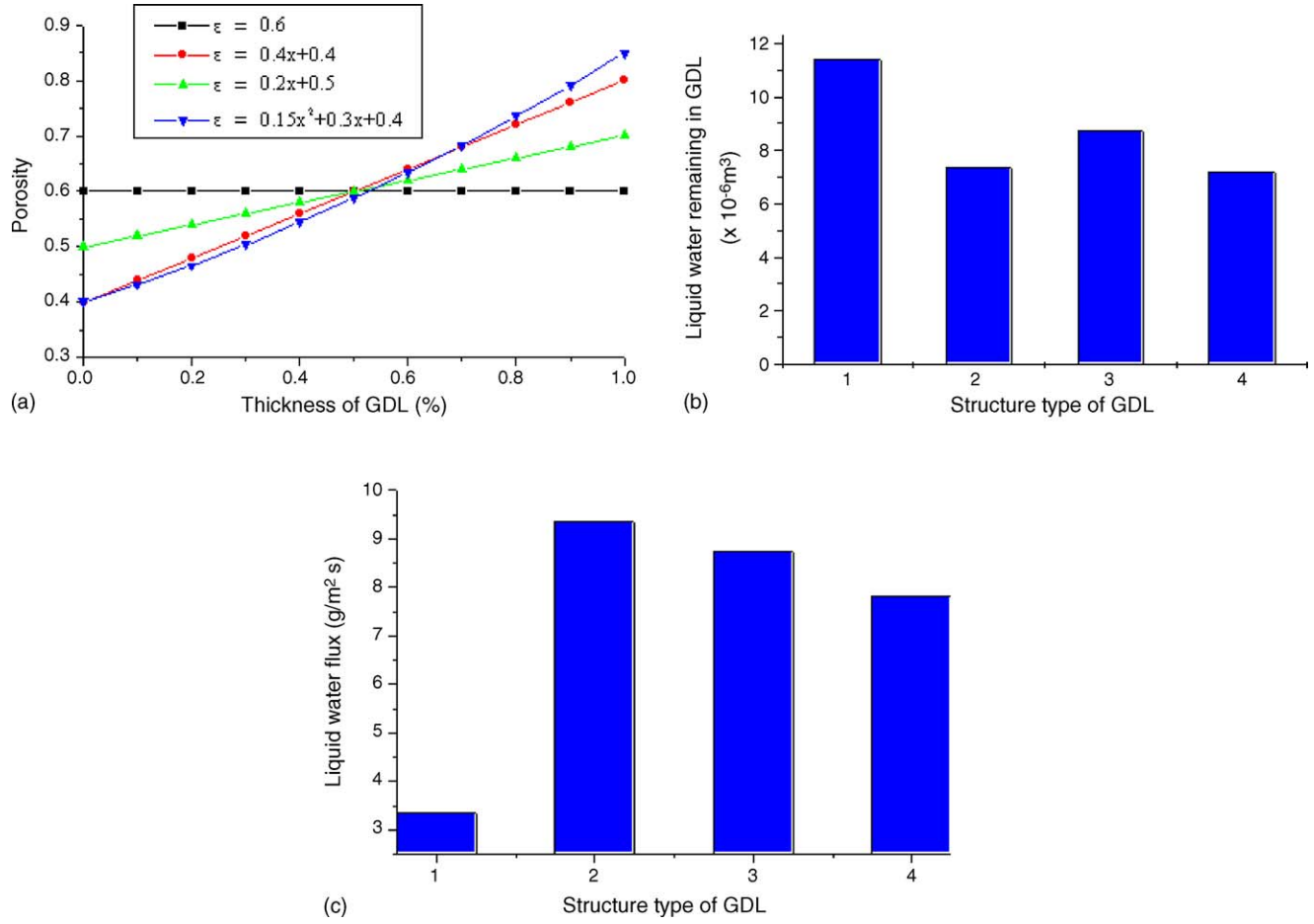

Fig. 5. Comparison of: (a) GDLs with same average porosity but different porosity structure; (b) liquid water remaining in GDL; (c) liquid flux through GDL.

and c) shows the liquid volume remaining in GDLs and liquid flux through GDLs, respectively, and x-coordinate of the figures is the structure type. As for the liquid volume remaining in GDL, the parabola type is the least, and the linear type $0.4x + 0.4$ is the second better, which decreases by 3.08% and 0.79% when compared with the single GDL (Fig. 5(b)). As for the liquid flux through a GDL, the linear type $0.4x + 0.4$ is the best, and the parabola type is the next, which decreases by 16.65% compared with the former (Fig. 5(c)). Considering that it is more difficult to prepare a parabolic GDL than to prepare a linear one, $0.4x + 0.4$ should be the best among the computed cases.

5. Conclusions

(1) Under steady conditions, the liquid water flux through the GDL increases with increase of contact angle and porosity, and with the decrease of the GDL thickness.
(2) When an MPL is placed between the catalyst layer and the GDL, the liquid saturation is redistributed across the MPL and GDL, thus improving the liquid water draining performance; the liquid water flux through the GDL increases with increase of MPL porosity and decrease of MPL thickness; keeping the total thickness of the GDL and MPL the same, when the MPL is thinned to 3 μm, the liquid water flux increases considerably, i.e. flooding of the MEA will not be easy to happen.
(3) A GDL with a gradient in porosity is more favorable for liquid water removal from the catalyst layer to the gas channel; for the GDLs with the same equivalent porosity, the larger the gradient is, the more easily the liquid water is removed; of the computed cases, a GDL with a linear porosity $0.4x + 0.4$ is the best.

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