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Two-phase flow in the mixed-wettability gas diffusion layer of proton exchange membrane fuel cells

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HIGHLIGHTS

- Two-phase flow in mixed-wettability GDLs are investigated using a 3D VOF model.
- The GDL microstructures of PTFE spatial distribution are digitally reconstructed.
- The VOF predictions are compared with the LBM and experimental results.
- The effects of the PTFE distribution on water dynamics are studied.

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Gas diffusion layer
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Capillary pressure
Volume of fluid
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ABSTRACT

Polytetrafluoroethylene (PTFE) is widely employed to improve the hydrophobicity of gas diffusion layer (GDL) in proton exchange membrane (PEM) fuel cells. In this study, the effects of different PTFE loadings on the relationship of the capillary pressure P_c and water saturation s in the mixed-wettability GDL, i.e. P_c-s, are investigated using a three-dimensional (3D) volume of fluid (VOF) model. The simulated P_c-s curves are presented and compared with results obtained from the lattice Boltzmann model (LBM) and experiments. The good agreement between the VOF predictions and experiment data is achieved, indicating that the mixed wettability in the PTFE treated GDL is an important feature to understand two-phase behaviors in fuel cells. The homogeneous and heterogeneous PTFE distributions resulted from two PTFE drying methods (i.e. the vacuum and air dryings, respectively) are studied. It was found that the air drying GDL yields a high PTFE concentration near the water inlet and reduces water imbibition near the inlet. The simulated P_c-s correlation from VOF model was compared with standard Leverett correlation.

1. Introduction

Proton exchange membrane (PEM) fuel cells are promising alternative power devices of electric vehicles due to their outstanding merits such as high efficiency, negligible emissions and high power density. Water management is a major bottleneck for the improvement of fuel cell efficiency and durability [1]. In PEM fuel cells, the gas diffusion layer (GDL) is a key component which provides pore paths for reactant diffusion and product water removal. Excessive liquid water in GDLs may hinder reactant transport and increases the mass transport polarization [2]. Thus, effective water removal is important to ensure high efficient operation of fuel cells [3]. This issue requires comprehensive understanding of liquid water transport in GDLs [4].

In general, GDLs are fibrous media, and carbon paper and carbon cloth are popular GDL materials [5,6], as shown in Fig. 1. The inherent hydrophilicity of graphite (its static contact angle $\theta \approx 75$–86° [7–9]) may resist water removal in GDLs. A common strategy of facilitating water removal is to add hydrophobic agents, such as Polytetrafluoroethylene (PTFE) [10–15]. In fabrication, it is difficult to coat PTFE homogeneously in all the inner surfaces, causing the treated GDL to show mixed wettability, i.e. the part of the GDL inner surface is hydrophobic, and the rest is hydrophilic. In addition, PTFE loadings (typically 5–30 wt%) [12,13], loss during mechanical compression or freezing cycles [14,15], degradation [16–19], and drying methods (e.g. air versus vacuum drying) [20] will impact the PTFE distribution and mixed wettability in GDLs.

Several experiments were conducted to assess the GDL's mixed wettability and measure the correlation between the capillary pressure P_c (defined as $P_c = P_l - P_g$) and water saturation s in GDLs [21–26]. Gostick et al. [21,22] employed the standard porosimetry (MSP)

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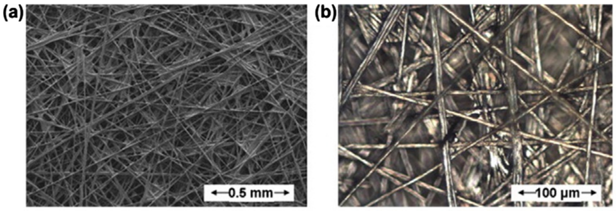
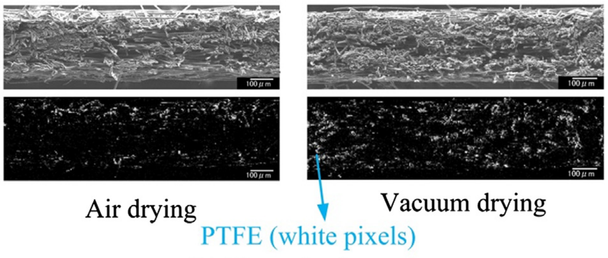


(a) In-plane



(b) Through-plane

Fig. 1. Scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDS) images of Toray-090 carbon paper. (a) In-plane [5] and (b) through-plane [20].

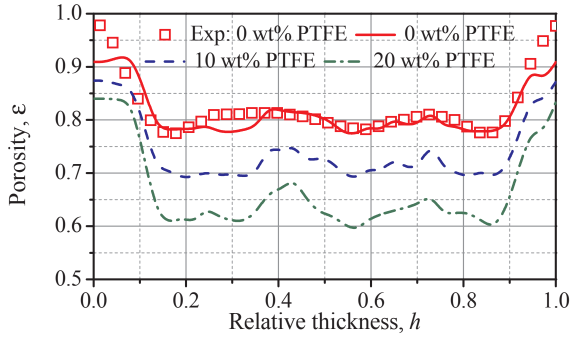


Fig. 3. Comparisons of simulated and experimental local porosity distribution [51] along the through-plane direction for GDLs.

method to measure the P_c-s correlation under various GDL thickness, PTFE loading, and compression. They found water withdrawal occurs at a negative capillary pressure and observed hysteresis between water injection and withdrawal processes. Kumbur et al. [24] measured the P_c-s curves for SGL 24 series GDLs using the MSP technique. They derived a new K function to replace the J function in the standard Leverett correlation, which takes into account the PTFE loading (ranging from 5 wt% to 20 wt%). Hao and Cheng [26] developed a micro-fluidic device to investigate the P_c-s curves of the Toray-090 carbon paper coated with different PTFE loadings (10 wt% and 30 wt%). They determined the parameters in the Leverett J correlation using experimental data.

Numerical study is an efficient tool to investigate the complex two-phase transport in GDLs, such as the multiphase mixture (M2) model [27–31], two-fluid model [32], lattice Boltzmann method (LBM) [26,33–36], pore network method (PNM) [37–40] and volume of fluid (VOF) method [41–45]. Wang and Chen [27] compared their M^2 model prediction (based on the standard Leverett correlation) of the liquid water through-plane profile with the high-resolution neutron radiography data, and showed an acceptable agreement in the GDL regions. Wang and Chen [28] further explored the spatial variation in GDL properties including porosity, permeability, and wettability, and derived a generalized formula based on the standard Leverett correlation to account for the impacts of the property spatial variation. They also explained local water accumulation observed in high-resolution neutron radiography of GDLs and achieved a good agreement with the experimental water profile. The impact of land compression was also discussed and compared with experiment. Wu et al. [29] employed a M^2 model to explore how the arrangement pattern of the protrusive GDL affects the fuel cell performance. They found that the small density of protrusive GDL distribution can enhance the fuel flow into the catalyst layer with the smallest pressure drop. Si et al. [32] adopted a two-fluid model which incorporated a validated Leverett function (K function) in ref. [24] to investigate the effect of different PTFE loadings, compression pressure and micro-porous layer (MPL) on the cell performance. They found liquid water is hard to be removed from the GDL with poor hydrophobicity. Hao and Cheng [26] developed a three-dimensional (3D) LBM model to perform pore scale simulations of air-water flow in the mixed-wettability carbon paper GDL. They validated

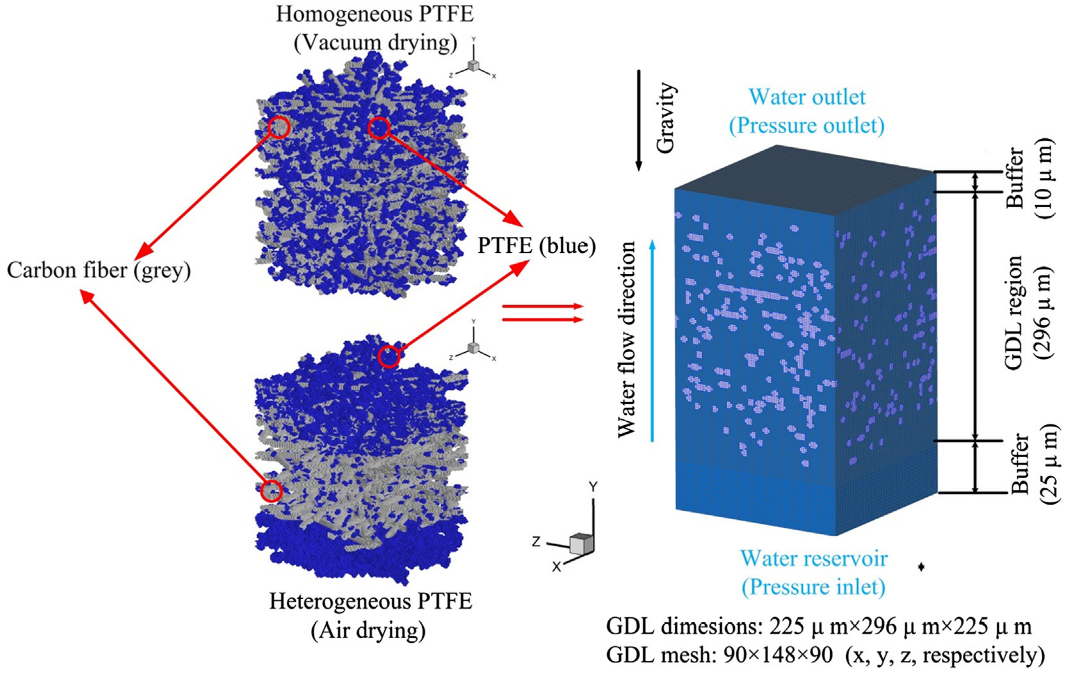


Fig. 2. Computational domain and boundary conditions of present two-phase GDL model.