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A unit-cell approach for determining the effective thermal conductivity of the polymer electrolyte membrane fuel cell microporous layer

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ABSTRACT

In this study, we investigated the use of unit-cells comprised of nano-scale spherical particles to model the microporous layer (MPL) of polymer electrolyte membrane (PEM) fuel cells. We determined the effective thermal conductivity of various unit-cells based on body-centred cubic (BCC) and face-centred cubic (FCC) orientations, developed for commercially available MPL materials SGL-10BB and SGL-10BC. Informed by previous work done by the authors, unit-cells are constructed with constant particle size and filling radius, while varying the particle separation distance. Here it was found that the effective thermal conductivity of the MPL is heavily dependent on particle spacing due to the high thermal conductivity of the carbon particles. Also, it was found that SGL-10BB unit-cells are slightly more conductive than SGL-10BC. The results of this study can be used in the stochastic representation of the MPL, and can be extended to measurements of oxygen diffusion and electrical conductivity.

1. Introduction

Polymer electrolyte membrane (PEM) fuel cells are electrochemical power generation devices with many advantages, such as maintaining high efficiency during operation, operating with a high power to volume ratio [1–3], and having the ability to operate with zero local greenhouse gas emissions [4]. However, before PEM fuel cells can reach widespread adoption, effective thermal management must be achieved in order to optimise performance and durability levels through the design of cell components [5–7]. The ability to model each component with the highest possible accuracy is key to improving the design and performance of the PEM fuel cells and thereby increasing its widespread adoption.

This study focuses on the microporous layer (MPL) of the PEM fuel cell. The MPL is a nano-porous material found between the PEM fuel cell gas diffusion layer (GDL) and catalyst layer (CL). The MPL is composed of particle agglomerations built from carbon particles ranging in diameter from approximately 10–100 nm [8,9], bound together with polytetrafluoroethylene (PTFE) [10,11]. The MPL is a relatively new material structure used in PEM fuel cell operation with growing interest as studies have shown that the incorporation of an MPL improves cell performance [12–15]. Though the specific relationships between the MPL structure and PEM fuel cell performance are somewhat unknown, it has been hypothesised that the MPL improves cell performance by:

- minimizing ohmic resistances between the GDL and CL due to surface pores in the MPL being smaller than that of the GDL [16]
- improving water management due to its:
  - ○ highly hydrophobic nature, permeating water through the MPL cracks and large pores [17]
  - ○ larger temperature gradients, affecting the humidity gradient in the through-plane direction of the MPL, helping diffuse water vapour away from the reaction site [18]
- Improving structural integrity of the cathode by protecting the membrane from overhanging GDL fibres which may puncture the membrane, causing reactant crossover.

Due to the random nature of the MPL particle agglomerations [8–10,19], the variability in the MPL structure, and scale of the structural features [10,19], developing an accurate representation of the MPL has associated challenges. In particular, the resolution required to effectively model the contact region between particles (a dominant mechanism in thermal conductance between particles [8]) has not been achieved in literature [19,20]. The current models found in literature are based on the stochastic reconstruction of the entire MPL material [9,10,21] or deterministic reconstructions of the MPL based on computed tomography [19,20,22]. The impact of the nature of contact between the particles on thermal conductance has also been studied by the authors group in an earlier work [8], where it was found that the MPL should be modelled with

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material-specific inputs regarding the particle size and nature of contact. Incorporating higher resolution information is necessary for modelling heat transfer between MPL particles and nano-pores found between the particles, but has been disregarded in the past since the resolution of computed tomography reconstructions of the MPL are incapable of imaging the nano-structures with such resolution [19,20,23].

Therefore, reconstruction of computed tomography does not provide the required resolution to fully characterise the MPL. For stochastically generated models, such high resolutions require significant time to generate (and solve for heat transfer) a representative volume of the MPL (~2.5 μm) at a resolution of ~2–5 nm/voxel, and this time is not practical for evaluating numerous MPL materials. An alternative method is through the use of unit-cell analysis.

In this study, we present unit-cell analysis (UCA) as an alternative technique for modelling conductive and convective properties through the MPL, considering the resolution issues with modelling the MPL structure. This allows for considering high-resolution transport properties at the nano-scale while minimizing the time required for macro-scale representative domain analysis and accounting for cracks and large pore spaces commonly found in the MPL interaction with the GDL. The technique of UCA has been used to model the GDL by Sadeghi et al. [24–26], who considered fibre interactions to be repeating structures.

In treating the MPL as a packed bed of carbon particles, the thermal conductivity can be modelled using the unit cell approach. A review of some other methods of modelling packed bed of particles was presented by Tsotsas and Martin [27]. However, this approach has not been applied to the investigation of thermal conductivity through the MPL. Therefore, the objective of this study is to obtain specific unit-cell configurations for SGL-10BB and SGL-10BC informed by the material specific results obtained in [8], which can then be used as building blocks to reconstruct the MPL stochastically. These unit-cells can also be used to populate low-resolution material reconstructions (such as obtained from X-ray CT [28]) which may have been previously viewed as insufficient for modelling heat transfer (along with other transport phenomena). A schematic of how UCA can be applied in the construction of nano-resolved MPL structures is shown in Fig. 1 (in 2-dimensions for illustrative purposes).

2. Unit-cell reconstruction of MPL particles

The unit-cell reconstructions utilised to model MPL particles are based on body-centred cubic (BCC) and face-centred cubic (FCC) orientations. As can be seen in Fig. 2, the BCC unit-cell contains particles positioned at the volumetric centre of the unit-cell and eight corners. The FCC unit-cell is similar to the BCC structure, with eight 1/8th particles located at the eight corners of the unit-cell. However, instead of having a particle at the volumetric centre, six half-particles are centred at the six faces of the unit-cell. Visual representations of BCC and FCC structures, assuming point contact, are displayed in Fig. 2, for various unit-cell side lengths, **a**, measured in voxels. Increasing the voxel resolution (voxel/nm) used in the unit-cell reconstruction improves the accuracy of the reconstruction (Section 2.1) and the accuracy of the calculated effective thermal conductivity (Section 2.2).

2.1. Voxel resolution selection based on unit-cell reconstruction accuracy

One of the first considerations in the development of the unit-cells is the resolution used, corresponding to the number of voxels utilised in the reconstruction. As can be seen in Fig. 2, as the voxel count increases, related to side length **a**, the particles appear to be more spherical. The accuracy of the reconstruction can be quantified by using the solid volume fraction, *svf*, which is defined as the amount of solid volume in a unit-cell divided by the total unit-cell volume. Eqs. (1) and (2) show the analytical solid volume fractions for the BCC and FCC structures, assuming the particle diameters are all equal. The unit-cell side length, $\boldsymbol{a}$, can be measured in terms of the particle radius, *r*, given that the particles are touching along the body-diagonal for BCC structures ($\mathbf{a}_{\mathrm{BCC}}\sqrt{3} = 4r$) and along the face-diagonal for FCC structures ($\mathbf{a}_{\mathrm{FCC}}\sqrt{2} = 4r$).

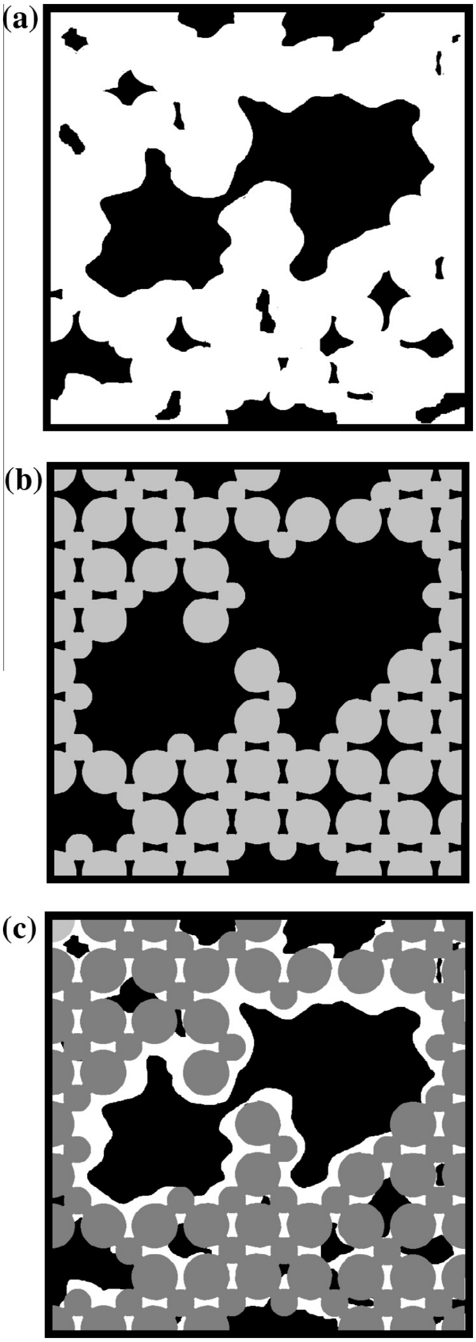

Fig. 1. Schematic of an MPL cross-section, approximately 1 μm by 1 μm, showing the use of unit-cells for population low-resolution MPL reconstructions in 2D; (a) schematic of an MPL cross-section, (b) schematic of a unit-cell populated cross section, and (c) both images overlaid.