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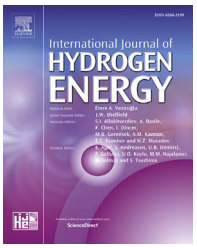

# Apparent contact angles of liquid water droplet breaking through a gas diffusion layer of polymer electrolyte membrane fuel cell

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ABSTRACT

The lattice Boltzmann method is used to simulate the three-dimensional dynamic process of liquid water breaking through the gas diffusion layer (GDL) in the polymer electrolyte membrane fuel cell. An accurate method is introduced to analyze asymmetric droplet shape. Ten micro-structures of Toray GDL were built based on a stochastic geometry model. It was found that asymmetric droplets are produced on the GDL surfaces. Their local apparent contact angles vary with different view angles and geometries. They are different to the idealized contact angles by symmetric simplification. It was concluded that the apparent contact angles are influenced by GDL structures and view angles. This information can help to bridge the gap between mesoscale and cell-scale simulations in the field of fuel cell simulation.



## Introduction

Polymer electrolyte membrane fuel cell (PEMFC) have attracted increasing attention over the past few decades because they have zero emissions [1]. Liquid water is produced under the typical operating temperatures (around 343 K) on the cathode side [2,3]. The liquid water should be transported through the gas diffusion layer (GDL) and removed with reactant gas flow in the gas channel (GC) [4,5].

According to literature, a number of computational fluid dynamics (CFD) simulations are performed for the droplet flows in the GC using volume of fluid (VOF) methods [6]. The hysteresis of droplets in the GC is studied with flow in GC, in which the contact angle of the droplet and breakthrough area (or contact area) on the GDL-GC interface are two input parameters in VOF models [7–9]. The values of the contact angles and breakthrough area are not very clear: the contact angle is usually assumed to be the ideal droplet on homogeneous GDL surfaces [10], or based on the sessile drop experiment [11], while the breakthrough area is set to the regular shape with specific sizes [12]. In the sessile drop experiment, one droplet is set on a sample surface and its shape simplified to be symmetric. The circle method is usually applied when analyzing the idealized contact angles based on the symmetry

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simplification which is not realistic [13]. Two issues cause inaccuracies in VOF simulations. The first is that the shape of droplets breaking through GDL is asymmetric due to the irregular surface of GDL [14]. Another issue is that the contact area of droplets with GDL is connected to a saturated region inside of the GDL (liquid droplets are not just pure-contact GDL solid surfaces). These detailed problem can be solved by the theory of multi-scale simulation that the detailed information can be obtained using one lower scale simulation method [15]. The detailed results will support the upper scale simulations. On the experiment side, it is argued that the irregularity of GDL surface have more influence than the hydrophobicity of components (such as Polytetrafluoroethylene (PTFE)) on GDL carbon fibers surfaces [16]. But the influence of GDL irregularity is still unclear in quantity [17]. One suitable simulation tool is therefore selected to simulate two-phase flow through GDL to support VOF simulations and verify the argument through simulation.

Three methods are favored with respect to two-phase flow in GDL: the pore-network modeling (PNM), a continuum based approach such as the finite volume method (FVM) and the lattice Boltzmann method (LBM). For PNM method, the reconstructed pore-network or topologically equivalent pore-network need to be provided [18,19]. The flow inside of porous media are studied, but it is difficult to study the situation outside of porous media using this method. In the continuum-based approach, the tendency of the saturation curve along the water flow direction (membrane to GC) can be roughly predicted based on the differential equations of mass and energy conservation [20–23]. Jiao's group [24,25] studied the two-phase behavior in the GC by solving conservation equations. In comparision with the first two methods, LBM can trace droplet interface, while PNM can trace the liquid-gas interface inside of GDL. LBM is also more helpful when investigating water transport in the GDL because the real GDL porous structures and non-slip boundary conditions can be easily implemented [26]. For the PEMFC, LBM was already applied on investigating transport properties, droplet behavior and reactive flow with some opportunities and challenges [27,28]. Therefore, in this study, the LBM is chosen to simulate liquid water through GDL and the droplets produced on the GDL surface. However, there are multiple two-phase flow models including the Rothman-Keller (RK) model, the ShanChen pseudo-potential model and the free energy model [29–31]. The first of these is the RK model using density gradient to separate phases [32]. The second is the pseudo-potential model proposed by Shan and Chen in which a pseudo-potential is introduced and applied to a multiphase and multicomponent system [33,34]. The third model is the free energy model proposed by Swift et al. [35], in which phase effects are introduced directly in the collision steps. In comparison with these models, the ShanChen model is more commonly applied due to it being easier to understand and implement [36]. In order to improve numerical stability and accuracy, the models using a multi-relaxation-time (MRT) approach were extended [37]. According to the literature, a number of LBM simulations were performed with respect to GDL applications by using one of the two phase LBM models. Tao's group [38,39] carried out LBM simulations on water transport and distribution in GDL. Kang's group [40,41] recently worked on the multi-field LB simulations which coupled components of PEMFC with different 2D grid styles. Sundén's group [42] compared the through-plane diffusibility correlations in GDL by the multi-components LBM. Li et al. [43] simulated the impact of regular artificial GDL surface micro-structures on dynamic water droplet behaviors in GC. Kim et al. [44] performed the LBM simulation on liquid water transport through GDL in artificial random structures. Jiang's group [45,46] used the free energy LBM to simulate two-phase and flow behaviors in GDL with considering PTFE effects. Mukherjee et al. [47] studied the influence of pore structure and partially wetability of water transport through catalyst layer (CL) and GDL. Meng's group investigated the relationship between GDL structure and liquid water entering the GC. They simulated the liquid water flowing through the artificial or irregular pores in GDL and the droplets behaviors in GC [48–50]. In our group, we had analyzed the single phase flow properties (such as permeability, tortuosity and porosity) in reconstructed GDL for high temperature polymer electrolyte membrane fuel cells (HT-PEMFC) [51–53]. Our recent work has focused on low-temperature polymer electrolyte membrane fuel cells (LT-PEMFC; also referred to as PEMFC) and two-phase flow inside of the same GDL structures which were also used in HT-PEMFC in our previous work. Some micro-structures (such as MPL or GDL) are reconstructed [54]. In this work, the GDL geometries are built based on a stochastic geometry model [55,56].

In a summary of introduction, it is clearly seen a scientific research gap in multi-scale modeling (between micro-scale simulation in GDL and cell-scale VOF simulation in GC) of PEMFC. The contact angle (output result of micro-scale simulation and one of important input parameters in cell-scale simulations) is one of good term to bridge this gap. In the micro-scale simulations of the present paper, the D3Q19 ShanChen pseudo-potential multiphase multicomponent LBM model was selected to simulate liquid water transport through the GDL and asymmetric apparent contact angles at the GDL-GC interface. The simulations are performed using the multi-relaxation-time (MRT) approach with the exact difference method (EDM) force scheme. The asymmetric drops are analyzed by the sub-pixel polynomial fitting (SPPF) method. The fundamentals of the model and SPPF are outlined in Section Algorithms and validated in Section Validation. Simulation setups and conditions are then introduced in Section simulation setups and conditions. Finally, the results are shown in Section Results and discussion and conclusions are summarized in Section Conclusion.

## Algorithms

In this section, the algorithms of LBM using the MRT approach are roughly reviewed. One of the developed force schemes (the way to couple pseudo-potential in the ShanChen LBM model), EDM is then explained. Finally, a post-process method for analyzing asymmetric droplet shape in our results is presented, which is known as the sub-pixel polynomial fitting (SPPF) method.

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