Modeling the Effective Thermal Conductivity of an Anisotropic and Heterogeneous Polymer Electrolyte Membrane Fuel Cell Gas Diffusion Layer

by

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Abstract

In this thesis, two numerical modeling methods are used to investigate the thermal conductivity of the polymer electrolyte membrane (PEM) fuel cell gas diffusion layer (GDL). First, an analytical model is used to study the through-plane thermal conductivity from representative physical GDL models informed by microscale computed tomography imaging of four commercially available GDL materials. The effect of the heterogeneity of the through-plane porosity of the GDL and polytetrafluoroethylene (PTFE) treatment is studied and it is noted that the high porosity surface transition regions have a dominating effect over the addition of PTFE in impacting the overall thermal conductivity. Next, the lattice Boltzmann method (LBM) is employed to study both the in-plane and through-plane thermal conductivity of stochastic numerically generated GDL modeling domains. The effect of GDL compression, binder content, PTFE treatment, addition of a microporous layer (MPL), heterogeneous porosity distributions, and water saturation on the thermal conductivity are investigated.
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Abbreviations & Nomenclature

Acronyms

CFD  computational fluid dynamics
DM   diffusion media
EDS  energy dispersive X-ray spectrometry
GDL  gas diffusion layer
LBM  lattice Boltzmann method
lu   lattice unit
MPL  microporous layer
PEM  polymer electrolyte membrane
PTFE polytetrafluoroethylene
PTL  porous transport layer
SEM  scanning electron microscopy
µCT  micro-computed tomography

Variables

A    cross-sectional area
a,b  major and minor semi-axis of elliptical contact area
C    contact point
C1,C2,C3  D3Q15 predicted thermal conductivity fitting parameters
c    physical sound speed
c_s  lattice sound speed
d    fibre diameter
E    Young’s modulus
e    discrete velocity set of energy particles
F    force
g    distribution function
g_eq equilibrium distribution function
H    height of domain
k    thermal conductivity
L    length of domain
1  length of fibre
N  number of fibres
P  pressure
q  heat flux
R  thermal resistance
T  temperature
t  time
Δt  simulation time step
u  fluid velocity
V  volume
W  width of domain
x  spatial coordinate
Δx  lattice spacing

**Greek Letters**

α, β  parameters used to describe PTFE location in analytical model
γ  compression ratio
δ  thickness of the GDL in the direction of interest
ε  porosity
f(ε)  structural function
K(η)  complete elliptical integral
θ  orientation angle
Λ  number of layers
λ  PTFE weight content
ξ  strain
ρ  density ratio
τ  dimensionless relaxation time
ω  weighting factor
Chapter 1 Introduction

1.1 Preamble
Polymer electrolyte membrane (PEM) fuel cells are promising clean energy conversion technologies for portable, automotive, and stationary applications due to their potential for high power density at low operating temperatures. Achieving proper heat management within a PEM fuel cell is critical for improving its performance and lifetime. Numerical modeling is a powerful means of designing the PEM fuel cell, and in particular, designing the microscale features of individual components. However, accurately modeling the heat transfer rates and temperature distributions within a fuel cell requires knowledge of the thermal transport properties of individual materials, in particular, the effective thermal conductivity. The main path for heat removal from the PEM fuel cell membrane to the current collectors is through the gas diffusion layer (GDL) and the rate of heat removal is therefore largely dependent upon the thermal transport properties of the GDL.

1.2 Motivation and Objective
The main objective of this study is to use numerical modeling methods to investigate the thermal conductivity of the PEM fuel cell GDL. The two modeling approaches that are investigated are an analytical model and a thermal lattice Boltzmann model. The analytical model is used to study the through-plane thermal conductivity. The effect of the heterogeneity of the through-plane porosity of the GDL and polytetrafluoroethylene (PTFE) treatment on the thermal conductivity is studied. Next, the lattice Boltzmann method (LBM) is used. A thermal fluid-solid conjugate heat transfer model capable of handling multiple phases is employed to study both the in-plane and through-plane thermal conductivity. The effect of GDL compression, binder content, PTFE treatment, addition of a microporous layer (MPL), heterogeneous porosity distributions, and water saturation on the thermal conductivity are investigated.

1.3 Contributions
The work presented in this thesis has led to the following contributions: two published journal papers, one submitted manuscript, and one manuscript in preparation:


Yablecki, J., Bazylak, A. “Modeling the Anisotropic Effective Thermal Conductivity of a Gas Diffusion Layer in a Polymer Electrolyte Membrane Fuel Cell with Modifications using the Lattice Boltzmann Method” (In Preparation)

1.4 Organization of Thesis

This thesis is organized into six chapters. In this first chapter, a general introduction, motivation and objective are given. Chapter 2 provides an in-depth literature review on thermal conductivity measurements of the GDL, including both experimental and numerical approaches, as well an introduction to LBM and the development of the thermal LBM framework used in this thesis. A brief background on PEM fuel cells and the thermal and water management issues they face is also presented. In Chapter 3, the through-plane thermal conductivity is investigated with an analytical model. The effect of the heterogeneity of the through-plane porosity on the thermal conductivity is investigated for untreated and PTFE-treated carbon paper GDLs. In Chapter 4, a thermal lattice Boltzmann model is used to study the through-plane and in-plane thermal conductivity of numerically generated stochastic modeling domains of the GDL. The model is used to investigate the effect of GDL compression, binder and PTFE content, addition of the MPL, water saturation, and through-plane porosity heterogeneity on the thermal conductivity. A conclusion and future works are presented in the last two chapters.
Chapter 2 Background and Literature Review

2.1 Introduction
In this chapter PEM fuel cells are introduced and a description of the GDL and heat transfer within PEMs is presented. Previous studies on the effective thermal conductivity of the GDL, both experimental and numerical, are discussed. An overview of LBM and previous studies with modeling of the GDL is reviewed. Lastly, the development of the thermal LBM framework employed in this thesis is examined.

2.2 Polymer Electrolyte Membrane Fuel Cells
PEM fuel cells are promising clean energy conversion technologies for portable, automotive, and stationary applications due to their potential for high power density at low operating temperatures. Hydrogen and oxygen PEM fuel cells electrochemically produce electricity with heat and water as the only by-products (see Figure 2.1). Despite the promise of PEM fuel cells, cost, durability, and reliability still remain the largest challenges to their design, operation, and introduction into the commercial market. In particular, achieving coupled thermal and water management remains an important challenge in the overall performance and durability of the fuel cell.

2.2.1 Gas Diffusion Layer
The PEM fuel cell GDL, otherwise known as the diffusion medium (DM) or porous transport layer (PTL), which is typically composed of a carbon-based fibrous textile, plays a key role in achieving proper thermal and water management. The GDL is porous to allow for reactant and product gases to diffuse to and from the catalyst layers and current collecting bipolar plates. The GDL is also hydrophobically treated with PTFE to promote the removal of excess liquid water. Although excess water can inhibit reactants from reaching the catalyst site, membrane dehydration can occur when there is an inadequate amount of water. A MPL is often added at the GDL/catalyst interface to aid in cell performance by assisting in liquid water management [1]. The MPL is composed of carbon or graphite particles mixed with a polymeric binder, which is usually PTFE.
2.2.2 Heat Transfer in PEM Fuel Cells

Proper heat management within the fuel cell is critical for its performance and lifetime. Coupled thermal and water management have been the focus of a number of non-isothermal PEM fuel cell models [2-7]. The two processes are inherently coupled as water phase change is temperature dependent, and the evaporation and condensation processes involve the absorption and release of latent heat, respectively. Heat produced from the electrochemical reaction and water phase change results in temperature gradients across a single cell and fuel cell stack. The temperature within the fuel cell can affect the relative humidity, membrane water content, saturation pressure, and reaction kinetics. Knowledge of the temperature throughout the cell is important for avoiding membrane dehydration and degradation at elevated temperatures and detrimental humidity levels [8]. Nafion membranes can be damaged and have significant and permanent decreases in ionic conductivity at elevated temperatures [9].

2.3 Effective Thermal Conductivity

Modeling of the heat transfer and temperature distribution in a fuel cell requires the knowledge of the thermal transport properties such as the thermal conductivity and thermal contact resistance. The main path from heat removal from the PEM membrane to the current collectors is through the GDL and the rate of heat removal is therefore largely dependent upon the thermal transport properties of the GDL [10].

The effective thermal conductivity is a property of porous media that accounts for the contributions of the thermal conductivity of each phase [11]. For determination of the effective thermal conductivity of the GDL, the two phases that are considered are carbon fibres and air with values of thermal conductivity of 120 W/m K and 0.03 W/m K, respectively [12]. Residual water in the GDL, the addition of a MPL and PTFE treatments also affect the value of the effective thermal conductivity.

Until recently, isotropic thermal transport parameters, in particular an isotropic effective thermal conductivity of the GDL, have been assumed in numerical modeling studies [11]. However, the GDL has been shown to be an anisotropic and heterogeneous [13] material with transport properties that vary significantly between the through-plane and in-plane directions. Figure 2.2
shows two SEM images of a Toray TGP-H-090 carbon paper GDL, where the anisotropic nature of the GDL can be seen from the two orientations shown.

In 2006, Pharoah et al. [11] surveyed 100 fuel cell modeling papers and found that every non-isothermal model employed a volume averaging, and hence isotropic, thermal conductivity. In recent work, Pfrang [14] et al. found there to be little data available on anisotropic thermal conductivity for the GDL and inconsistent values used for an isotropic thermal conductivity. They surveyed a number of non-isothermal fuel cell models and found values for the isotropic thermal conductivity to range from 0.15 W/m K to 65 W/m K. Pasaogullari et al. [7] first reported a non-isothermal two-phase PEM fuel cell model that considered the anisotropic properties of the GDL. The GDL anisotropy was reported to have significant impact on the temperature distribution and liquid water transport in the fuel cell.

2.3.1 Experimental Determination of Thermal Conductivity

The effective thermal conductivity of the GDL can be determined experimentally in-situ from the temperature distribution of an operating fuel cell [15], or from ex-situ experiments. In-situ experiments are not common due to the complexity of coupled processes within an operating fuel cell [8]. For ex-situ experiments transient and steady-state measurements are possible. Transient thermal conductivity measurements include laser flash radiometry techniques, and often require knowledge of other material properties including the density and heat capacity. For a porous and stochastic material like the GDL, where these properties are not readily known, steady-state experiments are more common due to the reliability of results and ease of measurement.

Ex-situ measurements generally consist of a constant one-dimensional heat flux applied to the GDL. Thermocouples and flux meters are used to measure the temperature and heat flux. The thermal conductivity is then calculated from Fourier’s law. The thickness of the GDL must also be measured and the precision of this measurement can have a large impact on the accuracy of the thermal conductivity measurements [8].

The first experimental measurements of the thermal conductivity of the GDL were reported by Vie and Kjelstrup in 2003 [15]. Since then a large number of experimental works [8, 16-20] have focused on through-plane effective thermal conductivity of the GDL and the effect that GDL material, residual water content, PTFE content, temperature, and compression have on
effective values of thermal conductivity. Experimental work on the in-plane thermal conductivity has been limited to two recent studies by Teestra et al. [21] and Sadeghi et al. [22].

2.3.2 Numerical Modeling and Thermal Conductivity

The GDL is a highly porous and stochastic material, and numerical modeling of the thermal conductivity has largely been limited to work with the GeoDict™, a commercial simulation software package, and a compact analytical model developed by Sadeghi et al. [23]. The compact analytical model developed by Sadeghi et al. was used to predict the through-plane [20, 23] and in-plane [22] effective thermal conductivities of non-woven carbon fibre paper GDL materials taking into account the effect of fibre compression. Pfrang et al. [14], Zamel et al. [12, 24, 25] and Veyret et al. [26] employed GeoDict to determine the anisotropic thermal conductivity of the GDL. Zamel et al. [24] employed GeoDict to construct a three-dimensional (3-D) pore morphology of dry carbon paper GDL with no PTFE by providing the geometric parameters of the GDL as the initial conditions. The work was extended by the same group to consider the effect of water saturation [25] on the thermal conductivity and to determine the thermal conductivity of the MPL by generating a model of its microstructure [12].

2.4 Lattice Boltzmann Method

The LBM is based on the discretization of the Boltzmann transport equation in space, time, and velocity of the particles under study. LBM is considered a particle-based mesoscopic method, as it can describe the time evolution and interaction of quasi particles of different natures [27]. The behaviour of these fictitious quasi-particles, as they move on a lattice and collide with each other, is determined in two distinct steps, namely: streaming and collision. By predicting the distribution of these particles in space and time, the macroscopic physical parameters can be predicted as functions of their distribution.

A large advantage to LBM is in its application for the study of flow in porous media or complex geometries [28, 29]. Along with having comparable accuracy to Navier-Stokes-based computational fluid dynamics (CFD) methods [30-32], the LBM’s inherent locality makes it suitable for distributed parallel computing [33]. The implementation of boundary conditions, even complex ones, is greatly reduced with LBM. Koponen et al. [34, 35], Van Doormaal and
Pharoah [27], and Nabovati et al. [36] have used the LBM to predict permeability of the fibrous porous materials. Mukherjee et al. [37, 38] used LBM to study the two-phase flow in PEMFCs from stochastic reconstructions of the catalyst layer and GDL. A 3-D two-phase LBM approach was used to model liquid water movement and flooding inside the structure.

Research in the area of thermodynamics using the LBM began in 1993. Prior to the introduction of the model presented by He et al. [39] in 1998, two approaches for thermal LBM had been used: the passive scalar and the multi-speed approaches. Both methods proved to be complicated to implement, and the multi-speed approach was constricted to a small temperature range with significant stability problems [39]. He et al. [39] introduced an energy double distribution model that could account for the viscous heat dissipation and compression work done by pressure that the passive scalar approach could not recover. It also solved the problem of numerical stability that was present in the multi-speed approach [39]. The model includes the derivation of two discrete evolution equations: the density distribution found in fluid LBM to account for the density and velocity fields, and an internal energy or thermal distribution to account for the temperature field, leading to an approach with double storage requirements [39].

Peng et al. [40] reduced the complexity of the model of He et al. [39] by introducing a model for incompressible flows that neglects the viscous and compressive terms. Following this work, Wang et al. [41] proposed a simplified fluid-solid conjugate heat transfer model that used a generalized heat source in a thermal LBM framework. Their work considered 2-D fluid-solid conjugate heat transfer for an incompressible fluid and laminar flow with a single relaxation time collision operator. The model presented by Wang et al. [41] accounted for continuity of temperature and heat flux at the solid/fluid interface required for local thermal equilibrium [42] without the additional computational costs encountered in conventional CFD. This model was extended by the same group to look at the effective thermal conductivity of a random porous material [43] and a fibrous material [44] in two dimensions. This model was also extended to study the 3-D effect of the thermal conductivity of a porous material [45], a carbon fibre-in-oil material and phase changing material containing carbon fibres [46]. The results from these studies [44-47] were in good agreement with previously reported literature results.
2.5 Figures

Figure 2-1: PEM Fuel Cell Schematic.
Figure 2-2: SEM image of Toray TGP-H-090 carbon paper GDL (a) side view and (b) top view.
Chapter 3 Analytical Model

3.1 Introduction

In this chapter, an analytical model is used to determine the effective through-plane thermal conductivity of commercially available GDL materials based on the heterogeneous porosity profiles previously published in [13, 48]. The effects of the heterogeneous porosity distribution, GDL compression, and PTFE content on the effective thermal conductivity are investigated. The results of the model are compared with experimental data presented by Burheim et al. [16].

3.2 Motivation and Objective

The rate of heat removal in a PEM fuel cell is largely dependent upon the thermal transport properties of the GDL. Although a number of analytical correlations exist for the effective thermal conductivity of a composite material with varying geometries [42], the GDL thermal conductivity can be estimated but not accurately represented by a single analytical correlation due to the anisotropic [19] and heterogeneous nature [13] of the fibres within the GDL. Burheim et al. [16] attributed experimentally found trends in the through-plane thermal conductivity with the GDL thickness of carbon paper materials to this heterogeneity.

Through a compact analytical model of the GDL, Sadeghi et al. [23] calculated the through-plane effective thermal conductivity of the GDL using an idealized repeating unit cell to represent the GDL structure. In this chapter, this analytical model is extended to consider a more stochastic geometry. Heterogeneous porosity profiles of commercially available carbon paper GDL materials previously published in [13, 48] are employed to create representative physical GDL models.

3.3 Model Development

The approach to determining the effective through-plane thermal conductivity of the GDL consists of two steps: the representative physical GDL model and the thermal resistance model. Based on the findings of Sadeghi et al. [9, 10] where heat transfer was found to be dominated by fibre-to-fibre contact, the analytical model will only consider heat transfer through fibre-to-fibre
contact points. Using inputs of porosity, GDL compression pressure, and in the case of treated materials, PTFE distributions, the representative physical GDL model is used to determine the number of fibre-to-fibre contact points throughout the domain and the contact area dimensions of the contact points. These dimensions are then used as inputs to the thermal resistance model for determining the effective thermal conductivity of the GDL.

### 3.3.1 Fibre Representation

To model the effective through-plane thermal conductivity of the GDL, a representative modeling domain, shown in Figure 3.1, with width, $W$, and length, $L$, was constructed for a number of randomly oriented fibres, $N_t$, in the $x$-$y$ plane. The fibres are immersed in quiescent air, each with a length and diameter of $l$ and $d$, respectively. Layers of the fibres are stacked vertically through the entire thickness, $H$, of the GDL modeling domain. The geometric parameters of the modeling domain are shown in Table 3.1.

The through-plane porosity distributions reported in [13] of four GDL materials are employed in this work to establish the number of fibres, in each layer, $t$, of the modeling domain. The porosity of each layer can be found from:

$$\varepsilon_t = 1 - \frac{V_{solid}}{V_{total}}$$  \hspace{1cm} (3.1)$$

where $V_{solid}$ is the total volume of the solid carbon fibres, and $V_{total}$ is the total volume of modeling domain. Based on the volume of the modeling domain used and the prescribed porosity of each layer, the number of fibres required in each layer, $N_t$ is determined from:

$$N_t = (1 - \varepsilon_t) \frac{LWt}{\pi d^2} \frac{4}{l}$$  \hspace{1cm} (3.2)$$

where $d$ and $l$ are the diameter and length of the carbon fibre, respectively.

In the model, the number of contact points, $C_f$, for a single fibre in layer, $t$, and the adjacent layer, $t+1$, is a function of the porosity of both layers (i.e. number of fibres in each layer, $N_t$) and the orientation angle of the fibre, $\theta$. The fibres are oriented in the modeling domain with a uniform distribution of randomly chosen angles between 0 and 90° (in the $X$-$Y$ plane). The maximum number of contact points for a given fibre in each layer occurs when the fibre orientation angle is 90°, as shown in the periodically ordered fibre arrangement (Figure 3.2). The
minimum number of contact points is seen when $\theta$ is $0^\circ$ between the two layers. A non-linear relationship was determined for the number of contact points for a single fibre in a layer with porosity, $\varepsilon$, based on its orientation angle (Figure 3.3a). The average number of contact points for a single fibre, $C_f$, in a layer with porosity, $\varepsilon$, was determined from this relationship and is used as an input into the model. The average number of contact points, $C_{f,\text{avg}}$, was found to vary linearly with porosity and is interpolated for each porosity value, $\varepsilon$, (Figure 3.3b), when generating the thermal resistance model.

3.3.2 Compression

In practice the PEM fuel cell is compressed during assembly. It is important to consider the effect of this compression on both the individual carbon fibres and on the overall GDL. Here, it assumed that the fibres do not bend under compression, and the only deformation of the fibres is at the contact area between two fibres. The application of a load will create a contact area between two fibres that is finite but very small when compared with the dimensions of the fibres [17]. The overall thickness of the GDL will change due to compression. These considerations are described in detail in the following subsections.

3.3.2.1 Fibre Compression

When two solids are in contact in reality, an imperfect interface is formed as surfaces are neither perfectly smooth nor flat [49]. The solids under consideration in this work are cylindrical and as such, will create a non-conforming interface. Surface roughness can also cause imperfect contact between two solids as microgaps will appear where there is an absence of solid-to-solid contact. For simplicity in this model, the solids are assumed to be perfectly smooth, and the presence of microgaps is ignored. Based on the assumption that both fibres are smooth, a smooth, non-conforming contact area between the two fibres is formed, which is a function of the force applied on the fibres, $F$, the Young’s modulus of the fibre (210 GPa), $E_f$, and the angle between the two fibres, $\theta$. The material properties of the fibres are shown in Table 3.2. When cylindrical fibres contact each other eccentrically, the contact region is considered elliptical [49]. The Hertzian theory of contact is used to predict the shape of contact between solids and how it grows under an increasing load [49]. Following the analytical modeling approach for the GDL presented by Sadeghi et al. [23], the application of the Hertzian contact theory for non-
conforming smooth cylinders is used to define the elliptical contact region, described by the major and minor semi axis, \( a \) and \( b \). The detailed formulation for this theory can be found in Sadeghi et al. [9] and Johnson [17]. In reality, microgaps may decrease the effective thermal conductivity of the GDL.

It is assumed that fibres will not bend and new contact points will not be created under compression. However, under compression, it is assumed that the existing contact area between two fibres will increase. The contact load, \( F \), for each contact point can be expressed in terms of the pressure within the GDL, \( P_{GDL} \), the cross-sectional area of the unit cell defined by \( L \) and \( W \), and the number of contact points for a given layer, \( C_t \) [23]:

\[
F_{max,t} = \frac{P_{GDL} LW}{C_t} \tag{3.3}
\]

### 3.3.2.2 GDL Compression

In this work, the porosity profiles and material thicknesses for the uncompressed materials measured in [13] are employed. To account for the compression of the GDL, Hooke’s law is used to determine the compressed thickness of the material. Assuming the deformation of the GDL is elastic, \( \xi \), the strain from the GDL compression is given by:

\[
\xi = \frac{P_{GDL}}{E_{GDL}} \tag{3.4}
\]

where \( E_{GDL} \) is the Young’s modulus of the GDL material. The compressed thickness, \( H_c \), is determined from:

\[
\xi = \frac{H_o - H_c}{H_o} \tag{3.5}
\]

where \( H_o \) is the uncompressed thickness of the GDL. A value of 17.9 MPa for the Young’s modulus of Toray carbon paper was used [1].

It is assumed that compression is applied uniformly within the GDL. It is important to note that, this is not the case during fuel cell operation as there will be higher compression in areas under the lands of the bipolar plate than in areas under the channels [8].

### 3.3.3 PTFE Treatment

For the GDL materials considered that have PTFE treatment, the method of incorporating the
PTFE in the analytical model is adopted from previous modeling of the effective thermal conductivity in the in-plane direction by Sadeghi et al. [22]. The bulk porosity with the addition of PTFE, $\varepsilon$, is calculated with the general form of the equation:

$$\varepsilon = \varepsilon_0 - \rho \frac{\lambda(1 - \varepsilon_0)}{(1 - \lambda)} \quad (3.6)$$

where $\varepsilon_0$ is the bulk porosity of the material before PTFE is added, $\lambda$ is the weight fraction of PTFE, and $\rho$ is the density ratio between carbon fibre and PTFE [50]. A value of 0.9 is used for $\rho$ [1].

In recent experimental work by Rofaiel et al. [51], the relative through-plane distribution of PTFE in Toray-TGP-H-090 was quantified using scanning electron microscopy (SEM) energy dispersive X-ray spectrometry (EDS) imaging. Results from this work revealed an almost symmetrical presence of PTFE in the paper GDL material with a lower accumulation in the centre region and peaks of accumulation towards the surfaces. The experimentally measured PTFE distributions from the previously published work of Rofaiel et al. [51] are employed in this model to inform the non-uniform distribution of PTFE.

The total weight fraction of PTFE in a GDL sample, $\lambda$, is known a priori from supplier specifications, and when combined with the relative through-plane distribution of PTFE provided by Rofaiel et al. [51], the heterogeneous distribution of PTFE in the through-plane direction of the GDL can be extracted. In other words, the non-uniform weight fraction of PTFE at each layer, $\lambda_t$, in the through-plane direction of the domain is determined for this investigation from supplier specifications combined with the measurements reported in [51].

To fully define the modeling domain, the number of fibres, $N_t$, as well as the weight fraction of PTFE, $\lambda_t$, must be prescribed at each through-plane position (layer) of the GDL. The through-plane porosity distributions reported in [13, 48] were employed; however, Fishman et al. reported porosity distributions that accounted for both PTFE and carbon fibres. Therefore, in order to extract $N_t$, it is first necessary to determine the porosity at each through-plane position of the material in the absence of PTFE addition (fibre porosity), $\varepsilon_{0,t}$, by using Equation 5. Once $N_t$ is extracted, the same procedure for Fibre Representation and Fibre Compression outlined in Sections 3.3.1 and 3.3.2.1 for untreated GDL materials can be followed.
3.3.4 Thermal Model

The effective thermal conductivity is a porous media property that accounts for the contributions of the thermal conductivity of each phase present [11]. For the determination of the effective thermal conductivity of the GDL, the two phases that are generally considered are solid carbon fibres and gaseous air with values of thermal conductivity of 120 W/m K and 0.03 W/m K, respectively [24].

For the analytical model presented here, it is assumed that the only heat transfer is steady-state, one-dimensional (1-D) conduction in the through-plane direction of the GDL. With the calculation of the Grashof and Peclet numbers, Ramousse [19] showed that natural convection and convective heat transfer are negligible compared with conduction in the GDL. Radiative heat transfer can be neglected for temperatures below 1000 K [52], which is well above the operating range of a PEM fuel cell. Therefore, heat is transferred through the modeling domain from fibre-to-fibre contact only. The thermal resistance in the conduction along a fibre is ignored, as it is assumed to be negligible compared with the thermal constriction and spreading resistances [23].

A thermal resistance network or circuit can be constructed for 1-D heat transfer with no internal energy generation and with constant properties [53]. The thermal resistance, $R_{t,\text{cond}}$, for conduction in a plane wall is defined by:

$$R_{t,\text{cond}} = \frac{\Delta T}{q} = \frac{L}{kA} \quad (3.7)$$

where $\Delta T$ is the difference in temperature across the wall, $q$ is the heat flux, $L$ is the length of the plane wall, $k$ is the thermal conductivity of the wall, and $A$ is the cross sectional area of the wall. An equivalent thermal circuit with thermal resistances in parallel and series is analogous to an electrical circuit governed by Ohm’s law.

The dominant thermal resistance for heat conduction through the GDL is the thermal constriction and spreading resistance [20]. The thermal spreading resistance, $R_{sp}$, is equivalent to the thermal constriction resistance, $R_{co}$, and accounts for the thermal energy that is transferred between the two fibres at the contact interface. When the dimensions of the contact area are very small compared with the dimensions of the contacting bodies, the heat transfer through the contact area is constrained, and the solution can be modeled as a heat source on a half-space [54]. Heat
entering the half-space is constricted to flow through the small contact area (thermal constriction resistance), and heat leaving the half-space spreads out from the contact area (thermal spreading resistance) [54]. In the case of the GDL, the contact area between the two fibres is considered as the heat source, and the much larger carbon fibres are considered as the half space.

The thermal constriction resistance is then a function of the elliptical contact area calculated with the Hertizan contact theory and described by the major and minor semi-axis of the elliptical contact area, $a$ and $b$, respectively. Using the work of Yovanovich [55] that resulted in half-space solutions for different contact interfaces, Sadeghi et al. [23] expressed the thermal constriction resistance analytically to be:

$$ R_{co} = \frac{1}{2\pi k_s a} K(\eta) \quad (3.8) $$

where $K(\eta)$ is the complete elliptical integral and a function of the contact area dimensions, $a$ and $b$. The total thermal resistance at a fibre contact point, $R_{cp}$, is given by:

$$ R_{cp} = R_{co} + R_{sp} \quad (3.9) $$

The total thermal resistance for each layer, $R_t$ (Figure 3.4a), within the modeling domain can be found from the summation of each individual thermal resistance of each contact point in parallel:

$$ \frac{1}{R_t} = \sum_{0}^{c_t} \frac{1}{R_{cp}} \quad (3.10) $$

The total resistance in the modeling domain, $R_{total}$ (Figure 3.4b), can be found as a series summation of the thermal resistances of each layer, for a total of $A$ layers in the domain:

$$ R_{total} = \sum_{0}^{A} R_t \quad (3.11) $$

The effective thermal conductivity can be found from the total thermal resistance:

$$ k_{eff} = \frac{t_{total}}{R_{total}A} = \frac{H - t}{R_{total}WL} \quad (3.12) $$
When considering GDL materials treated with PTFE, the approach outlined above is followed, and only heat transfer through fibre-to-fibre contact is considered. The PTFE distribution within the GDL can vary with method of application, but for the purpose of this analytical model only PTFE that has accumulated at fibre contact points will be considered. An equivalent thermal resistance at each fibre-to-fibre contact point is constructed to include the addition of PTFE. The location of PTFE at a contact point and the equivalent thermal resistance diagram is given in Figure 3.5. The total thermal resistance at a fibre contact point, \( R_{cp} \), is given by:

\[
R_{cp} = \left[ (R_{PTFE})^{-1} + (R_{co} + R_{sp})^{-1} + (R_{PTFE})^{-1} \right]^{-1} \quad (3.13)
\]

The thermal resistance of the PTFE is given by:

\[
R_{PTFE} = \frac{1}{\pi k_{PTFE}} \left[ \int_{\alpha}^{\beta} \frac{u \, du}{1 - \sqrt{1 - u^2}} \right]^{-1} \quad (3.14)
\]

where \( \alpha \) and \( \beta \) are parameters developed by Sadeghi et al. to describe PTFE [22]. The proposed relations from Sadeghi et al. [11] describe the geometry of the portion of PTFE that has accumulated at a contact point from SEM images of carbon paper GDL with varying PTFE wt. content. The thermal conductivity of PTFE is 0.649 W/m K [22]. The parameter, \( \alpha \), gives the radius ratio of contact area to fibre area. A value of 0.1 was estimated from SEM images by Sadeghi et al. [22] and is also used in this study. The parameter \( \beta_t \) is the radius ratio of PTFE to carbon fibre and varies with the amount of PTFE with the following relationship in the through-plane direction [22]:

\[
\beta_t = 0.25 + 3(\lambda_t - 0.05) \quad (3.15)
\]

where \( \lambda_t \) is weight fraction of PTFE at each layer in the through-plane direction. As before, the total thermal resistance for each layer, \( R_t \), within the modeling domain can be found from the summation of each individual thermal resistance of each contact point in parallel. The total resistance in the modeling domain, \( R_{total} \), can be found as a series summation of the thermal resistances of each layer.
3.4 Results

The analytical model was implemented with heterogeneous through-plane porosity distributions for Toray carbon paper TGP-H-030, 060, 090, and 120 obtained through x-ray microscale computed tomography (µCT) experiments conducted in [13, 48]. The GDL materials considered were uncompressed and did not have a microporous layer (MPL). The spatial resolution for the µCT data gathered is 2.44µm [13, 48]; however, for the purpose of this model, measurements for porosity are interpolated from the experimentally determined data set at every 7.32 µm through the thickness of the GDL. The details of the microscale computed tomography visualization are presented in references [13, 48].

3.4.1 Untreated Paper

The effective through-plane thermal conductivity obtained from the analytical model is shown in Table 3.3 for three compression pressures, $P_{bp}$. The effective thermal conductivity results for varying compression pressures from the analytical model are also shown in Figure 3.6 and compared with experimental data for Toray carbon paper GDL materials presented by Burheim et al. [16]. The results from the analytical model are in agreement within 11.6%, 7.6%, and 4.0% of the experimentally obtained results from [16] for Toray TGP-H-060, 090, 120, respectively averaged over the range of compression pressures. Similar trends can be seen between both sets of data, as described below.

The effective thermal conductivity is observed to increase with increasing compression pressure. As noted by Burheim et al. [16], and shown in Figure 3.6, the effective thermal conductivity increases almost linearly with increasing compression pressure. The results from the analytical model also display another trend noted in the experimental work by Burheim et al. [16] for Toray carbon GDL material with varying thickness. As shown in Figure 3.6, the effective thermal conductivity increases with increasing GDL thickness (even as the average bulk porosity remains approximately constant) for all three GDL materials presented by Burheim et al. [16]. The results presented in Figure 3.6 for the analytical model follow this trend except for Toray TGP-H-060 and 090. The effective thermal conductivity of Toray TGP-H-060 is an average of 3% higher than Toray TGP-H-090. To further explore the reason for this trend, the effective thermal conductivity for each GDL material is presented as a function of through-plane position in Figure 3.7.
As shown in Figure 3.7, the thermal conductivity through the thickness of the GDL is strongly dependent upon the local value of the porosity, $\varepsilon_t$. For all four GDL materials presented, the Toray carbon paper materials display local porosity minima and maxima throughout the thickness [13]. At the location of a porosity minimum, a maximum local value of thermal conductivity is observed. The four materials investigated have a local maximum value of thermal conductivity between 2.5 and 3.0 W/m K.

Fishman et al. [13] noted that the heterogeneous porosity distributions for all four GDL materials are distinct and consist of three 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 region is between the two transitional surface regions (see Figure 3.8) [13]. After isolating the core region in the analytical model, the effective thermal conductivity of this region was determined. Core region thermal conductivity results are presented in Table 3.4 for a single compression pressure, $P_{bp}$, of 460 kPa and compared with the overall bulk effective thermal conductivities. The core thermal conductivity is 7.79, 2.87, 3.74, and 3.31 times higher than the overall bulk thermal conductivity for Toray TGP-H-030, 060, 090, and 120, respectively. Unlike the overall bulk thermal conductivity, the thermal conductivity of the core region is not dependent upon the material thickness.

### 3.4.2 PTFE Treated Paper

The analytical model was implemented with heterogeneous through-plane porosity distributions for Toray carbon paper TGP-H-060 obtained in [13, 48] for four PTFE weight contents; 0, 5, 10, and 20 wt. %. The effective through-plane thermal conductivity obtained from the analytical model is shown in Figure 3.9a at a single compression pressure, $P_{bp}$, of 460 kPa. The results in Figure 3.9a show an increase in the effective thermal conductivity initially as PTFE is added to the material (from 0 to 5 wt. %) and then decrease as the amount of PTFE increases above 5 wt. %. The results displayed in Figure 3.9a are counter-intuitive, as it was expected that the thermal conductivity would increase with increasing PTFE. As PTFE is added, additional pathways in the through-plane direction are created for heat transfer. To better understand the decrease in the thermal conductivity with increasing PTFE, an alternate method of applying PTFE in the analytical model was investigated.
In this alternate method, a single through-plane porosity distribution of Toray TGP-060 with 0 wt.% PTFE was employed. The desired amount of PTFE was subsequently added to the material following the approach outlined in Section 2.4 at each layer in the modeling domain. The effective through-plane thermal conductivity obtained from the analytical model is shown in Figure 3.9b for the alternate method at a single compression pressure, $P_{bp}$, of 460 kPa. The results in Figure 3.9b show that the effective thermal conductivity increases with increasing PTFE content. An increase of 24.2% with the addition of 20 wt.% PTFE is observed. These results will be discussed later in Section 4.2.

3.5 Discussion

3.5.1 Untreated Paper

The effective thermal conductivity is observed to increase with increasing compression pressure (Figure 3.6). This result has been previously shown in experimental [8, 20] and analytical work [20, 23] and can be explained with the analytical model presented. As the compression pressure increases, the elliptical contact area between two fibres increases. Increases in the contact area cause a decrease in the thermal constriction and spreading resistance, and subsequently, an increase in the overall thermal conductivity. Compression will also cause a decrease in the overall thickness of the GDL material that has been accounted for in this analytical model. It is important to note that while a value of 17.9 MPa was employed in this work for the Young’s modulus of the GDL, within the literature, there is a large variation [1, 56]; however, the model was not found to be significantly sensitive to the compressed thickness, and the trends reported in this work were not affected by the value used.

Fishman et al. [13] noted that the heterogeneous porosity distributions for all four GDL materials are distinct but each display three distinct segments: two transitional surface regions and a core region. For the thinnest GDL investigated, Toray TGP-H-030, this transitional surface region accounts for approximately 66% of its total thickness [13]. Fishman et al. [13] observed that the transitional surface region accounts for 45%, 33%, and 28% of the material thickness for Toray TGP-H-060, 090, and 120, respectively. The variation of the overall effective thermal conductivity with GDL thickness observed in Table 3.3 can be attributed to the relative amount of transitional surface region. The thermal conductivity appears to be strongly affected by the
higher porosity values in the transitional surface regions, regardless of the overall bulk porosity value or the local maximum thermal conductivity value. This can be further explained by the results in Table 3.4 that compare the overall bulk thermal conductivity ($k_{\text{eff,bulk}}$) with the thermal conductivity of the core region ($k_{\text{eff,core}}$). The thermal conductivity of the core region is not dependent upon the material thickness but appears to depend on the porosity of the material.

The discrepancy between the analytical results and the experimental results from literature in Figure 3.6 stems from the small variability in the experimentally obtained porosity profiles of commercially available materials. Batch-specific variations associated with the manufacturing process have been observed in these commercially available materials [57]. In this work, one sample of each GDL was employed as an input in the analytical model. Here, it can be seen that the results in Figure 3.7 from the analytical model are quite sensitive to the porosity values in the transitional regions, or locations of porosity maximums.

### 3.5.2 PTFE Treated Paper

The addition of PTFE in the GDL was expected to lead to an increase in the effective through-plane thermal conductivity for a single compression value. Even though the thermal conductivity of PTFE is significantly lower than that of carbon fibre, the PTFE is expected to provide an additional pathway for heat transfer at a fibre contact point in the analytical model. The results from the analytical model for Toray TGP-H-060 (see Figure 3.9a) show an initial increase in the thermal conductivity with the addition of 5 wt. % PTFE, followed by a counter-intuitive decrease with increasing PTFE content (from 5 – 20 wt. %).

An alternate method of PTFE application in the analytical model was used to further understand this trend. This method involves a single heterogeneous porosity distribution (0 wt.% PTFE) upon which PTFE is added, and the results for the thermal conductivity (Figure 3.9b) show an increase in the thermal conductivity with increasing PTFE content (0 – 20 wt. %). It was observed for the untreated paper GDL materials (Figure 3.6) that the thermal conductivity was strongly affected by the heterogeneous porosity distributions and the higher porosity values in the transitional surface regions, regardless of the overall bulk porosity value or the local maximum thermal conductivity value. The porosity profiles of Toray TGP-H-060 with 0, 5, 10, and 20 wt. % PTFE presented by Fishman et al. [48] are shown in Figure 3.10. The addition of
PTFE affects the overall shape of the through-plane porosity distribution of the material, the slope of the transitional surface regions, and the values of the porosity maxima and minima. The slopes of the transitional surface regions increases with increasing PTFE wt. content (Figure 3.10). It is expected that the larger gradients in the porosity distribution at the transitional surface regions, with their associated high porosities and high thermal resistances, strongly impact the overall thermal conductivity of the PTFE treated GDL. From the results of the alternate method of PTFE application, it is observed that the surface transition regions of the porosity distributions dominate over the addition of PTFE in their impact on the overall thermal conductivity. The decrease in thermal conductivity with increasing PTFE content (from 5 – 20 wt. %) in Figure 3.9(a) is therefore attributed to the dominating thermal resistances of the surface transition regions of the GDL.

3.6 Conclusion

An analytical model to determine the effective thermal conductivity of the GDL was presented. Representative physical GDL models informed by microscale computed tomography imaging of four commercially available uncompressed GDL materials [13, 48] were employed to define the thermal model. The effect of the heterogeneous porosity distribution, GDL compression, and PTFE content on the effective thermal conductivity was investigated. The model predictions for untreated Toray carbon paper GDL materials were compared with recent experimental work by Burheim et al. [15] where two district trends were noted between both sets of data. The first trend noted was an almost linear increase in the effective thermal conductivity with increasing bipolar plate compaction pressure. The effective thermal conductivity was also seen to increase with increasing GDL thickness as bulk porosity remained almost constant. This trend was attributed to the heterogeneous porosity profiles of the material as the thermal conductivity appears to be strongly affected by the higher porosity values in the transitional surface regions, regardless of the overall bulk porosity value or the local maximum thermal conductivity value. The analytical modeling approach allows this trend to be investigated further by isolating the core region and studying its effective thermal conductivity. The effective thermal conductivity of the core region was found to be independent of the material thickness.

Two methods of applying PTFE in the analytical model were investigated to isolate the impact of the PTFE addition and the through-plane porosity distribution on the effective thermal
conductivity. An initial increase in the thermal conductivity with the addition of 5 wt. % PTFE, followed by a decrease with increasing PTFE content (from 5 – 20 wt. %) was noted when four heterogeneous porosity distributions were employed in the analytical model. However, an overall increase of 24.2% in the effective thermal conductivity was noted with the addition of 20 wt. % PTFE when alternate method of PTFE application was employed, thus indicating that the overall thermal conductivity has a strong dependence on the heterogeneous porosity distributions of the treated GDL materials (in particular, the surface transition regions). The outcomes of this work provide insight into dominating effect of heterogeneity and anisotropy of the GDL on the thermal management required for improved PEM fuel cell performance.
3.7 Tables

Table 3-1: Unit cell geometry properties employed to generate the representative physical model.

<table>
<thead>
<tr>
<th>Fibre diameter, $d$</th>
<th>Fibre length, $l$</th>
<th>Unit cell width, $W$</th>
<th>Unit cell length, $L$</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.32 µm</td>
<td>325 µm</td>
<td>1500 µm</td>
<td>1625 µm</td>
</tr>
</tbody>
</table>
Table 3-2: Carbon fibre properties employed for representative physical model.

<table>
<thead>
<tr>
<th>Thermal Conductivity, $k$</th>
<th>Poisson’s ratio, $v$</th>
<th>Young’s Modulus, $E_f$</th>
</tr>
</thead>
<tbody>
<tr>
<td>120 W/m K [19]</td>
<td>0.3 [23]</td>
<td>210 GPa [23]</td>
</tr>
</tbody>
</table>
Table 3-3: Effective thermal conductivity of untreated Toray paper GDLs for four compression pressures.

<table>
<thead>
<tr>
<th>$P_{GDL}$ (kPa)</th>
<th>H (µm)</th>
<th>$\varepsilon_{bulk\text{-}avg}$ (%)</th>
<th>$k_{eff}$ (W/m K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Toray TGP-H-030</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>460</td>
<td>114.11</td>
<td>82.50</td>
<td>0.2165</td>
</tr>
<tr>
<td>930</td>
<td>111.03</td>
<td>82.01</td>
<td>0.3233</td>
</tr>
<tr>
<td>1390</td>
<td>108.02</td>
<td>81.51</td>
<td>0.4099</td>
</tr>
<tr>
<td>Toray TGP-H-060</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>460</td>
<td>213.96</td>
<td>81.62</td>
<td>0.4505</td>
</tr>
<tr>
<td>930</td>
<td>208.18</td>
<td>81.11</td>
<td>0.6039</td>
</tr>
<tr>
<td>1390</td>
<td>202.54</td>
<td>80.58</td>
<td>0.7327</td>
</tr>
<tr>
<td>Toray TGP-H-090</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>460</td>
<td>290.03</td>
<td>82.00</td>
<td>0.4268</td>
</tr>
<tr>
<td>930</td>
<td>282.2</td>
<td>81.50</td>
<td>0.5945</td>
</tr>
<tr>
<td>1390</td>
<td>274.55</td>
<td>80.98</td>
<td>0.7317</td>
</tr>
<tr>
<td>Toray TGP-H-120</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>460</td>
<td>349.46</td>
<td>78.13</td>
<td>0.6111</td>
</tr>
<tr>
<td>930</td>
<td>340.03</td>
<td>77.52</td>
<td>0.8329</td>
</tr>
<tr>
<td>1390</td>
<td>330.81</td>
<td>76.89</td>
<td>0.9851</td>
</tr>
</tbody>
</table>
Table 3-4: Effective thermal conductivity of untreated Toray paper GDLs: bulk and core values.

<table>
<thead>
<tr>
<th></th>
<th>$\varepsilon_{\text{bulk,avg}}$</th>
<th>$\varepsilon_{\text{core}}$</th>
<th>$k_{\text{eff, bulk}}$ (W/m K)</th>
<th>$k_{\text{eff, core}}$ (W/m K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Toray TGP-H-030</td>
<td>0.825</td>
<td>0.742</td>
<td>0.2165</td>
<td>1.686</td>
</tr>
<tr>
<td>Toray TGP-H-060</td>
<td>0.816</td>
<td>0.794</td>
<td>0.4505</td>
<td>1.294</td>
</tr>
<tr>
<td>Toray TGP-H-090</td>
<td>0.820</td>
<td>0.801</td>
<td>0.4268</td>
<td>1.596</td>
</tr>
<tr>
<td>Toray TGP-H-120</td>
<td>0.781</td>
<td>0.760</td>
<td>0.4519</td>
<td>1.496</td>
</tr>
</tbody>
</table>
3.8 Figures

Figure 3-1: Schematic illustrating a section of the representative physical model, which consists of solid, continuous fibres oriented in a planar direction (a) x-y plane orientation, and, (b) isometric view.
Figure 3-2: Schematic illustrating a periodically ordered fibre arrangement with $\theta = 90^\circ$. 
Figure 3-3: (a) Contact points per fibre in layer, $t$, within the defined unit cell with a porosity of 80% as a function of the orientation angle, $\theta$, and, (b) average number of contact points as a function of porosity, $\varepsilon$. 

```plaintext
Figure 3-3: (a) Contact points per fibre in layer, $t$, within the defined unit cell with a porosity of 80% as a function of the orientation angle, $\theta$, and, (b) average number of contact points as a function of porosity, $\varepsilon$. 
```
Figure 3-4: (a) Equivalent thermal resistance network of a single layer with \( C \) number of contact points, and, (b) the equivalent thermal resistance network through the GDL thickness with \( \Lambda \) number of layers.
Figure 3-5: (a) Schematic illustrating a single fibre-to-fibre contact point for treated carbon paper, and (b) the equivalent thermal resistance network for (a).
Figure 3-6: Comparison of measured average effective thermal conductivity with compression pressure with values extracted from Burheim et al. [16].
Figure 3-7: Through-plane effective thermal conductivity and porosity distributions for untreated (a) Toray TGP-H-030, (b) Toray TGP-H-060, (c) Toray TGP-H-090, and (d) Toray TGP-H-120.
Figure 3-8: Through-plane effective thermal conductivity and porosity distribution for Toray TGP-H-090 depicting core and transitional surface regions defined by Fishman et al. [13].
Figure 3-9: Through-plane effective thermal conductivity with 0, 5, 10, and 20 wt.% PTFE using (a) four district porosity profiles presented in Fishman et al. [13] for Toray TGP-H-060 and, (b) a single porosity profile of Toray TGP-H-060 with 0 wt.% PTFE employed to generate representative physical GDL models of treated GDL materials.
Figure 3-10: Through-plane porosity distributions of Toray TGP-H-060 from Fishman et al. [48] employed to generate representative physical GDL models.
Chapter 4 Lattice Boltzmann Model

4.1 Introduction
In this chapter, the in-plane and through-plane effective thermal conductivity of non-woven carbon paper PEM GDLs is determined using a thermal LBM framework. Using stochastic modeling domains of the GDL in two and three dimensions, the effective thermal conductivity of the GDL is evaluated over the relevant range of porosity values. The effect of GDL compression, binder and PTFE content, addition of a MPL, water saturation, and heterogeneous through-plane porosity distributions on the resulting thermal conductivity is investigated.

4.2 Motivation and Objective
The GDL is a highly porous and stochastic material, and numerical modeling of the thermal conductivity has largely been limited to work with GeoDict™, a commercial simulation software package, and the compact analytical model developed by Sadeghi et al. [23] and presented in Chapter 3. While the analytical model was able to provide insight into the through-plane thermal resistance and thermal conductivity it has limits in its application of studying the thermal conductivity of the GDL. It is not able to easily accommodate different geometric inputs and the anisotropy of the GDL.

The mesoscopic approach of LBM and the ease of implementing solid-fluid boundary conditions in porous media have made LBM an appropriate modeling method for the porous GDL of PEM fuel cells. LBM has previously been used to study the permeability and water management in PEM fuel cells in a number of studies [37, 38]. The main objective of this chapter is to apply a thermal LBM framework to determine the through-plane and in-plane thermal conductivity of the GDL.

4.3 Methodology
For this work, the determination of the effective thermal conductivity of the GDL consists of three steps, namely: i) stochastic GDL modeling domains, followed by ii) thermal lattice
Boltzmann modeling of heat flow through the domains, and then iii) using the Fourier heat equation to predict the effective thermal conductivity of the GDL.

4.3.1 Lattice Boltzmann Methodology

In this work, the model presented by Wang et al. [41] was adopted to model the effective thermal conductivity of the reconstructed GDL samples. The effective thermal conductivity was predicted with a 2-D nine-speed (D2Q9), and a 3-D fifteen-speed, (D3Q15) two-phase single relaxation time LBM models. The radiative and convective heat transfer modes are neglected in this work for simplicity. With the calculation of nominal Grashof and Peclet numbers in GDL structures, Ramousse [19] showed that natural convection and convective heat transfer are negligible compared to heat conduction. Radiative heat transfer is also shown to be negligible in GDL structures for temperatures below 1000 K [52], which is well above the operating range of a low temperature PEM fuel cell. There is no phase change or heat source term inside the domain. Thermal contact resistance between the touching fibres is neglected. Only the conduction heat transfer mode is considered; hence, a single thermal distribution function can be used. The governing equation for pure thermal conduction in a porous two-phase domain is given by:

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

(4.1)

where \( \vec{x} \), \( \Delta \vec{x} \), \( t \), and \( \Delta t \) are spatial coordinate, lattice spacing, time, and simulation time step, respectively. \( \tau_{a} \) is the dimensionless relaxation time that is related to the thermal conductivity of each phase, and \( \alpha \) denoted solid and liquid phases. \( g_{\alpha} \) and \( g_{\alpha}^{eq} \) are distribution function and equilibrium distribution function of fictitious energy particles of phase \( \alpha \). The lattice spacing (\( \Delta \vec{x} \)) is related to the simulation time step as \( \Delta \vec{x} = \vec{e} \Delta t \), where \( \vec{e} \) is the discrete velocity set of energy particles.

Since the fluid velocity (\( \vec{u} \)) is assumed to be zero inside the pores of the GDL sample, the equilibrium distribution equations of energy particles can be simplified. For the D2Q9 lattice, they are given as follows:
\[ g_i^{eq} = -\omega_i T \frac{\vec{v} \cdot \vec{u}}{c^2} = 0 \quad i = 0 \]
\[ g_i^{eq} = \omega_i T \left[ \frac{3}{2} + \frac{3}{2} \frac{\vec{e}_i \cdot \vec{u}}{c_s^2} + \frac{9}{2} \frac{\vec{e}_i \cdot \vec{u}}{c_s^4} - \frac{3}{2} \frac{\vec{v} \cdot \vec{u}}{c_s^2} \right] = \frac{3}{2} \omega_i T \quad i = 1 - 4 \quad (4.2) \]
\[ g_i^{eq} = \omega_i T \left[ 3 + 6 \frac{\vec{e}_i \cdot \vec{u}}{c_s^2} + \frac{9}{2} \frac{\vec{e}_i \cdot \vec{u}}{c_s^4} - \frac{3}{2} \frac{\vec{v} \cdot \vec{u}}{c_s^2} \right] = 3 \omega_i T \quad i = 5 - 8 \]

where \( T \) is the macroscopic temperature, \( c_s \) is the lattice sound speed, and \( \omega \) 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 lattice sound speed is \( 1/\sqrt{3} \) for both D2Q9 and D3Q15 lattices. The weighting factors, \( \omega \), for the D2Q9 model are:

\[ \omega_i = \frac{4}{9} \quad i = 0 \]
\[ \omega_i = \frac{1}{9} \quad i = 1 - 4 \quad (4.3) \]
\[ \omega_i = \frac{1}{36} \quad i = 5 - 8 \]

For the D3Q15 model the equilibrium distribution equations are given as:

\[ g_i^{eq} = 0 \quad i = 0 \]
\[ g_i^{eq} = \frac{3}{2} \omega_i T \quad i = 1 - 6 \quad (4.4) \]
\[ g_i^{eq} = 3 \omega_i T \quad i = 7 - 14 \]

The weighting factors, \( \omega \), for the D3Q15 model are:

\[ \omega_i = \frac{2}{9} \quad i = 0 \]
\[ \omega_i = \frac{1}{9} \quad i = 1 - 6 \quad (4.5) \]
\[ \omega_i = \frac{1}{72} \quad i = 7 - 14 \]

In this work, the standard labelling of D2Q9 and D3Q15 lattices was followed. The discrete velocity set, \( \vec{e}_i \), for the D2Q9 is given as:

\[ \vec{e}_i = (0,0) \quad i = 0 \]
and the $\vec{e}_i$, for the D3Q15 is as follows:

\[
\vec{e}_i = (0,0,0) \quad i = 0 \\
\vec{e}_i = (\pm 1,0,0), (0,\pm 1,0), (0,0,\pm 1) \quad i = 1 - 6 \\
\vec{e}_i = (\pm 1,\pm 1,\pm 1) \quad i = 7 - 14
\] (4.7)

Each phase’s relaxation time, $\tau_\alpha$, depends on macroscopic thermal conductivity of its respective phase and the lattice parameters. The dimensionless relaxation times for the D2Q9 model are:

\[
\tau_\alpha = \frac{3}{2} \frac{k_\alpha}{c^2 \Delta t} + \frac{1}{2} \quad (4.8)
\]

and for the D3Q15 model:

\[
\tau_\alpha = \frac{9}{5} \frac{k_\alpha}{c^2 \Delta t} + \frac{1}{2} \quad (4.9)
\]

where $k_\alpha$ is the thermal conductivity of phase $\alpha$ and $c$ is the physical sound speed. The lattice parameters, $\Delta x$ and $\Delta t$, are chosen to ensure that $\tau_\alpha$ remains in the stable range of $0.5 < \tau_\alpha < 2$, indicated by Wang et al. [41]. The lattice spacing ($\Delta x$) 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:

\[
T_\alpha = \sum_i g_{i,\alpha} \quad \vec{q}_\alpha = \left( \sum_i \vec{e}_i g_{i,\alpha} \right) \frac{\tau_\alpha - 0.5}{\tau_\alpha} \quad (4.10)
\]

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

\[
k_{\text{eff}} = \frac{\delta \int q \, dA}{\Delta T \, A} \quad (4.11)
\]
where $A$ is the cross section area, and $\delta$ is the thickness of the GDL in the direction of interest, and $\Delta T'$ is the temperature differences implemented between the two sides 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 [58] for velocity and pressure boundary conditions. D'Orazio and Succi [59] 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.

4.3.2 GDL Modeling Domains
The LBM simulations presented in this work were based on numerically generated modeling domains of carbon fibre paper GDL materials. The geometries generated are stochastic and a number of simulations for each case under consideration were performed. It is expected that there will be variations between each GDL modeling domain that can lead to variability in the predicted results for the thermal conductivity; however, consistency in certain parameters is required. To effectively compare the parameters under consideration, control over the porosity was required. Repeatability in the generation of the domains and the ability to compress the materials, add additional phases (binder, PTFE, and microporous layer), and control the through-plane porosity distribution is required. Section 4.3.2.1 describes the method employed for generating the GDL modeling domains and Sections 4.3.2.2 – 4.3.2.5 outline the additional parameters incorporated within the modeling domains.

4.3.2.1 Stochastic Fibre Placement Algorithm
In order to represent the stochastic nature of the GDL, a stochastic fibre placement algorithm was employed. The computational domain used was a cuboid with dimensions for the length and width ($x$ and $y$) and a thickness ($z$). The fibre placement algorithm uses the following assumptions commonly found in GDL literature [60]:

- Fibres are straight, cylindrical, and have a given length of the width, or length of the domain.
• 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 the generated GDL modeling domains, each fibre has a diameter of 8 $\mu$m to represent a real GDL sample, where the fibres typically have a diameter between 7 to 12 $\mu$m [11]. Each fibre has a randomly defined origin position ($x_o$, $y_o$, $z_o$) within the domain. The fibres extend outwards in the $x$-$y$ plane at a random angle direction between 0° and 180°. Periodic boundary conditions in the $x$- and $y$-directions are used in the algorithm.

Two common approaches to the fibre placement algorithm in fuel cell literature currently exist. The first method (Method 1) follows the stochastic fibre placement algorithm described above with fibres placed at a random location within the domain until a desired porosity is achieved. In the second method (Method 2), the same method is followed except the $z$-component ($z_o$) of the fibre origin position is suppressed and selected such that the fibres are placed in discrete layers with a defined porosity. In Method 2, the layers are stacked in the through-plane position until the desired GDL thickness is achieved.

Preliminary results for the predicted thermal conductivity using LBM were done on GDL modeling domains that followed Method 2 for the fibre placement algorithm. These results are described in more detail in Section 4.4.3. While this method for generating GDL domains provided results that were in good agreement with previously reported thermal conductivity values, the method was found to have limitations when considering compression. It was also thought to be an over simplification of the GDL structure that could be over emphasizing the contact between GDL fibres. While, the GDL fibres are preferentially oriented in the $x$-$y$ plane (see Figure 2.2), they are not found in discrete layers.

Method 1 for the fibre placement algorithm also has limitations when considering its application to this work. Regions of high porosity can exist with this method to the degree that there may be layers in the through-plane direction with a porosity of 100%. These regions will have a high thermal resistance that will impact the overall predicted through-plane thermal conductivity value and give potentially unrealistic results. As the domain size is increased, the porosity will
become more uniform, but the computational costs will also increase. To overcome the limitations of both of these methods for this work, a third fibre placement algorithm (Method 3) was developed that combines the two original methods.

In Method 3, each fibre has a randomly defined origin position within the domain with the z-component of the fibre suppressed. The fibres are placed in the through-plane direction at a predefined z-coordinate to ensure that there are no layers in the through-plane direction with a porosity of 100%. The z-coordinate, $z_i$, is determined by:

$$z_i = \frac{(i - 1)}{N} \times Z \quad (4.12)$$

where $N$ is the total number of fibres placed within the domain, $Z$ is the height of the domain, and $i$ represents the fibre number. The total number of fibres placed in the domain, $N$, is determined by generating a domain and using the first method for the fibre placement algorithm to fill the domain until the desired porosity is achieved. A sample domain is shown in Figure 4.1.

### 4.3.2.2 Reduced Compression Model

During the operation of a PEM fuel cell, the GDL will experience compressive forces that have been experimentally shown to affect the through-plane thermal conductivity [8, 16, 18, 20]. The reduced compression model developed by Schulz et al. [61] was used to simulate compression in the GDL modeling domains. Starting with a GDL modeling domain generated with Method 3 described in Section 4.3.2.1, the reduced compression algorithm assumes that the $x$ and $y$ position of each solid voxel remains fixed. The domain is compressed by a given compression ratio, $\gamma$, defined by:

$$\gamma = \frac{\text{Height of compressed sample}}{\text{Initial height}} \quad (4.13)$$

In order to compress the domain, the material is fixed at the bottom plane ($z = 0$) and a load is applied on the opposite surface ($z = Z$). Transverse strain is neglected. Each solid voxel is shifted from its original height, $h$, to a new height, $h'$, calculated from the compression ratio, $c$:

$$h' = [(1 - c)h] \quad (4.14)$$
where the square brackets are used to indicate that $h'$ is rounded off to the nearest integer. Since solid material cannot be lost in this process, the model assumes that the solid voxels cannot penetrate into each other [61]. A sample domain, first uncompressed and then compressed to a ratio of $c = 0.75$ and then to a ratio of $c = 0.5$ is shown in Figure 4.2.

4.3.2.3 Binder and PTFE

Carbonaceous binder, a thermoset resin [1, 60], is commonly added to carbon paper GDL materials during the manufacturing process. When added to the GDL, the binder has been observed to behave like wetting fluid, having a low static contact angle with the carbon fibres and generally accumulating at the intersection between fibres. PTFE is generally added to the GDL in an aqueous solution by either dipping the GDL into the solution or spraying the GDL with the solution and allowing it to dry [1, 16, 60]. Since both binder and PTFE are assumed to behave like a wetting fluid when added to the GDL, both can be modeled in the GDL modeling domains with the same algorithm.

The algorithm employed mimics the behavior of a wetting fluid by filling the smallest pores within the GDL modeling domain first. A similar approach has been employed in previous numerical work by [24, 62]. The binder is added with the algorithm first, followed by the PTFE. The bulk porosity with the addition of binder or PTFE, $\varepsilon$, is calculated with the general form of the equation:

$$
\varepsilon = \varepsilon_0 - \rho \frac{\lambda (1 - \varepsilon_0)}{(1 - \lambda)}
$$

(4.15)

where $\varepsilon_0$ is the bulk porosity of the material before binder or PTFE is added, $\lambda$ is the weight fraction of binder or PTFE, and $\rho$ is the density ratio between carbon fibre and binder or PTFE [50]. When considering PTFE, a value of 0.9 is used for $\rho$ [1]. The binding material is assumed to be composed of the same material as the carbon fibres [22, 24]. As such, a value of 1.0 is used for $\rho$. A sample domain first untreated, with the addition of 30% binder, and with the addition of 20% PTFE is shown in Figure 4.3.
4.3.2.4 Microporous Layer
The microstructure of the MPL is two orders of magnitude lower than the GDL [1]. This, along with the presence of cracks (on the order of microns) makes modeling the MPL structure together with the GDL unfeasible. The approach employed to study the GDL/MPL assembly was to add the MPL as a homogeneous material [1] with a known thermal conductivity value into the GDL modeling domains. The two physical parameters that are controlled when adding the MPL into the domains are the depth that the MPL has intruded into the GDL and the thickness that the MPL extends off of the top face \((z = Z)\) of the GDL. An example of a modeling domain of a GDL/MPL assembly is shown in Figure 4.4.

4.3.2.5 Residual Water
Water was added to the GDL modeling domains using 3-D pore network modeling simulations with invasion percolation. The pore network models are conducted by another member of the research group (Hinebaugh, J.). A pore growth algorithm [63] was used to extract a topologically equivalent pore network from the pore space. The invasion percolation drainage algorithm [63] was performed on the GDL modeling domains until breakthrough is achieved. Two inlet conditions are used for each of the GDL modeling domains. For the first inlet condition, the flooded inlet condition, any pore on the bottom face is included in the inlet. The second inlet condition, the one point injection inlet condition, has a circle in the centre of the bottom face with an area of 1% of the bottom face. Any pore in contact with this circle is included in the inlet. The two inlet conditions produce distinct saturation patterns (See Figures 4.5 and 4.6) and on average, distinct total saturation values. The final GDL domains consist of the following four phases; carbon fibres, carbonized resin or binder, air, and water (Figure 4.7).

4.3.2.6 Heterogeneous Porosity Profiles
The heterogeneous through-plane porosity distributions reported in [13], and utilized in Chapter 3 with the analytical model were used to generate GDL modeling domains for the LBM simulations. Following Method 3 for the stochastic fibre placement algorithm, the GDL modeling domains were generated but with a modification to account for the heterogeneous nature of the through-plane porosity distribution.
The mean porosity of the materials was determined and used to calculate the total number of fibres to be placed within the domain, \( N \). Equation 4.12 is only valid for a uniform porosity distribution. In order to modify the equation to handle a varying porosity input but follow the same procedure, the \( z \)-coordinate, \( z_i \), is determined based on the ratio of the local value of the through-plane porosity to the mean through-plane porosity. This ratio allows each fibre to be placed within the domain at a specified location with a non-uniform spacing between adjacent fibres.

### 4.4 Results and Discussion

The methodology outlined in the Section 4.3 was used to predict the effective thermal conductivity of GDL samples using 2-D and 3-D GDL modeling domains. The method was first validated by simulating the case of a dual component material and comparing the results with the analytical solution for simplified cases of parallel and series configurations. The simulation parameters were verified by performing simulations with a porous GDL modeling domain and varying the lattice spacing and domain size. Simulations with 2-D and 3-D GDL modeling domains were evaluated with the parameters discussed in Sections 4.3.3.2 – 4.3.3.5.

All the simulations were parallelized using OpenMP and run on the General Purpose Cluster of SciNet supercomputing facilities (Compute Canada).

#### 4.4.1 Validation

A number of analytical correlations exist for determining the thermal conductivity of a composite material. The simplest analytical correlations used to estimate the thermal conductivity are volume averaging methods that include parallel and series thermal resistances of each phase [42], defined as:

\[
k_{\text{eff, series}} = \frac{k_s k_f}{\varepsilon k_s + (1 - \varepsilon) k_f} \quad k_{\text{eff, parallel}} = \varepsilon k_f + (1 - \varepsilon) k_s \quad (4.16)
\]

where \( \varepsilon \) is the porosity (volume of fluid divided by total volume). Values employed in the LBM simulations for the thermal conductivities of carbon fibres and air at 80 °C are \( k_s = 120 \text{ W/m K} \) [19] and \( k_f = 0.03 \text{ W/m K} \) [53], respectively. Simulations were performed for the basic structure
of a dual-component material in a parallel and series configurations and the predicted values of effective thermal conductivity were compared with the analytical solutions.

A lattice spacing, $\Delta x$, of 2 $\mu$m/lattice units (lu) was selected for the simulations, which yields four lattice points across fibre diameters for fibres of diameter of 8 $\mu$m. 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. Lattice temperatures of $T_{\text{hot wall}} = 1.5$ and $T_{\text{cold wall}} = 0.7$ were implemented in the simulations.

The size of the computational domain was studied for the case of a dual component material. If we consider the thermal conductivity of air and carbon fibres for the two phases of parallel and series configurations, the analytical prediction for the effective thermal conductivity (Eq. 4.16) are 60.015 and 0.059985 W/m K, respectively. The results from the LBM simulations and respective deviations are presented in Table 4.1 for the grid sizes studied.

In the LBM simulations, the temperature field was found to converge during the simulations at a much faster rate than the heat flux vector. This trend has been reported consistently for nano-scale thermal transport modeling using the LBM for phonon transport modeling [64] and molecular dynamic simulations [65]. The heat flux vector was monitored until a convergence criterion of $10^{-4}$ was reached for the absolute value of residuals summed over the entire domain. The accuracy of the solution increases with the size of the computational domain. This also results in a subsequent increase in the computational requirements. With a relative deviation under 1% or both the parallel and series configuration, a computational domain of 200×200 was chosen for the D2Q9 simulations. The size of the computational domain was also studied for the D3Q15 lattice, where a similar accuracy was observed at a lattice size of 60×60×60. Larger grids of at least 100×100×100 nodes, were chosen for the D3Q15 grids to obtain a more realistic GDL reconstruction. The chosen grid size varies with the parameter under consideration and will be discussed in each subsequent section.

The dual component series and parallel configurations are structured configurations; therefore, the same grid size may not produce lattice independent results for the stochastic porous case. Further 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 3-D GDL reconstruction with a lattice spacing of 2.0, 1.3, and 1.0 μm/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 domain sizes studied are reported in Table 4.2. The relative deviation is calculated based on the case of lattice spacing of 2.0 μm/lu. Since a small deviation is observed between the three cases (less than 1 %), a lattice spacing of 2.0 μm/lu was chosen for all subsequent simulations.

4.4.2 Preliminary D2Q9 Results

For the 2-D simulations, the anisotropy of the GDL was represented by two separate GDL reconstructions, namely an in-plane geometry and a through-plane geometry. The 2-D geometric reconstructions were generated by taking slices in the in-plane and through-plane directions of 3-D cubic GDL modeling domain generated with Method 2 of the fibre placement algorithm. Figure 4.8 shows an example of the two geometries created for the 2-D simulations.

The 2-D predictions of the through-plane and in-plane effective thermal conductivities are given in Figure 4.9. As expected, the effective thermal conductivity decreases with increasing the porosity for both cases. This trend in the results is in agreement with work by Zamel et al. [24]. As compression effects are not taken into account in the current study, the porosity is only increased by randomly adding the carbon fibres to the computational domain. The significantly higher thermal conductivity of the carbon fibres when compared with the air causes the increase in the effective thermal conductivity with decreasing porosity.

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. The predicted values of the in-plane thermal conductivity were in good agreement with the results published in PEM fuel cell literature summarized in Table 4.3; however, the results of the through-plane thermal conductivity were significantly lower than the published results. This difference can be attributed to the 2-D nature of the GDL reconstructions. The fibre-to-fibre contact is greatly underestimated in the 2-D slices of GDL samples that were used for through-plane simulations. A contour plot of
the heat flux magnitude for through-plane and in-plane configurations is shown in Figure 4.10. In the through-plane case, the regions of high heat flux are seen at the contact areas of adjacent fibres. Due to the much larger thermal conductivity of the fibre material, the preferential path for heat conduction is through the fibre-to-fibre contacts.

4.4.3 Preliminary D3Q15 Results

Simulations were performed for 3-D GDL modeling domain generated with Method 2 of the fibre placement algorithm with porosity values in the range of 0.70 to 0.86. The modeling domains employed in the simulations are cubic with dimensions of 200 μm. The results of the D3Q15 LBM simulations for the through-plane and in-plane effective thermal conductivity are given in Figure 4.11. Consistent with 2-D simulations, the effective thermal conductivity decreases with increasing porosity.

A comparison between the 2-D and 3-D simulations results shows a significant increase in the predicted through-plane thermal conductivity, while the predicted values of in-plane conductivity are in general agreement. For the case of the through-plane effective thermal conductivity, the fibres are better connected in the 3-D reconstructed structures, which create a preferential path for heat transport, and thus a higher effective thermal conductivity. The predicted values of effective through-plane and in-plane effective conductivities are compared against reported values in the literature for porosity of 0.78 shown in Table 4.3.

The range of predicted values for the in-plane effective thermal conductivity did not significantly change between the 2-D and 3-D simulations; however, the large amount of scatter in the thermal conductivity that was noted in the 2-D results is not present in the 3-D results because of the smoothing effect of the third dimension (see Figure 4.12). The 3-D GDL reconstructions eliminate the dependence of the thermal conductivity on the random grid and fibre arrangement that is generated by slicing the cubic domain for the two-dimensional cases and provides an improved overall estimate based on porosity.

The results presented in Figure 4.11 were compared with an existing correlation presented by Zamel et al. [24]. Their work provided a fitting function for the data obtained from GeoDict
simulations for dry, uncompressed untreated carbon paper GDL materials. They proposed the following relationship for the effective thermal conductivity of the GDL:

\[
\frac{k_{\text{eff}}}{k_s} = 1 - f(\varepsilon) \frac{3\varepsilon}{3 - (1 - \varepsilon)}
\]  

(4.16)

where the structural function, \(f(\varepsilon)\), is given by:

\[
f(\varepsilon) = C_1 (1 - \varepsilon)^{C_2} \exp[C_3 (1 - \varepsilon)]
\]

(4.17)

where \(C_1\), \(C_2\), and \(C_3\) are the fitting parameters. This form of relation for thermal conductivity was used and fitted it to the data across the range of porosities studied. The best fit parameters obtained from fit to the data, as well those reported by Zamel et al. [24], are reported in Table 4.4. A comparison of the results and fitting curves obtained with the LBM simulations in this study and the results presented in [24] is shown in Figure 4.13. Reasonable agreement between the two sets of data can be seen.

### 4.4.4 D3Q15 Results with a new method for fibre placement

Simulations were performed for 3-D GDL reconstructions generated with Method 3 of the stochastic fibre placement algorithm. The modeling domains employed in the simulations are cubic with dimensions of 200 \(\mu\text{m}\). The results of the D3Q15 LBM simulations for the through-plane and in-plane effective thermal conductivity are given in Figures 4.14 and compared with the results from Section 4.4.3.

There is relatively good agreement between the two sets of data in the porosity range (70 – 90\%) considered. For both the through-plane and in-plane results, the new method of fibre placement yields slightly lower thermal conductivity values. This is expected as the old method places fibres in discrete layers and as a result may be over emphasizing the contact between GDL fibres. Both sets of data follow the same trend of increasing thermal conductivity with decreasing porosity.

### 4.4.5 Compression

Simulations were performed for the GDL modeling domains with a compression ratio in the range of 1.0 to 0.50 and an uncompressed porosity of 80.0\%. The computational domains used
have dimensions for the length and width \((x \text{ and } y)\) of 200 \(\mu\text{m}\) and an uncompressed thickness \((z)\) of 300 \(\mu\text{m}\). Through-plane and in-plane effective thermal conductivity results are given in Figure 4.15(a). Each point on the graph represents the average of 10 modeling domains with the standard deviation shown.

Both the through-plane and in-plane thermal conductivity increase with increasing compression as seen in Figure 4.15(a). The same trend was noted in Chapter 3 with the analytical model for the through-plane thermal conductivity. An almost linear increase with compression is noted for the in-plane thermal conductivity, where an increase in the thermal conductivity by a factor of 1.6 is noted between the uncompressed and 50% compressed results. The through-plane thermal conductivity is more affected by compression, especially at compression ratios above 25%, and an increase by a factor of 14 is noted between the uncompressed and 50% compressed results.

A decrease in the compression ratio (increase in compression pressure on the GDL) will cause a decrease in the porosity, structural changes within the GDL, and an increase in fibre contact areas in the through-plane direction. Similar trends for the through-plane thermal conductivity have been reported in experimental and numerical [8, 16, 20, 23, 24], however, there is minimal information on the in-plane thermal conductivity reported in PEM fuel cell literature. Experimental results reported in literature, which use quantified compression values rather than the compression ratio used in this work would result an arbitrary comparison.

Changes in the fibre structure with increasing compression affect the through-plane thermal conductivity more than the in-plane. In the in-plane direction heat is conducted along fibres and in the through-plane direction heat is conducted between fibre contact regions (as discussed in detail in Chapter 3). To look closer at the effects of compression on the geometry of the GDL and the thermal conductivity, the results from Section 4.4.4 are plotted on the same graph as the results from the compression study in Figure 4.15(b). For the through-plane thermal conductivity at high porosity values (75 – 80%), there is no difference between the uncompressed and compressed results at the same porosity. As the porosity is lower (60 – 70%) the compressed results are higher than the uncompressed at the same porosity values, indicating that compression effects are not just changes in the porosity but changes in the fibre structure.

A comparison for the in-plane results is not possible, as there appears to be another phenomena occurring. As the compression ratio is increased, the height of the GDL modeling domain is
decreased, but for the uncompressed results the modeling domain height is constant. To better understand what is happening with the in-plane results, the in-plane thermal conductivity is plotted versus the aspect ratio \((z/x)\) of the modeling domain for three porosity value in Figure 4.16. The in-plane thermal conductivity increases linearly as the aspect ratio of the modeling domain increases. Increases in the height of the modeling domain (z-direction) will create more fibres for heat to be conducted along.

4.4.6 Binder and PTFE

Simulations were performed for the GDL modeling domains with a binder weight content between 0 to 50% and a porosity of 80%. The computational domains used have dimensions for the length and width \((x\) and \(y\)) of 200 µm and a thickness \((z)\) of 300 µm. It is assumed that the binder is composed of the same material as the carbon fibres [22, 24], and as such, a thermal conductivity value of \(k_b = 120\) W/m K [16] was employed for the binder material. Through-plane and in-plane effective thermal conductivity results are given in Figure 4.17. Each point on the graph represents the average of 10 modeling domains with the standard deviation shown.

Both the through-plane and in-plane thermal conductivity increase with increasing amounts of binder. The addition of binder causes a significant increase in the through-plane thermal conductivity by creating preferential pathways for heat transfer in this direction. The actual amount of binder found in carbon paper GDL materials is not known. Values reported in fuel cell literature range from 5 – 50% [17, 24, 60, 62]. Toray paper is thought to have more binder [22, 24] as noted by its higher thermal conductivity at the same porosity as other carbon paper GDL materials. In this work, an increase in the through-plane thermal conductivity by a factor of 6 is noted with the addition of 50% binder. This might be higher than in practice, as the placement of binder may be overemphasized. While the binder is known to be preferentially located in the intersections between fibres, in reality, not all of the binder may be located in these regions.

Simulations were performed for the GDL modeling domains with a PTFE weight content between 0 to 30% and an untreated porosity of 80.0%. A thermal conductivity value of 0.25 W/m K was employed in the LBM simulations for the PTFE [16]. Two different cases of PTFE distributions were considered (uniform and heterogeneous distribution). The predicted through-
plane and in-plane thermal conductivity as a function of PTFE weight content is presented in Figure 4.18. Each point on the graph represents the average of 10 modeling domains.

The through-plane thermal conductivity increases with increasing amounts of PTFE for the uniform and heterogeneous PTFE distribution. The addition of 30% wt. PTFE resulted in a 16% and 6% increase in the through-plane thermal conductivity with a uniform and heterogeneous PTFE distribution, respectively. The addition of PTFE causes an increase in the through-plane thermal conductivity, but it is not as significant as the addition of binder. While binder and PTFE are located in the same regions within the GDL, the significantly lower thermal conductivity of the PTFE reduces its impact on through-plane heat transfer. The increase noted in the LBM simulations is comparable, but lower than the results from the analytical model where an overall increase of 24.2% in the through-plane thermal conductivity was noted with the addition of 20 wt. %

The addition of PTFE had a significantly lower impact on the in-plane thermal conductivity. An increase of 1.6% in the in-plane thermal conductivity was noted with the addition of 30% wt. PTFE. This is in agreement with previous experimental and numerical work by Sadeghi et al. [22]. Their findings showed that varying the PTFE content of the carbon paper GDL materials under consideration did not affect the predicted in-plane thermal conductivity values.

4.4.7 Microporous Layer

Thermal conductivity values for the MPL reported in literature range from 0.03 – 1.0 W/m K [8]. The MPL does not exist as an independent structure [12], and as such, the thermal conductivity of the MPL cannot be measured alone. Measurements for the thermal conductivity of a GDL/MPL assembly are conducted and compared with measurements for a GDL [66]. The same issues with measuring the thickness of the MPL occur. The thickness of the MPL has been reported to be between 50 – 90 µm [1, 8]. Thickness measurements are conducted by measuring a GDL only and then a GDL/MPL or by removing the MPL from the GDL [8].

Simulations were performed for the GDL modeling domains with a MPL thickness between 0 to 90 µm and a GDL porosity of 80%. The computational domains used have dimensions for the length and width (x and y) of 200 µm, a GDL thickness (z) of 300 µm, and an MPL intrusion
depth of 20 µm. A thermal conductivity of 0.3 W/m K was used for the MPL [66]. Through-plane and in-plane effective thermal conductivity results are given in Figure 4.19. Each point on the graph represents the average of 10 modeling domains with the standard deviation shown.

The addition of the MPL causes a decrease in the through-plane thermal conductivity of 22% and negligible increase in the in-plane thermal conductivity. Further increases in the thickness of the MPL do not affect the thermal conductivity. The lower thermal conductivity of the MPL causes it to act like a thermal resistor in the through-plane direction and increase the overall thermal resistance of the GDL/MPL assembly when compared with a GDL alone. This result is in agreement with previously reported work in literature [8, 66, 67].

Simulations were also performed for the GDL modeling domains with a MPL intrusion depth between 0 to 40 µm and a GDL porosity of 80%. The same parameters for the computational domain as Figure 4.19 were used. Through-plane and in-plane effective thermal conductivity results are given in Figure 4.20. Each point on the graph represents the average of 10 modeling domains with the standard deviation shown. As shown in Figure 4.20, the intrusion depth of the MPL does not have an effect on the through-plane or in-plane thermal conductivity.

The effect of compression on a GDL/MPL assembly was investigated. Simulations were performed for the GDL modeling domains with a compression ratio between 0 to 50% and an uncompressed GDL porosity of 80%. The computational domains used have dimensions for the length and width (x and y) of 200 µm, a GDL thickness (z) of 300 µm, an MPL thickness of 50 µm, an MPL intrusion depth of 20 µm, and a GDL porosity of 80%. A thermal conductivity of 0.3 W/m K was used for the MPL [66]. It is assumed that at the compression values used, the MPL will not compress [66], and the reduced compression algorithm is only applied to the GDL. Through-plane and in-plane effective thermal conductivity results are given in Figure 4.21. Each point on the graph represents the average of 10 modeling domains. The results are compared with the predicted thermal conductivity values from Section 4.4.5 for the compression of a GDL.

The through-plane thermal conductivity of a GDL/MPL assembly decreased with increasing compression. The decrease in effective thermal conductivity with increasing compression is counter intuitive, and requires a detailed explanation. The GDL/MPL material can be considered a composite of three regions: GDL, GDL and MPL, and MPL, as shown in Figure 4.22. Due to an increased heat flux within the GDL region, the effective thermal conductivity of the GDL
region increases. However, as noted with the addition of the MPL in Figure 4.19, the MPL is acting as a dominating thermal resistor to through-plane heat transfer. The dominance of the MPL is further highlighted after compression since the structure of the MPL does not change and its thermal resistance, which remains constant throughout compression, composes the majority of the composite material resistance. As can be seen in Equation 4.11, the effective thermal conductivity depends on the thickness of the material, as well as the heat flux. When considering the entire composite material, the effective thermal conductivity decreases due to the decreasing overall material thickness. The decrease in effective thermal conductivity of the overall material is seen because the increased effective thermal conductivity of the GDL region is dominated by the large thermal resistance of the MPL combined with the decreasing material thickness. The in-plane thermal conductivity of the GDL/MPL assembly exhibited the same increase with compression as a GDL without the MPL. It should be noted that this result is highly dependent on the geometric representation of the GDL/MPL material along with its material properties (thermal conductivity).

4.4.8 Water

The LBM simulations were performed on a total of 20 GDL modeling domains. The computational domains used have dimensions for the length and width (x and y) of 600 µm and a thickness (z) of 300 µm. A thermal conductivity value of $k_w = 0.58 \text{ W/m K}$ [16] is employed for water. For each of these domains, the through-plane and in-plane effective thermal conductivity was determined for the cases of: a dry material, a flooded inlet condition, and a one point injection inlet condition. The predicted thermal conductivities are presented for the flooded inlet condition and the one point injection inlet condition in Figure 4.22. The dry thermal conductivity plotted on both figures is the average of the 20 GDL modeling domains. Since we are plotting the average value of 20 simulations, some data points for the saturated simulations are below this average. This applies especially to the in-plane simulations. In spite of this, for every GDL modeling domain, thermal conductivity increased with saturation.

The effective thermal conductivity of the GDL increases with increasing water saturation for both of the inlet conditions as seen in Figure 4.22. The increase with increasing saturation is more significant for the through-plane thermal conductivity. For a GDL modeling domain with a total saturation of 24.4% (flooded inlet condition), increases of 20.8% and 5.4% are noted for the
through-plane and in-plane thermal conductivities, respectively. This same trend was reported in numerical work by Zamel et al. [25], where an increase in the through-plane thermal conductivity by a factor of 3 times was noted for a fully saturated GDL, and an increase of just 5% was observed for the in-plane thermal conductivity in a fully saturated GDL.

Saturation has a larger effect on the through-plane thermal conductivity due to the mechanisms of heat transfer in the GDL. Water in the GDL is located in the pores at the intersection of carbon fibres. While the thermal conductivity of the water is lower than that of the carbon fibres, it is still higher than air and as such, will aid in diminishing the thermal resistance in the through-plane direction. This effect is not as significant in the in-plane direction as heat is transferred along fibres and the additional material is not aiding in this process. The results illustrate the importance of fibre orientation on the prediction of the thermal conductivity value.

In experimental work on the through-plane thermal conductivity, Burheim et al. reported increases of 40 – 70% for 25% saturation [8] and an increase by a factor of 4 for a saturation of 60 – 70% for carbon paper GDL materials [16]. While increases in the thermal conductivity were noted with increasing saturation in this work, the increases from this work were not as high as those reported in [8]. The discrepancy can be attributed to the saturation patterns. The pore network models employed in this work used an invasion percolation algorithm that saturated the GDL until breakthrough occurred. While multiple pathways for water exist using this method, there is only one pore at the outlet that is filled (a single breakthrough location). The location of water in the through-plane position can be seen in Figure 4.15. In the experimental set-up for the work by Burheim et al. [16], water is introduced into the GDL for 20 - 30 seconds via a vacuum system, thus likely providing a more uniform and unrealistic through-plane water distribution than what the numerical model would predict.

4.4.9 Heterogeneous Porosity Profiles

The effect of the heterogeneous through-plane porosity distributions on the effective thermal conductivity was investigated. The properties of the four Toray carbon paper GDL materials are presented in Table 4.5. The predictions for the through-plane and in-plane effective thermal conductivity of the four GDL materials are given in Table 4.6 for the cases of a heterogeneous
and uniform porosity distribution. Each value reported in Table 4.6 is the average of 10 LBM simulations with stochastic modeling domains.

A comparison between the simulation results for the through-plane thermal conductivity shows a notably higher thermal conductivity when a uniform porosity distribution is employed. The results for the through-plane thermal conductivity for heterogeneous porosity distributions 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 have on the through-plane thermal conductivity of the GDL. Both of these trends were also noted in Chapter 3 with the analytical model.

As noted in Chapter 3, the Toray carbon paper materials display local porosity minima and maxima throughout the thickness of the material. Even though bulk porosities are the same, the through-plane thermal conductivity differences between the uniform and heterogeneous porosity domains can be attributed to the minimum porosity regions in the heterogeneous porosity distributions. Through-plane thermal conductivity variations within the heterogeneous modeling domains can be attributed to differences in the porosity distributions of the four GDL materials considered. These variations in the thermal conductivity with GDL material are not as pronounced with the LBM simulations as they were in the results from the analytical model in Chapter 3. Due to computational limits, the domain size for the LBM simulations (400 μm x 400 μm x GDL thickness) is smaller than that used for the analytical model (1500 μm x 1650 μm x GDL thickness). The effect of the transitional surface regions between the four materials studied may be decreased from this smaller grid size used for the LBM simulations.

This difference between the simulation results for uniform bulk porosity and heterogeneous porosity distribution does not seem to affect the thermal conductivity in the in-plane direction; however, the thermal conductivity was noted to increase with increasing material thickness as previously noted in Section 4.4.5.

4.5 Conclusion
In this chapter, the anisotropic effective thermal conductivity of the PEM fuel cell GDL was determined using 2-D and 3-D two-phase conjugate fluid-solid thermal lattice Boltzmann models. The geometry of the GDL was generated numerically with a new algorithm for
stochastic fibre placement. It was shown that the through-plane and in-plane thermal conductivities strongly depend on the porosity and fibre orientation. It was also found that the anisotropic reconstruction of the GDL results in an anisotropic thermal conductivity tensor, with significantly larger values of the in-plane thermal conductivity than the through-plane thermal conductivity.

The 2-D GDL reconstructions resulted in a lower than expected value for the through-plane thermal conductivity. The 3-D reconstructions provided a more accurate through-plane thermal conductivity measurement and good agreement with previously reported values in the literature. The range of values for in-plane thermal conductivity did not significantly change between the 2-D and 3-D simulations, but the variability in the results was reduced with the 3-D GDL reconstruction.

GDL compression was modeled with the reduced compression model [61]. Both the through-plane and in-plane thermal conductivity increased with increasing compression, but compression had a more significant impact on the through-plane thermal conductivity. The effect of binder and PTFE on the thermal conductivity was investigated. Both were found to increase the through-plane thermal conductivity, but the binder with a larger thermal conductivity value had a larger impact. The in-plane thermal conductivity was not affected by the addition of PTFE.

The addition of the MPL caused a decrease in the through-plane thermal conductivity, but further changes in the depth or thickness of the MPL did not have an effect. The in-plane thermal conductivity did not change with the addition of an MPL. The through-plane thermal conductivity of a GDL/MPL assembly decreased with increasing compression. The MPL is acting as a thermal resistor to through-plane heat transfer, and its impact on the overall thermal conductivity increases with increasing compression. The in-plane thermal conductivity of the GDL/MPL assembly exhibited the same increase with compression as a GDL without the MPL.

The model was used to study the effect of residual water on the through-plane and in-plane thermal conductivities. Three-dimensional pore network modeling with invasion percolation was used to simulate water in the GDL modeling domains. It was shown that the through-plane and in-plane thermal conductivities increase with liquid water saturation. For a GDL modeling domain with a total saturation of 24.4% (flooded inlet condition), increases of 20.8% and 5.4% are noted for the through-plane and in-plane thermal conductivities, respectively. The anisotropic
nature of the GDL was emphasized by the results indicating the through-plane thermal conductivity increased more significantly with increasing saturation.

The model was used to study the effects of heterogeneous porosity distributions presented in [13]. 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.
### 4.6 Tables

Table 4-1: Computational domain size independence study results for LBM simulations from Section 4.4.1.

<table>
<thead>
<tr>
<th>Grid Size</th>
<th>Parallel LBM Prediction (W/m K)</th>
<th>Parallel Relative Deviation (%)</th>
<th>Series LBM Prediction (W/m K)</th>
<th>Series Relative Deviation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100×100</td>
<td>60.009</td>
<td>0.00010</td>
<td>0.06096</td>
<td>0.01629</td>
</tr>
<tr>
<td>140×140</td>
<td>60.0079</td>
<td>0.00012</td>
<td>0.06079</td>
<td>0.01344</td>
</tr>
<tr>
<td>180×180</td>
<td>60.0069</td>
<td>0.00013</td>
<td>0.06070</td>
<td>0.01186</td>
</tr>
<tr>
<td>200×200</td>
<td>60.0069</td>
<td>0.00013</td>
<td>0.06047</td>
<td>0.00814</td>
</tr>
</tbody>
</table>
Table 4-2: Lattice spacing independence study results for LBM simulations from Section 4.4.1.

<table>
<thead>
<tr>
<th>Domain Size</th>
<th>Lattice Spacing (μm/lu)</th>
<th>Predicted Effective Through-Plane Thermal Conductivity (W/m K)</th>
<th>Relative Deviation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100x100x100</td>
<td>2.00</td>
<td>0.4640</td>
<td>-</td>
</tr>
<tr>
<td>150x150x150</td>
<td>1.33</td>
<td>0.4684</td>
<td>0.948</td>
</tr>
<tr>
<td>200x200x200</td>
<td>1.00</td>
<td>0.4674</td>
<td>0.732</td>
</tr>
</tbody>
</table>
Table 4-3: Reported values of through-plane and in-plane thermal conductivities in the literature for a sample of porosity of 78%.

<table>
<thead>
<tr>
<th></th>
<th>Reported $k_{\text{eff}}$ (W/m K)</th>
<th>Porosity</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Through-plane</td>
<td>1.84</td>
<td>78%</td>
<td>[68]</td>
</tr>
<tr>
<td></td>
<td>1.7</td>
<td></td>
<td>[26]</td>
</tr>
<tr>
<td></td>
<td>1.4</td>
<td></td>
<td>[20]</td>
</tr>
<tr>
<td></td>
<td>0.5</td>
<td></td>
<td>[16]</td>
</tr>
<tr>
<td>In-plane</td>
<td>21 - 23</td>
<td>78%</td>
<td>[68]</td>
</tr>
<tr>
<td></td>
<td>17.5</td>
<td></td>
<td>[22]</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td></td>
<td>[24]</td>
</tr>
</tbody>
</table>
Table 4-4: Fitting parameters for Equation 4.16 and comparison with work in Zamel et al. [24] for a D3Q15 LBM simulation results from Section 4.4.3.

<table>
<thead>
<tr>
<th></th>
<th>$C_1$</th>
<th>$C_2$</th>
<th>$C_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>In-Plane – current work</td>
<td>0.999</td>
<td>-0.00129</td>
<td>0.4677</td>
</tr>
<tr>
<td>Through-Plane – current work</td>
<td>0.9454</td>
<td>-0.01675</td>
<td>0.8852</td>
</tr>
<tr>
<td>In-Plane – [24]</td>
<td>0.997</td>
<td>-0.009</td>
<td>0.344</td>
</tr>
<tr>
<td>Through-Plane – [24]</td>
<td>0.975</td>
<td>-0.002</td>
<td>0.865</td>
</tr>
</tbody>
</table>
Table 4-5: GDL material properties from Fishman et al. [13] employed to generate GDL modeling domains with a heterogeneous porosity distribution for Section 4.4.9.

<table>
<thead>
<tr>
<th>Material</th>
<th>Thickness (µm)</th>
<th>Bulk Average porosity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Toray TGP-H-030</td>
<td>118</td>
<td>82.93</td>
</tr>
<tr>
<td>Toray TGP-H-060</td>
<td>220</td>
<td>82.09</td>
</tr>
<tr>
<td>Toray TGP-H-090</td>
<td>300</td>
<td>82.60</td>
</tr>
<tr>
<td>Toray TGP-H-120</td>
<td>360</td>
<td>78.69</td>
</tr>
</tbody>
</table>
Table 4-6: LBM simulation results for the effective thermal conductivity with a heterogeneous porosity distribution from Section 4.4.9.

<table>
<thead>
<tr>
<th>Material</th>
<th>Bulk Average Porosity</th>
<th>Through-plane Effective Thermal Conductivity</th>
<th>In-plane Effective Thermal Conductivity</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Bulk Uniform Porosity (W/m K)</td>
<td>Heterogeneous Porosity (W/m K)</td>
</tr>
<tr>
<td>TGP-H-030</td>
<td>0.8192</td>
<td>0.4628</td>
<td>0.2338</td>
</tr>
<tr>
<td>TGP-H-060</td>
<td>0.806</td>
<td>0.6006</td>
<td>0.4081</td>
</tr>
<tr>
<td>TGP-H-090</td>
<td>0.8125</td>
<td>0.5244</td>
<td>0.2929</td>
</tr>
<tr>
<td>TGP-H-120</td>
<td>0.777</td>
<td>0.9706</td>
<td>0.7753</td>
</tr>
</tbody>
</table>
4.7 Figures

Figure 4-1: 3-D view of a carbon paper GDL modeling domain with a uniform porosity of 78%. The domain is a cube with sides of 400 μm in length.
Figure 4-2: A single GDL modeling domain that employs the reduced compression model. The domain has an uncompressed porosity of 78% and an uncompressed domain size of 200 μm x 200 μm x 300 μm, and is shown (a) uncompressed, (b) at a compression ratio, \( c \), of 75%, and (c) at a compression ratio of 50%.
Figure 4-3: GDL modeling domain with a porosity of 78%, (a) untreated, (b) with the addition of 30 wt.% binder, and (c) with the addition of 20 wt.% PTFE.
Figure 4-4: GDL modeling domain with a porosity 78% with a microporous layer. The GDL has dimensions of 300 μm x 300 μm x 300 μm. The microporous layer (shown in green) has a depth of 20 μm and a thickness of 50 μm.
Figure 4-5: Liquid water location after invasion percolation for a (a) flooded inlet condition (23.9 % total saturation and (b) one point injection inlet condition (12.6% total saturation).
Figure 4-6: Saturation distributions for (a) flooded inlet condition (23.9% total saturation) and (b) one point injection inlet condition (12.6%) from 3-D pore network model simulations.
Figure 4-7: GDL modeling domain with liquid water for a flooded inlet condition. Fibres and binder are shown in yellow and water in blue.
Figure 4-8: 2-D GDL reconstructions (a) through-plane, and (b) in-plane structures. These two structures are slices of the numerically generated 3-D GDL modeling domain in $z-x$ (or $z-y$) and $x-y$ planes, respectively.
Figure 4-9: Predicted effective thermal conductivity as a function of porosity from 2-D simulations on a D2Q9 lattice for: (a) through-plane, and (b) in-plane.
Figure 4-10: Surface contours of the heat flux vector based on 2-D LBM simulations on a D2Q9 lattice.
Figure 4-11: Predicted effective thermal conductivity of stochastic numerically generated 3-D GDL modeling domains as a function of porosity: (a) through-plane, and (b) in-plane.
Figure 4-12: Comparison of predicted values of the in-plane effective thermal conductivity using 2-D (on a D2Q9 lattice) and 3-D (on a D3Q15 lattice) LBM simulations as a function of porosity.
Figure 4-13: Dimensionless in-plane and through-plane effective thermal conductivities obtained from LBM simulations with curve fit given in Equation 4.16 (solid line), and reported relation of Zamel et al. [24] (dashed line).
Figure 4-14: Predicted through-plane and in-plane effective thermal conductivities as a function of porosity obtained from LBM simulations. The two methods of the stochastic fibre placement algorithm presented are compared.
Figure 4-15: Predicted through-plane and in-plane thermal conductivity: (a) as a function of compression ratio, and (b) as a function of porosity for uncompressed and compressed GDL modeling domains.
Figure 4-16: Predicted in-plane effective thermal conductivity as a function of aspect ratio for a porosity of 60%, 70%, and 80%.
Figure 4-17: Predicted effective thermal conductivity as a function of binder weight content: (a) through-plane and (b) in-plane.
Figure 4-18: Predicted effective thermal conductivity as a function of PTFE weight content for a uniform and non-uniform through-plane PTFE distribution: (a) through-plane, and (b) in-plane.

The non-uniform PTFE distribution is obtained from Rofaiel et al. [51].
Figure 4-19: Predicted effective thermal conductivity as a function of MPL thickness for a constant MPL intrusion depth of 20 μm: (a) through-plane, and (b) in-plane.
Figure 4-20: Predicted effective thermal conductivity as a function of MPL intrusion depth for a constant MPL thickness of 50 μm: (a) through-plane, and (b) in-plane.
Figure 4-21: Predicted through-plane and in-plane effective thermal conductivity as a function of the compression ratio. A comparison between the GDL and the GDL/MPL assembly is presented.
Figure 4-22: (a) Schematic representation of the GDL/MPL assembly and (b) equivalent thermal resistance network.
Figure 4-23: Predicted through-plane and in-plane effective thermal conductivity as a function of the water saturation for: (a) a flooded inlet condition and (b) one point injection inlet condition.
Chapter 5 Conclusion

In this thesis, two modeling methods were investigated to study the thermal conductivity of a PEM fuel cell GDL. A literature survey on the GDL of the PEM fuel cell, the experimental and analytical determination of the thermal conductivity, and LBM were presented. First, an analytical model capable of determining the through-plane thermal conductivity of representative physical GDL models, informed by microscale computed tomography imaging of four commercially available GDL materials was presented. Next, a thermal lattice Boltzmann model was investigated to study the through-plane and in-plane thermal conductivity of the GDL with different geometric inputs. A new method for a stochastic fibre placement algorithm to create GDL modeling domains was presented. A summary of the two modeling methods is as follows:

- The analytical modeling approach provided insight into the mechanisms of heat transfer by providing a through-plane distribution of thermal conductivity and thermal resistance. This model accommodated large grid sizes at a low computational cost, but was limited by geometric constraints and was only able to consider heat transfer at existing fibre contact points. Without devising a new model, the in-plane thermal conductivity cannot be determined using this model.
- The LBM model was capable of performing a parametric study on the through-plane and in-plane thermal conductivity by incorporating different phases into the model (binder, PTFE, water, MPL) and GDL compression. It was able to provide information on the temperature and heat flux distribution in the modeling domain, but was limited to the grid size used. The LBM simulations were significantly more computationally expensive than the analytical model.

The main conclusions of this thesis relating to the thermal conductivity are:

- The two-dimensional in-plane LBM simulations provided reasonable estimates of the thermal conductivity at a significantly lower computational cost than the three-dimensional simulations. Adding the third dimension, however, dampens the effect of structure randomness and reduces the variance in the predicted data points.
The predicted values of effective through-plane thermal conductivity from 2-D LBM simulations are almost one order of magnitude smaller than those predicted from three-dimensional simulations.

The GDL is highly anisotropic. The in-plane thermal conductivity is an order of magnitude larger than the through-plane. Fibre distribution and orientation, in addition to porosity, play a significant role in the resulting thermal conductivity value.

Both the through-plane and in-plane thermal conductivities increase with increasing compression. Compression has a stronger impact on the through-plane thermal conductivity.

The impact of GDL heterogeneity on the thermal conductivity was investigated with both models. The through-plane thermal conductivity is strongly affected by the higher porosity values in the transitional surface regions, regardless of the overall bulk porosity value or the local maximum thermal conductivity value.

The presence of carbonized binding material in the GDL leads to increases in both the through-plane and in-plane thermal conductivities.

The addition of PTFE causes an increase in the through-plane thermal conductivity. This increase was less significant when a heterogeneous through-plane PTFE distribution was considered compared to a uniform through-plane PTFE distribution.

The analytical modeling results revealed that the surface transition regions of the porosity distributions have a dominating effect over the addition of PTFE in impacting the overall thermal conductivity.

The addition of the MPL causes a decrease in the through-plane thermal conductivity, but further changes in the depth or thickness of the MPL did not have an effect. The in-plane thermal conductivity does not change with the addition of an MPL.

The through-plane thermal conductivity of a GDL/MPL assembly decreased with increasing compression. The MPL is acting as a thermal resistor to through-plane heat transfer and its impact on the overall thermal conductivity increases with increasing compression. The in-plane thermal conductivity of the GDL/MPL assembly exhibited the same increase with compression as a GDL without the MPL.

The through-plane and in-plane thermal conductivities increase with liquid water saturation. The anisotropic nature of the GDL was emphasized by the results indicating
the through-plane thermal conductivity increased more significantly than that of the in-plane thermal conductivity with increasing saturation.

The outcomes of this work provide insight into the dominating effect of heterogeneity and anisotropy of the GDL on the thermal management required for improved PEMFC performance.
Chapter 6 Future Works

Based on the findings in this thesis, there are future areas of the thermal conductivity of the GDL and LBM that can be investigated.

The existing LBM model that has been developed is capable of solving the thermal conductivity of generated GDL modeling domain. With some small modifications to the LBM model, it can be extended to look at other properties of these GDL modeling domains, including but not limited to, permeability and diffusivity.

The thermal contact resistance between the GDL and bipolar plate or catalyst layer is another important thermal parameter in the study of temperature and heat transfer in a PEM fuel cell and has been determined in experimental work. Extending the thermal LBM framework and GDL modeling domains to include the calculation of this parameter would provide more insight into heat transfer within the GDL and PEM fuel cell.

It was found that the through-plane thermal conductivity is strongly dependent on porosity and the heterogeneous through-plane porosity. In this thesis, only one sample of each GDL material was assessed, and future work should include a larger number of samples to fully characterize the reported trends. Future work should also include compressed porosity profiles obtained from µCT experiments. This will allow for consideration of the effects of compression in different areas of the fuel cell, as it has been noted that the land and channel areas of the GDL will have different values of compression [16]. A comparison can be made between the results from the µCT experiments and the reduced compression model used in this thesis.
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