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Pore Structure Modeling of Flow in Gas Diffusion Layers of Proton Exchange Membrane Fuel Cells

The gas diffusion layer (GDL) in a proton exchange membrane (PEM) fuel cell has a porous structure with anisotropic and non-homogenous properties. The objective of this research is to develop a PEM fuel cell model where transport phenomena in the GDL are simulated based on GDL's pore structure. The GDL pore structure was obtained by using a scanning electron microscope (SEM). GDL's cross-section view instead of surface view was scanned under the SEM. The SEM image was then processed using an image processing tool to obtain a two-dimensional computational domain. This pore structure model was then coupled with an electrochemical model to predict the overall fuel cell performance. The transport phenomena in the GDL were simulated by solving the Navier-Stokes equation directly in the GDL pore structure. By comparing with the testing data, the fuel cell model predicted a reasonable fuel cell polarization curve. The pore structure model was further used to calculate the GDL permeability. The numerically predicted permeability was close to the value published in the literature. A future application of the current pore structure model is to predict GDL thermal and electric related properties. [DOI: 10.1115/1.4005613]

Introduction

The gas diffusion layer (GDL) plays an important role in maintaining proton exchange membrane fuel cell performance [1]. For example, the GDL provides mechanical support to the membrane, serves as the pathway for reactant gas and product water to flow, and conducts electrons and heat. To improve fuel cell performance, optimizing GDL properties such as porosity, permeability and polytetrafluoroethylene (PTFE) coating has attracted lots of research attention [2].

The GDL is usually made of highly porous carbon cloth or highly porous carbon paper. To simulate the flow in the porous GDL, the macro scale model such as Darcy's law is often used. The GDL properties, such as porosity and permeability, are usually the input parameters for those macro-scale equations. Both the porosity and permeability are highly anisotropic. They also vary because of the assembly compression [3]. Therefore, it is desirable to develop a realistic pore structure of the GDL where the Navier-Stokes equations can be solved directly without the GDL porosity and permeability.

The pore structure model has been widely used in the geology and medical research [4-9]. Until recently, a few efforts have been made in developing the model for the flow in the GDL. Nam and Kaviany [10] assumed that the fibers in the GDL were infinitely long and the fibers were overlapped to form a twodimensional screen. A solid GDL structure was constructed in such a way that the two-dimensional screens were continuously stacked and their positions were shifted by a random in-plane distance. Using the screen like structure for the GDL proposed by Nam and Kaviany [10], He et al. [11] predicted the GDL permeability. Gostick et al. [12] treated the GDL as a regular cubic network of pore bodies and pore throats, whose geometric parameters were calibrated with respect to experimental measurement. A similar approach was used by Wang et al. [13], Tabe et al. [14], Chapuis et al. [15] and Djilali [16]. Fluckiger et al. [17] filled GDL's fiber section with an idealized porous pattern to imitate its fiber and binder section, and left the rest of the space open to represent the void space. GDL's diffusivity was found anisotropic between the in-plane and through-plane directions. Park et al. [18] treated the GDL as a combination of fiber bundles and void space. Lattice Boltzmann method (LBM) was used to simulate the micro-scale flow in the porous electrode of a PEM fuel cell. The results showed that the GDL was not homogeneous and its permeability was strongly dependent on the fiber orientation. Doormaal and Pharoah [19] used the Monte Carlo method to generate an idealized GDL structure with fibers aligned in plane and angled between different layers. By changing the fibers filled into the geometry, GDL's porosity was varied and its influence on GDL permeability was presented. Recently, Wang et al. [20] developed a fuel cell model based on a stochastic-model-based reconstructed GDL structure.

The aforementioned GDL models assumed an ideally simplified pore configuration (cubic or shifted layers of square) of the GDL. A real pore structure of GDL is needed in order to understand transport phenomena in the GDL.

Using the technique which is commonly used in geological science to reconstruct the structure of rocks, clays or soil, both Lindquist et al. [21] and Johshi et al. [22] constructed a two-dimensional pore scale structure of the anode in a solid oxide fuel cell and used it in the Lattice Boltzmann Method (LBM) model. Park and Li [2] constructed a two-dimensional pore scale structure of a carbon paper using the surface view of GDL. The LBM method was then applied to model the multiphase and micro-scale flow through the GDL in a PEM fuel cell. GDL's permeability was obtained numerically. It was found to vary significantly with the carbon fiber's orientation with respect to the flow.

Other efforts have also been attempted to reconstruct three dimensional pore structure and investigate transport phenomena inside a GDL. The X-Ray CT technique or stochastic based digital generation has been used for this purpose [23–27]. Using the three-dimensional pore structure model, Schulz et al. [23] successfully studied the two phase flow behavior in the GDL under different mechanical loads. Hao and Cheng [28] even filled the stochastic generated pore scale structure with 15% PTFE coating to improve the accuracy of three dimensional digital structure of the GDL.

In summary, most of the previous pore structure models were developed based on an artificial structure of the GDL. In addition,

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Fig. 1 GDL SEM image-surface

most pore-scale models were developed purely for transport phenomena inside a GDL. Very few models were coupled with electrochemical reaction to predict the fuel cell performance. The objective of this paper was to develop a fuel cell model where a real pore structure of GDL was used. Instead of using GDL surface SEM image, the cross-sectional view was used to reconstruct GDL's pore structure. The GDL's permeability was also calculated using the pore structure model.

2 Gas Diffusion Layer Pore Structure Reconstruction

In order to obtain the detailed pore structure, the GDL was investigated under a scanning electronic microscope (SEM). Toray carbon fiber paper-TGP-H-120 was used as the testing sample because of its popularity in fuel cell application and its higher thickness in the cross-sectional view. To maintain its original fiber orientation, the GDL sample was put into liquid nitrogen to make it easy to cut and preserve its structure. Figure 1 shows the SEM image of the GDL surface magnified by 200 times, and Fig. 2 shows the SEM image of the GDL cross-sectional view magnified by 300 times. As a fuel cell operates, reactant gases diffuse through the surface of the porous GDL, and sweep through the electrode by diffusion and/or convection. Thus the cross view of the GDL should be the focus in order to study the transport phenomena in the GDL.

Using the Image Processing Toolbox in Matlab, the SEM image was first converted to grayscale image. The grayscale image was then converted into binary format using a thresh-hold value. The critical value was adjusted based on visual comparison with



Fig. 2 GDL SEM image-cross section view

CIP. 120. Cross: 20.0kV/ v300. 500m

Fig. 3 GDL SEM image converted into binary format-cross section view



Fig. 4 GDL pore scale structure contour line

original SEM image. The SEM image of Fig. 2 was converted into binary format with symbols of only "0" or "1" in each location. If the "0" is defined as the void space, then the "1" is defined as the solid space occupied by carbon fiber. The binary format image is shown in Fig. 3. By tracking the interface between the solid space and the void space, a series of lines were identified as shown in Fig. 3. These lines are the boundaries of the reconstructed computational domain as shown in Fig. 4. The reconstructed computational domain was then imported into COMSOL Multiphysics as the computational domain of GDL for PEM fuel cell modeling.

3 Proton Exchange Membrane Fuel Cell Model

Figure 5 shows the computational domain used to develop a two-dimensional half-cell fuel cell model. The domain includes the cathode GDL in a PEMFC with an interdigitated flow pattern. The GDL is 250 μ m thick × 1 mm wide, the flow channel is 0.5 mm wide × 0.5 mm deep, and the bipolar plate shoulder is 0.5 mm wide. The boundary at the bottom is the catalyst layer. The left and right boundaries are symmetrical due to the periodic gas channel configuration.

The present model was developed under the following major assumptions:

- (1) The fuel cell operates at a constant temperature.
- (2) The gas mixture behaves like ideal gases.
- (3) The gas flow is assumed to be laminar and incompressible in the modeling domain.

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(a) A 3D PEM Fuel Cell with Interdigitated Flow Fields



(b) A Cross-sectional View of PEM Fuel Cell (a)



(c) PEM Fuel Cell Model Computational Domain

Fig. 5 Computational domain of the pore structure based model, (*a*) a 3D PEM fuel cell with interdigitated flow fields, (*b*) a cross-sectional view of PEM fuel cell, (*c*) PEM fuel cell model computational domain

- (4) The catalyst layer is assumed to be an infinitely thin layer between the membrane and GDL.
- (5) Water was assumed to exist in vapor form only.
- (6) The gas mixture flows continuously through the reconstructed pore structure.

Using the reconstructed GDL pore structure, there is no need to use models such as Darcy's Law or Brinkman Equation to model the flow in the porous GDL. Instead, the Navier-Stokes equations and the continuity equation could be solved directly throughout the fluid domain:

$$\nabla \cdot u = 0 \tag{1}$$

$$\rho u \cdot \nabla u = \nabla \cdot \left[-pI + \eta (\nabla u + (\nabla u)^T)\right]$$
(2)

where *u* is the velocity vector, m s⁻¹, ρ is the fluid density, kg m⁻³, *p* is the pressure, Pa, and η is the dynamic viscosity, kg m⁻¹s⁻¹.

The multispecies mass transport described by the Maxwell-Stefan equation solves for the fluxes of each species in terms of mass fraction. The general form of the Maxwell-Stefan equation is shown below:

$$\nabla \cdot \left[-\rho w_i \sum_{j=1}^N D_{ij} \left\{ \frac{M}{M_j} \left(\nabla w_j + w_j \frac{\nabla M}{M} \right) + (x_j - w_j) \frac{\nabla P}{P} \right\} + w_i \rho u \right] = R_i$$
(3)

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where D_{ij} is the binary diffusion coefficient, m²/s, R_i is the reaction rate, kg/(m³s), x is the molar fraction, w is the mass fraction, M is the molecular mass, kg/mol, i and j represent different species O₂, H₂O or N₂. On the cathode side, only the mass fractions of oxygen and water were solved because the mass fraction of nitrogen can always be obtained from the mass balance equation as follows:

$$w_{N2} = 1 - w_{O2} - w_{H2O} \tag{4}$$

The catalyst layer was treated as an infinitely thin boundary between the GDL and the membrane. The Tafel equation given below was used to predict the distribution of the current density along the catalyst layer [29],

$$I = I_0 \frac{C^g y_{O_2}}{C_{O_2, ref}} \exp\left(\frac{\alpha_c F}{RT} \eta_c\right)$$
(5)

where I_0 is the exchange current density, $A \cdot m^{-2}$, η_c is the overpotential on the cathode, V, α_c is the cathode transfer coefficient, *F* is the Faraday's constant, and *C* is the concentration of gas, mol·m⁻³.

The governing equations were solved along with the following boundary conditions:

- (1) Flow inlet: The upstream pressure was prescribed at the flow inlet. The inlet gas species fractions were calculated using the data from the validation experiment.
- (2) Flow outlet: The back pressure at the outlet was set to the atmospheric pressure. The flow was assumed to be fully developed.
- (3) Impermeable walls and surfaces: A no-slip boundary condition was applied to the impermeable walls and surfaces, where the no-flux condition was set for the species equations.
- (4) Catalyst layer: Oxygen leaves the GDL at this interface due to electrochemical reaction, while water enters into the GDL domain due to both electrochemical reaction and electro-osmotic drag from the anode side to cathode side [30]. Thus at this boundary, the oxygen flux, water flux and total velocity are functions of the local current density, and they were given as:

Oxygen:
$$N_{O_2} = -\frac{I}{4F}$$
 (6)

Water:
$$N_{H_2O} = (0.5 + \alpha) \frac{I}{F}$$
 (7)

Normal Velocity:

$$u_{normal} = \frac{I}{F} \frac{M_{H_2O}\left(\frac{1}{2} + \alpha\right) - M_{O_2}\frac{1}{4}}{\rho}$$
(8)

where *N* is the inward molar flux, mol/($m^2 \cdot s$), and α is the number of water molecules dragged across the PEMFC membrane for each electron transferred.

4 Results and Discussions

The PEM fuel cell GDL pore structure model was implemented into a commercial software package COMSOL Multiphysics. A total of 121,240 triangular meshes were generated in the computational domain. The discretized governing equations were solved using the stationary SPOOLES solver. A total of 1,131,812

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Table 1 Base case model parameters

Parameter name	Value	Unit
Channel width	0.5	mm
BP shoulder width	0.5	mm
GDL thickness	250	μm
Inlet channel pressure	1.0133	atm
Inlet mass fraction of oxygen	0.233	1
Inlet mass fraction of Nitrogen	0.767	1
Inlet mass fraction of water	0	1
Temperature	60	°C
Back pressure	1	atm
Gas dynamic viscosity	2.03×10^{-5}	Pa⋅s
Exchange current density	1×10^{-2}	A/cm ²
Transfer coefficient of the ORR	0.5	1
Cathode reaction overpotential	0.4	V
Binary diffusion coefficient $D_{O2 N2}$	2.75×10^{-5}	m ² /s
Binary diffusion coefficient $D_{02 H20}$	3.50×10^{-5}	m^2/s
Binary diffusion coefficient D_{H2O} N2	3.50×10^{-5}	m^2/s



Fig. 6 Base case model polarization curve comparison

degrees of freedom were solved. The base case parameters used in the models are listed in Table 1. The fuel cell operating potential was calculated as

$$V_{cell} = V_{oc} - \eta_c - \eta_a - I \cdot R_{cell} \tag{9}$$

where V_{cell} is the fuel cell operating voltage, V, V_{oc} is the open circuit voltage, V, η_c is the cathode overpotential, η_a is the anode overpotential, *I* is the fuel cell operating current density, A/m², and R_{cell} is the electrical resistance of the fuel cell, $\Omega \cdot m^2$, and it includes the electrical resistances in both membrane and catalyst layers in the current study. The ohmic losses in the GDL were small and assumed negligible. The contact electrical resistance between the GDL and BP were ignored.

The current fuel cell model was validated by comparing the predicted polarization curve with experimental data used in Ref.

[29]. The experiment was conducted at 60 °C with atmospheric back pressure. The active cell area is 9 cm². The Nafion 115 membrane with proton conductivity of 7 S/m was used. The catalyst layer was about 60 to 70 μ m with electric conductivity of 53 S/m. The anode overpotential is small, and its value was estimated as a linear function of the current density [29].

Figure 6 compares the polarization curves between the experimental data and numerical simulation. The current fuel cell model developed based on the pore structure GDL predicted the fuel cell performance with a reasonable accuracy.

The pore structure model enables us to investigate the details of the flow inside the GDL and how the flow affects the fuel cell performance. Figure 7 shows the pressure distribution in the GDL in a contour plot. Figure 7 also shows the velocity distribution in vectors to represent velocity's magnitude and direction. Because the interdigitated flow is driven by the pressure difference between the upstream and downstream channels, the pressure decreases along the flow direction. The pressure shows a sharp drop in the throat area, which corresponds to a higher velocity represented by the length of the velocity vectors.

Figure 8 shows the mass fraction of oxygen in the cathode GDL. As oxygen passes through the GDL, it is consumed due to the electrochemical reaction. Therefore the oxygen mass fraction decreases in both in-plane and through-plane directions. Water mass fraction, however, shows the opposite trend as shown in Fig. 9 because water is produced in the catalyst layer and dragged from the anode side due to electro-osmotic effect.

5 Characterize the GDL Permeability by Pore-Structure Model

The pore structure network model was also used to determine the permeability of a porous material numerically in both in-plane and through-plane directions. According to Darcy's Law, the permeability of a porous material is calculated as

$$\kappa = -\frac{Q \cdot \mu \cdot L}{A \cdot \Delta P} \tag{10}$$

where Q is the volumetric flow rate through the cross-section of the domain, m³/s, which was obtained by integrating the result of the Navier-Stokes equations in the pore structure domain directly, ΔP is the pressure drop from inlet to outlet, Pa, μ is the dynamic viscosity, Pa·s, and L is the geometry length, m.

Figure 10 shows the computational domain along with boundary conditions. The X axis represents the in-plane direction, and the Y axis represents the through-plane direction. As shown in Fig. 10, the two sets (X and Y) of the boundary conditions were used separately to characterize GDL's permeability in either inplane or through-plane direction.

Table 2 lists the predicted in-plane and through-plane permeability of the GDL, respectively. Mathias et al. [31] reported that the in-plane permeability of Toray 060 is in the range of $(5-10) \times 10^{-12}$ m². Gostick et al. [3] reported the Toray 090's inplane permeability to be 8.99×10^{-12} m², and the in-plane



Fig. 7 Pressure and velocity distribution in cathode GDL

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Fig. 9 Water mass fraction in cathode GDL



Y: Inlet Pressure= 2 atm

Fig. 10 Computational domain for porous GDL permeability

permeability of a GDL was approximately twice as high as its through-plane permeability. The current pore structure model predicted both the in-plane and through-plane permeability in the magnitude of 10^{-12} , and the in-plane permeability is approximately three times the through-plane permeability. This might be due to the fact that the GDL pore structure domain used in this calculation was taken from a small portion of the GDL sample. If the entire GDL structure is obtained, a more accurate result is expected. In summary, it is concluded that the current pore structure provides the structure of the structure of the structure of the structure provides the structure of the structure port of

 Table 2 Calculated GDL permeability in the in-plane and through-plane directions

Direction	Permeability (m ²)
In-plane Through-plane	$\begin{array}{c} 9.53 \times 10^{-12} \\ 2.78 \times 10^{-12} \end{array}$

ture model predicts Toray 120 GDL's permeability with a good range and relative magnitude.

6 Conclusions

A two-dimensional pore structure of a GDL was reconstructed from the GDL SEM image. Toray carbon fiber paper TGP-H-120 with 5% PTFE coating was chosen as the testing sample because of its popularity and higher thickness in the cross-sectional direction. The SEM was used to obtain both surface image and crosssectional image of the sample. The 2D cross-section SEM image was converted into binary format and the contour lines were identified by using the Matlab Imaging Processing Toolbox. These contour lines were then imported into COMSOL Multiphysics software. The reconstructed geometry was then used as the computational domain of a fuel cell numerical model, which coupled the fluid flow with electrochemical reaction to predict fuel cell performance. The Navier-Stokes equations were solved directly on the pore structure without using any empirical numerical

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model such as Darcy's Law. Therefore, the permeability or porosity of the porous material is not required. The pore structure model predicted a reasonable fuel cell performance compared with the experimental testing. For the GDL in an interdigitated flow channel fuel cell, the flow is driven by the pressure difference between the upstream and downstream channel. The pressure decreases along the flow direction, and pressure shows a sharp drop in the throat area. The detailed information within the pore structure, such as oxygen and water mass fraction, was also presented.

The pore structure was also used to numerically determine the permeability of the GDL using Darcy's Law. The predicted permeability agrees with the published value. The GDL has different permeability in in-plane and through-plane directions. The current pore structure model predicted the permeability in the magnitude of 10^{-12} m², and the in-plane permeability is approximately three times that of the through-plane permeability. It is concluded that the current pore scale model predicts Toray 120 GDL's permeability with a good range and relative magnitude. The further application of the presented method in this section includes the prediction of GDL thermal conductivity and electrical conductivity, which will contribute to improving PEM fuel cell modeling.

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