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Characterization of transport phenomena in porous transport layers using X-ray microtomography

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HIGHLIGHTS

- Xray tomography is used to image commercial porous transport layers for fuel cells (85).
- 3D image-based modeling is used to quantify porosity, permeability, & diffusivity (85).
- Porous PTLs show a large variation in transport properties; dense PTLs do not (81).
- Cracks in PTLs play a key role in estimating transport properties from 3D images (84).

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ABSTRACT

Among different methods available for estimating the transport properties of porous transport layers (PTLs) of polymer electrolyte membrane fuel cells, X-ray micro computed tomography (X-μCT) imaging in combination with image-based numerical simulation has been recognized as a viable tool. In this study, four commercially-available single-layer and dual-layer PTLs are analyzed using this method in order to compare and contrast transport properties between different PTLs, as well as the variability within a single sheet. Complete transport property datasets are created for each PTL. The simulation predictions indicate that PTLs with high porosity show considerable variability in permeability and effective diffusivity, while PTLs with low porosity do not. Furthermore, it is seen that the Tomadakis-Sotirchos (TS) analytical expressions for porous media match the image-based simulations when porosity is relatively low but predict higher permeability and effective diffusivity for porosity values greater than 80%. Finally, the simulations show that cracks within MPL of dual-layer PTLs have a significant effect on the overall permeability and effective diffusivity of the PTLs. This must be considered when estimating the transport properties of dual-layer PTLs. These findings can be used to improve macro-scale models of product and reactant transport within fuel cells, and ultimately, fuel cell efficiency.

1. Introduction

The porous transport layer (PTL) is a critical component within polymer electrolyte membrane fuel cells (PEMFCs) as it facilitates fluid flow of the reactants and by-products, electron conductivity from the catalyst layer to the bipolar plates, heat conduction, and mechanical support for the membrane assembly [1]. PTLs, made mostly of carbon-based materials, can be classified as either single or dual layer. The single-layer PTL, consisting of carbon fibres and 5–30% PTFE loading, has a thickness between 150 and 500 μm and an effective pore size of approximately 10–30 μm. The reasons for PTFE loading are to increase hydrophobicity of the carbon fibre mat and to bind the carbon fibres together. The dual-layer PTL contains an additional micro porous layer consisting of carbon particles mixed with PTFE that is coated onto the single-layer PTL. Within the literature, there is no convention on naming of sub-layers within the PTL. In this paper, the first layer (carbon fibre + PTFE) will be named GDL, while the second layer will be named MPL. Thus, a single-layer PTL consists of only GDL, while a dual-layer PTL consists of both GDL and MPL.

The main properties related to transport phenomena [2] of PTLs include porosity, permeability, and effective diffusivity [3]. Porosity,

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ε, is defined as the relative void volume within a material [4],

$$\varepsilon = 1 - \frac{V_{PTL}}{V_{total}}, \tag{1}$$

where V_{PTL} is the volume of PTL material contained within a bounding volume V_{total}. Porosity is the main parameter of a porous medium, strongly correlating with all other properties of interest [5]. Permeability is defined as the structural resistance to a pressure driven flow. The well-established formula to define this characteristic is Darcy's law,

$$-\nabla P = \frac{\mu}{k}\vec{v}, \tag{2}$$

where k is the absolute permeability of the porous medium, μ is the viscosity of the flow, $\vec{v}$ is the velocity of the fluid and P is the pressure. Permeability is a structural property, and thus neither the viscosity, nor the applied pressure, nor the velocity affect its value. However, for Darcy's law to be valid, flow must occur in the low Reynolds number ($Re < 1$) regime, as is the case in PEMFCs. Effective diffusivity, $f(\varepsilon)$, is defined as the structural resistance against the movement of species from a region of high concentration to a region of low concentration [6],

$$f(\varepsilon) = \frac{D_o}{D_{eff}}, \tag{3}$$

where D_0 is the bulk diffusion coefficient of a chemical species, and D_{eff} is the apparent diffusion coefficient of a given porous medium. As PTLs are very thin in one direction as compared to the other two directions, these transport properties are anisotropic, *i.e.* they have different values in the “in-plane” directions (across the thickness) and the “through-plane” direction (along the thickness).

X-ray micro-computed tomography (X-μCT) is a non-destructive imaging technique that can provide the internal 3D structure of porous media. Recently, this method has been used extensively to quantify porosity in PTLs [4,7–14]. A thorough literature review on this subject was provided in Ref. [10]. There are two main challenges when using X-μCT to quantify porosity in PTLs. The first challenge relates to binary segmentation, which is used to numerically separate PTL material from the background. Different thresholding techniques have been used in the literature [10,13–15] to perform this task. Recently, Odaya et al. [13] showed that for high resolution 3D images of PTLs, a direct selection of a global threshold parameter is the most efficient while remaining quite accurate. The second challenge is identification of the bounding volume in which the material is enclosed. This difficulty is due to the fact that the surface of PTLs is quite rough. Hasanpour et al. [10] proposed a robust method to identify the surface of highly porous media, known as the Rolling Ball method, and demonstrated its applicability to PTLs.

X-μCT has also been used to characterize transport phenomena in PTLs. In a study by Becker et al. [16], numerical methods were applied to the 3D X-μCT images to quantify permeability, effective diffusivity, and electrical conductivity, which were then compared to experimental measurements. The effective diffusivity and permeability results showed very good agreement; however, the electrical conductivity results deviated from the experimental results, especially in the through-plane direction. Further, it was found that the through-plane effective diffusivity is lower than the in-plane value by a factor of two. Fishman et al. [17] used the porosity distribution data from X-μCT in combination with an analytical model (the so-called Tomadakis-Sotirchos [18] equations), to show the anisotropic nature of permeability and effective diffusivity within PTLs. Nabovati et al. [19] studied the effect of the heterogeneous porosity distribution on permeability of PTLs, concluding that higher through-plane heterogeneity leads to higher in-plane permeability. Instead of directly using the X-μCT data to calculate permeability, this study used the porosity distribution data from X-μCT to make a stochastic model with a simplified 3D structure. In another study, James et al. [20] performed X-μCT on compressed PTLs to obtain 3D structures at 0%, 20% and 40% compression. The structures were then used as the input geometry for modeling diffusive flow, showing that the effective diffusivity of a sample decreases with increased compression. Furthermore, since higher compression leads to a decrease in porosity, samples with low porosity showed low effective diffusivity [20].

Although the use of X-μCT to analyze transport phenomena in PTLs has shown to be quite effective, a systematic study comparing different PTLs has not yet been carried out. At present, the optimum size of the PTL representative volume element for simulating permeability and effective diffusivity is also unknown. A study conducted by Bernard et al. [21] on Al-Cu alloys showed that sample size plays a critical role when simulating transport phenomena in porous media using X-μCT datasets. In addition, in dual-layer PTLs, it is known that the MPL has a significant effect on permeability and effective diffusivity. In general, the MPL contains lower overall porosity and very small pores (approximately 100 nm in size [22]), causing the permeability and effective diffusivity values to decrease by one order of magnitude [23] as compared to single-layer PTLs. However, the MPL also contains large cracks (formed during the coating process) that can be hundreds of microns in size [24]. The effect of cracks within the MPL on dual-layer PTL transport phenomena is unknown.

In this study, a comprehensive analysis of the transport properties (porosity, permeability, and effective diffusivity) of three commercially-available single-layer PTLs (SGL 35BA, SGL 35BC, Toray 090 and Freudenberg H2315 I6) is carried out using X-μCT imaging and image-based numerical simulations. The effect of cracks within the MPL on the transport phenomena in a dual-layer PTL (SGL 35BC) is also examined. The research focused on both providing quantitative data for validation of analytical equations, and investigating property variation within the same sheet. Although the transport properties of PTLs has been previously investigated, to date there has not yet been a systematic study of multiple products from multiple vendors, nor a study on property variability. The results provide a complete dataset that can be used to inform macro-scale simulations of gas flow in PEMFCs, and to provide new recommendations for improving diffusion media.

2. Experimental methodology

2.1. Materials

Four commercially available PTLs were investigated, SGL 35BA and SGL 35BC (donated by SGL Carbon group, Wiesbaden, Germany), as well as Toray 090, and Freudenberg H2315 I6 (purchased from College Station, TX, United States). SGL 35BA is a single-layer PTL with 5 wt% PTFE loading and a specified thickness of 300 μm. SGL 35BC is a dual-layer PTL with 5 wt% PTFE loading and a specified thickness of 325 μm [25]. The Toray 090 and Freudenberg H2315 I6 were selected due to their thickness and PTFE loading, which are similar to SGL 35BA, enabling a direct comparison between different manufacturing processes.

2.2. Scanning procedure

The tomographic imaging was performed using a Zeiss