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Review article

Multi-phase models for water and thermal management of proton exchange membrane fuel cell: A review

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HIGHLIGHTS

- The coupling of two-phase flow in channels and other phenomena is still a challenge.
- The effect of thermal management on water transport is perhaps underestimated.
- The agglomerate model of CL is suggested to be added in 3D CFD two-phase model.
- The 3D CFD multi-phase simulations in cell level and stack level are suggested.
- Current 3D CFD simulations are far from practical demand in design optimization.

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ABSTRACT

The 3D (three-dimensional) multi-phase CFD (computational fluid dynamics) model is widely utilized in optimizing water and thermal management of PEM (proton exchange membrane) fuel cell. However, a satisfactory 3D multi-phase CFD model which is able to simulate the detailed gas and liquid two-phase flow in channels and reflect its effect on performance precisely is still not developed due to the coupling difficulties and computation amount. Meanwhile, the agglomerate model of CL (catalyst layer) should also be added in 3D CFD model so as to better reflect the concentration loss and optimize CL structure in macroscopic scale. Besides, the effect of thermal management is perhaps underestimated in current 3D multi-phase CFD simulations due to the lack of coolant channel in computation domain and constant temperature boundary condition. Therefore, the 3D CFD simulations in cell and stack levels with convection boundary condition are suggested to simulate the water and thermal management more accurately. Nevertheless, with the rapid development of PEM fuel cell, current 3D CFD simulations are far from practical demand, especially at high current density and low to zero humidity and for the novel designs developed recently, such as: metal foam flow field, 3D fine mesh flow field, anode circulation etc.

1. Introduction

In order to achieve the goal of energy saving and emission reduction, PEM (proton exchange membrane or polymer electrolyte membrane) fuel cell, a typical electrochemical device that converts chemical energy into electricity with low to zero emissions, has received considerable attention in past decades [1]. It is expected to be applied in a lot of applications, such as automobile, backup power, portable electronics, etc. [2]. However, it still has not been largely utilized limited by the performance, durability and cost. Fig. 1 (a) shows the schematic of a small PEM fuel cell stack with four single cells. For a single cell, it consists of PEM in between, CL (catalyst layer), MPL (micro-porous layer, sometimes not used), GDL (gas diffusion layer) and BP (bipolar

plate) in both anode and cathode. Hydrogen and air (sometimes with water vapor for humidification) are supplied to anode and cathode flow channels, respectively, and then the electric current produced by electrochemical reaction flows to external load through the two current collectors on anode and cathode sides in the stack. At the same time, coolant water is pumped into coolant channels to keep PEM fuel cell operating within reasonable temperature range.

Under normal operation, PEM has to absorb enough water to maintain high ionic conductivity, which explains that water vapor is usually added to inlet gas despite that external humidifier increases system complexity and cost. On the other hand, water is produced in cathode CL due to ORR (oxidant reduction reaction) and EOD (electro-osmotic drag) also pushes water from anode to cathode side. Owing to

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Nomenclature

a_{Pt}	The specific platinum surface area per unit catalyst volume ($m^2 m^{-3}$)
C	Molar concentration ($mol m^{-3}$)
D	Diffusion coefficient ($m^2 s^{-1}$)
EW	PEM equivalent weight
F	The Faraday's constant
F_s	The surface tension source term ($N m^{-3}$)
f	Interfacial drag coefficient
H	Henry's coefficient ($Pa m^3 mol^{-1}$)
i_0^{ref}	Exchange current density ($A m^{-2}$)
J	Reaction rate ($A m^{-3}$)
J_{ion}	Ionic current density ($A m^{-2}$)
$J_{0,a}^{ref}$	Anode reference volumetric current density ($A m^{-3}$)
$J_{0,c}^{ref}$	Cathode reference volumetric current density ($A m^{-3}$)
K	Intrinsic permeability (m^2)
k	Relative permeability
k_c	Electrochemical reaction constant (s^{-1})
m_C	The carbon loading ($kg m^{-2}$)
m_{Pt}	The platinum loading ($kg m^{-2}$)
N	Number of agglomerates
P	Pressure (Pa)
P_c	Capillary pressure (Pa)
R	Universal gas constant ($J mol^{-1} K^{-1}$)
$Ratio_{PtC}$	Platinum ratio
r_{agg}	Agglomerate radius (m)
r_{Pt}	The Pt particle radius (m)
	Relative humidity
S	Source term ($kg m^{-3} s^{-1}$ or $mol m^{-3} s^{-1}$)
S_{ac}	The specific surface area per unit mass ($m^2 kg^{-1}$)
s	Liquid saturation
T	Temperature (K)
u	Velocity ($m s^{-1}$)

Greek letters

α	Phase volume fraction or transfer coefficient
δ	Thickness (m)
ε	Porosity
ε_1	Pt surface ratio

$\varepsilon_{Pt/C}$	Carbon supporting platinum particle volume fraction
η	Overpotential (V)
θ	Contact angle ($^\circ$)
κ_e	Electric conductivity ($S m^{-1}$)
κ_{ion}	Ionic conductivity ($S m^{-1}$)
λ	Membrane water content
μ	Dynamic viscosity ($kg m^{-1} s^{-1}$)
ρ	Density ($kg m^{-3}$)
σ	Surface tension coefficient ($N m^{-1}$) or electric conductivity ($S m^{-1}$)
φ_e	Electric potential (V)
φ_{ion}	Proton potential (V)
χ	An auxiliary variable
ω	Electrolyte volume fraction

Subscripts and superscripts

0	Standard state
a	Anode
act	Activation state
agg	Agglomerate
ave	Average value
c	Cathode
CL	Catalyst layer
d	Membrane water
e	Electrical
eff	Effective
g	Gas phase
H	Henry's law
H ₂	Hydrogen
ion	Ionic
l	Liquid
mem	Proton exchange membrane
O ₂	Oxygen
out	Outlet
Pt	Platinum
ref	Reference state
sat	Saturation state
T	Temperature

low operation temperature of PEM fuel cell (60–90 °C), the water in PEM fuel cell is much likely to exist in liquid state, which may cause “water flooding” phenomenon [3] and then reduce the performance of PEM fuel cell severely. In PEM fuel cell operation, the irreversible heat and entropic heat generated by electrochemical reaction, Joule heat caused by electronic and ionic transport resistances and latent heat in water condensation and evaporation will lead to uneven temperature distribution. High operation temperature will decrease water absorption in PEM and thus decrease its durability, and the reversible voltage and efficiency are also low. On the contrary, low operation temperature is unfavorable for the electrochemical reaction kinetics and ionic transport in PEM. Besides, low temperature is more likely to result in “water flooding” phenomenon because of the low water saturation pressure. Nevertheless, almost all the transport parameters in PEM fuel cell are dependent on temperature. As a consequence, the complicated and highly interrelated water and thermal management is a vital factor affecting the performance and durability of PEM fuel cell.

The transport phenomena in PEM fuel cell concerning water and thermal management are very complex, including gas and liquid two-phase flow, gas species transport, water condensation and evaporation, membrane water absorption and desorption, electrochemical reactions, electron and proton transport, heat transfer and so on. Given the high

cost and complexity in conducting experiments, developing model to optimize water and thermal management has been a popular choice since the pioneer work done by Bernardi and Verbrugge [4,5] and Springer [6]. However, the transport phenomena in different layers of PEM fuel cell are in different length scales according to the Knudsen number [7], as shown in Fig. 1 (b). In general, the VOF (volume of fluid) method [8] takes the advantages of tracking the gas and liquid phase interface in macroscopic scale and widely utilized to simulate the gas and liquid two-phase flow in channel and GDL. The LBM (lattice-Boltzmann method) [9] (mesoscopic scale) exhibits excellent numerical stability and constitutive versatility and has advantages in treating interfacial dynamics and complex geometries, which makes it advantageous to simulate the two-phase flow in porous electrodes. Further, to simulate the transfer phenomena in microcosmic scale (e.g. proton transport in PEM), the MD (molecular dynamics) [10] or QM (quantum mechanics) [11] methods are usually adopted.

Moreover, there are also significant differences in time scales between the various transport phenomena in PEM fuel cell operation. For instance, the time step of VOF is usually 10^{-6} – 10^{-3} s, but the formation of slug flow in channels may take several minutes and even hours [2]. Limited by the coupling difficulties and computation amount, it is still impractical to develop a PEM fuel cell model including detailed gas and

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