Journal of Power Sources 248 (2014) 83–90

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Effect of porosity heterogeneity on the permeability and tortuosity of gas diffusion layers in polymer electrolyte membrane fuel cells

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HIGHLIGHTS

- Stochastic, 3D carbon fiber material models generated with controlled porosity heterogeneity.
- Lattice Boltzmann method employed to measure bulk hydrodynamic properties.
- Porosity heterogeneity contributed a negligible effect on modeled transport, compared to porosity and material composition.
- Tortuosity increases with binder & PTFE fraction, which was unexpected by the authors.
- Permeability increases with permeability and binder & PTFE fraction.

ARTICLE INFO

Article history:
Received 23 June 2013
Received in revised form
1 September 2013
Accepted 15 September 2013
Available online 24 September 2013

Keywords:
Porosity heterogeneity
Binder & PTFE
Permeability
Tortuosity
Gas diffusion layer
Lattice Boltzmann method

ABSTRACT

In this paper, we study the effect of porosity heterogeneity on the bulk hydrodynamic properties (permeability and tortuosity) of simulated gas diffusion layers (GDLs). The porosity distributions of the heterogeneous reconstructed samples are similar to those previously reported in the literature for Toray TGP-H 120™ GDLs. We use the lattice Boltzmann method to perform pore-level flow simulations in the reconstructed GDL samples. Using the results of pore-level simulations, the effect of porosity distribution is characterized on the predicted in- and cross-plane permeability and tortuosity. It was found that porosity heterogeneity causes a higher in-plane permeability and lower in-plane tortuosity, while the effect is opposite in the cross-plane direction, that is a lower cross-plane permeability and a higher cross-plane tortuosity.

We further investigate the effect of adding poly-tetra-fluoro-ethylene (PTFE) & binder material to the reconstructed GDL samples. Three fiber volume percentages of 50, 75, and 100% are considered. Overall, increasing the fiber volume percentage reduces the predicted in- and cross-plane permeability and tortuosity values. A previously reported relationship for permeability of fibrous materials is fitted to the predicted permeability values, and the magnitude of the fitting parameter is reported as a function of fiber volume percentage.

1. Introduction

Hydrogen-based power systems, specifically fuel cells, have attracted significant attention in the last decade due to their low local CO_2 emission, high efficiency, and versatility. Among different types of fuel cells, polymer electrolyte membrane (PEM) fuel cells, which typically have energy conversion efficiencies over 50%, are a promising option for the transportation sector, distributed power generation, and backup power generation, to name a few applications [1–4].

Considerable advancements have been made towards the commercial development of PEM fuel cells in areas such as reduced platinum loading, higher current density, and reduced costs; however, there still exist challenges that should be removed before PEM fuel cells can become commercially viable. Mass transport issues such as liquid water flooding at high current working conditions leads to inefficient performance [5]. Overcoming these operational challenges requires more research on characterization and performance-evaluation of PEM fuel cell materials, which will

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http://dx.doi.org/10.1016/j.jpowsour.2013.09.061

inform new material designs for improved efficiency and product reliability.

Computational modeling, next to experimental studies, has been widely used to study mass, energy, and charge transport at different length scales inside PEM fuel cells. The numerical studies can be categorized into two groups, namely: (i) studies that use first principles, thus, can only include small system sizes comparable to length scales of the transport phenomenon under study, and (ii) studies that use models and relations to predict transport phenomena inside multiple components of a fuel cell, and predict the cell-level behavior. These models usually include larger combined domains, and need effective transport properties of each part of the cell as an input. The aim of many of the works under the first category is to predict effective transport properties of individual components of a PEM fuel cell, which will be used in the studies of the second type.

One particular part of a PEM fuel cell, which is the focus of this paper, is the gas diffusion layer (GDL). The GDL is a layer consisting of carbon fibers in either woven or non-woven formats. The thickness of a GDL is typically between 200–400 μm, with fiber diameters in the range of 7–10 μm. In a conventional PEM fuel cell, the GDL is placed between the micro porous layer (MPL) and a graphite plate that includes gas channels. The produced water inside the cell passes through the GDL to be guided out of the system via gas channels. The GDL should be of overall high thermal and electrical conductivity, and should be structured in a way that prevents excess water accumulation.

The GDL influences many aspects of a PEM fuel cell's operation, thus, accurate prediction of its effective transport properties is crucial in understanding the cell performance. El-Kharouf et al. [6] presented a comprehensive review of available ex-situ measurement techniques for determining a range of GDL properties.

A review of available methods for bulk porosity measurement techniques is presented by Cindrella et al. [7]. Determining effective thermal conductivity of GDL samples has also been subject of many previous studies [8–13]. Another important bulk transport property of a GDL is its absolute permeability. Bulk permeability prediction in GDL samples has been widely investigated numerically and experimentally [14–19]. All these works focus on determining the bulk permeability, and do not discuss the effect of pore-level structural parameters on the permeability. Recently, Fishman et al. [20] showed that porosity distribution through the thickness of GDL samples is not uniform. Through rigorous analysis of computed tomography (CT) scan images of samples from different manufacturers, they reported typical porosity distribution profiles of different types of GDLs. In another work, Fishman and Bazylak [21] studied the effect of poly-tetra-fluoro-ethylene (PTFE) and binder material addition on the overall through-thickness porosity profiles of GDL samples. They reported that PTFE and binder material accumulate in low porosity areas, where the density of fibers is higher.

It is known that the heterogeneous porosity distribution affects the overall transport properties of porous material [22,23], however, this dependance has not been quantified. Fishman et al. [24] applied the available relations for permeability/tortuosity of porous materials to the measured porosity profile inside the GDL, and mapped the porosity profile into permeability/tortuosity profiles. In this work, we show that their method of calculating the resultant bulk permeability is only accurate for the in-plane direction.

In this work, we use pore-level flow simulations to investigate how the heterogeneous porosity distribution of the GDL affect overall bulk hydrodynamic properties (*i.e.*, permeability and tortuosity). In order to conduct a parametric study, GDL samples are reconstructed over the entire range of relevant bulk porosity values with homogeneous and heterogeneous porosity profiles. The heterogeneous profile is similar to that reported previously by Fishman et al. [20,21] from CT scan imaging of core section of Toray TGP-H 120™ GDL samples. Furthermore, the effect of the PTFE & binder addition on bulk hydrodynamic properties of GDL samples has also been systematically investigated. PTFE and binder are treated as one solid material.

We have followed the lattice Boltzmann method (LBM) [25] approach for solving the steady-state three-dimensional single-phase fluid flow in the reconstructed geometries. Due to its capability in easily handling complex geometries, the LBM has been used widely to model fluid flow in reconstructed porous structures [17,26–29]. In a recent work, Froning et al. used the same methodology to study the compression effect of the predicted permeability in three-dimensional reconstructed media [30]. In all these works, the porous structure is reconstructed by randomly placing cylindrical fibers in a three-dimensional domain. However, while randomly placing the fibers in the planes, we control the through-plane porosity profile. This enables us to systematically study the effect of porosity heterogeneity on bulk properties. Furthermore, we also investigate the effect of Binder & PTFE addition.

2. Methodology

In this work, steady-state three-dimensional fluid flow was simulated using LBFlow,[1] which is a single-relaxation-time implementation of the LBM [25].

The LBM is a mesoscopic approach for modeling fluid hydrodynamics, where fictitious particles represent fluid volumes. The macroscopic hydrodynamic parameters (*e.g.*, density and velocity) are calculated based on the moments of the distribution function of these fictitious particles on each lattice site. The LBM algorithm includes two steps, namely: (i) streaming step, where the distribution functions move in the direction of their assigned discrete velocity set, and (ii) the collision step, where new distributions are calculated on each lattice site based on pre-defined collision rules. The collision step includes the effect of the external forces and the boundary conditions. The details of the numerical methodology and method validation for LBFlow are presented elsewhere [27,31–33].

The computational domain of the porous region is specified as a three-dimensional binary array of solid and fluid nodes. Physical properties of working fluid are set to those of water at 20 °C. The coordinate system is defined such that the z-direction is aligned with the cross-plane direction, and the x–y plane is the plane parallel to the carbon fibers of the GDLs. A pressure gradient, in the form of a body force, is applied in the direction of each main axis, and the obtained three-dimensional flow field is averaged to yield a volume-averaged velocity vector, $\overline{\boldsymbol{u}}$. The permeability tensor ($\boldsymbol{K}$) is then calculated using Darcy's law, as follows:

$$\overline{u}_i = \frac{K_{ij}}{\mu}\frac{\mathrm{d}p}{\mathrm{d}x_j} \tag{1}$$

where $\overline{u}_i$ is the volume-averaged velocity in the direction i, K_{ij} is the ij^{th} component of the permeability tensor, μ is the working fluid viscosity, and dp/dx_j is the applied pressure gradient in the direction j. Considering the structure of GDL samples in this work, we assumed that off diagonal elements of the permeability tensor (K_{ij}, $I \neq j$) has negligible value in comparison to its diagonal elements (K_{ij}, $I = j$).

[1] Available from http://www.lbflow.co.uk.