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On the permeability of gas diffusion media used in PEM fuel cells

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Abstract

The current work investigates laminar channel flow in square serpentine channels mounted on a permeable gas diffusion media (GDM) as in the case of a PEM fuel cell. Computational fluid dynamics is used to determine the importance of convective transport as a function of GDM permeability. Both isotropic and orthotropic permeabilities are considered and it is found that the in-plane permeability is the parameter of importance. For typical flow field dimensions and fuel cell operating conditions, it is found that convective transport will occur in the GDM when the permeability exceeds $1 \times 10^{-13}\,\mathrm{m}^2$. This value is lower than that reported experimentally, meaning that convection is significant in fuel cell GDM. Currently, virtually all fuel cell modelling work either implicitly or explicitly ignores this effect. Finally, it is noted that the in-plane permeability is the relevant parameter in these configurations. This is particularly relevant in the case of GDM with micro-porous layers which decrease the through-plane permeability by several orders of magnitude but do not change in the in-plane permeability.

Keywords: PEM fuel cell; Gas diffusion media; In-plane permeability; Through-plane permeability; Serpentine channels; Computational fluid dynamics

1. Introduction

The gas diffusion media (GDM) in a polymer electrolyte fuel cell is a criticial component which is located between the flow field plate and the catalyst. This media must transport reactant gases from the channel to the catalyst, transport electrons from the bipolar plate to the catalyst, remove reaction products (liquid and gas) from the catalyst and transport heat from the catalyst to the cooling channels in the bipolar plate. As such, the GDM is a critical link between the electrochemical reaction and the system engineering needed to construct a fuel cell. A good introduction to the function and characterization of GDMs is given by Mathias et al. [1].

In a pressure driven channel flow, the pressure drops monotonically in the direction of flow in order to balance the wall shear. In a laminar flow, this pressure gradient required to drive the flow is very small, but given a long enough channel, it can become quite significant. Serpentine flow fields result in very long channel lengths in relatively small areas, and as a result, the largest pressure difference across a section of GDM occurs between any two unconnected bends (as at the bottom of Fig. 1). When a GDM is substituted for one wall of the channel, these pressure differences have the potential to drive fluid through the GDM from one channel to the next, and in the linear regime, this behaviour is described by Darcy's law

$$v = \frac{k}{\mu}\frac{\Delta P}{l} \tag{1}$$

where v is the velocity, k the permeability, ΔP the pressure drop, l the thickness of the material and μ is the fluid viscosity. It should be clear that the pressure differences developed in serpentine channels results in an in-plane pressure gradient through the GDM, as opposed to a through-plane pressure gradient.

Values of GDM permeability vary greatly in the literature, particularly those reported in the modelling literature. Experimental determination of through-plane permeabilities are reported by Mathias et al. [1] to be of the order of 5–10 $\times 10^{-12}\,\mathrm{m}^2$. Williams et al. [2] report values of 0.8–3.1 $\times 10^{-11}\,\mathrm{m}^2$ for bare paper and showed that the addi-

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tion of micro-porous layers decreases this value by approximately two orders of magnitude. Prasanna et al. [3] report 1–8 × 10^{-11} m^2 and show a significant decrease as the teflon loading is increased. In modelling papers, the range is larger, spanning nine orders of magnitude, from 4.73×10^{-19} m^2 [4] to 1×2^{-10} m^2 [5], with many papers in the middle of this range [6,7].

Recently, some authors [8,9] have considered the flow in serpentine channels in the absence of gas diffusion media and while this is one limit, it is very likely that the GDM and channel flows are strongly coupled in a way that has a large impact on fuel cell performance. To date, this issue has only been explored in the patent literature [10].

The objective of this paper is to investigate the effect of GDM through-plane and in-plane permeability on the flow and pressure drop in serpentine channel/GDM systems, and to discuss the importance of flow field matching with GDM characteristics. Very little work to date has included the effect of GDM anisotropy, and accordingly, a further objective is to study the effect of anisotropic GDM permeability.

2. Computational model

The steady incompressible continuity equation and Navier–Stokes equations for momentum were solved using the commercial code Fluent 6.1, in serpentine square channel mounted on a permeable gas diffusion media as in Fig. 1. A grid refinement study was carried out and it was found that a grid of 214,000 nodes (20 × 20 in the square cross-section) resulted in a change of less than 0.5% in pressure drop compared to a much finer grid.

The GDM thickness was held constant at 250 μm, the channel was square and 1 mm, and two channel lengths were explored: $L = 40$ mm and 80 mm. The flow rate was varied corresponding to Reynolds numbers between 100 and 400. The lower value corresponds to a reasonable operating point at moderate current density, and the higher value is approaching the limit where the flow in serpentine channels in the absence of a GDM becomes unsteady [9].

Following virtually all CFD modelling of fuel cells, source terms consistent with Darcy's law are applied to the momentum equation in the GDM. The flow is assumed periodic across the domain represented in Fig. 1, and a constant pressure gradient is applied in the *x*-direction in order to achieve the desired Reynolds number. This approach was verified against a model consisting of five serpentine channels, and it was confirmed that at $Re = 100$ the periodic solution is completely developed by the third period.

3. Results

3.1. Isotropic permeability

In the first instance, the GDM was considered to be an isotropic material. Fig. 2 shows the pressure drop across the periodic domain as a function of GDM permeability. For low permeability, the pressure drop is indistinguishable from the no GDM asymptote, but as the permeability increases above 10^{-13} m^2, the channel pressure drop begins to decrease. The pressure difference continues to drop sharply until approximately 5×10^{-10} m^2, where another asymptote is approached. The decrease in pressure required to drive the flow can be explained with reference to Fig. 3, which shows the percentage of flow which passes through the channel area at the periodic boundaries. The balance of the flow remains in the GDM. As the permeability increases, less and less flow

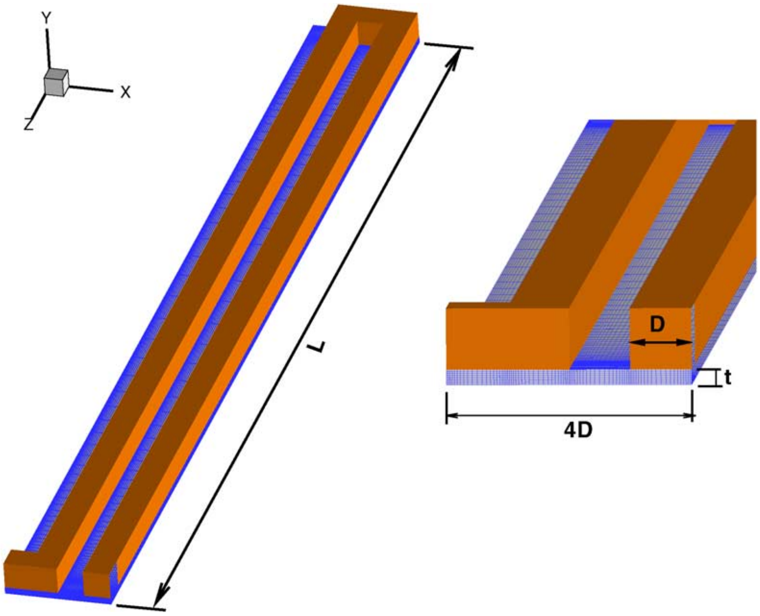

Fig. 1. Periodic channel geometry.

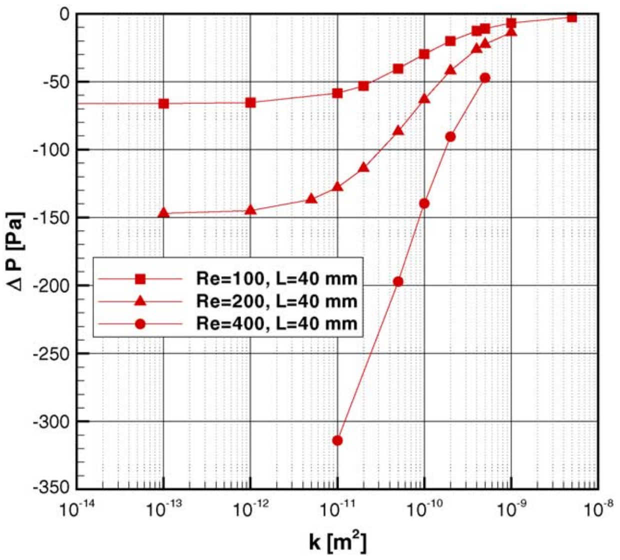

Fig. 2. Channel pressure drop as a function of permeability and *Re*.

passes through the channel, and more flow remains in the GDM where there is a much larger cross-section for flow. The net result is that the overall pressure drop is dramatically lower at low permeability.

As the Reynolds number increases, the channel pressure drop increases, and it appears that the onset of convective flow in the GDM occurs at lower permeabilities. It is important to note that virtually all experimental data for GDM permeability is greater than the lower threshold value of 10^{-13} m^2. This suggests that convective flows in the GDM are important even at the relatively low flow rate represented by $Re = 100$. As the flow rate increases, an increasing amount of the flow is transported through the GDM, and the importance of convective transport in the GDM is increased.

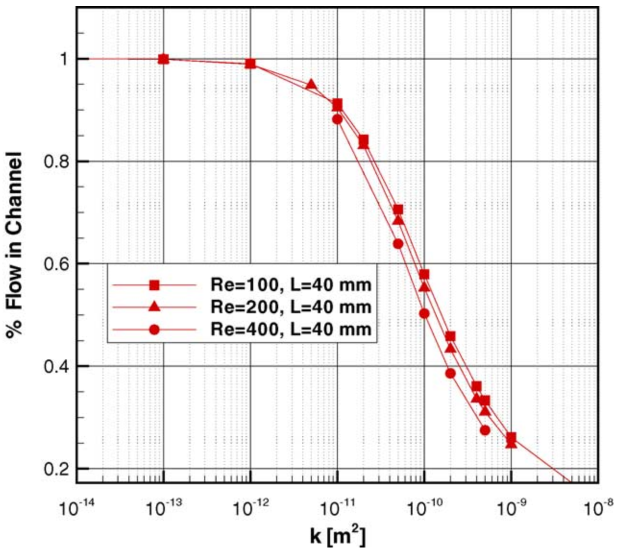

Fig. 3. Percentage of total flow rate which passes through the channel at the periodic boundary as a function of *Re*. The remainder of the flow passes through the GDM.

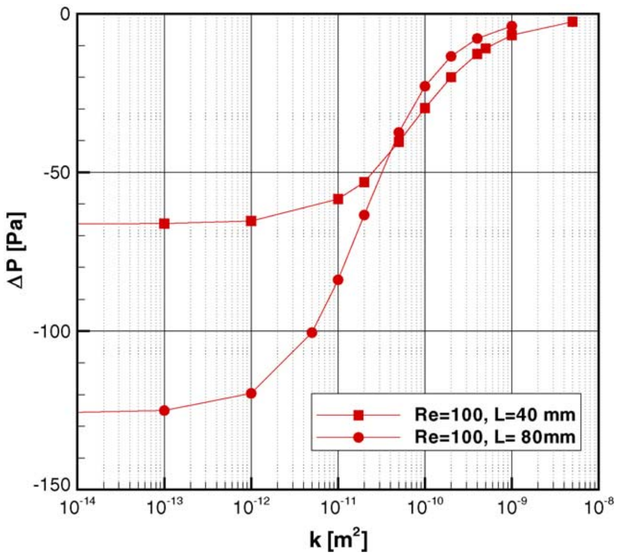

Fig. 4. Channel pressure drop as a function of channel length ($Re = 100$).

3.2. The effect of channel length

As the flow channels get longer, the maximum pressure difference between channels increases, resulting in larger driving forces for convective flows through the GDM. Fig. 4 compares the pressure drop across the domain as a function of GDM permeability for $L = 40$ mm and 80 mm.

As the channel length increases, the pressure drop across the domain increases substantially, and the onset of convective transport occurs at lower permeability. The pressure drop curve for the longer channel crosses the curve for the shorter channel as the permeability increases because significantly more flow passes through the GDM in every case (Fig. 5). This flow in the GDM passes through a cross-sectional area proportional to L, and hence the average velocity is much lower in the case of a longer channel. The net effect is an even lower pressure drop as the permeability increases. This effect, which is perhaps counter intuitive, is solely a result of the interaction between the flow channel and the GDM, and hence is not considered when the flow channel is examined in isolation.

3.3. Orthotropic permeability

Even without the addition of micro-porous layers, discussed in the next section, there is a strong possibility that the permeability of GDMs is not isotropic. These materials are made either by pressing chopped carbon fibres into a paper, or by weaving bundles of fibres into a cloth. In either case, the structure is very clearly different in the through-plane and in-plane directions. It is important then to consider this effect on the transport in fuel cells.

An orthotropic media is a media which has the same properties in a plane, and different properties perpendicular to that plane. This is quite a reasonable approximation in a fuel

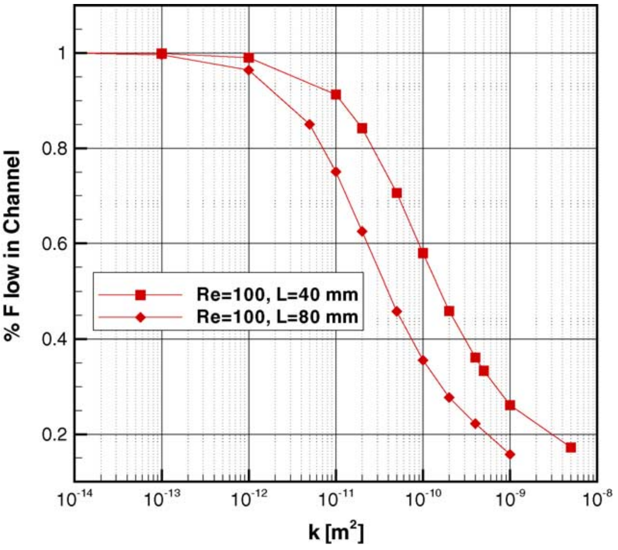

Fig. 5. Percentage of total flow rate which passes through the channel at the periodic boundary for two different channel lengths. The remainder of the flow passes through the GDM.

cell GDM, considering that how the materials are made. The through-plane permeability is given by $k_\perp$ while the in-plane permeability is given by $k_=$.

Additional computations were carried out to explore varying the through-plane permeability at constant in-plane permeability. The effect of through-plane conductivity variations is explored at both $k_= = 1 \times 10^{-11}\,\mathrm{m}^2$ and $1\ \times 10^{-10}\,\mathrm{m}^2$. The resulting channel pressure drops in a $L = 40$ mm channel at $Re = 100$ are given in Fig. 6, and it is clear that in-plane permeability is the dominant parameter, especially at the lower permeability which is typical of reported experimental data.

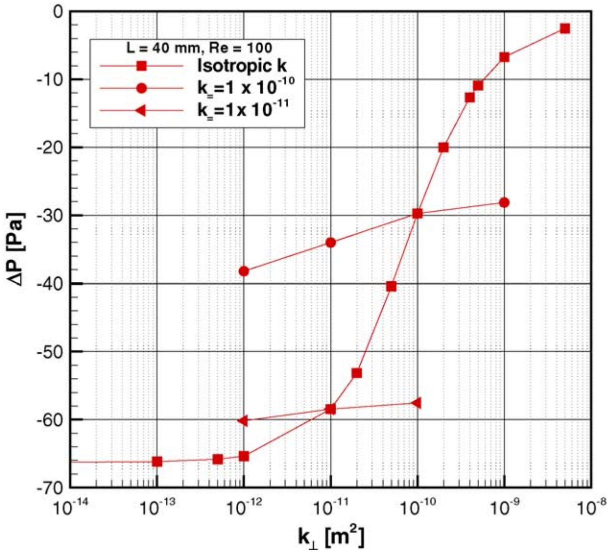

Fig. 6. The effect of orthotropic permeability on channel pressure drop ($Re = 100$ and $L = 40$ mm).

The slight dependance on through-plane permeability is due to the fact that gases must first be transported a short distance in the through-plane direction before they can move in the in-plane direction. This applies directly to a homogeneous material with differing values of permeability in each direction, but would differ substantially if the through-plane permeability was drastically lower due to the addition of a nearly impermeable layer at the catalyst GDM interface. This scenario corresponds to addition of a micro-porous layer.

3.4. Implications for GDM with micro-porous layers

Many GDM employ a thin micro-porous layer on the face adjacent to the membrane as a potential means of improving water removal and electronic contact, and thus increasing cell performance. A typical such structure is shown in Fig. 7. The micro-porous layer is sufficiently dense that regular pores are not evident even at five times the scale shown here. Transport is evidently through the cracks that are clearly shown in Fig. 7(a). Both the flip side view (Fig. 7(b)) and the edge view (Fig. 7(c)) clearly indicate that the permeability of bulk of the material remains unaltered. The addition of a thin micro-porous layer amounts to adding a very thin layer with a significantly decreased permeability in series with the raw GDM. Clearly, the through-plane permeability, $k_\perp$, of the combined system will be significantly reduced, but the in-plane permeability, $k_=$, of the bulk material, will be largely unaltered.

The results of the preceding section show that the through-plane permeability is significantly less sensitive than the in-plane permeability, yet most measurements are taken of the through-plane permeability as it is experimentally more convenient. Unfortunately, a micro-porous layer causes the permeability to drop by up to four orders of magnitude [2] compared to the bare GDM. While the bare materials have a permeability that places them clearly in the regime where convective transport is significant, the permeability decrease in the case of a micro-porous layer would seem to indicate that convective transport would no longer be important. This is in fact not the case, since the micro-porous layer only affects the permeability locally (it is in series with the bare material). It will have absolutely no effect on the transport mechanisms discussed above, as the pressure gradients are in the plane of the GDM and the micro-porous layer is never placed between the channel and the GDM. Further, if the material is assumed to be isotropic but non-homogeneous (i.e. it has a different permeability in the bare paper and in the micro-porous layer), then the through-plane permeability of the total system is completely irrelevant, and the only parameter of interest is the bare GDM permeability.

This argument is supported by recent experimental work looking at the role of convection on the limiting current for a bare paper and for two different micro-porous layers on the same paper [11]. In this work, there is a nearly constant offset between the different cases as the cathode flow rate (and hence convective transport in the the GDM) is increased. This strongly suggests that the convective transport through the identical substrates is very nearly the same in each case, and that the variation in limiting current is due to different transport resistances in the micro-porous layers themselves.

3.5. Implications for fuel cell modelling

There is a very substantial body of literature in PEM fuel cell modelling, and the issue of convective transport in the GDM has been either explicitly or implicitly neglected in almost every case. A notable exception is in the case of interdigitated flow fields which use a geometry that forces convective transport in the GDM (i.e. [12]). Convective transport is explicitly neglected by not solving for momentum transport [13] and is implicitly neglected by setting in two ways: either by setting the permeability below $1 \times 10^{-13}\,\mathrm{m}^2$ [14,15], or by modelling a short straight channel section of a fuel cell [16,7].

Unduly neglecting convective transport impacts the results in important ways. In the first case, the gas concentrations in the GDM will be incorrect since convective transport serves to bring more oxygen rich gases from the channel into the GDM, and to draw water vapour (and liquid) out into the channel. Additionally, this transport is from an upstream channel through the GDM towards a downstream channel which on average has a lower oxygen concentration due to the electrochemical reaction. Not only does this have the potential to radically alter species concentrations, but it also breaks the symmetry which is the usual boundary condition when considering a short straight section.

Also, as shown above, the pressure drop in the channels is substantially over-predicted when convective transport is neglected. While the actual pressure drops are small in practice, this has the potential to affect the pressure difference across the electrolyte as well as the reaction rates which are functions of pressure.

4. Conclusions

The effect of GDM permeability on convective transport has been explored. It was found that convection effects become important beyond a threshold permeability value of approximately $1 \times 10^{-13}\,\mathrm{m}^2$. This is particularly significant since virtually all experimental measurements of the permeability of bare GDM material are greater than this value. The GDM and the flow channel are an integrated system, and must be considered together.

The effect of orthotropic permeability was also explored, and it was determined that the in-plane permeability is the dominant parameter, which is not surprising since serpentine channels result in pressure drops which in the plane of the GDM.

The addition of a micro-porous layer decreases the permeability of the GDM by several orders of magnitude, and this decrease is not relevant to convective transport. It is essential

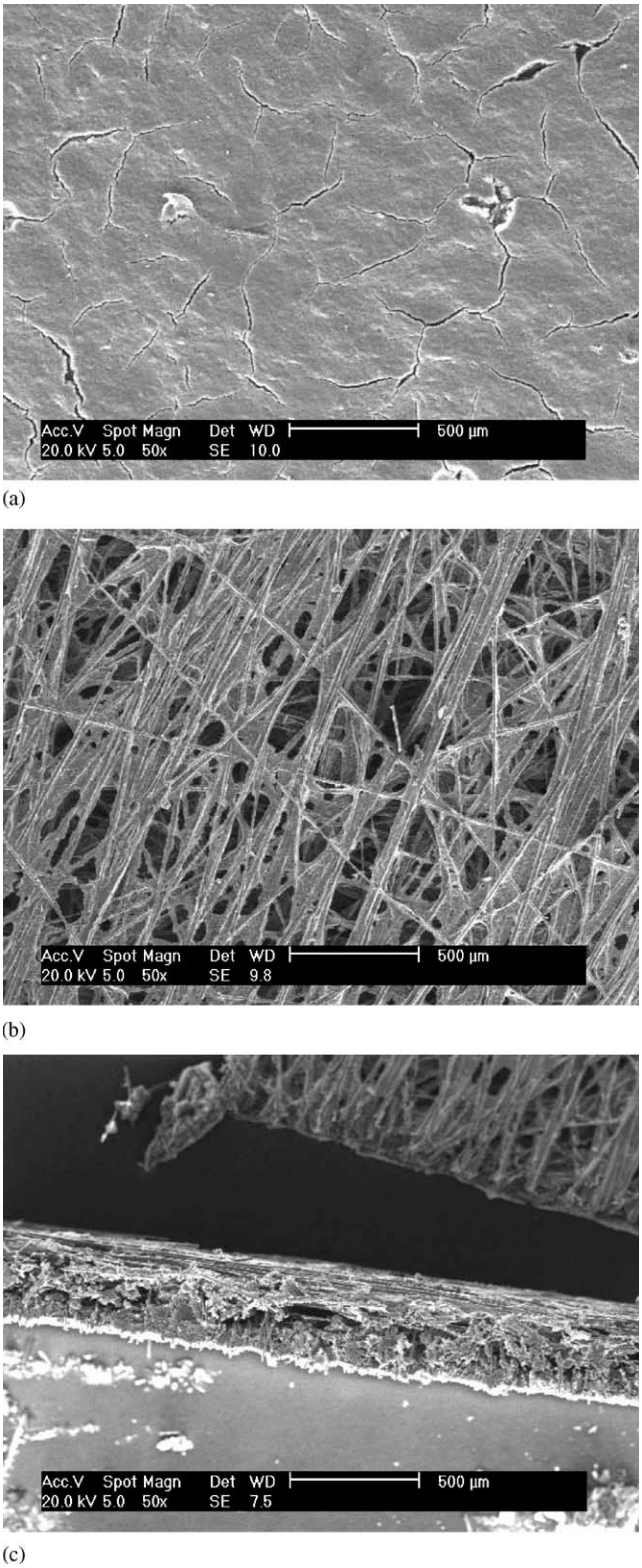

Fig. 7. A typical GDM with a micro-porous layer. (a) Face with the micro-porous layer, (b) opposite face (no micro-porous layer) and (c) edge view: the thin micro-porous layer is the bright line on the bottom face.

that either the in-plane permeability, or the permeability of the bare GDM be used in this case.

Finally, virtually all modelling work has either implicitly or explicitly neglected the effect of convection in the GDM, and this is not justified.

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