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LATTICE BOLTZMANN MODELING OF THE EFFECTIVE THERMAL CONDUCTIVITY OF AN ANISOTROPIC GAS DIFFUSION LAYER

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ABSTRACT

In the present work, the anisotropic and heterogeneous effective thermal conductivity of the gas diffusion layer (GDL) is determined using a thermal lattice Boltzmann model. A three-dimensional fifteen speed (D3Q15), two-phase (fibre and air) model with single relaxation time is presented to determine the effective thermal conductivity in the through-plane and in-plane directions. The model solves the energy transport equation through a reconstructed fibrous GDL geometry. The geometry of the GDL was reconstructed using microscale computed tomography imaging of commercially available GDL materials. Simulated thermal conductivities determined in this work can provide insight into the effect of heterogeneity and anisotropy of the GDL on the thermal management required for improved PEM fuel cell performance.

INTRODUCTION

Thermal management has a crucial contribution to the performance and durability of polymer electrolyte membrane (PEM) fuel cells. Temperature gradients across a single fuel cell or fuel cell stack can be attributed to heat generation from electrochemical reactions and water phase changes. The gas diffusion layer (GDL) plays a vital role in PEM fuel cell thermal management by providing the main pathway for heat removal from the membrane electrode assembly. The effective properties, with the thermal conductivity of GDL in particular, are important for understanding heat transfer and temperature distribution within a PEM fuel cell.

The effective thermal conductivity of the GDL is most commonly determined using ex-situ experiments due to the complexity of coupled processes within an operating fuel cell [1]. Experimental studies have [1-6] largely focused on the determination of the through-plane thermal conductivity and its relation to the GDL structure, residual water content, polytetrafluoroethylene (PTFE) content, and compression. The GDL is a highly porous and stochastic material, which poses

challenges when attempting to use CFD and similar computational methods. Numerical modeling of the GDL thermal conductivity has largely been limited to work with GeoDict™, a commercial simulation software package, and a compact analytical model developed by Sadeghi et al. [6, 7]. Sadeghi et al. developed an analytical model to study the through-plane thermal conductivity over a range of porosity values and under compression. A similar study was presented by the same group [8] to study the in-plane thermal conductivity over a range of PTFE content. Zamel et al. [9] developed a numerical model with GeoDict for the through-plane and in-plane thermal conductivity of dry carbon paper GDL. The effect of porosity, fibre distribution, and compression were studied and a correlation for the effective thermal conductivity of a dry, uncompressed, untreated GDL was proposed. Pfrang et al. [10] calculated the thermal conductivity of three commercially available GDLs with GeoDict based on three-dimensional (3-D) reconstructions from x-ray computed tomography visualizations.

Lattice Boltzmann method (LBM), a mesoscopic approach to fluid dynamics, has recently been shown to be a powerful modeling tool for fluid and heat flow simulation in porous media [11, 12]. Koponen et al. [13], Van Doormaal and Pharoah [14], Hao and Cheng [15], and Nabovati et al. [16] have used LBM to predict permeability of fibrous porous materials. A simplified fluid-solid conjugate heat transfer LBM model developed by Wang et al. [17] was used to study the thermal conductivity of a random porous material [18], a fibrous material [19], the 3-D effect of the thermal conductivity of a porous material [20], a carbon fibre-in-oil material, and phase changing material containing carbon fibres [21]. Conventional methods pose challenges in modeling structural complexities, such as randomly orientated fibres in the GDL structure [21], since the grid refinement needed to reproduce the complex structures is computationally expensive. The mesoscopic approach of LBM provides an appropriate modeling method for

porous GDL and PEM fuel cells due to the ease of implementing solid-fluid boundary conditions in relation to traditional methods.

Recent work by Fishman et al. [22] indicates that the porosity of the GDL is heterogeneous. Burheim et al. [2] have attributed experimentally-observed trends in the through-plane thermal conductivity with the thickness of carbon paper GDL to this heterogeneity. It is important to investigate the effect of heterogeneous porosity distributions on both the through-plane and in-plane effective thermal conductivity of the GDL in more detail. In this work, an LBM model is used to determine the effective through-plane and in-plane thermal conductivity of four commercially available GDL materials using stochastic reconstructions. The effects of GDL compression on the effective thermal conductivity will also be investigated.

METHODOLOGY

Determination of the GDL effective thermal conductivity consists of three steps: i) stochastic reconstruction of the GDL structure, ii) thermal lattice Boltzmann modeling of heat flow through the sample, and iii) using the Fourier heat equation to predict the effective thermal conductivity.

GDL Reconstruction

The LBM simulations presented in this paper are based on numerical reconstructions of carbon fibre paper GDL materials. The fibre orientation within the GDL contributes to anisotropic and heterogeneous material properties with significantly varying transport parameters in the through-plane and in-plane directions. The determination of the transport parameters is further complicated by the stochastic and porous nature of the GDL.

In order to represent the GDL stochastic nature in the geometric reconstructions, a stochastic fibre placement algorithm is employed. The computational domain used is a cuboid with dimensions for the length and width (x and y) of 200 μm . The thickness of the domain (z dimension) varies for the various domains and materials under consideration. The following assumptions are employed with our fibre placement algorithm (commonly found in GDL literature [23]):

- Fibres are straight, cylindrical, and infinitely long.
- Fibres are preferentially oriented in the x-y plane (where the through-plane is defined in the z-direction).
- All fibres have the same diameter.
- Fibres are allowed to intersect.

For our numerical reconstructions, each fibre has a diameter of 8 μm to represent a realistic GDL sample, where the fibres typically have a diameter between 7 to 12 μm [24]. Each fibre has a randomly defined origin position within the domain and extends outwards in the x-y plane at a random angle direction between 0° and 180°. The numerical reconstructions consist of domains with a uniform bulk porosity and heterogeneous porosity distribution. The reconstruction of these domains is further discussed in the subsequent text.

Uniform Bulk Porosity

The four Toray carbon paper GDL materials presented in [22] are reconstructed with a single bulk porosity and domain thickness value. The properties of the four materials are presented in Table 1. The stochastic fibre placement algorithm is used to place the fibres in the cuboid domain. Fibres are added to the domain until the desired porosity value is achieved. A sample domain of the Toray TGP-H-060 is given in Figure 1.

Figure 1: Reconstructed Toray TGP-060 GDL with straight fibres and fibre radius of four lattice units with a uniform porosity

Heterogeneous Porosity Distribution

The heterogeneous through-plane porosity distributions reported in [22] are utilized for the GDL reconstructions. The porosity distributions of the domains are determined by the experimentally obtained heterogeneous through-plane porosity distributions acquired in [22] using x-ray microscale computed tomography (μCT) of the four Toray carbon paper GDL materials (Table 1). The spatial resolution of the μCT data gathered is 2.44 μm , which is sufficiently smaller than the diameter of the fibres used for the reconstructions. The details of the μCT visualization are presented in reference [22].

Table 1: GDL Material Properties

Material	Thickness (μm)	Bulk Average porosity (%)
Toray TGP-H-030	118	82.93
Toray TGP-H-060	220	82.09
Toray TGP-H-090	300	82.60
Toray TGP-H-120	360	78.69

The stochastic fibre placement algorithm is used to construct this domain. The z-component of the fibres is suppressed and discrete layers of fibres with a prescribed porosity are constructed. Fibres are added to each layer to

obtain the appropriate porosity. The layers are stacked in the through-plane direction until the desired material thickness is achieved. Sample GDL reconstructions of the four materials employed for our work are shown in Figure 2.

Figure 2: Reconstructed (a)Toray TGP-030, (b) Toray TGP-060, (c) Toray TGP-090, and (d) Toray TGP-120, GDL all with straight fibres and fibre radius of four lattice units with a heterogeneous porosity distribution from [22].

Lattice Boltzmann Method (LBM)

In this particular work, the model presented by Wang et al. [17] is adopted to simulate the effective thermal conductivity of the GDL using a three-dimensional fifteen speed (D3Q15), two-phase (fibre and air) model. Wang et al. [17] proposed a simplified fluid-solid conjugate heat transfer LBM model and demonstrated its applicability to studying the thermal conductivity of porous media [17-21]. The model is able to account for continuity of temperature and heat flux at the solid/fluid interface required for local thermal equilibrium [25] without the additional computational costs encountered in conventional CFD. The governing equation for pure thermal conduction in a porous two-phase domain is given by:

$$g_{i,\alpha}(\vec{x} + \Delta\vec{x}_i, t + \Delta t) - g_{i,\alpha}(\vec{x}, t) = -\frac{1}{\tau_\alpha} [g_{i,\alpha}(\vec{x}, t) - g_{i,\alpha}^{eq}(\vec{x}, t)] \quad (1)$$

where $\vec{x}$, $\Delta\vec{x}$, t , and Δt are spatial coordinate, lattice spacing, time, and simulation time step, respectively. τ_α is the dimensionless relaxation time that is related to the thermal

conductivity of each phase, α . g_α^{eq} is the equilibrium distribution function of the distribution function, g_α , given as:

$$\begin{aligned} g_i^{eq} &= 0 & i &= 0 \\ g_i^{eq} &= \frac{3}{2} \omega_i T & i &= 1 - 6 \\ g_i^{eq} &= 3 \omega_i T & i &= 7 - 14 \end{aligned} \quad (2)$$

where T is the macroscopic temperature and ω is the weighting factor. The equilibrium distribution equations are independent of the material phase, and only depend on lattice structure and value of local macroscopic temperature. The weighting factors, ω , for the D3Q15 model are:

$$\begin{aligned} \omega_i &= \frac{2}{9} & i &= 0 \\ \omega_i &= \frac{1}{9} & i &= 1 - 6 \\ \omega_i &= \frac{1}{72} & i &= 7 - 14 \end{aligned} \quad (3)$$

Each phase's relaxation time, τ_α , depends on macroscopic thermal conductivity of its respective phase and the lattice parameters. The dimensionless relaxation times for the model are:

$$\tau_f = \frac{9}{5} \frac{k_f}{c^2 \Delta t} + \frac{1}{2} \quad \tau_s = \frac{9}{5} \frac{k_s}{c^2 \Delta t} + \frac{1}{2} \quad (4)$$

where k is the thermal conductivity, c is the physical sound speed, and the subscripts f and s denote the fluid and solid phases, respectively. Values of thermal conductivities for carbon fibres and air at 80 °C are $k_s = 120$ W/m K [5] and $k_f = 0.03$ W/m K [26], respectively. The lattice parameters, Δx and Δt , are chosen to ensure that τ_α remains in the stable range of $0.5 < \tau_\alpha < 2$, indicated by Wang et al. [17]. The lattice spacing (Δx) is calculated using the number of lattice nodes across diameter of the fibres and their physical diameter.

The temperature and heat flux on each lattice node are calculated by:

$$T_\alpha = \sum_i g_{i,\alpha} \quad \vec{q}_\alpha = \left(\sum_i \vec{e}_i g_{i,\alpha} \right) \frac{\tau_\alpha - 0.5}{\tau_\alpha} \quad (10)$$

where e_i is the discrete velocity. Once the temperature and heat flux values are determined on each node, the effective thermal conductivity can be calculated using the Fourier heat equation as follows:

$$k_{eff} = \frac{\delta \int q dA}{\Delta T A} \quad (12)$$

where A is the cross section area, and δ is the thickness of the GDL in the direction of interest, and ΔT is the temperature difference implemented between the two boundaries of the GDL.

Dirichlet or isothermal boundary conditions are imposed on the south ($z = 0$) and north ($z = Z$) walls for the through-plane simulations, and on the side walls ($x = 0$ and $x = X$) for the in-plane simulations. A bounce-back approach of the non-equilibrium distributions was proposed by Zou and He [27] for velocity and pressure boundary conditions. D’Orazio and Succi [28] demonstrated that this approach can be applied to thermal boundary conditions as well, hence, we adopted it in this work to implement the temperature boundary conditions on the two walls of interest. Periodic boundary conditions are implemented in the other two directions.

RESULTS AND DISCUSSION

The LBM method explained in the previous section is used to predict the effective thermal conductivity of GDL samples using 3-D reconstructed geometries. All of the simulations were parallelized using OpenMP and run on the General Purpose Cluster of SciNet supercomputing facilities (Compute Canada).

Validation

The present lattice Boltzmann method has validated for the case of dual component material in a parallel and series configuration in our previous work [29]. Model validation was deemed necessary to ensure that simulations would yield lattice independent results with the GDL reconstructions. The through-plane effective thermal conductivities of the same GDL reconstruction with a bulk uniform porosity value and a lattice spacing of 2.0, 1.3, and 1.0 $\mu\text{m}/\text{lu}$ were determined. A decrease in the lattice spacing causes a subsequent increase in the domain size and number of lattice points across a fibre diameter. The predicted values of through-plane thermal conductivity for different domain sizes are reported in Table 2. The relative deviation is calculated based on the 2.0 $\mu\text{m}/\text{lu}$ lattice spacing case. Since a small deviation is observed between the three cases (less than 1 %), a lattice spacing of 2.0 $\mu\text{m}/\text{lu}$ was chosen for all subsequent simulations.

Table 2: Lattice Spacing Independence Study for LBM Simulations

Domain Size	Lattice Spacing ($\mu\text{m}/\text{lu}$)	Predicted Thermal Conductivity ($\text{W}/\text{m K}$)	Relative Deviation (%)
100×100×100	2.00	0.4640	-
150×150×150	1.33	0.4684	0.948
200×200×200	1.00	0.4674	0.732

A lattice spacing 2.0 $\mu\text{m}/\text{lu}$ yields four lattice points across an 8 μm fibre diameter. A lattice spacing of 2.0 $\mu\text{m}/\text{lu}$ will also yield a computational domain of 590,000 to 1,800,000 grid points, depending on the material under consideration. The isothermal temperatures used for the boundary conditions are non-dimensionalized. The purpose of setting a temperature difference across the domain is to create a heat flux that can be used to calculate the effective thermal conductivity. Normalized

lattice temperatures of 1.5 and 0.7 were implemented in the simulations for the hot wall, T_{hotwall} , and cold wall, T_{coldwall} , respectively.

Heterogeneous Porosity Profiles

Using the GDL reconstructions, the effect of the heterogeneous through-plane porosity distributions on the effective thermal conductivity was investigated. The predictions for the through-plane and in-plane effective thermal conductivity of the four GDL materials reconstructed are given in Table 3. Each value reported in Table 3 is the average of 10 simulations with different reconstructed stochastic domains.

The significant difference between the through-plane and in-plane thermal conductivities highlights the anisotropic nature of the GDL and the significant role that fibre distribution and orientation plays in the determination of the effective thermal conductivity. This trend in the results is in agreement with experimental work by Sadeghi et al. [6, 8] and numerical work by Zamel et al. [9]. Sadeghi et al. [8] reported measurements for the in-plane effective thermal conductivity of uncompressed carbon paper were 12 times higher than for the through-plane effective thermal conductivity. The thermal conductivity of the fibre material is significantly higher than that of air, making them the preferential path for heat conduction throughout the GDL material. The fibre orientation becomes very important for the resulting effective thermal conductivity value, as highlighted by the large difference between the in-plane and through-plane results.

A comparison between the simulation results for the through-plane thermal conductivity for the domains with a uniform bulk porosity and a heterogeneous porosity distribution shows a notably lower thermal conductivity for the case of the heterogeneous porosity distributions (20.1 – 49.5%). The results for the through-plane thermal conductivity for the domains with a heterogeneous porosity distribution also vary with the GDL thickness, or material type, with small changes (between 0.8 – 4.6%) in the bulk porosity. Both of these noted trends show the strong effect that porosity and fibre placement has on the through-plane thermal conductivity of the GDL.

The Toray carbon paper materials display local porosity minima and maxima throughout the thickness of the material. Fishman et al. [22] noted that the heterogeneous porosity distributions for all four GDL materials are distinct but each display three different segments: two transitional surface regions and a core region. The transitional surface region extends linearly between the outer surfaces and the local porosity minima and the core area is between the two transitional surface regions. Even though bulk porosities are the same, the through-plane thermal conductivity differences between the two GDL reconstructions can be attributed to the minima porosity regions in the heterogeneous porosity distributions. Through-plane thermal conductivity differences within the four materials with heterogeneous porosity distributions can be attributed to differences in the porosity distributions of the different GDL materials. Since through-plane thermal conductivities are strongly dependant on porosity

Table 3: LBM Simulation results for the effective thermal conductivity. Values given in parentheses are one standard deviation.

Material	Through-plane Effective Thermal Conductivity			In-plane Effective Thermal Conductivity	
	Bulk Average Porosity	Uniform Porosity	Heterogeneous Porosity	Uniform Porosity	Heterogeneous Porosity
TGP-H-030	0.8192	0.46 (0.07)	0.23 (0.04)	3.4 (0.4)	3.6 (0.3)
TGP-H-060	0.8060	0.6 (0.1)	0.4 (0.1)	6.6 (0.6)	6.7 (0.2)
TGP-H-090	0.8125	0.5 (0.3)	0.3 (0.1)	7 (1)	7.7 (0.4)
TGP-H-120	0.7770	1.0 (0.1)	0.78 (0.07)	13.3 (0.8)	13.7 (0.4)

and only one sample of each material was assessed in this study, future work should include a larger number of samples to fully characterize reported trends.

A difference in the results for the thermal conductivity in the in-plane direction between the two GDL reconstructions is not observed; however, the in-plane thermal conductivity was noted to increase with increasing material thickness. Increasing the thickness of the GDL increases the number of pathways for heat transfer, indicating thermal conductivity is dependent on fibre orientation within the GDL. Future work should include a more complete study on the effect of the material thickness on the in-plane thermal conductivity over a range of porosities.

CONCLUSION

In this work, the anisotropic effective thermal conductivity of four commercially available Toray carbon paper GDL materials was determined using a two-phase conjugate fluid-solid thermal LBM model. The model was used to study the effects of heterogeneous porosity distributions presented in [22] and GDL compression on the through-plane and in-plane thermal conductivities of numerically reconstructed GDL materials. It was shown that the through-plane and in-plane thermal conductivities strongly depend on the porosity and fibre orientation. The anisotropic nature of the GDL was emphasized by the results indicating the in-plane thermal conductivity was over an order of magnitude higher than the through-plane for all domains investigated. The heterogeneous porosity distributions had an effect on the through-plane thermal conductivity, resulting in lower values (20.1 – 49.5%) than simulations that used a uniform bulk porosity value. The in-plane thermal conductivity was not affected by the heterogeneous porosity distribution, but did increase with increasing material thickness.

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