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# Review on microstructure modelling of a gas diffusion layer for proton exchange membrane fuel cells

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ABSTRACT

A gas diffusion layer (GDL) is the key component in a proton exchange membrane fuel cell (PEMFC), where transportation of reactants and oxidants to electrodes and removal of water from the cell occur. Accurate prediction of the effective transport properties of GDL is important in understanding its effects on PEMFC performance. However, prediction of GDL behavior is challenging because of the complex geometries involved. Hence, microstructure modelling of GDL is highly beneficial in this condition. This article reviews numerous research endeavors that focused on GDL modelling and the parameters that affect the GDL microstructure. This review aims to understand how each parameter affected the GDL performance. The highlighted parameters in this article are fiber diameter, GDL thickness, porosity, and the effect of polytetrafluoroethylene (PTFE).

## 1. Introduction

The proton exchange membrane fuel cell (PEMFC) is one of the most common types of fuel cells that show promise in green energy technology. This cell directly and efficiently converts chemical energy stored in hydrogen fuel to electrical energy, with water as the only by-product [1]. Hence, PEMFCs show the great potential to reduce energy consumption, pollutant emissions, and dependency on fossil fuels. However, the major constraints to PEMFC commercialization are durability and cost [1]. Since the last two decades, researchers have expended considerable efforts toward improving and advancing fundamental research on PEMFC technologies to achieve marketable products. Several main challenges in PEMFC components related to durability are the performance and effectiveness of the GDL.

Gas diffusion layers (GDL) are porous media that serve as one of the electrode components for membrane-electrode assembly (MEA) in PEMFCs. The main functions of GDL are for gas permeation to catalyst layer and water removal during fuel cell operation. GDL is a porous layer composed of randomly oriented carbon fibers that are either woven or non-woven [2]. The typical range of GDL thickness is between 200 and 400 μm, with fiber diameter in the range of 7–10 μm [3]. GDL is a vital component that performs a key function in PEMFC performance. Although GDL is not an electrochemical reaction site, it functions crucially to provide the reactants good access to the catalytic sites and effectively remove the reaction product, that is, water, from the electrode [4]. In essence, GDL should effectively transport the gas reactants from the flow channel to the catalyst layer, have high electronic conductivity, have a surface that enhances good electronic contact, and have a proper wetting characteristic for low-temperature applications [5].

PEMFC operation is highly influenced by GDL. Thus, proper prediction of the effective transport properties of GDL is important in understanding cell performance. Although several reports have evaluated the performance of PEMFC experimentally, employing numerical methods is more convenient to better understand the effective parameters in designing and optimizing the functions of fuel cells to improve fuel cell technologies [6]. Numerous experimental and modelling studies have been performed to investigate transport behavior and water distribution within the GDL.

However, accurate prediction of the transport properties of the GDL is challenging because of the complex geometries involved [7,8]. Specifically, GDL carbon fibers form a thin layer of stacked cylinders that extend far more into the in-plane direction than the through-plane direction [9]. In addition, when the GDL material is used in PEMFC, compression is applied by the flow field, which causes to different GDL microstructures in the regions under channel and under land [10].

Different GDL structures show substantial difference in water distribution in the GDL due to hydrophobic wettability characteristic [11]. Numerous studies have been conducted to investigate water transport in PEMFCs in recent years; however, the behavior of liquid

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water in a GDL at a pore-level is poorly understood. Experimental methods are not convenient towards a good understanding at a microscopic level because of the micro size of the GDLs porous structure. Two-phase computational model currently has been popular to investigate the fluid behavior and flooding phenomenon in PEMFC in various degree complexities [12–18]. However, all the above mentioned model are examples of macroscopic two-phase model facing with deficiency of realistic two-phase correlation, and hence they fail to incorporate the influence of the structural morphology of GDLs on liquid water transport behavior. Due to the lack of reliable two phase correlations, these models often deploy a generic curve-fitted capillary pressure-saturation data originally obtained by Udell [19] in the form of Leverett-J function from imbibition in water-wet unconsolidated sand [20]. In order to improve the reliability of the available two-phase PEMFC models, it is necessary to understand linkage between principle microstructure and two-phase characteristics of the GDLs in PEMFC.

Microstructure is important in GDL performance; hence, considering the microstructure properties that contribute to optimal transport properties is crucial. Microstructure modelling is an activity in which the microstructural features of materials are predicted and characterized. This specific activity will provide microstructure input data to upscale modelling as a function of process parameters needed for effective medium and performance modelling.

The microstructure modelling approach has a relevant potential to contribute to the improvement of the current GDL. Virtual geometries are a better alternative to resolve the high expense of tomography experiments, where computer-process-X-rays is used to produce tomographic image of a cross section through a solid object. One feature that all approaches have in common is that the generated geometries show the same characteristics as the original structure. The complex fiber structure can be simplified by applying assumptions, such as the fiber is assumed to be a straight cylinder, as long as the GDL with those assumptions can still satisfy the original functions. Researches on the GDL composition and structure have also been conducted. Simulations focusing on the GDL microstructure were conducted by Mukherjee and Wang, who are trying to develop a model that represented the structure of a real GDL [21]. Using this model, the tandem reproduced the pore structure and evaluated the GDL capillary pressure. Moreover, the two researchers evaluated the effects of GDL compression caused by cell clamping [22]. Schulz et al. modelled the relationship between GDL morphology and flooding behavior by capillary pressure [23].

## 2. Parameters of GDL microstructure

Knowing and understanding the physical properties of GDL is important in developing a GDL microstructure model. These properties constitute the variable parameters in constructing a 3D model of GDL. The present paper discusses the four main key parameters for a GDL microstructure, namely, porosity, thickness, diameter of fiber and polytetrafluoroethylene (PTFE) effect.

### 2.1. Porosity

Porosity is the volume ratio of void space in a material [24]. Porosity can also be defined as the ratio of non-fiber volume to the total volume [25]. Bulk porosity in a GDL is described as the total pore volume divided by the summation of the total solid volume and the pore volume.

Knowing how porosity impacts PEMFC performance is important. A previous report stated that the porosity distribution through the thickness of GDL samples is not uniform [26]. As a result, the overall transport properties of the porous material are affected by this heterogeneous porosity distribution. Nabovati et al. [3] showed that porosity heterogeneity increases the in-plane permeability and decreases the overall cross-plane permeability. Numerous studies on determination on the porosity geometry have been conducted. Igathinathane et al. [27] successfully developed an image processing method to determine the dimensions of the particles and their size distribution. Diego et al. [28] proposed a method to calculate the average pore radii on samples with spherical pores. He et al. [108] performed porosity calculations on a Toray carbon paper substrate; however, the sample did not possess a microporous layer (MPL) and the method provided only allowed for calculations on 2D porosity. Farmer et al. [24] used the area per layer technique through scanning electron microscopy (SEM) image processing to investigate the porosity of GDL in the presence of MPL and wetting agent. The method was able to calculate porosity in both GDL and MPL. This method is widely used to determine the surface geometry of GDL. The pore structures of GDL highly affect the performance in the mass transport region of PEMFC. GDL structure consists of a substrate (carbon cloth) coated with carbon powder, hydrophobic agent, and pore formers [29]. Connecting the performance characteristics with a comprehensive evaluation of porosity of the GDL is necessary. Hiramitsu et al. [30] shows that improved cell voltage can be obtained for even a slight amount of GDL pore micronization at the CL/GDL interface. Luo et al. [31] developed a pore network model, in which a porous medium is represented by a network of pores connected by narrower regions called throats.

Yan et al. [32] has investigated the effect of GDL porosity towards fuel cell performance as shown in Fig. 1. Based on the presented result, GDL with higher porosity provides better performance than GDL with lower porosity because a higher porosity provides higher limiting current density. This effect is attributed to the greater space for diffusion, which provides efficient distribution and transport of the reaction gases and the emerging water between the reaction zone and gas distributor [33]. However, in other studies, Obayopo et al. [34] found that the increasing effect decreases as the porosity increases. Porosity beyond 0.6 does not have a tangible effect on the fuel cell polarization curve.

Water tends to accumulate in regions with relatively high porosity because of lower associated capillary pressures [25,35], consequently causing water flooding in the layer [36]. In addition, high porosity affects thermal conductivity [25] by inducing a higher contact resistance in GDL [37]. In other words, high porosity may cause a performance drop despite the increase in limiting current density. Hence, the porosity should be sufficient and optimized to supply an effective amount of reactant and maintain higher electronic and thermal conductivity. This finding is supported by Brandon et al. [38]. Hence, considering the durability issues in the PEMFC structure, reasonable porosities should range between 0.4 and 0.6 [39].

### 2.2. Thickness

The GDL in a PEMFC serves three important functions, namely,

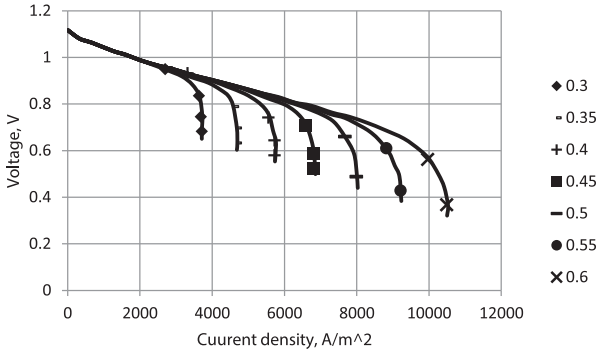


**Fig. 1.** Comparison of cell performance curves for models with porosity in range $0.3 \le \varepsilon 1 \le 0.6$ [32].