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Effective gas diffusion coefficient in fibrous materials by mesoscopic modeling

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ABSTRACT

This paper presents a method to establish a relationship between internal microstructure and the effective gas diffusion coefficient in fibrous materials via a mesoscopic modeling approach and, when possible and based on the analysis, to propose user-friendly formulas as functions of structural parameters for practical engineering applications. The entire numerical framework includes two main parts: a random generation-growth method to reconstruct the digital microstructures of fibrous materials based on experimental statistical information of the actual structure, and then a high-efficiency lattice Boltzmann algorithm for modeling the gas diffusion process through porous structures. The predictions are then validated by existing experimental data for both dry and saturated fibrous materials. Owing to the unique robustness of the developed modeling approaches, we are then able to conduct a parametric analysis, more detailed than ever, of the influences on the system effective diffusion coefficient in fibrous materials by such important parameters as structural anisotropy, system water content, microstructure morphology and the layering space in a laminated fibrous system. These results may improve our understanding of gas diffusion in fibrous materials, and this method may serve as a tool for easy estimation of effective diffusivity, leading to the optimal design of fibrous materials.

1. Introduction

Diffusion of a substance in another media is a spontaneous movement of the substance caused by its concentration gradient, and governed by Fick's laws of diffusion, or at a microscopic view, as the random walk of the diffusing particles self-propelled by thermal energy [1]. The most unique characteristics in diffusion are the small length scale concerned, limited in between micrometer and millimeter, and the trifling amount of mass involved [1]. Such characteristics of transporting mass in tiny amount over minor spatial scale render the diffusion process less visible, but fundamentally essential and ubiquitous in many physical, chemical phenomena and beyond. In fact, the process of diffusion is so prevalent that, together with reaction, it was considered the "possible basis of modern micro- and nanotechnologies" [2]. What is more vital, at such minuscule spatial scale, the interfacial effects become much more dominant in determining the system properties.

For practical applications, a thorough understanding of the mechanisms involved in a given diffusion process is crucial for design and optimization. For instance, gas diffusion in fibrous materials, one of the common occurrences such as mass transport through fibrous screen filters in many filtering processes, and through gas diffusion layer (GDL) in fuel cells, is considered essential in many engineering fields [3–5]. A deep understanding of how the fibrous microstructures affect the diffusive flux can lead to significant improvements in product design [6]. Macroscopically a porous medium by definition is a two-phase (solid and air) system, and the gas diffusion efficacy in porous media is usually considered a function of the system porosity ε, the internal tortuosity τ, and the pore size distributions [6], and has been widely studied via theoretical, experimental and numerical methods [7–11]. Below we will present a very brief overview of previous work on diffusion in fibrous materials in terms of these different methods respectively.

In theoretical aspect, both deterministic modeling based on Fick's diffusion equation, and stochastic approaches rooted in random walk picture have been widely applied to investigate various

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diffusion processes. Several studies have recommended different models to predict the behaviors of gas diffusion through fibrous materials. For instance, the simplest model is proposed by assuming a fibrous material as the assembly of a bundle of tortuous channels, and demonstrated in a normalized form that the effective diffusivity is related to the bulk diffusivity in the void in terms of both porosity and tortuosity. Although the porosity here is easy to be calculated or measured, the practical applicability of this model is crippled by the difficulty in accurately determining the value of tortuosity. Bruggeman [12] also presented a model based on the effective medium approximation. However, this model was derived from a system with uniformly packed spherical particles rather than media with more intricate internal structures. Li et al. [13] treated a fibrous material as a system formed by cylindrical fibers arranged either in square or hexagonal configuration. Then by varying the width of the gap between fibers, the effect of the system porosity was investigated. Their model however over-predicted the material diffusivity, and one possible cause is that the gas concentration was assumed in their analysis to be constant at any cross-section of the material. Recently, Shou et al. [14] proposed a fractal model for gas diffusion through nanofibrous and microfibrous materials including both the Fick's diffusion and Knudsen diffusion. However, too many simplifications were made such as the pores in fibrous media form a bundle of tortuous open channels with statistically fractal-like sizes. Thus this fractal model remains to be validated by experimental data in more different fibrous structures beyond the specific highly anisotropic nanofibrous webs and GDLs considered in their work.

Experimentally, the effective diffusivity of fibrous materials as defined can be determined in principle by measuring the diffusive flux and the directional concentration gradient. Penman [7] measured the cross-plane diffusion coefficient of a steel wool sample as early as in 1940. In a similar way, Bateman et al. [15] obtained the effective diffusivity of NO gas penetrating through a 2D cellulosic filter. Gibson et al. [16] used a dynamic diffusion test cell method to measure the effective diffusivity of vapor in fibrous media by getting the change of relative humidity in the vapor. The dynamic diffusion cell is also used by LaManna and Kandlikar [17] to investigate the effects of micro-porous layer (MPL) coatings and sample thickness on the effective water vapor diffusion coefficient in GDLs.

In terms of computational simulation and numerical approaches, owing to the rapid development of computational techniques, a variety of major numerical schemes have been developed for analyzing the mass transport in fibrous media. The single component gas flow and effective permeability in fibrous materials has been widely studied usually by combining stochastic method for structure generation and highly efficiency numerical schemes like the lattice Boltzmann method [18,19]. In contrast, there is much less work on the gas diffusion in fibrous structures as another important mass transport process. Tomadakis and Sotirchos [20] used Monte Carlo method to simulate gas diffusion in 1D, 2D and 3D regular fiber networks and calculated the effective diffusivity. Becker et al. [21] numerically reconstructed a fibrous structure from a 3D image, and then developed a relationship between the effective diffusion coefficient and the saturation of the GDL. Zamel et al. [22] used GeoDict software to generate the structure of a carbon paper, and imported the structure into a commercial CFD to compute the effective diffusion coefficient of fibrous media.

However, a review of the existing work uncovers that most of the previous studies focus on specific and regular microstructures of the media, ignoring the structural variations and the intricate system geometries. For instance, in a fibrous media of given porosity, when parameters like fiber size and orientation vary, it can give rise to vastly different internal porous structures and hence diverse diffusion behaviors. A general approach dealing with the diffusive physics in fibrous materials as a whole, i.e., demonstrating the effects of fiber and structure parameters on the system effective diffusion coefficient is highly desirable, yet non-existent to our best knowledge; this therefore becomes the main objective of this present work. It will be shown at the end that attributed to the unique robustness of our new modeling approaches, we are able to investigate, more detailed than ever, the influences on the system effective diffusion coefficient in fibrous materials of such important parameters as the fiber orientation hence the structural anisotropies, system water content, microstructure morphology and the layering space in a laminated fibrous system.

This article is organized as follows: In Section 2, we introduce the theoretical foundation and numerical framework of our approach, including a random generation growth method for microstructure reproduction for fibrous, granular as well as partially saturated porous media; the governing equations for gas diffusion; and a lattice Boltzmann solver for the governing equations. In Section 3, the present method is validated with existing theoretical solutions and experimental data from other publications. Then in Section 4, the structure effects (liquid saturation, types of structures, orientation angle of fiber, and the layer-spacing in a laminated structure) on gas diffusion in porous media are analyzed. Concluding remarks based on this study are finally made in Section 5.

2. Numerical framework

2.1. Reconstruction of microstructures

To study the microstructure effect via computational modeling, we have to reconstruct the microstructures of porous materials in the computer. Generally speaking, the microstructures of actual porous media have some significant features, often being stochastic with statistical characteristics and rarely if ever regular and remaining constant. To photo-realistically reproduce the intricate details of such microstructures is both impractical and unnecessary. In fact, if our interest is on the steady state as in this case of fluid diffusion, the macroscopic (effective) transport properties of such systems are actually governed by the statistical average values of the parameters involved. As a result, we adopt a multi-parameter random generation-growth algorithm to reconstruct the random microstructures using the statistical average information from the real porous materials [23,24]. The equivalent structure generated this way with finite parameters will reflect the major characteristics of the actual system. The another major issue in tackling such porous media is the morphological difference, and different morphologies have demonstrated very significant impact on the effective energy transport properties, such as on thermal conductivity [25]. Microstructures of porous media may be roughly divided into three categories in morphology: fibrous, granular and network structures [25]; we hence designed the reconstruction methods for each type correspondingly. To consider the liquid saturation effect when water coexists in a system, an algorithm describing the multiphase distributions will be presented, as detailed in the following paragraphs.

For a fibrous structure, the algorithm assumes that each fiber is represented by a straight cylinder with given diameter d and length l, and located by its core position (the geometrical center) and an orientation angle pair (ϕ,θ) as shown in Fig. 1 [26]. When describing the size of a fiber, fiber aspect ratio $p = l/d$ is often used, as also in this work, and a larger p value represents a slenderer shape (longer length or thinner diameter). The generation process for a three dimensional fibrous microstructure is conducted as follows: (***i***) randomly distributing fiber seeds (center) in a given grid system based on the seed distribution probability s_d, whose value is determined by the fiber number density; (***ii***) assigning a random