Water Flow Characteristics and Related Effects in PEMFC

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Abstract: Water management in proton exchange membrane fuel cells (PEMFC) is a topic of great importance for the optimization of these systems. Effective proton conductivity calls for moderate moisture content in the membrane, while uneven water distribution can lead to instability of the whole flow field, thereby decreasing the performance of the fuel cell. In the present study, a simplified two-tier hybrid structure is used to investigate the impact of the dynamic behavior of liquid water on the current density of the PEMFC. Simulation results show that water droplets attached to wall sides tend to increase current density. Visualization experiments confirm the existence of liquid droplets and the enhancement of current density, while indicating that the best performance and stability of fuel cell are attained for a cathode air flow rate of 300 ml/min.

Keywords: PEMFC, water management, simulation, airflow rate, visualization.

1 Introduction

Water management in proton exchange membrane fuel cell (PEMFC) is a significant issue during the process of use. Appropriate quantities of water which remain in the membrane lead to a better proton conductivity. Meanwhile, redundant water in the gas diffusion layer and channel flow should be removed, which will result in a greater pressure drop and a block of reactive gas in flow channel. However, it is an enormous challenge to hinder the production of liquid water, especially in conditions of high current density. Liquid water causes gas-liquid two-phase flow, which has been proved, has a great impact on the performance of the PEMFC.

Many studies have focused on gas-liquid two-phase flow from experiments and simulations. Anderson et al. [Anderson, Blanco, Bi et al. (2012)] emphasized on two-phase flow in channels had a great impact on the overall performance of the fuel cell. Former studies [Ding, Bi, Wilkinson (2011); Le, Zhou, Shiu et al. (2010); Gurau, Zawodzinski and Mann (2008)] indicated that liquid water varies in different behavior under different operating conditions from experiments. Such as: fog flow, water drip and liquid film and liquid plunger. However, due to the limitations of visualization technology, it is still on the ropes to make clear the quantitative causality between hydrodynamics of two-phase flow and the performance of PEMFC. Meanwhile, model calculation and numerical simulation can get quantitative results. Development of numerical model for PEMFC dates back to Springer [Springer (1993)], Bernardi et al. [Bernardi and Verbrugge (1991)]. The early model

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assumed water only in the gaseous phase in channel flow, and two-phase flow did not take into account since 2000. After that, phase transition, multiphase species transition and electrochemical reaction were considered.

In recent decades, three-dimensional models based on computational fluid dynamics (CFD) were widely used, which were primarily effectuated by commercial CFD software. Wöhr et al. [WöHr, Bolwin, Schnurnberger et al. (1998)] presented a CFD model which contained electrochemical reaction, current density, fluid dynamics, and multi-component transition. With FLUENT, Akhtar et al. [Akhtar and Kerkhof (2011)] successfully simulated a three dimensional model of PEMFC with single straight channel and Mazumder et al. [Mazumder and Cole (2003)] got a liquid water transition model. Dutta et al. [Dutta, Shimpalee and Zee (2000)] studied steady flow of micro-size parallel flow channel, discussed the influence of flow characteristics and oxygen distribution along the flow path. However, these studies ignored the effect of liquid water. Pasaogullari et al. [Pasaogullari and Wang (2005)] pointed out the inevitability of water vapor condensation in anode and cathode and excessive liquid water was drove to cooling channel by pressure difference. Jiao et al. [Jiao and Zhou (2008)] studied the distribution of water in PEMFC with serpentine flow field. Zhu et al. [Zhu, Sui and Djilali (2008)] used a straight flow channel with a hole for water entrance to simulate the airflow, gas density and viscosity, and surface tension of the droplets.

To sum up all the studies above, thermal chemical reaction, flow field of multicomponent mass transfer process, as well as liquid water flow motion characteristics of the numerical model were discussed in detail, but not in one model.

This paper proposed a simplified two-tier hybrid structure model to comprehensively study flow characteristics of liquid water, mass transfer and chemical reactions in flow channel of PEMFC. Also visualization experiment of self-assembled single fuel cell with parallel flow field was investigated to verify the simulation results.

2 Numerical theory of two-phase flow

The volume-of-fluid (VOF) numerical model of FLUENT has been developed to describe gas-liquid two-phase flow in flow channels. VOF model may be the only and best topdown model to study the detailed behavior of liquid water in PEMFC. The transport of liquid water in gas diffusion layer (GDL) is quite important, while VOF support that, even on the micron-sized level. However, only a few researchers like Park et al. [Park and Li (2008)] had used the VOF model to simulate water transport phenomena in GDL. Liquid and gas phase in the VOF model share the same set of equations of momentum, the interface of two-phase is controlled by the computing element, which is calculated from volume fraction of fluid k:

$$S_{k}(\mathbf{x}, \mathbf{y}, \mathbf{z}) = \begin{cases} 0(\text{out of computing domain}) \\ 1(\text{inside of computing domain}) \\ 0 - 1(\text{on the boundary of computing domain}) \end{cases}$$
(1)

where S_k is the volume fraction of fluid k: $S_k+S_g=1$

While phase transitions, electrochemical reactions, gravity and heat transfer are ignored, twophase, gas or liquid VOF model can be expressed as the following conservation equation. The mass (continuity) equation is expressed as follows:

$$\frac{\partial}{\partial t}(\rho) + \nabla \cdot (\rho \vec{u}) = 0 \tag{2}$$

The momentum conservation equation is expressed as follows:

$$\frac{\partial}{\partial t}(\rho\vec{u}) + \nabla \cdot (\rho\vec{u}\vec{u}) = -\nabla p + \mu \nabla \cdot (\nabla(\vec{u}) + (\nabla\vec{u}^{\mathrm{T}})) - \frac{2}{3}\rho\vec{g} + \vec{F}$$
(3)

Volume fraction equation of liquid is expressed as follows:

$$\frac{\partial}{\partial t}(\rho S_k) + \nabla \cdot (\rho S_k \vec{u}) = 0 \tag{4}$$

where p is the static pressure, ρ is the mixture density, μ is the dynamic viscosity, \vec{F} is the power source term correlate to surface tension, ρ and μ . The density and dynamic viscosity in each computational cell is then:

$$\rho = \rho_g + S_{lq} \left(\rho_{lq} - \rho_g \right) \tag{5}$$

$$\mu = \mu_g + S_{lq} \left(\mu_{lq} - \mu_g \right) \tag{6}$$

The surface tension force can be calculated by continuous interfacial force. The pressure drop across the surface depends on the surface tension coefficient σ :

$$\Delta p = \sigma \frac{1}{R} \tag{7}$$

$$\vec{F}_{\rm vol} = \sigma \kappa \frac{\rho \nabla S}{1/2 \left(\rho_{\rm g} + \rho_{\rm lq}\right)} \tag{8}$$

where σ denotes the surface tension coefficient, 1/R is the surface curvature, and R is the radius. K is local interface gradient.

$$\kappa = \nabla \cdot \vec{n} = \nabla \cdot \left(\vec{n}_{\rm w} \cos(\theta) + \vec{t}_{\rm w} \sin(\theta) \right) \tag{9}$$

where \overline{n} is the unit vector normal to the interface between the two phases near the solid surfaces, \overline{n}_w and \overline{t}_w are the unit vectors normal to the solid surfaces, θ is the static contact angle. What should be noted is the shape of interface between the two phases is decided by wetness. In addition, properties such as porosity of porous medium are ignored, which still work in this flow channel. Flow water is excluded to a large extent by the higher pressure gradient and hydrophobicity of GDL decision. Water drainage is mainly decided by high pressure gradient and hydrophobicity of GDL.

3 Liquid water behaviors in flow channel

This two-phase simulation considered not only flow characteristics, but also mass transfer

in GDL and electrochemical reaction of catalyst layer. Effect of reactive gas airflow and wettability of solid surfaces or GDL are also considered. At the same time, pressure drop, current density and reaction mass transport will be studied as well.

3.1 Model description

GDL was a porous medium material constituted by carbon fiber, which was contacted with the surface of flow channel. The surface micro-structure on the flow has important implications for liquid water behavior, so the GDL was designed as two-tier hybrid structure in this section.

The whole calculation model consisted of a flow channel, a two-tier hybrid GDL, and catalyst layer, as shown in Fig. 1. Catalyst layer was regarded as a thin layer, where electrochemical occurred. The first layer of GDL, as shown, was a solid zone with holes. Porosity was equal to the proportion of holes' area. The wheels within wheels design suited for the simulation of water behaviors on cellular structure, as the first layer contacted with flow channel. On the contrary, this structure took more computing resources, so the second layer was designed as solid with uniform porosity, permeability, and gas diffusion rate, same as other studies did. This two-tier hybrid structure of GDL met the needs of both precise simulation and time saving. Finally, the size of the gas channel was set to 1 mm×1 mm×30 mm, and the thickness of the first layer was set to 1 mm, and the second diffusion layer was set to 0.254 mm.



Figure 1: Numerical model of simplified two-tier hybrid structure

3.2 Modeling assumption

- (1) Reactant gas is assumed as ideal gas.
- (2) The flow of the fluid was under the conditions of constant temperature, unsteady state, laminar and incompressible. Temperature was an important factor for proper functioning of PEMFC, especially for water condensation and evaporation, permeability and diffusivity during the fluid transmission, proton conductivity of membrane and reaction rate in catalyst layer.
- (3) Studies were now considering constant temperature VOF model. Model in this article was

half-battery model, which can ignore the influence of temperature on the conductivity.

(4) The flow characteristic of liquid water was a key consideration, so the mass transfer between liquid water and vapor was ignored.

3.3 Boundary conditions

(1) Velocity, component mass fraction at the inlet:

 $u = u_{\text{in}}, v = v_{\text{in}}, Y_{\text{o}} = 0.23, Y_{\text{n}} = 0.77, S_{\text{liquid}} = 0, p = p_{\text{in}}$

where the subscript o means oxygen, and n means nitrogen.

(2) Conditions at the outlet:

$$\frac{\partial u}{\partial x} = 0, \frac{\partial v}{\partial x} = 0, \frac{\partial w}{\partial x} = 0, \frac{\partial P}{\partial x} = 0, \frac{\partial Y_o}{\partial x} = 0, \frac{\partial Y_n}{\partial x} = 0, \frac{\partial S_{\text{liquid}}}{\partial x} = 0$$

(3) Wall sides of GDL ($z \le 2$ mm, $y = \pm 1$ mm) were assigned as symmetry.

(4) Solid surfaces boundaries were set as: $\frac{\partial Y_o}{\partial n} = 0, \frac{\partial Y_n}{\partial n} = 0, u = 0, v = 0, w = 0$

(5) Water outlet of subface of GDL was set as: $u = 0, v = v_{in}, w = 0, Y_o = 0, S_{liquid} = 1$

(6) Catalyst layer boundaries were set as: $u = 0, v = 0, w = 0, \frac{\partial Y_o}{\partial n} = -\frac{M_o}{4F}J$

where M_o is the molar mass of oxygen, F is the Faraday constant, J is current density of cathode, which can be calculated by Butler-Volmer equation, and can be expressed as:

$$J = J_{\rm ref} \left(\frac{C_o}{C_{0,\rm ref}}\right)^r \exp\left(-\frac{\alpha F}{RT}\eta\right)$$
(10)

where J_{ref} is reference exchange current density, α is the Transfer Factor, R is the gas constant, η is the local overpotential, r is cathode concentration coefficient.

(7) Effective diffusion coefficient in GDL:

$$D_{\rm eff} = \left[\varepsilon \left(1 - S_{\rm liquid} \right) \right]^{1.5} D_{\rm o} \tag{11}$$

Operating condition and physical properties of the numerical model were shown in Tab. 1.

Parameters	Values
Inlet gas flow-rate $u_{in}(\mathbf{m} \cdot \mathbf{s}^{-1})$	2, 1, 0.6
Inlet mass fraction of oxygen $Y_{o,in}$	0.23
Inlet mass fraction of nitrogen Y _{n,in}	0.77
Inlet mole fraction of vapor Yvapor,in	0
Contact angle of GDL $\theta_{GDL}(^{\circ})$	150

 Table 1: Basic parameters

Contact angle of channel $\theta_{CHAL}(^{\circ})$	70
Porosity of GDL ε	0.4
Permeability of GDL κ (m ²)	1.0×10 ⁻¹¹
Temperature <i>T</i> (K)	333
Pressure <i>P</i> (Pa)	101325
Gas constant R(J·mol ⁻¹ ·K ⁻¹)	8.314
Faraday constant F(C·mol ⁻¹)	96487
Surface tension $\sigma(N \cdot m^{-1})$	0.0625
Overpotential $\eta(V)$	0.5
Diffusivity of oxygen $D_0(\mathbf{m}^2 \cdot \mathbf{s}^{-1})$	2.84×10 ⁻⁵
Transfer ratio of cathode ς	0.5
Transfer ratio of anode <i>r</i>	1
Reference exchange current density/ Reference concentration of oxygen (A·mol ⁻¹)	120

3.4 Numerical calculation

There were 723,320 regular hexahedral grids in this study, which were gridded from Gambit software, and calculated by Fluent. Pressure-based Fluent solver was used to simulate the unsteady incompressible flow. Equation of velocity and pressure were calculated by PISO method, and interfacial flow of liquid water and gas were calculated by explicit formulation equation of VOF. Mass transfer processes of GDL and flow channel were simultaneously calculated by mass equation. UDFs of Fluent were used to write the effective gas diffusion coefficient of GDL. Time step was set to 1.5×10^{-6} s, and all the residual values were set to 10^{-7} .

3.5 Liquid water behaviors in flow channel

As shown in Fig. 2, case 1 was calculated as an essential condition, where flow velocity was 2 m/s. Contact angle of the channel plates was 45° , which stood for the hydrophilcity of channel plates. While, contact angle of the GDL was 150° , which stood for hydrophobicity. Liquid water formatted from electrochemical reaction in catalyst layer, and entered flow channel through GDL, and came to be spherical crown under the surface tension at t=0.0013 s. When the spherical crown gained, the space left shrank, which gradually raised the velocity of airflow. This larger airflow strengthened shear stress and pressure on the liquid surface, which made spherical crown become bigger. When the liquid drop became large enough, shear stress and pressure then drove the spherical crown move forward, which was fast than the formation of water from GDL, and leave flow channel (t=0.0023 s). As the hydrophilcity of flow channel, some liquid water would adsorb to the surface. This phenomenon somehow decreased clogging and pressure drop in flow channel, more droplets will be attracted to the top of the flow channel and become liquid membrane.

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Figure 2: Flow characteristics of liquid water under airflow rate of 2 m/s

As shown in Fig. 3, the airflow velocity was set to 1 m/s. Liquid drop from GDL became lager (Fig t=0.0012 s) than case 1. Surface tension was now heavy enough to keep the drop from fracture. At t=0.0014 s, liquid drop came into contact with the side wall and started to move. At t=0.0017 s, it adsorbed to the corner between side wall and top surface, formed one-fourth oval water film. When shear stress and pressure gained heavy enough, water film left flow channel along with the airflow.



Figure 3: Flow characteristics of liquid water under airflow rate of 1 m/s

Case 3 was presented in Fig. 4, where airflow velocity was 0.6 m/s, lower than all above, which would cause even bigger liquid drop and water film. Due to the instability of flow channel, most liquid water stuck to one wall side, and a small part of liquid attached to the other side (t=0.0012 s). The water drop contacted both side walls and went though the flow channel much slower.



Figure 4: Flow characteristics of liquid water under airflow rate of 0.6 m/s

All cases showed typical four flow behaviors in the whole process: water drop growths, separation from GDL, adsorption to surfaces, water film formation, which are conform to actual behaviors. It's a wise choice to remove liquid water in flow channel in time. Also, it's easy to find that liquid water drops were more quickly expelled at higher airflow velocity as we known. But the relationship of water drop situations and PEMFC performance still need to be studied to find the suitable airflow velocity.

3.6 Influence to pressure drop from liquid water

Distribution of liquid water on the flow channel would have a greater impact on pressure drop, which directly affected the performance of PEMFC [Mortazavi and Tajiri (2015)].

$$\Delta P = \frac{128\nu F_c U_{in} L_c}{\pi D_c^4} \tag{12}$$

It can be seen from Fig. 5, overall distribution of the pressure drop is similar under three different inlet velocities, but the pressure drop values and the occurred times are quite different. With the increased time, pressure drops become wider. Because of the liquid water drop's growth, flow channel blocked and left space shrank. Pressure drop increased to a maximum value until liquid drop separated, when there were most quantities of liquid water in the channel, which affected the mass transfer reaction most. Then the pressure drop relatively declined rapidly, because at this time, liquid water drop adsorbed to the surface and form water film. The formation of water film blocked the flow channel slightly, which similarly increased the pressure drop as the liquid drop, but genteelly.



Figure 5: Pressure drops in relative time under different flow rates

3.7 Influence to current densities from liquid water



Figure 6: Current densities in relative time under different flow rates. (a) 2 m/s, (b) 1 m/s, (c) 0.6 m/s

Current density-time relationships under different airflow velocities were plotted in Fig. 6. A reduction of current densities can be found at the time two-phases flows occurred at the beginning. This is because when liquid water appeared in flow channel, it decreased the contacting area for reactant gas. All subsequent curves decreased, as liquid water drops grew to summit with the pressure drops. The current densities rebounded at the time liquid water absorbed to surface, and reached the pecks when separation. Then the current densities decreased quickly again at the time of rapidly remove of liquid water drops.

4 Visualization experiment



Figure 7: Visualized PEMFC assembly diagram



Figure 8: Self-built PEMFC performance test system

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According to former studies [Bozorgnezhad, Shams, Kanani et al. (2015); Liu and Pan (2012); Banerjee and Kandlikar (2014)], transparent material was adopted for end plates of PEMFC to observe the inner flow characteristics. Cathode side was chosen for observation and analysis as liquid water mainly existed on cathode side. Meanwhile, poor water management parallel flow field was selected for obvious water behaviors. Visualization single fuel cell was assembled as Fig. 7. Two mm stainless steel plates with through-holes were applied as bipolar plates, which were clamped between PMMA plates and MEA. Performance was evaluated measuring I-V curves with a self-built system as Fig. 8, which included hydrogen generator, electromagnetic flowmeter, customized bubbling humidification system, and electronic load. Parameters for single cell tests were shown in Tab. 2.

1	e	
Parameters	Values	
	Anode	Cathode
Inlet pressure $P/(K \cdot Pa)$	250	160
Airflow rate (ml/min)	800	60,300,600,800
Operation temperature (K)	60	
Operation voltage (V)	0.3	

 Table 2: Operating conditions



Figure 9: Water distributions under different airflow rates

Liquid water distributions under different airflow rates were presented at Fig. 9. Because of the humidification of reactant gas, all inlet air was fog flow at first. It is not hard to find that liquid water drops attached to channel wall at low airflow velocities, especially under the condition of 60 ml/min, where plenty of liquid plungers appeared. With the increase of airflow rate, liquid droplets were discharged constantly, little liquid water drops was left under 800 ml/min. But no phenomenon of attachment to both up wall and side wall (as present in Figs. 2, 3) had been found, which may result from the difference hydrophily of PMMA (θ =45°) and stainless steel (θ =70°).

Current densities under different airflow rates were plotted in Fig. 10. Under 60 ml/min, liquid water mainly existed in flow channel as liquid plungers, thus the performance of PEMFC stayed at a low level. While when the airflow velocity increased to 300 ml/min, most liquid plungers turned to water drip, the current density steadily improved. When the airflow rate was suddenly aggrandized to 600 ml/min, most liquid drops were teared to small drops and excluded out the flow channels, the current density reached at peck instantly, which was corresponding to the result in Fig. 6. Similar with current density variation under 800 ml/min, the performance of fuel cell under 600 ml/min emerged to attenuation quickly with elapse time and finally worse than that of 300 ml/min, which also given an index that certain adsorbed liquid water drops on channel walls can improve the PEMFC performance.



Figure 10: Current density variations with the increased airflow rates

5 Conclusions

Liquid water behaviors under different airflow velocities were emphasized with a simplified two-tier hybrid structure flow channel. Visualization experiment of self-assembled single fuel cell with parallel flow field was investigated, and conclusions were as follows:

(1) Liquid water behaviors in flow channel have a tempestuous relationship with external operating conditions, the greater flow velocity of reactant gas drives faster separation of water drop from GDL and a more rapid movement through the flow channel, and vice versa. Low airflow condition will promote formation of liquid plunger, while high velocity liquid film.

- (2) Certain content of adsorbed liquid water drops on channel walls can improve current density in a region. Moderation airflow rate is necessary for the best performance of PEMFC.
- (3) Visualization experiment verified the existence of water drip has improved the PEMFC current density and indicated that the best performance and stability of fuel cell under cathode air flow rate at 300 ml/min.

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