Validation methodology for PEM fuel cell three-dimensional simulation

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Abstract

For modeling and simulation of proton exchange membrane (PEM) fuel cell, validation has been an essential and challenging task. This study implements a comprehensive validation including both overall cell performance and local distribution characteristics under different operating conditions with experimental data from two public sources. Polarization curve, cell ohmic resistance, current density distribution and temperature distribution are all involved. A “three dimensional + one dimensional” (“3D+1D”) model is adopted which simplifies part of cell components in order to boost the calculation efficiency. The validation methodology is clarified by listing those

undetermined model parameters and analyzing their "accessibility" as well as correlations with the three kinds of voltage losses (activation, ohmic and mass transfer). It is found that the control regions of ohmic voltage loss and concentration voltage loss overlap among a wide current density range, which may lead to misjudgment in the validation process. The details of parameter adjustment are also shared. Simulation results of the two validation tests both obtain decent agreement with the experiments and reflect consistent variation trends as the condition changes. The liquid water in gas channel is proved to have a double effect on cell performance and should be taken into careful consideration especially under low humidification and high current density working conditions.

Keywords

PEM fuel cell; Validation; Three-dimensional simulation; Current density distribution; Channel liquid water

Nomenclature

A	specific surface area (m^{-1})
C	gas molar concentration (mol m^{-3}) /specific heat capacity (J mol^{-1} K^{-1})
D	diffusivity (m^2 s^{-1})
EW	equivalent weight of ionomer (kg mol^{-1})
F	faraday's constant (C mol^{-1})

H	henry's constant (Pa m^3 mol^{-1})
h	latent heat of water (J mol^{-1})
i	exchange current density (A m^{-2})
j	electrochemical reaction rate in current form (A m^{-3})
K	intrinsic permeability (m^2)
k	relative permeability/thermal conductivity (W m^{-1} K^{-1})
M	molar mass (kg mol^{-1})
m	specific mass (kg m^{-2})
P	pressure (Pa)
R	universal gas constant (J mol^{-1} K^{-1})/transport resistance (s m^{-1})
RH	relative humidity
S	source term (kg m^{-3} s^{-1}, W m^{-3} …)
s	liquid water saturation
T	temperature (K)
$\vec{u}$	velocity vector (m s^{-1})
V	output voltage (V)
Y_i	gas species mass fraction

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2 **Greek letters**

α	transfer coefficient

δ	thickness (m)
ε	porosity or phase fraction
$\zeta_{pt/c}$	platinum weight percentage of Pt/carbon catalyst
$\zeta_{im/c}$	mass ratio of ionomer to carbon
η	overpotential (V)
θ	correction coefficient
κ	charge 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})
φ	charge potential (V)

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2 **Subscripts and superscripts**

0	standard state
a	anode
act	active
ave	average
BP	bipolar plate
CL	catalyst layer

c	cathode/carbon support
ECSA	electrochemical active surface area
eff	effective
ele	electronic
GDL	gas diffusion layer
g	gas
i	gas species
im	ionomer
im/c	ionomer/carbon
ion	ionic
local	local characteristic
lw	liquid water
MEM	membrane
MPL	microporous layer
m	mass
mix	gas and liquid mixture
mw	membrane water
n	number of computing node
O_2	oxygen
pt	platinum

ref	reference
sat	saturation state
T	temperature
*	correction

1. Introduction

Proton exchange membrane (PEM) fuel cell is a device that converts chemical energy into electricity through electrochemical reaction and has been widely considered as a promising choice for automobile power source [1]. Recently, Toyota has released its new version of fuel cell vehicle (FCV), MIRAI second generation [2], which confirms the potential and illustrates that the technology development of PEM fuel cell has advanced to a new stage. As the power density of PEM fuel cell stack has jumped to higher level (4-5 kW L^{-1}) and keeps rising, the current density at rated operating condition may attain 3 A cm^{-2} or higher, which means much quicker reaction consumption, water and heat generation, and so could bring unforeseeable problems. Consequently, it is of great significance to gather a deep understanding about the underlying transport phenomena occurring in the fuel cell as working condition evolves. Numerical simulation usually functions as an efficient tool in the pre-development stage [3]. Simulation tools with high efficiency, decent validity and good applicability are urgently needed to ensure the effectiveness of preliminary evaluation for designing and optimization work.

As for the macroscale PEM fuel cell models, it is typical to make trade-off between complexity and efficiency, as the scale of simulation domain varies from one-dimensional (1D) to three-dimensional (3D) [4] along with different assumptions and simplifications. Consequently, validation process is indispensable to ensure the validity of utilizing these models, qualitatively or quantitatively. 3D modeling and simulation benefits from the capability of reproducing the geometry structure of bipolar plate (BP) and is preferred by studies with designing purpose [5, 6]. However, the validation requirements of 3D simulation tools concomitantly become higher because it presents more comprehensive information, some of which also enters the detectable scope of laboratory. Usually, polarization curve is used for verification as the most common characterization method. However, evidence suggests that one polarization curve is not nearly enough to prove the reliability of a model with a host of given parameters, i.e., different parameter combinations can draw similar results [7, 8]. Actually, multi-conditional comparison has been gradually accepted by more and more numerical modeling researchers [9, 10, 11], e.g., adopting two or more polarization curves at different operating conditions or cell configurations to guarantee the quality of validation procedure. Another feasible way to obtain more credible results is to compare the cell ohmic loss as well as output voltage at different working current density [12]. Even so, the validation indexes mentioned above are all lumped or global features of PEM fuel cell, which contain very limited information of local characteristics.

In order to conduct more comprehensive and convincing validation, comparing the spatial distribution of key parameters, e.g., current density, is another important means. Actually, methods of spatially resolved measurement with segmented bipolar plate (BP) [13] or specially designed membrane electrode assembly (MEA) [14] have been proposed by prior work such as printed circuit board method [15], resistance array method [16], Hall sensor method [17], et al. And there has been quite a few valuable data which are available for simulation researchers. However, relatively scant studies that have implemented distribution validation are reported on PEM fuel cell modeling and simulation. Ju and Wang [18] performed early comparison of current distribution between simulation results and measured data. They used 9 average values of segmental current density along the channel. Li et al. [19] underlined the in-plane difference of current distribution between channel and land. They found that the position of current density peak would change as operating conditions varied, indicating the importance of understanding local characteristics. Hakenjos et al. [20] measured the current density and the high frequency resistance (HFR) of a 45 segmented flow field plate (3 rows of 15). They validated their model at three different air flow rates and only the 50 sccm case showed a distinct gap between simulation and measurement. There is no doubt that higher spatial resolution of segment directly improves the accuracy of measurement. Nevertheless, it also adds to the difficulty and complicity, not only for experiment setup but also for model validation. Carnes et al. [21] increased the number of segments to

100 on a 50 cm^2 single cell with 10×10 arrangement. They compared their model results with experimental data under different inlet humidity and working temperature. The validation results showed acceptable trend with less than 30% local error. Similarly, Zhang et al. [22] compared two sets of experimental data from different sources (including Carnes et al. [21]) with their simulation results of two cell domain, respectively. Fink et al. [23] conducted current distribution validation using a simulation domain of metallic BP and considered the effect of manufacturing assembly and welding spots. Tardy et al. [24] validated the liquid water heterogeneities of a large area PEM fuel cell by extracting liquid water thickness from neutron imaging measurement. It is also worth mentioning that both Fink et al. and Tardy et al. made some simplification on the 3D transport in MEA to facilitate the computation process. This is kind of in common with this study and will be clarified in next section. Hao et al. [25] gave a comprehensive validation of current, temperature and liquid water distribution under different working conditions with different cathode platinum (Pt) loadings. The experimental data originated from a U.S. Department of Energy (DOE) project [26] and the measuring points was set by 4×25 arrangement. The current and temperature are simultaneously measured on anode side and cathode side, respectively. The completeness of Hao's work is marvelous and stands out from most of peer studies. However, the ointment is that only part of the cases showed good agreement on the comparison of current density and temperature contours. Distinct difference between experiment and simulation could still be seen under low cathode inlet humidity and low

Pt loadings. Of course, there should still be some unknown factors, either for the understanding of transport mechanism or the uncertainty of experiment operation.

Validation plays an essential part in simulation studies and could be challenging and time-consuming. To the authors' knowledge, only a tiny quantity of PEM fuel cell modeling studies has implemented model validation with adequate data and in-depth analysis. And the details of model validation are seldom mentioned. In this study, a comprehensive validation will be carried out including both overall (polarization curve and ohmic loss) and local characteristics (current and temperature distribution) based on two different experimental data sources with a "3D+1D" PEM fuel cell model. Different inlet relative humidity, operating temperature and flow arrangement are all involved, which is also rare in the open literature. The validation methodology will be clarified by listing those undermined model parameters and elucidating the adjustment strategy. At last, the effect of liquid water in gas channel will be discussed based on the simulation results.

2. Implementation method

2.1 Computational domain

Fig. 1 shows the flow field layouts of two computational domains for this study. In the first experiment, current density distribution was measured with the Hall-Effect sensor by separating the active area into 10 segments as depicted in Fig. 1a [27]. Co-flow

arrangement was adopted with two kinds of serpentine flow fields. For the second data source in Fig. 1b [25], current distribution and temperature distribution were monitored at the same time at anode BP surface and cathode BP surface with 4×25 measuring points, respectively. The zig-zag gas channels and the straight coolant channels are reproduced. For simplicity, the part of bipolar plate around the curving manifold is removed. All the geometry dimension is kept consistent with the reference as much as possible, using directly available or estimated parameters.

2.2 "3D+1D" PEM fuel cell model

Model Assumptions

- In-plane transport in microporous layer (MPL), catalyst layer (CL) and membrane (MEM) is neglected;
- GDL and MPL are seen as homogeneous porous media;
- Catalyst layer is composed of evenly dispersed platinum-carbon-ionomer agglomerates [28];
- Ideal gas; Laminar flow; Steady state.

The "3D+1D" modeling method adopted in this study has been clarified with details in our previous work [29] and the model framework is exhibited in Fig. 2. The 3D part contains bipolar plate, gas channel (GCH), coolant channel (not schematized here) and gas diffusion layer (GDL), where 3D mesh is also drawn. The rest components (MPL,

CL and MEM) are mathematically embedded into the model code with 1D description. The two parts are bridged through two extra layers of mesh connected to anode and cathode separately, in which the 1D solutions are obtained and data are exchanged between the 3D computational fluid dynamics (CFD) model and the 1D sub-model. For every computing node set at the component interface (MPL-CLs, CL-MEMs), equations are built among three adjacent nodes (numbered 0, 1, 2) and two components (numbered 0, 1). The involved conservation equations are listed as below.

3D domain,

Mass $$\nabla \cdot \left(\rho_{\mathrm{g}} \vec{u}_{\mathrm{g}} \right) = S_{\mathrm{m}} \tag{1}$$

Momentum $$\nabla \cdot \left(\rho_{\mathrm{g}} \vec{u}_{\mathrm{g}} \vec{u}_{\mathrm{g}} \right) = -\nabla P + \nabla \cdot \left(\mu_{\mathrm{mix}} \left(\nabla \vec{u} + \nabla \vec{u}^{\mathrm{T}} \right) - \frac{2}{3} \mu_{\mathrm{mix}} \nabla \vec{u} \right) + S \tag{2}$$

Species $$\nabla \cdot \left(\rho_{\mathrm{g}} \vec{u}_{\mathrm{g}} Y_{\mathrm{i}} \right) = \nabla \cdot \left(\rho_{\mathrm{g}} D_{\mathrm{i,eff}} \nabla Y_{\mathrm{i}} \right) + S_{\mathrm{i}} \tag{3}$$

Energy $$\nabla \cdot \left(\rho_{\mathrm{g}} C_{\mathrm{pg,eff}} \vec{u}_{\mathrm{g}} T \right) = \nabla \cdot \left(k_{\mathrm{eff}} \nabla T \right) + S_{\mathrm{T}} \tag{4}$$

Liquid water saturation in flow channel

$$\nabla \cdot \left(\rho_{\mathrm{lw}} \vec{u}_{\mathrm{lw}} s \right) = \nabla \cdot \left(\rho_{\mathrm{lw}} D_{\mathrm{lw}} \nabla s \right) + S_{\mathrm{lw}} \tag{5}$$

Liquid pressure in porous electrodes

$$0 = \nabla \cdot \left(\rho_{\mathrm{lw}} \frac{K k_{\mathrm{lw}}}{\mu_{\mathrm{lw}}} \nabla P_{\mathrm{lw}} \right) + S_{\mathrm{lw}} \tag{6}$$

Electronic potential

$$0 = \nabla \cdot \left(\kappa_{\mathrm{ele,eff}} \nabla \phi_{\mathrm{ele}} \right) + S_{\mathrm{ele}} \tag{7}$$

1D domain,

Species $$0=\sum_{n=0}^{1} D_{\mathrm{i,eff}}^{\mathrm{n}} \frac{C_{\mathrm{i}}^{\mathrm{n}+1}-C_{\mathrm{i}}}{\delta^{\mathrm{n}}}+\sum_{n=0}^{1} 0.5\delta^{\mathrm{n}} S_{\mathrm{i}}^{\mathrm{n}} \quad (8)$$

Temperature $$0=\sum_{n=0}^{1} k_{\mathrm{i,eff}}^{\mathrm{n}} \frac{T^{\mathrm{n}+1}-T}{\delta^{\mathrm{n}}}+\sum_{n=0}^{1} 0.5\delta^{\mathrm{n}} S_{\mathrm{T}}^{\mathrm{n}} \quad (9)$$

Liquid pressure in porous electrodes

$$0=\sum_{n=0}^{1} \frac{\rho_{\mathrm{lw}} K^{\mathrm{n}} k_{\mathrm{lw}}^{\mathrm{n}}}{\mu_{\mathrm{lw}}} \frac{P_{\mathrm{lw}}^{\mathrm{n+1}}-P_{\mathrm{lw}}^{\mathrm{n}}}{\delta^{\mathrm{n}}}+\sum_{n=0}^{1} 0.5\delta^{\mathrm{n}} S_{\mathrm{lw}}^{\mathrm{n}} \quad (10)$$

Membrane water content in ionomer

$$0=\sum_{n=0}^{1} \frac{\rho_{\mathrm{im}} D_{\mathrm{mw}}}{EW} \frac{\lambda^{\mathrm{n+1}}-\lambda^{\mathrm{n}}}{\delta^{\mathrm{n}}}+\sum_{n=0}^{1} 0.5\delta^{\mathrm{n}} S_{\mathrm{mw}}^{\mathrm{n}} \quad (11)$$

Electronic potential

$$0=\sum_{n=0}^{1} \kappa_{\mathrm{ele,eff}}^{\mathrm{n}} \frac{\phi_{\mathrm{ele}}^{\mathrm{n+1}}-\phi_{\mathrm{ele}}^{\mathrm{n}}}{\delta^{\mathrm{n}}}+\sum_{n=0}^{1} 0.5\delta^{\mathrm{n}} S_{\mathrm{ele}}^{\mathrm{n}} \quad (12)$$

Ionic potential

$$0=\sum_{n=0}^{1} \kappa_{\mathrm{ion,eff}}^{\mathrm{n}} \frac{\phi_{\mathrm{ion}}^{\mathrm{n+1}}-\phi_{\mathrm{ion}}^{\mathrm{n}}}{\delta^{\mathrm{n}}}+\sum_{n=0}^{1} 0.5\delta^{\mathrm{n}} S_{\mathrm{ion}}^{\mathrm{n}} \quad (13)$$

where ε, ρ (kg m^{-3}), $\vec{u}$ (m s^{-1}), s, P (Pa), μ (kg m^{-1} s^{-1}), Y, D_{eff} (m^2 s^{-1}), C_{p} (J mol^{-1} K^{-1}), T (K), k_{eff} (W m^{-1} K^{-1}), K (m^2), k_{lw}, κ_{eff} (S m^{-1}), ϕ (V), $C_{\mathrm{i}}^{\mathrm{n}}$ (mol m^{-3}), δ (m), EW (kg mol^{-1}) and λ are the porosity, the density, the superficial velocity vector, the liquid water saturation, the pressure, the viscosity, the species mass fraction, the effective gas diffusivity, the specific heat capacity, the temperature, the effective heat conductivity, the intrinsic permeability, the liquid phase relative permeability, the charge conductivity, the charge potential, the gas molar concentration, the component thickness, the equivalent ionomer weight and the membrane water content, respectively.

The $\vec{u}_{\mathrm{lw}}$ in the convection term of liquid water saturation equation in gas channel is derived from the gas velocity with the correction of viscosity ratio and local saturation values [30].

$$\frac{\vec{u}_{\mathrm{lw}}}{\vec{u}_{\mathrm{g}}} = \frac{\mu_{\mathrm{g}}}{\mu_{\mathrm{lw}}}\left(\frac{s}{1-s}\right)^{3} \tag{14}$$

Leveret-J function [31] is adopted to obtatin the diffusion coefficient of channel liquid water and also to calculate liquid water saturation from liquid pressure in porous electrodes.

$$P_{\mathrm{c}} = \sigma\cos\theta\left(\frac{\varepsilon}{K}\right)^{0.5} J\left(s\right) \tag{15}$$

$$J\left(s\right) = \begin{cases} 1.42\left(1-s\right) - 2.12\left(1-s\right)^{2} + 1.26\left(1-s\right)^{3} & \theta < 90° \\ 1.42s - 2.12s^{2} + 1.26s^{3} & \theta > 90° \end{cases} \tag{16}$$

The electrochemical reaction rate is calculated by the Butler-Volmer equation and the cathode part is revised with agglomerate correction [28],

$$j_{\mathrm{a}} = i_{0,\mathrm{a}}^{\mathrm{ref}} A_{\mathrm{pt}}^{\mathrm{eff}} \theta_{\mathrm{T,a}} \left(\frac{RTC_{\mathrm{H_2}}}{H_{\mathrm{H_2}} C_{\mathrm{H_2}}^{\mathrm{ref}}}\right)^{0.5} \left[\exp\left(\frac{2F\alpha_{\mathrm{a}}\eta_{\mathrm{a}}}{RT}\right) - \exp\left(-\frac{2F(1-\alpha_{\mathrm{a}})\eta_{\mathrm{a}}}{RT}\right)\right] \tag{17}$$

$$j_{\mathrm{c}} = \frac{RTC_{\mathrm{O_2}}}{H_{\mathrm{O_2}}} \left(\frac{C_{\mathrm{O_2}}^{\mathrm{ref}}}{i_{0,\mathrm{c}}^{\mathrm{ref}} A_{\mathrm{pt}}^{\mathrm{eff}} \theta_{\mathrm{T,c}} \left[\exp\left(\frac{4F\alpha_{\mathrm{c}}\eta_{\mathrm{c}}}{RT}\right) - \exp\left(-\frac{4F(1-\alpha_{\mathrm{c}})\eta_{\mathrm{c}}}{RT}\right)\right]} + \frac{R_{\mathrm{local}}}{4FA_{\mathrm{im}}}\right)^{-1} \tag{18}$$

where j (A m^{-3}), i (A m^{-2}), A (m^{-1}), θ_{T}, R (J mol^{-1} K^{-1}), H (Pa m^{3} mol^{-1}), F (C mol^{-1}), α, η (V) and R_{local} (s m^{-1}) are the electrochemical reaction rate, the exchange current density, the specific area, the temperature correction coefficient, the universal gas constant, the Henry's coefficient, the Faraday constant, the transfer coefficient, the

overpotential and the local gas transport resistance. S_m (kg m^{-3} s^{-1}), S_u (N m^{-3}), S_i (kg m^{-3} s^{-1}), S_T (W m^{-3}), S_{lw} (kg m^{-3} s^{-1}), S_{ele} (A m^{-3}) and S_{ion} (A m^{-3}) are corresponding source terms of above conservation equations. The subscripts "g", "lw", "mix", "i", "ele", "ion", "eff", "im", "pt" and "mw" represent gas, liquid water, mixture of gas and liquid, gas species (hydrogen, oxygen and water vapor), electric, ionic, effective value, ionomer, platinum and membrane water, respectively. The geometry parameters and operating conditions are given in Table 1.

2.3 Validation methodology

In order to build a PEM fuel cell model, there are usually a multitude of input parameters and correlations regarding transport properties or electrochemical mechanisms. Due to the nature of unobservability or unpredictability for most of microscale transport phenomena, undetermined coefficients and correction parameters are usually necessary. These parameters are attached with physical meaning to describe those factors which surely exist but remain unclear. On the one hand, a model needs adequate degrees of freedom to be capable of adapting to the variation of practical conditions. On the other hand, those parameters or correlations are often introduced or derived based on relevant research findings or convincing data, and thereby, should follow their valid range or application scope. Apart from the complexity of model itself, reasonable and subtle adjustment of the undetermined parameters functions as the most important part to accomplish the validation process.

Normally speaking, the core of model validation is to capture the change rule of all the (ideally) or the currently-dominant voltage losses (activation, ohmic and mass transfer or concentration) by recognizing and adjusting those associate parameters. The most difficult task is to coordinate all the undetermined parameters within reasonable range, i.e., without violating general knowledge or physical laws, at the same time yielding results with correct trends. Table 2 has listed the undetermined parameters of the "3D+1D" model in this work, which are also shared by other macroscale PEM fuel cell models (may not be completely same because of different model framework or assumptions). Their typical ranges for usage are also given. And the accessibility is lumped index that varies from 0 to 5 and represents multiple meanings, including whether the parameter can be obtained, accurately measured or roughly, in-situ or ex-situ, i.e., how much room it has for adjustment and whether the mechanism is clearly understood. Larger value of accessibility means the parameter can be measured or accessed with less uncertainty or deviation, i.e., the parameter could be adjusted in a more limited range. It should be noted that though smaller accessibility represents more freedom for parameter adjustment, the setting of values could not be more cautious and must obey physical laws and existing credible theories. The relationship between these parameters and the three kinds of voltage losses is also annotated to help choose suitable parameters according to the practical demand, e.g., adjusting only one kind of voltage loss or two kinds at the same time, slightly or sharply. It should be noted that Table 2 is

concluded based on authors' knowledge and public data and may have deviations. Nevertheless, it provides a more intuitional guide of adjusting strategy. In Table 3, values and expressions of key model parameters used for the two validation tests in this study are given.

3. Results and discussion

3.1 Validation 1

As shown in Fig. 3, the simulated polarization curves give good agreement with experimental data under both higher and lower inlet humidification. And Fig. 4 depicts the comparison of current density distribution of two cases between simulation and experiment, at 1.25 A cm^{-2} and 0.81 A cm^{-2} under different conditions respectively. The consistency of the variation trend is also quite well. It can be seen that the current density decreases from inlet to outlet in the high humidity case (Fig. 4a) and the tendency reverses for the low humidity case (Fig. 4b). This indicates that the former is at the stage that mass transfer voltage loss begins to dominate, and the latter is in the ohmic control region. However, if only judging from the polarization curve, view can be easily drawn that the 1.25 A cm^{-2} case is still in the ohmic control region because the slope of the curve nearly maintains the same, i.e., the obvious downward trend originated from oxygen shortage doesn't appear. And the reason should be related to the 50% relative humidity of cathode inlet and the co-flow arrangement. Under relatively higher current density, the area near the cathode outlet is susceptible to

oxygen shortage. On the other hand, the membrane hydration becomes better as current density increases near the inlet region, alleviating the drying effect of unsaturated air. Consequently, the combined influence of ohmic loss and concentration loss keeps the slope almost unchanged. This demonstrates that the ohmic control region and mass transfer control region overlap among a relatively wide current density range, as highlighted in Fig. 3. It also proves the importance of raising the validation requirement with other characteristics like cell ohmic resistance and current density distribution.

3.2 Validation 2

To further test the validation methodology, we choose two cases of different operating temperatures (40 ℃ and 60 ℃) from Hao et al.'s work [25]. The results are shown as in Fig. 5 and 6, together with output voltage and cell resistance. The 100 measuring points are picturized into partitioned contour and the simulated current density on the BP surface is also treated into 100 segments for the convenience of comparison. The working current density is fixed at 1.5 A cm^{-2}. The overall consistency of the temperature distribution and the 25 average values (see Fig. 5b, 5c and 6b, 6c) along the air flow direction is fairly well but part of the current density distribution still manifests relatively obvious deviation, especially for the inlet region. The cause could originate from the unknown gap between the real inlet condition and the ideal state of simulation setup to some extent. However, for both experiment and simulation, it can be seen from Fig. 5a and 6a that the current density peak at 60 ℃ becomes closer to the

cathode inlet and the current density near the outlet decreases distinctly compared with the 40 ℃ case. This is mainly because that the inlet oxygen partial pressure is lower at higher temperature for both fully humidified air, which means the effect of mass transfer voltage loss increases. For these two different operating temperatures, current density distribution, temperature distribution, output voltage and cell ohmic resistance are validated at the same time and the agreement is also good. It should be noted that most of the undetermined model input parameters are kept the same as listed in Table 3, which means the model complexity stands well with the two validation tests and the applicability of the current methodology is good.

3.3 Effect of liquid water in gas channel

The proper consideration of liquid water in gas channel has been a challenging and tough task for PEM fuel cell models [32], which also accounts for the deviation between model prediction and experimental measurement to a large extent. The space and time scales of those two-phase models which consider phase interaction, dynamic contact angle or wall adhesion, are too small for macroscale models and the demand of computing capability could be unaffordable even for a small cell domain. Instead, a continuous liquid water saturation is usually solved to consider the existence of liquid water as written in Eq. (5).

Fig. 7 shows the liquid water saturation contours in cathode gas channel of the above 4

validation cases. It can be seen that the liquid water saturation gains higher values around corners and near the outlet region, where gas flow is obstructed or oxygen is in short supply. And it has a close correlation with cell working conditions. There is nearly no water accumulation in gas channel for the low inlet relative humidity and low current density case (see Fig. 7b). In Fig. 7c and 7d, the liquid water saturation distribution pattern is very similar for the two cases. The reason is that the inlet air is kept saturated and the current density is the same. Fig. 8 compares the difference of solving the liquid water saturation equation in gas channel or not for the two validation tests. Interestingly, the two polarization curves under higher inlet humidification in Fig. 8a are almost the same while the ohmic loss shows apparent discrepancy, which means the liquid water in gas channel has a double effect on cell ohmic resistance and gas transport resistance at the same time. On the one hand, the liquid water in channel hinders the water removal from the electrode and adds to the gas blockage. On the other hand, the retained water improves the ionic conductivity to a certain degree under not fully humidified condition. The variation trend of current density distribution for the 1.25 A cm^{-2} case turns opposite in Fig. 8b also proves the influence of channel liquid water and again the importance of validating current density distribution. For the 60 ℃ case of validation 2 (see Fig. 8c), it can be seen that the cell output voltage becomes higher even the ohmic resistance increases and the current density distribution gets more uniform without the existence of liquid water in channel. This indicates that its influence on gas transport overcomes the ohmic part. These results illustrate that it is of great significance to

consider the liquid water in gas channel. In addition to the negative effect on mass transfer loss, which is also focused by most of preceding studies, the positive effect on ionic ohmic loss should also be brought to the forefront, especially under low humidification and high current density conditions.

4. Conclusion

In this study, a comprehensive validation process for PEM fuel cell three-dimensional simulation is accomplished and the validation methodology is investigated and analyzed. Polarization curves, ohmic loss, current density distribution and temperature distribution under different inlet humidification and operating temperature are compared with experimental data from two studies and decent agreement are achieved simultaneously. Both co-flow and counter-flow arrangement are included. The core part of model validation, that recognizing, adjusting and coordinating the undetermined parameters, is clarified by listing their typical range, accessibility and relationship with three kinds of voltage losses. By comparing the current density distribution, the ohmic voltage loss and the concentration voltage loss manifest a combined influence region across a relatively wide current density range, which is easy to mislead the validation process and strongly proves the necessity of multi-index comparison. The validation details are also shared by giving the setting of key parameters. Additionally, the liquid water saturation in gas channel is found to have a double effect on cell ohmic loss and concentration loss, which should be carefully considered under low humidification and

high current density conditions. The validation work in this study may still have doubtful points on current model assumptions, e.g., evenly distributed contact resistance and inadequate spatial discretization for one-dimensional domain. And the proposed methodology contains some subjective and empirical understandings such as the definition of "accessibility" and the adjustment of individual correction coefficients. Nevertheless, one thing is for sure that model validation should be as comprehensive as possible and the demand of improving the model complexity would inevitably increase in order to keep pace. With a mutual promotion, the misleading and the uncertainty caused by parameter combination could be gradually reduced. The results and analyses for validation methodology in this study are expected to provide some instructions for researchers and engineers in the field of PEM fuel cell design, to facilitate the application of three-dimensional modeling and simulation.

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Figure and table captions

Figure 1 Flow field layouts of the two computational domains for model validation. (a) Active area: 50 cm^2; Anode: triple serpentine flow field; Cathode: quadruple serpentine flow field [27]. (b) Active area: 40 cm^2; Anode: 11 zig-zag gas channels and 12 straight coolant channels; Cathode: 22 zig-zag gas channels and 12 straight coolant channels [25].

Figure 2 Framework of the "3D+1D" PEM fuel cell model.

Figure 3 Comparison of polarization curves between simulation and experiment [27] under higher inlet humidification, anode 100%/cathode 50% relative humidity (RH 100%/50%) and lower inlet humidification, anode 25%/cathode 25% relative humidity (RH 25%/25%) in validation test 1.

Figure 4 Comparison of current density distribution between simulation and experiment [27] from two cases in validation test 1. (a) 1.25 A cm^{-2} under higher inlet humidification. (b) 0.81 A cm^{-2} under lower inlet humidification.

Figure 5 Comparison of (a) current density distribution, (b) temperature distribution and (c) 25 average values along the air flow direction of the 40 ℃operating temperature case between simulation and experiment [25] in validation test 2. The current density

distribution and temperature distribution are simultaneously measured on the end plane of anode and cathode bipolar plates, respectively.

Figure 6 Comparison of (a) current density distribution, (b) temperature distribution and (c) 25 average values along the air flow direction of the 60 ℃ operating temperature case between simulation and experiment [25] in validation test 2.

Figure 7 Liquid water saturation contours in cathode gas channel of the 4 validation cases. (a) 1.25 A cm^{-2} case under 100%/50% inlet humification; (b) 0.81 A cm^{-2} case under 25%/25% inlet humidification; (c) 1.5 A cm^{-2} case at 40 ℃ operating temperature; (c) 1.5 A cm^{-2} case at 60 ℃ operating temperature.

Figure 8 Comparison of whether the liquid water saturation in gas channel is considered. (a) Polarization curve and ohmic loss under the 100%/50% inlet humification in validation test 1; (b) Current density distribution of the 1.25 A cm^{-2} case; (c) 25 average current density values of the 60 ℃ operating temperature case in validation test 2.

Table 1 Geometry parameters and operating conditions.

Table 2 Undetermined parameters of the “3D+1D” model in this study.

Table 3 Values and expressions of key model parameters used for validation.