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Effective permeability of gas diffusion layer in proton exchange membrane fuel cells

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ABSTRACT

In gas diffusion layers (GDLs) of proton exchange membrane fuel cells (PEMFCs), effective permeability is a key parameter to be determined and engineered. In this study, through-plane (TP) and in-plane (IP) flow behaviors of GDLs are investigated analytically based on a scaling estimate method. The TP permeability and IP permeability of unidirectional fibers are determined first, based on that the minimum distance and the inscribed radius between fibers are adopted as the characteristic lengths for normal and parallel flows, respectively. The permeabilities of two-dimensional (2D) and three-dimensional (3D) GDLs are estimated by a proper mixture of the local TP and IP permeabilities of fiber alignments. The mechanistic model agrees closely with experimental and numerical results over a wide porosity range. With the new model, the influences of porosity and fiber orientation on flow behaviors are analyzed.

1. Introduction

Proton exchange membrane fuel cell (PEMFC) has received much attention as one of the leading candidates for the power sources of stationary, mobile, and portable devices [1–3]. The gas diffusion layer (GDL) of PEMFCs is a fibrous medium for transferring reactants and products. For example, the produced water in the cathode side should be removed effectively to avoid condensing and blocking the porous GDLs [4]. Therefore, the effective permeability of water and vapor strongly affects the performance of the PEMFC [2,4].

Darcy's law is the best-known equation to characterize the laminar flow through a porous medium like the GDL, which linearly relates the volume-averaged velocity with the pressure gradient via the effective permeability [5]:

$$\langle u \rangle = -\frac{K}{\mu}\nabla p, \tag{1}$$

where K is the permeability tensor of the medium, μ is the fluid viscosity, ∇p is the pressure gradient and $\langle u \rangle$ is the average velocity. The permeability tensor K lumps all complex interactions between fluid flows and fibers. Based on Darcy's law, the TP and IP permeabilities of commercial GDLs with varied compressive loads were experimentally measured by Gostick et al. [6] and Becker et al. [7].

Later, several compact models were reported to predict the Darcy permeability coefficient of GDLs. Among them, Kozeny–Carman (KC) equation is broadly used by considering the GDL as consisting of bundles of tortuous tubes [8]. Following the KC assumption, Tomadakis and Robertson [8] proposed the permeability model in terms of porosity and tortuosity, viz.,

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$$K/r^2 = \frac{\varepsilon^3}{8\tau(1-\varepsilon)^2}, \quad (2)$$

where ε is porosity, r is fiber radius, and τ is the tortuosity of the medium. Later, a fractal technique, which statistically quantifies the pore size distribution in GDLs, was applied to calculate the TP permeability of fibrous layers [9,10]. However, the applicability of above models is sometimes tarnished by the difficulty in accurately determining the value of tortuosity.

In another parallel approach, limited flow in a unit cell is analytically explored [11–13]. The unit cell, which is regarded as existing repeatedly throughout the unidirectional fiber arrays, is approximated to have the same permeability with the GDL system. With proper boundary conditions defined, accurate solutions are available by solving Stokes equation:

$$-\nabla p + \mu \nabla^2 u = 0. \quad (3)$$

The lubrication approximation, which considers the velocity normal to flow direction negligible, was widely used to determine permeability for fiber alignments at the relatively low porosity range [11–13]. Recently, Clague et al. [14] presented a simple formula of the TP permeability on the basis of the scaling estimate rule, which regards the minimum distance between fibers as the critical parameter upon the fact that the normal flow is mainly controlled by the narrow slots formed between fibers. Later, Sobera and Kleijn [15] modified the scaling estimate model by introducing the ratio between the minimum to total open area.

Most models on the permeability of more realistic two-dimensional (2D) or three-dimensional (3D) GDLs are conducted by computational simulations, because an exact determination of permeability is almost impossible in these complex fibrous structures [12]. After the fibrous structures were generated digitally from the tomographic imaging, transport equations were solved numerically by powerful solvers [16–18]. Based on the fitting of numerical results, Hao and Cheng [18] suggested an empirical formula to determine the permeability of GDLs coated with PTFE films, which are randomly distributed between fibers. Numerical simulations echo experimental measurements with varied constructed structures and fast calculation, but they cannot fully reveal the underlying relationship between the permeability and microstructures of the GDL. The pioneering attempt to theoretically model the 3D flow behaviors was conducted by Jackson and James [19], who argued that the permeability of the 3D fibrous system was equivalent to an unweighted mixture of local permeabilities of aligned fibers normal and parallel with the principal flow direction.

In this paper, we employ a modified scaling estimate method to model TP flows through fiber alignments, and extend this technique to IP flows for the first time. Based on different mixing laws, 2D and 3D models for the TP and IP permeabilities are proposed and then compared to experimental and numerical data. The remainder of the paper is split into three parts. In Section 2, the scaling estimate technique is modified to determine the permeability of unidirectional fiber arrays and mixing laws are then used to estimate the permeability of GDLs. In Section 3, we compare the calculated permeabilities with experimental and numerical results and analyze the affecting parameters. Finally, concluding remarks are stated in the last section.

2. Model generation

Unidirectional circular fibers with square (Fig. 1(a)) and hexagonal (Fig. 1(b)) packing arrangements are firstly studied. The lubrication theory was commonly used to characterize fluid flows in narrow gaps [11–14], which refer to the narrow open channels between fibers in Fig. 2. The velocity component in vertical direction (y-direction) is approximated negligible comparing with that in horizontal direction (x-direction) [13]. Stokes equation of Eq. (3) can be simplified as:

$$\frac{dp}{dx} - \mu \frac{d^2 u}{dy^2} = 0. \quad (4)$$

Clague et al. [14] selected the half distance $h_{\min}$ between cylindrical fibers as the characteristic length, over which the flow velocity varies dramatically. The scaling estimate is given by Ref. [14]:

$$\nabla p \sim \mu \frac{\langle u \rangle}{h_{\min}^2}, \quad (5)$$

where '$\sim$' means 'scale as', and ∇p is the pressure gradient against the total length of the cell.

Later, Sobera and Kleijn [15] argued that it is more proper choosing the actual velocity $u_0 = \langle u \rangle / \chi$ at the narrow slot as the characteristic velocity, where χ is the ratio between the minimum to the total frontal areas. However, this correlation agrees unsatisfactorily with experimental results in a wide range of porosities as shown in Section 3. We believe that this is a consequence of neglecting the effect of the width shape of the narrowest gap between fibers, where most flow resistances exist [14]. Indeed, it is more reasonable to scale the pressure difference with the flow velocity against a length difference instead of the total cell length, because the velocities are not uniform-like or even in the same magnitude along the flow direction. A more generalized local scaling estimate is therefore presented, viz.,

$$dp \sim \mu \frac{u(x)}{h^2(x)} dx, \quad (6)$$

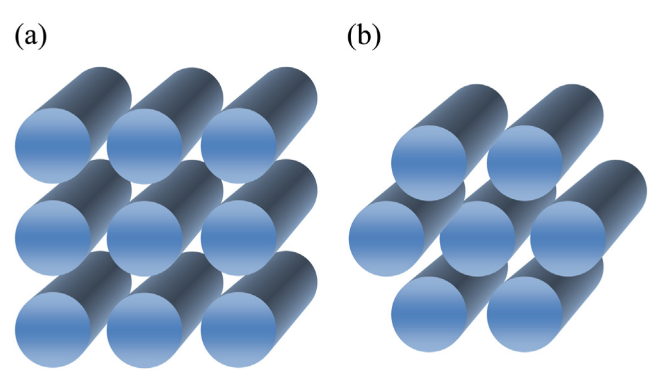

Fig. 1 – (a) Schematic of circular fibers in a square packing; (b) schematic of circular fibers in a hexagonal packing.