ISSN 2004-2965 Energy Proceedings, Vol. 24, 2021

International Conference on Applied Energy 2021
Nov. 29 - Dec. 5, 2021, Thailand/Virtual
Paper ID: 0652

Optimal design of Polytetrafluoroethylene distribution in a gas diffusion layer for proton exchange membrane fuel cell by Lattice Boltzmann method

Yulin Wang[1,2]*, Haokai Xu [1], Xiangling Liao[1], Xiaodong Wang[3]**

[1]Tianjin Key Lab of Refrigeration Technology, Tianjin University of Commerce, Tianjin, 300134, China.

[2]State Key Laboratory of Engines, Tianjin University, Tianjin 300350, China.

[3] Research Center of Engineering Thermophysics, North China Electric Power University, Beijing 102206, China..

ABSTRACT

The performance of a proton exchange membrane fuel cell exhibits strong association with liquid transport in a gas diffusion layer. The content and distribution of Polytetrafluoroethylene are key factors that determine liquid transport behaviors in a gas diffusion layer. In this study, by employing a stochastic algorithm, a two-dimensional microstructure of a representative carbon paper type gas diffusion layer was reconstructed. Subsequently, the influence of Polytetrafluoroethylene content and various proposed gradient distributions of Polytetrafluoroethylene in the reconstructed gas diffusion layer on liquid transport behaviors was examined by implementing a two-phase Lattice Boltzmann method. The results supported the findings that an increased content of Polytetrafluoroethylene in gas diffusion layer favored liquid removal, but an extremely high one could cause a remark decrease of the corresponding porosity of the gas diffusion layer, hence weakening mass diffusion. An optimal gradient design of Polytetrafluoroethylene could enhance water removal performance of a gas diffusion layer reflected by a reduced liquid water saturation and liquid phase steady-state time, meanwhile could ensure an excellent mass diffusion with a relatively high effective porosity of gas diffusion layer, thereby benefitting fuel cell performance. The study here could provide guideline for the design of high performance of a fuel cell with a gradient gas diffusion layer.

Keywords: proton exchange membrane fuel cell, gradient distributions of Polytetrafluoroethylene, liquid transport in gas diffusion layer, Lattice Boltzmann method, fuel cell performance.

NONMENCLATURE

Abbreviations	
PEMFC	Proton Exchange Membrane Fuel Cell
GDL	Gas Diffusion Layer
MPL	Microporous Layer
CL	Catalytic Layer
GC	Gas Channel
LBM	Lattice Boltzmann Method

1. INTRODUCTION

Proton exchange membrane fuel cell (PEMFC) is a device that converts the chemical energy stored in reactants into electrical energy through electrochemical reaction. PEMFC has the advantages of high efficiency, fast response and high power density. The performance of fuel cell depends on the transmission of liquid water in gas diffusion layer (GDL), microporous layer (MPL) and catalytic layer (CL). As the only by-product of electrochemical reaction, water is very important to the performance of PEMFC. Too low water content will lead to low proton conductivity, while too much will lead to electrochemical activity in CL, hinder fluid flow in gas channel (GC), and finally degrade performance[6].

GDL has two main functions: one is to transport the accumulated water from the fuel cell, and the other is to provide a way for the deposition of reaction gas into the CL. GDL is a single-layer structure composed of a certain number of carbon fibers, and then multiple single layers are superimposed and compressed to form porous materials. In order to generate two paths in the diffusion layer - hydrophobic reaction gas pathl and hydrophilic liquid water transfer path, GDL needs to be placed in a certain mass fraction of PTFE solution for hydrophobic treatment, achieve more hydrophobic surface physical properties (contact angle is about 100 ° ~ 150 °).

GDL is a complex micron scale three-dimensional microstructure. In terms of specific parameters, the thickness of GDL is about 150 to $400\,\mu m$. The diameter of carbon fiber is generally $7\,\mu m$, the pore size is about $80\,\mu m$. The transport process of liquid water in GDL is affected by microstructure, especially the carbon paper GDL treated with PTFE, which increases the complexity of liquid water flow. Nowadays, it is difficult to study the liquid water transport behavior of microstructure. Generally, microscopic visualization technology can be used for research. Visualization technology mainly includes nuclear magnetic resonance imaging and radiographic testing technology, such as neutron imaging Electron microscopy and X-ray technology. Z et al. [1] observed the carbon paper GDL containing PTFE through high-resolution micro-scale computer tomography. The research shows that the invasion of PTFE leads to the reduction of local pores near the surface area and the uneven distribution of the overall material. However, this kind of method can not theoretically explain the flow phenomenon and behavior. At the same time, due to the opacity of the material. The visualization depth is limited to a few fiber diameters, and the transmission of liquid water in the full form of GDL cannot be truly observed.

In addition, some numerical methods have been developed to study the performance of GDL. The numerical methods to study GDL from a macro perspective are mainly volumetric fluid method and finite element method. Wang et al. [2] used volumetric fluid method to study the effect of gradient pore design and wettability on liquid flow in GDL, so as to obtain the best GDL pore design and improve the drainage performance of fuel cell. However, the macro numerical method can not study the interaction between multiphase flow process and GDL performance. Therefore, the pore scale numerical method at the micro level is developed and run. Among many multiphase flow pore scale simulation methods, the lattice Boltzmann method (LBM) is favored because of its ability to deal with complex structures and capture the dynamic evolution of phase interfaces. Y et al. [3] used pseudopotential LBM to study the transmission of liquid water in MPL and GDL, and studied the effects of hydrophilic fiber percentage and GDL compression ratio on the formation and breakthrough of liquid water. However, when reconstructing the GDL structure, y et al. [3] designed some carbon fibers as PTFE materials to form hydrophilic and hydrophobic paths, but did not obtain the real GDL structure containing PTFE. At the same time, LBM is rarely used to simulate the gas-liquid two-phase flow with different contact angles in the GDL porous medium structure in the current research.

Based on the above problems, a random two-dimensional multiphase flow transport model is established in this study. The pseudo potential LB method proposed by Shan and Chen is used to construct different contact angles to study the gas-liquid two-phase flow behavior in carbon paper GDL containing PTFE. In addition, the real structure of PTFE is reconstructed based on random algorithm, the real shape of PTFE in GDL is presented, and different gradient distributions of PTFE are designed. On the premise of ensuring the same porosity and pore structure, the influence of PTFE distribution on liquid fluid flow is studied, and the dynamic transport behavior of liquid water in hydrophobic porous media is predicted, so as to improve the drainage and effective porosity of GDL, to improve the overall performance of PEMFC.

2. MODELING AND COMPUTING DOMAIN

GDL presents the characteristics of layered structure. Carbon fibers are staggered to form a single-layer structure, and then multiple single layers are super-imposed and compressed to form a porous structure. PTFE is wrapped in the outer layer of carbon fiber, and the position with small pores of carbon fiber is easy to be infected by PTFE.

2.1 uniform distribution of PTFE concentration

In this study, the GDL two-dimensional porous media region is constructed by using the random algorithm to simulate the liquid hydrodynamic behavior. The two-dimensional GDL interface carbon fibers are circular, the intersecting carbon fibers are randomly distributed in the plane, and the calculation domain is $200\mu m \times 100\mu m$. The side of the calculation domain adopts periodic boundary conditions, the liquid water inlet adopts velocity boundary conditions, and the velocity is set to 8×10^{-5}, where the resolution of each lattice is $1\mu m$. In this study, the simulated carbon paper GDL is treated with $0\,wt.\%$, $5\,wt.\%$, $10\,wt.\%$, $15\,wt.\%$ and $20\,wt.\%$ PTFE solution respectively. The initial porosity of GDL is 0.6, the contact angle between liquid water and GDL particles is 50°, and the contact angle between liquid water and PTFE is 130°. In GDL, capillary pressure and relative permeability are the main characteristics of gas-liquid two-phase flow characteristics, and water vapor is mainly mass transferred by diffusion, The mass transfer driving force of liquid water is capillary pressure, which is equal to the difference between liquid phase pressure and gas phase pressure:

$$P_c = P_g - P_l \quad (1)$$

The effect of different PTFE concentration on gas-liquid two-phase flow in GDL was studied by breaking through the steady-state time of carbon paper GDL model with different PTFE content by liquid water. The optimal PTFE content was selected by comparing the effective porosity and liquid saturation.

2.2 PTFE gradient distribution

Determine the optimal PTFE content, design concentration gradient, ensure a certain average concentration content, unevenly distribute the PTFE content along the flow plane, ensure the same carbon fiber structure and consistent porosity, and study the influence of PTFE distribution on liquid water transport and breakthrough. Generally, the gas phase pressure in the gas diffusion layer is basically consistent, It can be approximately considered that the capillary pressure is mainly determined by the liquid phase pressure, and s is defined as the volume fraction of the liquid in the total pore space in the porous medium:

$$s = \frac{V_l}{V_{proe}} \quad (2)$$

The best concentration gradient design is selected by comparing the liquid saturation and gas-liquid two-phase steady-state time.

3. LATTICE BOLTZMANN METHOD

3.1 *fluid flow model*

In this study, the pseudo potential multiphase LB model is used to study the gas-liquid two-phase flow and simulate the transport behavior of liquid water in GDL. $DnQm$ is used, where n represents the dimension and m represents the number of velocity lattice chains. The evolution equation of distribution function $f_i(x,t)$ is:

$$f_i(x+e_i\Delta t, t+\Delta t) - f_i(x,t) = -\frac{1}{\tau}[f_i^k(x,t) - f_i^{eq,k}(x,t)] \quad (3)$$

among Δt is the time step, e_i is the discrete speed, $D2Q9$ model is used in this study, and $f_i^k(x,t)$ is the kth density distribution function of lattice position x and lattice time t. The equilibrium distribution function $f_i^{eq,k}(x,t)$ is as follows:

$$f_i^{eq,k} = \omega_i \rho^k [1 + \frac{1}{(c_s)^2}(e_i \bullet u^{eq}) + \frac{1}{2(c_s)^4}(e_i \bullet u^{eq})^2 - \frac{1}{2(c_s)^2}(u^{eq})^2] \quad (4)$$

For the $D2Q9$ lattice model used in this study, c_s is the sound velocity, $c_s = c/\sqrt{3}$ (where $c = \Delta x \,/\, \Delta t$) Ref. [4-5].

F^k includes fluid solid force, fluid fluid force and external force:

$$F^k = F_{1k} + F_{2k} + F_{3k} \quad (5)$$

For the particle of the k-th component at lattice point x, the flow-flow force F_{1k} is expressed as:

$$F_{1k}(x) = -\psi_k(\rho_k(x)) \sum_{x'} \sum_{\bar{k}} G_{k\bar{k}}(x,x') \psi_k(\rho_k(x))(x'-x) \quad (6)$$

among $\psi_k(\rho_k(x))$ represents the effective density, defined as $\psi_k(\rho_k(x)) = \rho_0[1 - \exp(-\rho_k)\rho_0]$. $G_{k\bar{k}}$ represents the intensity of flow flow interaction, which controls the insolubility and surface tension of two-phase fluid, considering only the nearest particle force. The fluid-solid force F_{2k} is expressed as:

$$F_{2k}(x) = -\rho_k(x) \sum_{x'} G_{ks}(x,x') n_s(x')(x'-x) \quad (7)$$

Where n_s is the indicator function, when $n_s = 1$, it is a solid, when $n_s = 0$, it is a pore, and G_{ks} represents the strength of fluid solid interaction. Different contact angles can be obtained by adjusting.

4. RESULTS AND DISCUSSION

4.1 force verification of pseudo potential multiphase LB model

In the pseudo potential LB model, in order to truly simulate the physical phenomena of two-phase fluid flow, it is necessary to calibrate F_{1k} controlling the flow flow force and F_{2k}, $G_{k\bar{k}}$ and G_{ks} controlling the flow solid force to determine the strength of flow flow and flow solid interaction. Two numerical experiments were carried out to verify: bubble test to verify the flow-solid force F_{1k}, and static contact angle test to determine the flow-solid force F_{2k}.

4.1.1 Bubble test

Surface tension is caused by the adhesion of liquid molecules at the liquid gas interface. It is an important factor for two-phase fluid flow in porous media. It is closely related to capillarity and wetting phenomenon. For vacuoles suspended in gas, according to Laplace theorem, the pressure difference at the liquid gas interface is related to the radius of bubbles:

$$\Delta P = \frac{\sigma}{R} \quad (8)$$

among σ Is the surface tension, ΔP is the pressure difference inside and outside the bubble.

In order to verify the surface tension of the model, the dynamic evolution behavior of bubbles located in the center of the liquid is simulated, and all boundary surfaces are set as periodic boundary conditions. In the verification, we recorded the pressure difference on the liquid-gas interface (expressed in lattice units) and the radius of bubbles in the final equilibrium state (expressed in lattice units). Through verification, it conforms to Laplace's law.

4.1.2 Static contact angle test

When the multiphase fluid is stationary on the horizontal solid wall, it is observed that there is an angle at the interface between the liquid phase and the wall, which is called the static contact angle. When the static contact angle is less than 90°, it is said that the solid wall is hydrophilic or wet. At this time, a small amount of liquid on the solid surface tends to form film diffusion; When the static contact angle is greater than 90 °, it is said that the solid wall is hydrophobic or non humid, and liquid droplets are formed on the solid surface.

Simulate the evolution of the shape of the liquid on the solid wall. When the liquid is stable, record the shape of the liquid and the angle on the liquid-solid interface to obtain the contact angle.

4.2 Effect of PTFE uniform distribution

In this section, the two-dimensional model of porous media microstructure of carbon paper GDL was reconstructed by random algorithm. Five different uniform PTFE contents were added to investigate the dynamic behavior of two-phase fluid in GDL with different PTFE contents.

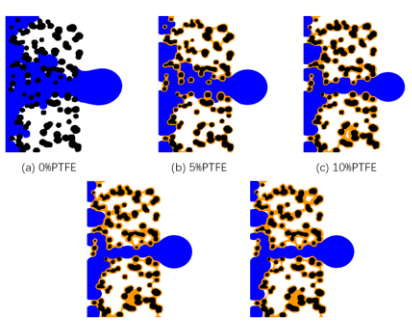

Fig. 1. Two-phase flow with different uniform PTFE content

In GDL, capillary force is the main driving force of liquid water, and other forces including viscous force, inertial force and gravity can be ignored. After entering GDL, driven by capillary force, liquid seeps while looking for macropores. By observing the flow behavior of liquid water, we can study the dynamic characteristics of liquid water clusters at the moment of breakthrough. When liquid water breaks through and flows out of GDL, a fixed liquid flow channel is formed, and subsequent liquid breaks through this flow path first (as shown in Fig. 1).

The results show that the addition of PTFE will reduce the porosity of GDL. PTFE covers the surface of carbon fiber. At the same time, PTFE preferentially invades the position where the pores of carbon fiber are small. The position where liquid water can flow is blocked and cannot be broken through, resulting in the decrease of liquid saturation. With the increase of PTFE content, the liquid saturation decreases, At the same time, the liquid behavior of liquid water on PTFE surface is different from that on carbon fiber surface. The dynamic characteristics of liquid water are recorded, which shows that the wettability of GDL has an important influence on the distribution and dynamic behavior of liquid water. As shown in the figure:

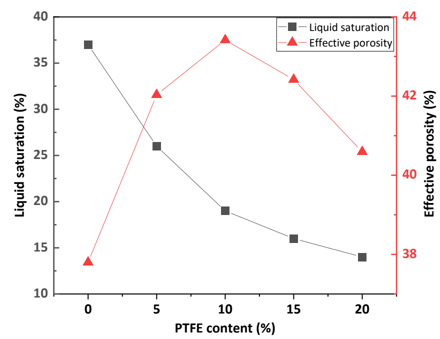

Fig. 2. Relationship between liquid saturation, effective porosity and PTFE content

Fig. 2 shows that with the increase of PTFE content, the drainage performance of GDL is enhanced, the liquid saturation continues to decrease, and the steady-state time of two-phase fluid continues to decrease with the increase of PTFE content, and then gradually tends to be flat, but the corresponding porosity de-creases, and the corresponding effective porosity first increases and then decreases. When the PTFE content reaches $10wt.\%$, the effective porosity reaches the best value of 43.416, and the liquid saturation is $s = 0.19$, When the gas-liquid two-phase flow in GDL reaches equilibrium, the liquid steady-state time is $t_1 = 0.69$ and the gas steady-state time is $t_2 = 1.08$.

4.3 Effect of PTFE with gradient content distribution

Through the two-phase flow behavior when PTFE content is evenly distribu ted in the previous section, it is obtained that the effective porosity reaches the best when PTFE content reaches $10wt.\%$. In this section, five PTFE gradient designs ($10wt.\%$, $6wt.\%$ and

$14wt.\%$, $8wt.\%$ and $12wt.\%$, $12wt.\%$ and $8wt.\%$, $14wt.\%$ and $6wt.\%$) are studied. GDL is designed to be divided into two concentration gradients along the liquid flow direction to obtain better drainage performance, As shown in the figure:

Fig. 3 shows the dynamic behavior of liquid water flow in the GDL with PTFE gradient design. With the extension of evolution time, the fluid continuously flows into the pores in the GDL driven by capillary force. The different structure of porous media leads to the change of liquid water breakthrough behavior. The distribution effect of PTFE can be seen from Fig. 3, especially the effect of inlet content effect. With the increase of PTFE content, The smaller the liquid saturation.

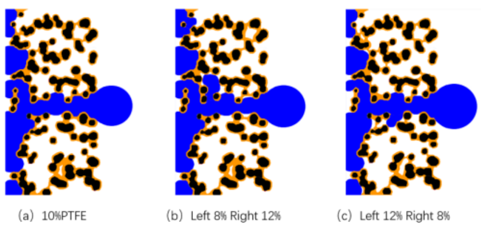

Fig. 3. Two-phase flow of gradient PTFE

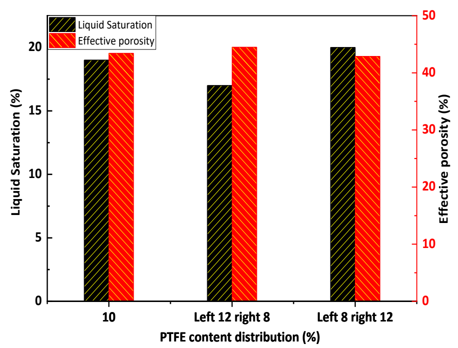

Fig. 4. Relationship between liquid saturation, effective porosity and PTFE content

As shown in Fig. 4, the results show that when the PTFE content at the inlet is large, the porosity of the inlet interface decreases, the breakthrough point of liquid water decreases, the breakthrough speed increases, the liquid saturation decreases, and the overall effective porosity of GDL increases, but the main flow path of liquid water through GDL has little effect. When the content of PTFE at the inlet is $14wt.\%$ and the content at the outlet is $6wt.\%$, the effective porosity is 45.241, which is 4.2% higher than the uniform distribution of PTFE 10wt.%, and the liquid steady-state time is reduced to $t_1 = 0.64$, which effectively improves the drainage performance of GDL.

5. CONCLUSION

In this paper, the random algorithm is used to reconstruct the two-dimensional model of GDL porous media. By reconstructing GDL with five different PTFE contents and five different PTFE distributions, the influence of PTFE content and distribution on the two-phase fluid flow in GDL is studied by LB method, and the flow behavior of liquid water under all simulated conditions is given.

The simulation results show that when the PTFE content is evenly distributed, the PTFE content has an effect on the breakthrough time of liquid water. The more PTFE content, the shorter the breakthrough time of liquid water, but the effective porosity decreases. When the content reaches $10wt.\%$, the effective porosity of GDL reaches the extreme value.

In gradient distribution, the distribution position of PTFE has an impact on the flow behavior of liquid water. When the average content is certain, when PTFE is distributed more on the left (CL), the breakthrough time of liquid water is short, the liquid saturation decreases and the effective porosity increases. The distribution of PTFE on the left has a great impact on the drainage performance of GDL.

ACKNOWLEDGEMENT

The authors are grateful for the financial support from the National Natural Science Foundation of China (No. 52176084).

REFERENCE

[1] Fishman Z, Bazylak A. Heterogeneous through-plane porosity distributions for treated PEMFC GDLs I. PTFE effect. Journal of The Electrochemical Society, 2011, 158(8): B841.

[2] Wang Y, Wang X, Wang X, Liu T, Zhu T, Liu S, et al. Droplet dynamic characteristics on PEM fuel cell cathode gas diffusion layer with gradient pore size distribution. Renewable Energy, 2021, 178: 864-874.

[3] Ira Y, Bakhshan Y, Khorshidimalahmadi J. Effect of wettability heterogeneity and compression on liquid water transport in gas diffusion layer coated with microporous layer of PEMFC. International Journal of Hydrogen Energy, 2021, 46(33): 17397-17413.

[4] Chen L, Luan H, He Y, Tao W. Numerical investigation of liquid water transport and distribution in porous gas diffusion layer of a proton exchange membrane fuel cell using lattice Boltzmann method. Russian journal of electrochemistry, 2012, 48(7): 712-726.

[5] Zhou C, Guo L, Chen L, Tian X, He T, Yang Q. Pore-Scale Modeling of Air–Water Two Phase Flow and Oxygen Transport in Gas Diffusion Layer of Proton Exchange Membrane Fuel Cell. Energies, 2021, 14(13): 3812.

[6] Wang Y, Wang X, Fan Y, He W, Guan J, Wang X. Numerical Investigation of Tapered Flow Field

Configurations for Enhanced Polymer Electrolyte Membrane Fuel Cell Performance. Applied Energy, 2022, 306: 118021.

