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Fractal model for predicting the effective binary oxygen diffusivity of the gas diffusion layer in proton exchange membrane fuel cells

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ABSTRACT

We propose an analytical model to predict the effective binary oxygen diffusivity of the porous gas diffusion layer (GDL) in proton exchange membrane fuel cells (PEMFCs). In this study, we consider the fractal characteristics of the porous GDL as well as its general microstructure, and we adopt the Bosanquet equation to derive effective diffusivity. The fractal characterization of GDL enables us to model effective diffusivity in a continuous manner while taking into account the effect of pore size distribution. Comparison to two other theoretical models that are generally accepted in the simulation of PEMFCs shows similar trends in all three models, indicating that our proposed model is well founded. Furthermore, the predicted effective binary oxygen diffusivities of two samples show that after treatment with polytetrafluoroethylene (PTFE), the effective binary diffusivity of the GDL decreases. Based on the parametric effect analysis, we conclude that effective binary diffusivity is negatively correlated with tortuosity fractal dimension but positively correlated with the fractal dimension of pore area, porosity, or mean pore diameter. The proposed model facilitates fast prediction of effective diffusivity as well as multi-scale modeling of PEMFCs and thus facilitates the design of the GDLs and of PEMFCs.

1. Introduction

The performance of proton exchange membrane fuel cells (PEMFCs) is influenced by fluid flow, mass transport and electrochemical reactions, among other factors. Approaching the limiting current density, concentration polarization results from the limit of oxygen transport through the air cathode in PEMFCs. The near-zero oxygen concentration at the cathode occurs due to the low diffusivity of oxygen, which is about three or four times lower than that of hydrogen [1]. This mass transport limitation is controlled by the gas effective diffusivity of the gas diffusion layer (GDL) under the same flow field design; as the value of effective diffusivity decreases, the oxygen flux is reduced. The performance of PEMFCs is therefore strongly related to oxygen effective diffusivity, which is also one of the key material parameters of the GDL and must be taken into account in the simulation of mass transport in PEMFCs.

In recent years, there has been an increasing need to engineer GDLs in such a manner that some of the transport processes can be controlled [2]. In our previous work [3–5], we have studied the gas permeability, effective thermal conductivity, and effective self-diffusivity of hydrogen within the GDL as well as demonstrated the relationship between the microstructure of the porous GDL and these transport properties. In this study, we investigate the effective binary diffusion of oxygen within the GDL and develop a predictive model of oxygen effective diffusivity by considering the real microstructure of the GDL using fractal methods.

2. Methods for determining the effective diffusivity of porous media

GDLs are commonly constructed using carbon fibers in either woven or paper form. These materials are porous to permit gas transport and are electric conductive. The diffusivity of a porous material is strongly dependent on its microstructural features, such as the tortuosity and interconnectivity of the pore system. Experimental measurements, numerical simulations, empirical equations, and analytical solutions are usually employed to determine effective diffusivity.

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2.1. Experimental measurements

Many experimental measurement methods are used to directly determine the diffusivity of porous media.

Bartelt-Hurt and Smith [6] have used a steady-state, one-flow sorbent-based technique to determine the effective air diffusion coefficient for trichloroethene in undisturbed soil cores. Elberling [7] has adopted conductimetric technology to evaluate diffusion coefficients in cemented porous materials. Two kinds of measurement techniques are used for gas diffusivity in porous media: macroscopic and microscopic methods [8]. The former include permeability, time lag, efficiency factor, frequency response, sorption rate measurement, Wilke–Kallenbach and chromatographic methods, while the latter include pulse-field gradient (PFG) NMR and quasielastic neutron scattering. Li et al. [8] have proposed a new microscopic measurement method for diffusivity in porous materials termed temporal analysis of products.

Large volumes of experimental results are available in the literature. However, reported diffusivities determined by different methods often significantly differ [8]. Worse yet, measurements are time consuming and expensive, and measured results are difficult to explain because it is non-trivial to experimentally study detailed physical phenomena within the pores. To this end, many numerical methods have been used to investigate the mechanism inside the pores.

2.2. Numerical simulations

In numerical studies, a geometrical pore model of a representative elementary volume must be established first. Then, effective diffusion mechanisms can be modeled in a continuous or discrete manner. In continuous models, finite difference [9], finite element [10] or spectral methods [11] can be chosen to solve the diffusion equations. For example, Fick's law is often used to describe gas diffusion. The lattice Boltzmann method [12] can also be used in continuous modeling. Discrete modeling methods include random walks [13], Monte Carlo methods [14,15], molecular gas dynamics [16] and particle tracing methods [17].

There are two kinds of geometrical pore model: non-fractal and fractal pore models. Zhu et al. [11] have used a grain growth model for 2D simulation; in this model, the solid phase consists of spheres or grains and the spaces between the grains compose the pores. Armatas [18] has developed a hyphenated 3D dual site-bond model network using Monte Carlo methods. Tomadakis and Sotirchos [14] have built 2D and 3D random models for fiber materials and used Monte Carlo methods to obtain the effective diffusivities of the porous fiber media. Pore shape and porosity are taken into account when building these models; therefore, the models can be used for any porous medium with a specific pore shape or distribution. Liu et al. [10] have processed binary images of thin soil sections using Adobe Photoshop to find the connected pore network for simulation. It is very difficult to deal with images exhibiting poor connectivity of pore networks, which is the case for many real porous materials. Vogel et al. [9] have used serial sections with a resolution of 0.1 mm to reconstruct a 3D pore model of soil. Their work is based on serial 2D images, reflecting the real space of porous media. But it is difficult to acquire serial images of porous media and reconstruct an accurate 3D model.

Because many natural pores have fractal-like properties, many pore models have recently been developed that take this into consideration. Anderson et al. [19] have constructed a 2D random Sierpinski carpet, a kind of fractal, to approximate the representative elementary volume of soil. Borrelli et al. [20] have used a 3D Menger sponge model, which is also a kind of fractal, in a simulation of diffusion mechanisms. Liang et al. [21] have created a pore fractal model with an arbitrary given porosity and an arbitrary fractal dimension using random walk methods. These studies took porosity and fractal characteristics into account. However, the shapes of Sierpinski carpet or Menger sponge models are regular and different from those of most natural porous media. Visual inspection shows that the shapes of these models are not similar to those of real pores.

With a geometrical pore model selected through numerical simulation, effective diffusivity can be predicted and parametric effects can be studied. However, because pore models vary with the microstructure of porous media, they must always be reconstructed for different porous materials. Moreover, numerical simulation is too time consuming to achieve fast and convenient prediction of diffusivity, so empirical equations are usually used.

2.3. Empirical equations

Many predictive models of effective gas diffusivity have been developed for porous media. Generally, the effective diffusivity is modified to account for the effects of the microstructure of porous media, as follows:

$$\frac{D_{\text{eff}}}{D_0} = f(\varepsilon, \phi, \tau, \cdots), \tag{1}$$

where D_{eff} is the effective gas diffusivity in a porous medium, D_0 is the gas diffusivity in free air, ε is the air-filled porosity, ϕ the total porosity, and τ is the tortuosity.

Some models are based on ε only. The most frequently used gas diffusivity model that is independent of soil type was suggested by Penman [22]:

$$\frac{D_{\text{eff}}}{D_0} = 0.66\varepsilon \tag{2}$$

In some models, both ε and ϕ are considered. Moldrup et al. [23] have proposed the so-called Buckingham–Burdine–Campbell model gas diffusivity model, which is soil type-dependent:

$$\frac{D_{\text{eff}}}{D_0} = \phi^2 \left(\frac{\varepsilon}{\phi}\right)^{2+3/b}, \tag{3}$$

where the term $2+3/b$ is an analog to the Burdine–Campbell tortuosity model for describing unsaturated hydraulic conductivity.

Taking water phase into consideration, Aachib et al. [24] have given a dual phase model:

$$D_{\text{eff}} = \varepsilon\tau D_0 + \varepsilon_w \tau_w D_w^0, \tag{4}$$

where ε_w is water-filled porosity, τ_w is the tortuosity in the water phase, and D_w^0 is the free diffusivity in water.

Generally, the empirical constants in empirical models do not indicate any specific physical meanings, and their values vary significantly among researchers. From the above discussion, we can see that most of these models are related to only one or two of the microstructural parameters of porous media. The drawbacks of using empirical laws lie in the fact that the basic phenomena remain poorly understood.

2.4. Analytical solutions

It is challenging to develop predictive models, which must appropriately characterize the transport properties of porous media to make meaningful predictions. Using analytical solutions should be an indispensable tool for exploring new architectures as well as designs of porous media and would contribute to a better understanding of the phenomenology compared to the use of other methods.

Gas transport in the GDL is typically described by the Bruggeman relation [2,25], which is analytic expression that belongs to the class

of solutions referred to as effective medium approximations:

$$\frac{D_{\text{eff}}}{D_{\text{c}}} = \varepsilon^{1.5}, \tag{5}$$

where D_{c} is the composite diffusivity. However, Pharoah et al. [2] have pointed that the Bruggeman relation will over-predict the concentration of oxygen in the catalyst layer when diffusion is an important mechanism.

In most studies [26–29], the effects of both the porosity (ε) and tortuosity (τ) of the capillary pathway are taken into account. The corresponding diffusivity is

$$\frac{D_{\text{eff}}}{D_{\text{c}}} = \frac{\varepsilon}{\tau} \tag{6}$$

For the diffusion of a single gas in porous media with very fine pores, both wall and molecule collisions redirect paths. The composite diffusivity D_{c} depends on the bulk and Knudsen diffusion components, commonly described by the Bosanquet equation [27]:

$$D_{\text{c}} = \left(\frac{1}{D_{\text{A}}} + \frac{1}{D_{\text{K}}(\lambda)}\right)^{-1}, \tag{7}$$

where D_{A} is the self-bulk diffusivity of gas A and $D_{\text{K}}(\lambda)$ is the Knudsen diffusivity. In the case of multi-gas diffusion in porous materials, D_{A} must be replaced by the binary bulk diffusivity D_{AB} of the mixture of gases A and B to obtain the composite diffusivity. For a smooth cylindrical pore in classical Knudsen theory, analytical solutions for regular-shaped pore geometries with relatively simple initial and boundary conditions can be obtained, and the Knudsen diffusivity is given by [26]:

$$D_{\text{K}}(\lambda) = \frac{2}{3}\lambda\left(\frac{2k_{\text{B}}T}{\pi m}\right)^{1/2}, \tag{8}$$

where λ is the pore diameter, m is the molecular mass, T is the temperature (K), and k_{B} is the Boltzmann constant (1.3806×10^{-23} J molecule^{-1} K^{-1}). It has been confirmed that the Bosanquet treatment of transition diffusion remains accurate for random pore structures [27].

In some studies [27], the Bosanquet equation has been used on the base of the principle of number-averaged pore diameter for the Knudsen diffusivity. However, it is more accurate to consider the pore size distributions of porous media. For a porous medium with a wide pore size distribution, the composite diffusivity is given by [30,31]:

$$D'_{\text{c}}\varepsilon = \varepsilon_1 D_{\text{B}} + \varepsilon_2 D_{\text{BK}} + \varepsilon_3 D_{\text{K}}, \tag{9}$$

where D_{B} is the bulk diffusivity, D_{BK} is the transitional diffusivity, and D_{K} is the Knudsen diffusivity, and ε_1, ε_2, and ε_3 are the porosities of the large pores, medium ones, and fine pores, respectively. Here, D'_{c} is related to the effective diffusivity as follows [30]:

$$D_{\text{eff}} = \frac{D'_{\text{c}}\varepsilon}{\tau} \tag{10}$$

This is a discrete expression of the composite diffusivity.

A continuous expression for the composite diffusivity is given by [28]:

$$D_{\text{c}} = \int_0^\infty \left(\frac{1}{D_{\text{AB}}} + \frac{1}{D_{\text{K}}(\lambda)}\right)^{-1} f(\lambda)d\lambda, \tag{11}$$

where $f(\lambda)$ is the pore size density function.

Obviously, the discrete model is very simple, but the continuous model is more accurate. For the continuous model, the pore size density function must be found. Armatas et al. [28] have used experimental data to obtain the function. Combined with experimental data, however, Eq. (11) cannot be used for rapid prediction. Furthermore, it is difficult to determine the pore size distribution of most porous materials.

Fortunately, as mentioned above [16,19,21], because many natural porous media have fractal-like properties, fractal geometry can be used to quantitatively describe porous media. In addition to the role that fractal geometry plays in the quantification of porous media, it can also be used to describe the relationship between microstructure and physical processes occurring in porous media.

Many studies have attempted to determine the relationship between the effective diffusivities and microstructures of porous media by using fractal methods [16,19,21,32–34]. In most of these studies, porosity is generally considered to have a significant effect on diffusivity, whereas the effect of pore size distribution is ignored. However, the permeability of some porous systems with high porosity is surprisingly poor because of their small pore size (pore sizes ranging between one nanometer and one micrometer) [35]. Thus, the effect of pore size distribution on material properties should be taken into account. For porous media with wide pore size distributions, the Knudsen diffusion should also be considered.

The aim of this study is to present a theoretical model of effective binary oxygen diffusivity for the GDLs of PEMFCs that takes into consideration microstructure parameters such as porosity, pore size distribution, and tortuosity. Our previous work has shown that microstructure and pore size distributions of GDLs in PEMFCs have fractal characteristics [36]. Thus, the diffusivity of the GDL can be studied by fractal theory, which has great capability for characterizing random porous media. In the following sections, a theoretical model of diffusivity will be deduced and the effects of microstructure parameters on diffusivity will be studied.

3. Fractal prediction model of the diffusivity of the GDL

When air is supplied, the gas diffusion of the cathode GDL is a binary diffusion for the gas mixture of oxygen and nitrogen. Here, we focus on the oxygen effective diffusivity within the GDL. According to experimental data from mercury porosimetry and BET absorption, the GDL exhibits a wide range of pore diameters from 10^{-5} m to 10^{-8} m [36]. Because the mean free path of gas molecules is about 10^{-7} m, the Knudsen number ranges from about 10^{-2} to 10, and both Knudsen and bulk diffusion should be taken into account for the GDL.

We use a model with curved capillaries of different diameters. This model was first proposed by Pitchumani and Ramakrishnan [37]. In this model, two fractal dimensions are used: the tortuosity fractal dimension D_{t}, which represents the extent of convolution of the capillary pathways for fluid flow through a medium, and the pore area fractal dimension D_{p}, which shows the scaling relationship between the measurement of a fractal object and the metrical yardstick.

Based on this model, the function $f(\lambda)d\lambda$ can be expressed as [5]

$$f(\lambda)d\lambda = \frac{4\varepsilon(2-D_{\text{p}})\lambda^{-D_{\text{p}}+1}\lambda_{\text{max}}^{D_{\text{p}}-2}}{1-(\lambda_{\text{min}}/\lambda_{\text{max}})^{2-D_{\text{p}}}}d\lambda, \tag{12}$$

where λ_{max} is the maximum pore diameter and λ_{min} is the minimum pore diameter. On the other hand, the tortuosity τ is given by [5]

$$\tau = \left(\frac{L_0}{\lambda}\right)^{2D_{\text{t}}-2}, \tag{13}$$

where L_0 represents the linear length of the capillary pathways in the direction of diffusion direction.

According to Section 2.4, considering Eqs. (9)–(11), we can obtain the effective diffusivity as follows:

$$D_{\text{eff}} = \int_0^\infty \frac{\varepsilon}{\tau}\left(\frac{1}{D_{\text{AB}}} + \frac{1}{D_{\text{K}}(\lambda)}\right)^{-1} f(\lambda)d\lambda, \tag{14}$$

where $f(\lambda)d\lambda$ represents the ratio of the void volume in pores that have a radius between λ and $\lambda + d\lambda$ to the total volume. By using Eq.

Table 1
Model parameters and sample properties.

Parameter	Sample 'a'	Sample 'b'	Description
λ_{max}	8×10^{-5} m	7×10^{-5} m	Maximum pore diameter
λ_{min}	3.079×10^{-8} m	1.487×10^{-8} m	Minimum pore diameter
ε	0.55	0.78	Porosity
L_0	1.9×10^{-4} m	1.9×10^{-4} m	Gas diffusion layer thickness
D_{AB}	3.2×10^{-5} m^2 s^{-1}	3.2×10^{-5} m^2 s^{-1}	Oxygen diffusivity in gas
T	353 K	353 K	Temperature
D_t	1.14	1.14	Tortuosity fractal dimension
D_p	1.9669	1.9276	Pore area fractal dimension

(14) and fractal theory, we also take into account the relationship between τ and λ.

Substituting Eqs. (8), (12) and (13) into Eq. (14), we arrive at the effective diffusivity:

$$D_{eff} = \frac{8\sqrt{2k_BT}\varepsilon^2 D_{AB}(2-D_p)L_0^{2-2D_t}}{\sqrt{\pi m}(\lambda_{max}^{2-D_p} - \lambda_{min}^{2-D_p})} \times \int_0^{\infty} \frac{\lambda^{2D_t-D_p}}{3D_{AB} + 2\lambda(2k_BT/\pi m)^{1/2}} d\lambda \quad (15)$$

and the relative effective diffusivity, a dimensionless variable:

$$f_1 = \frac{D_{eff}}{D_{AB}} = \frac{8\sqrt{2k_BT}\varepsilon^2(2-D_p)L_0^{2-2D_t}}{\sqrt{\pi m}(\lambda_{max}^{2-D_p} - \lambda_{min}^{2-D_p})} \times \int_0^{\infty} \frac{\lambda^{2D_t-D_p}}{3D_{AB} + 2\lambda(2k_BT/\pi m)^{1/2}} d\lambda, \quad (16)$$

where λ_{max} is the maximum pore diameter and λ_{min} is the minimum pore diameter.

Eq. (15) contains parameters with clear physical meanings and no empirical constants. The above general theoretical model can be used both for prediction of the effective binary diffusivity of a porous medium as well as for studying the effects of porosity and other microstructure parameters, such as the tortuosity fractal dimension D_t and the pore area fractal dimension D_p. From the viewpoint of microstructural characterization, this fractal model is closer to reality than common empirical models that contain few microstructural parameters (porosity or tortuosity) or other fractal models that ignore the significant impact of pore size distribution. Our model provides faster and easier predictions of effective diffusivity than numerical simulation methods can provide.

This model provides a bridge between microcosmic and macrocosmic models of PEMFCs, which are multi-scale objects. Based on this kind of model, we can perform research on multi-scale coupling to obtain more accurate results from simulations to facilitate the design of GDLs and PEMFCs.

4. Results and discussion

4.1. Model prediction and comparison

The effective binary oxygen diffusivity of the GDL can be predicted using Eq. (15). We use two samples: sample 'a' denotes TGP-H-060 carbon paper and sample 'b' denotes TGP-H-060 carbon paper treated with polytetrafluoroethylene (PTFE). In a previous study [3], we have determined two fractal dimensions. The operating parameters [1], oxygen diffusivity [1], and some microstructural data from mercury porosimetry [36] are listed in Table 1. Now we can substitute the real GDL parameters listed in Table 1 into Eq. (15) and obtain the integral result of Eq. (15).

In the model comparison and parametric study, the data for sample 'b' and the relative effective diffusivity are used. We compare the proposed fractal model (Eq. (16)) to two other theoretical models (Eqs. (5) and (6)) that are frequently used to model PEMFCs based on the relative effective diffusivity. According to fractal theory, Eq. (6) can be rewritten as

$$f_2 = \int_0^{\infty} \frac{\varepsilon}{\tau} f(\lambda) d\lambda = \frac{4\varepsilon^2(2-D_p)(\lambda_{max}^{2D_t-D_p} - \lambda_{min}^{2D_t-D_p})}{L_0^{2D_t-2}(2D_t-D_p)(\lambda_{max}^{2-D_p} - \lambda_{min}^{2-D_p})}, \quad (17)$$

which is the expression used in our comparison.

Fig. 1 shows the variation in the relative effective diffusivities predicted by the three models. All three models exhibit similar variation trends. The curve of the fractal model is beneath those of the other two models. As mentioned above, Pharoah et al. [2] have pointed out that the Bruggeman relation (Eq. (5)) will over-predict the concentration of oxygen in the catalyst layer when diffusion is an important mechanism. Our results show that the Bruggeman relation gives high oxygen diffusivity, which results in over-prediction.

Using the fractal model (Eq. (15)), we obtain predicted values for the effective diffusivity of samples 'a' and 'b' of 2.2445×10^{-5} m^2 s^{-1} and 1.0594×10^{-5} m^2 s^{-1}, respectively. The effective diffusivity of sample 'b' is lower than that of sample 'a'. The decrease in effective diffusivity of carbon paper after treatment with PTFE is due to the decrease in pore space and porosity.

4.2. Parametric effect

As mentioned above, the data for sample 'b' and the relative effective diffusivity f_1 are used for further parametric studies. D_p is related to ε by $\varepsilon = (\lambda_{min}/\lambda_{max})^{3-D_p}$ [34]. For a 2D space, this equation becomes $\varepsilon = (\lambda_{min}/\lambda_{max})^{2-D_p}$. We also consider this relationship in the parametric effect study.

The relative effective diffusivity shows a dependence on the mean pore diameter $\bar{\lambda}$ of sample 'b', as shown in Fig. 2. The value of f_1 increases sharply as the mean pore diameter increases. This increase should correspond to the transition from Knudsen-type diffusion to bulk-type diffusion.

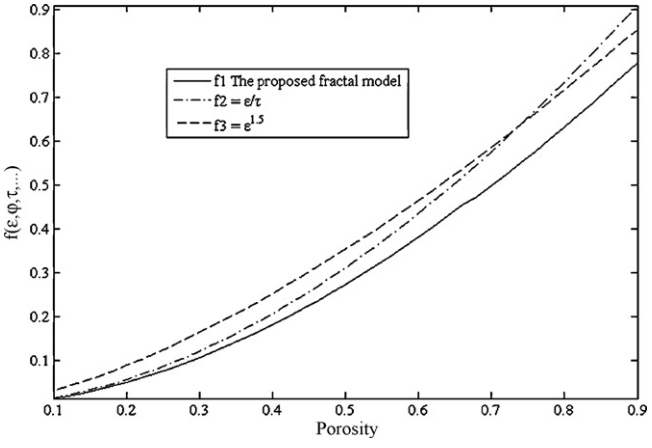

Fig. 1. Comparison between the effective diffusivity prediction models.

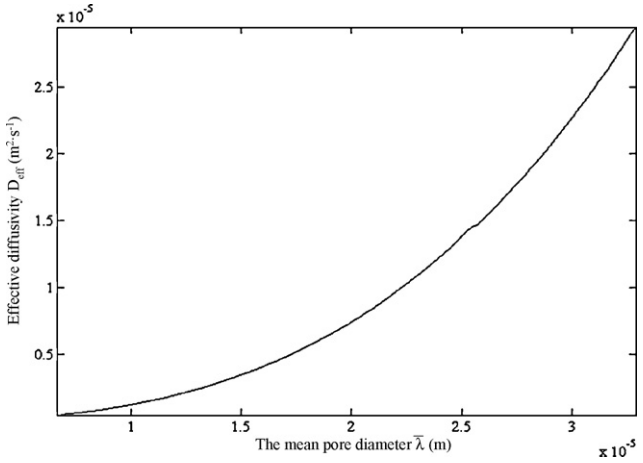

Fig. 2. Effect of mean pore diameter on the effective diffusivity of carbon paper.

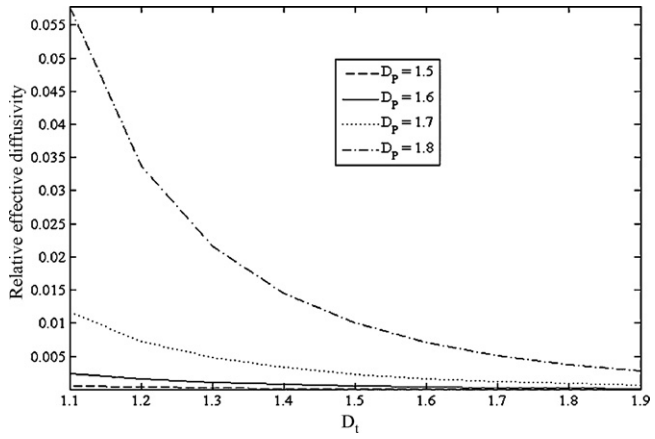

Fig. 3. Effect of D_t (tortuosity fractal dimension) on the effective diffusivity of carbon paper.

Fig. 3 shows that the relative effective diffusivity f_1 varies with the pore area fractal dimension D_p for different values of the tortuosity fractal dimension D_t. It can be seen that f_1 increases as D_p increases but decreases as D_t increases. The increase in D_p corresponds to the increase in pore area of cell sections of carbon paper, which increases the gas diffusion area, leading increased effective diffusivity.

Fig. 4 shows the variation in the relative effective diffusivity f_1 with porosity ε and tortuosity fractal dimension D_t. Relative effective diffusivity increases with increasing porosity ε of carbon paper because of the increase in the gas diffusion area, as with the effect of D_p on relative effective diffusivity. In addition, both Figs. 3 and 4 show that relative effective diffusivity decreases with increasing tortuosity fractal dimension D_t. This relationship can be attributed to increased flow resistance due to highly convoluted capillary pathways.

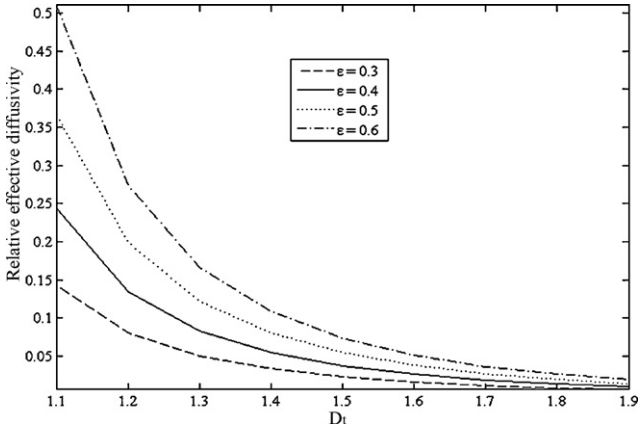

Fig. 4. Effect of porosity on the effective diffusivity of carbon paper.

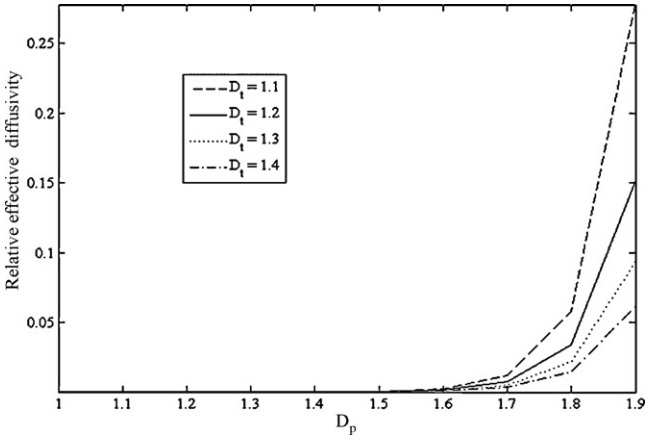

Fig. 5. Effect of D_p (pore area fractal dimension) on the effective diffusivity of carbon paper.

Fig. 5 illustrates the effect of the tortuosity fractal dimension D_t on the relative effective diffusivity f_1. This figure supports the same conclusion as that derived from Fig. 3.

5. Conclusion

The purpose of this study is to propose a simple analytical model to determine the oxygen binary effective diffusivity of the GDL. To this end, the relationship between the effective diffusivity and the microstructure of the GDL is considered using fractal methods.

To validate the proposed model, we compare it to two other theoretical models that are generally accepted in the modeling of PEMFCs based on two GDL samples. The similar variation trends yielded by the three models indicate the effectiveness of the proposed model.

Furthermore, the predicted effective oxygen diffusivities of the two samples show that after treatment with PTFE, the GDL exhibits lower effective diffusivity due to the decrease in pore space and porosity. From the parametric effect study, we conclude that effective diffusivity is negatively correlated with the tortuosity fractal dimension D_t, whereas it is positively correlated with the pore area fractal dimension D_p, the porosity ε, and the mean pore diameter $\bar{\lambda}$.

The proposed model not only facilitates rapid prediction of effective diffusivity but also provides a bridge between multi-scale models of PEMFCs. In future studies, we will leverage our previous work on the properties of the GDL to perform multi-scale modeling of PEMFCs and thus facilitate the design of the GDL and of PEMFCs.

Acknowledgments

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