EFFICIENT NUMERICAL INVESTIGATION OF FUEL CELL PERFORMANCE UNDER VARIOUS CONDITIONS

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ABSTRACT

The multi-resolution fuel cell simulation approach is employed in this study. For the membrane and diffusion layer, a full 3D modeling is employed; for the catalyst layer, 1D+2D, that is, at each location of the fuel cell plate, the governing equations are integrated only in the direction perpendicular to the fuel cell plate; for the flow channels, a quasi-ID method with high numerical efficiency and reasonable accuracy is employed. In addition, both the anode and cathode activation overpotential are computed according to the specified cell potential and reversible fuel cell potential. An industrial size single cell with a multi-channel serpentine flow pattern has been used to study the pressure and cathode inlet relative humidity effects on the fuel cell performance. The increase in pressure generally improves the fuel cell performance; the increase in cathode inlet relative humidity has an adverse affect on fuel cells at large current densities but a favorable effect at small current density.

INTRODUCTION

Fuel cells are electrochemical devices that directly convert chemical potential into electrical power. As a clean and efficient power source, fuel cells can be a potential replacement for traditional heat engine power sources, and thus have attracted numerous research efforts in the area of experiments, mathematical analysis, and numerical simulations. Especially, there have been extensive efforts devoted to PEM fuel cell modeling and simulation to study the fuel cell performance under various conditions (1-9). The research spans from the atomic and quantum level up to the fuel cell system, and even to the entire fuel cell power train. One can organize the fuel cell modeling and simulation activities into four levels: micro level, single cell level, stack level and system level. The current study will focus on single cell level, however, with the future integration with other levels in mind.

The key to the success of single cell simulation is (i) transport property measurements under operation conditions; (ii) microstructure characterizations at gas diffusion, catalyst and membrane layers; (iii) detailed reaction mechanism; and (iv) efficient integration of all layers with proper resolutions. The satisfaction of last condition is largely dependent upon algorithm design that takes advantage of typical geometry features and different flow transport scales in different directions and layers, and the efficient use of high performance computers.
The existing full 3D approach (6-7) can theoretically be employed to study the details of PEM fuel cells, but the simulation of a full size fuel cell and fuel cell stacks can be prohibitive due to the unbearable computational cost. Further, for the purpose of design optimization of fuel cells, the simulation turn around time has to be dramatically reduced to satisfy the industrial needs. On the other hand, some simplified models such as the empirical models (8), 1D models (1-2), 2D models (3-5) and quasi-3D model (9), can not be used for systematic study of various flow and coolant channel design and optimizations. For example, the correct dynamic pressure drop for different channel designs in both flow and coolant channels cannot be correctly predicted using the existing simplified methods. In this paper, it will be shown that the current multi-resolution strategy could be effectively used for simulation aided fuel cell design and performance evaluations.

The strategy of the multi-resolution fuel cell simulation is based on the following reasoning. For the membrane, a full 3D modeling is employed due to the fact that transport present in the membrane includes both convection and diffusion in all directions. The catalyst layer is modeled as 1D+2D, that is, at each location of the fuel cell plate, the governing equations are integrated only in the direction perpendicular to the fuel cell plate. The reason behind is that the catalyst layer is much thinner compared to other layers, and all variable gradients perpendicular to the plate are much larger than those in other directions. The diffusion layer is modeled as full 3D flow due to the dominance of diffusion process present. Finally, the most challenging part is the flow channel modeling. A full size high power single cell can have very long and complex flow channels. Current study takes advantage of the flow channel geometry and uses a quasi-1D method to model the flow channels. However, the interface between the flow channel and the diffusion layer needs extreme caution to satisfy conservation laws.

The integration of all layers has to be efficient and seamless. The computation work should be evenly and efficiently distributed among multiprocessors. To accomplish above goals, we have been developing our simulation capabilities based on Loci system (10), developed at Mississippi State University. The Loci system is an application framework that seeks to reduce the complexity of assembling large-scale finite-difference, finite-volume, or finite-element applications and automatically generates execution schedules for either shared or distributed memory parallel computing hardware. Unlike the traditional parallel software development, the numerical model implementers are exempt from low-level concerns such as message passing among processors and efficient memory allocations. In addition, Loci is especially useful in integrating multi-discipline simulation models by automatic consistency checks at the model interfaces, thereby, reducing implementation errors.

The results from the early works (11-13) have demonstrated good agreement between the full 3D and the multi-resolution approach. In this paper, an industrial size single cell with a multi-channel serpentine flow pattern was simulated. For the purpose of fuel cell optimization design, it is important to know the sensitivity of each variable. It is well known that water in the membrane layer plays a key role for the performance of PEM fuel cells because the proton conductivity in the membrane layer depends largely on the water content. Also, the operating pressure can affect the species reactions and transports in fuel cells. Therefore, the effects of the variations of pressure and the cathode inlet humidity on the fuel cell performance are to be analyzed. In addition, a new approach for...
accurately predicting the activation overpotential distribution of both anode and cathode and thus the fuel cell performance are to be developed.

FORMULATION

In this section, the models used for various components and interfaces in the current simulation are presented. Especially, the agglomerate oxygen reduction model employed for the cathode side, the interface model for the anode side, the porous media model for both the gas diffusion layers (GDL) and the membrane layer, and the quasi-1D model for the flow channels, as well as the related interface conditions will be discussed in detail.

Agglomerate catalyst layer model

The electrochemical reactions in the cathode catalyst layer can be symbolically expressed as:

\[ \text{O}_2 + 4\text{H}^+ + 4\text{e}^- \rightarrow 2\text{H}_2\text{O}. \]  \[1\]

A first order oxygen reduction mechanism is employed in association with the agglomerate model, in which the catalyst layer is treated as the collection of agglomerates consisting of carbon particle support and platinum particles surrounded by a thin layer of Nafion. In contrast to the macro-homogeneous model, which treats the catalyst layer as a homogenous phase, the agglomerate model itself integrates the mass diffusion transfer resistance when the current density is large, as a result, the agglomerate model is capable of predicting a voltage drop-off at large current densities (14). The cathode activation overpotential \( \eta_c \) is the dependent variables to be solved. The governing equation for it is presented as (14)

\[
\nabla \cdot (-\sigma \nabla \eta_c) = 4F \left[ \frac{1}{\delta} \frac{1}{aC_{\text{O}_2} D_{\text{eff},\text{nafion}}} + \frac{1}{kE} \right],
\]  \[2\]

where the reaction rate \( k \) is modeled using the simplified Butler-Volmer equation and expressed as:

\[
k = \frac{A_j_{\text{O}_2}}{4F} \exp \left( -\frac{\eta}{\beta F} \right),
\]  \[3\]

and the efficiency factor \( E \) is given as

\[
E = \frac{\tanh(mL)}{mL},
\]  \[4\]

where \( mL \) is the Thiele’s modulus.
\[ mL = L \sqrt{\frac{k}{C^*_{O_2} D_{eff,nafion}}} \]  \[5\]

In the above equations, \( A, j_0, \beta, \sigma, F, \delta, a, L, C^*_{O_2} \) and \( D_{eff,nafion} \) are the specific active surface area, exchange current density, cathodic transfer coefficient, effective ionic conductivities of Nafion, Faraday's constant, thickness of surrounding Nafion layer, exposed agglomerate area density, characteristic length of the agglomerate, Nafion oxygen saturation concentration and effective oxygen diffusivity in Nafion, respectively. Due to the thinness of the catalyst layer and high gas phase oxygen diffusivity, the gas phase oxygen concentration, used to calculate the oxygen saturation concentration inside the Nafion solution, is taken as the gas phase oxygen concentration at the catalyst and gas diffusion layer interface. Therefore, no explicit species and momentum transport equations are solved inside the catalyst layer, a practice also taken by other researches who employed the agglomerate model (14). The parameter values used for the current simulations are listed in Table I.

**Table I. Cathode catalyst layer parameter values**

| Symbol     | Parameter                                           | Values          |
|------------|-----------------------------------------------------|-----------------|
| \( A \)    | Specific exchange current density                   | \( 5 \times 10^4 \text{ A m}^{-2} \) |
| \( \beta \) | Cathodic transfer coefficient                       | 2               |
| \( \sigma \) | Effective ionic conductivities of Nafion           | 7 \text{ S m}^{-1} |
| \( \delta / a \) | Thickness of surrounding Nafion layer divided by Exposed agglomerate area density | 6.2 \times 10^{-12} \text{ m}² |
| \( L \)    | Characteristic length of the agglomerate            | 3 \times 10^{-6} \text{ m} |
| \( D_{eff,nafion} \) H | Effective oxygen diffusivity in Nafion times Henry’s law constant | 4 \times 10^{-15} \text{ m}² \text{ s}^{-1} |

**Anode catalyst layer interface model**

The electrochemical reactions in the anode catalyst layer can be symbolically expressed as:

\[ H_2 \rightarrow 2H^+ + 2e^- \] \[6\]

**Table II Anode catalyst layer parameter values**

| Symbol     | Parameter                                           | Values          |
|------------|-----------------------------------------------------|-----------------|
| \( i_{0,a}^{ref} \) | Anode reference exchange current density         | 6 \times 10^{-3} \text{ A m}^{-2} |
| \( C_H^{ref} \) | Anode hydrogen reference concentration            | 1.2 \text{ kmol m}^{-3} |
| \( \gamma_{H_2} \) | Hydrogen concentration parameter                   | 0.5             |
| \( \alpha_a \) | Anode anodic charge transfer coefficient            | 1               |
| \( \alpha_c \) | Anode cathodic charge transfer coefficient          | 1               |

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Hydrogen is transported through the GDL to the catalyst layer, and discharges electrons by the electrochemical reaction described in Eq. [6]. The electrons move through the external circuit to provide a useful current while the protons transport through the polymer electrolyte membrane to the cathode catalyst layer to produce water by the reduction of oxygen at the cathode catalyst layer. The local current density of the anode catalyst layer $i_a$ is modeled by the Butler-Volmer equation (1) as:

$$i_a = i_0^a \left( \frac{C_{H_2}}{C_{H_2}^\infty} \right)^{\gamma H_2} \left[ \exp \left( \frac{\alpha_a F}{RT} \eta_a \right) - \exp \left( - \frac{\alpha_c F}{RT} \eta_a \right) \right].$$  \[7\]

where $i_0^a$ is the anode reference exchange current density; $C_{H_2}^\infty$ is the anode hydrogen reference concentration; $\gamma H_2$ is the hydrogen concentration parameter; $\alpha_a$ is the anode anodic charge transfer coefficient; $\alpha_c$ is the anode cathodic charge transfer coefficient. The parameters used in above equations are listed in Table II.

**Gas diffusion layer (GDL) model**

The GDL is made of either carbon paper or carbon cloth with or without Teflon pretreatment. The GDL functions mainly as an electronic conductor and a reactant distributor. Therefore, the GDL should be porous and have high electronic conductivity. In addition, to avoid either flooding or membrane dehydration, the GDL is sometimes pretreated with Teflon at the catalyst interface side to increase the hydrophobicity of the GDL on the side in contact with the catalyst layer. Recently, people have employed an extra layer, called a micro GDL, between the GDL and catalyst layer at the cathode side. The micro GDL is usually characteristic of high hydrophobicity and finer pore sizes than common GDL. The advantages of using the micro GDL are the reduction of flooding effects through higher hydrophobicity, and the increased electronic conduction by reducing the contact resistance between the GDL and the catalyst layer.

Since the current study is focused on the demonstration of the multi-resolution simulation strategy, and its application to the simulation aided fuel cell design, the current GDL model only consists of the classic Darcy’s law for the velocity field and the gas phase species transport. The species transport equations are derived from the mass conservation laws. The following is the governing equations used for the simulation:

$$\varepsilon \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0, \quad [8]$$

$$\mathbf{u} = -\varepsilon \frac{K}{\mu} (\nabla p - \rho g), \quad [9]$$

$$\varepsilon \frac{\partial \rho Y_k}{\partial t} + \nabla \cdot (\rho \mathbf{u} Y_k) = -\nabla \cdot (\rho Y_k V_k), \quad [10]$$

where the diffusion velocity of the $k$th species $V_k$ is modeled using the Fick’s law:

$$Y_k V_k = -D_k^{\infty} \nabla Y_k. \quad [11]$$

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Here, \( \varepsilon, \rho, u, K, \mu, p, g, Y_k \) and \( D_k^{\text{eff}} \) denote the porosity, gas phase density, super-facial velocity, permeability, effective viscosity, pressure, gravity, the \( k \)th species mass fraction and the effective \( k \)th species diffusivity, respectively. Since the temperature variations present in a single cell are often negligible, the isothermal condition is assumed for simplicity, and the energy equation is thus reduced to a constant temperature condition and not shown here. The transport coefficients, such as the viscosity \( \mu \) and gas diffusivity \( D_k \), are computed using CHEMKIN developed at Sandia National Lab. The expression for \( \mu \) and \( D_k \) are as follows:

\[
\mu = \sum_{i=1}^{N_S} f_i \mu_i / \left( 0.353534 \sum_{j=1}^{N_S} f_j \left( 1 + \frac{\mu_j}{\mu_i} \left( \frac{W_j}{W_i} \right)^{1/2} \right)^2 \right),
\]

\[
D_k = \frac{P_a}{W_k} \frac{\sum f_j W_j f_j}{\sum f_j / D_{kj}},
\]

where \( N_S, f_j, p, p_a, W_k, \mu_j, \) and \( D_{kj} \) are the number of species, the \( j \)th species molar fraction, gas pressure, atmospheric pressure, the \( k \)th species molecular weight, the \( j \)th species viscosity and the binary diffusion coefficient of the species pair \( k \) and \( j \), respectively. Here, both \( \mu_j \) and \( D_{kj} \) are obtained through data fitting from the empirical CHEMKIN transport data library. The effective transport coefficients are obtained using the Bruggeman relation:

\[
D_k^{\text{eff}} = \varepsilon^{1.5} D_k.
\]

**Membrane model**

The water transfer inside membrane is modeled to account for the convection driven by a pressure gradient, electro-osmotic drag of water from the anode to the cathode and back diffusion from the cathode to the anode. Due to the electro-neutrality assumption and the homogeneous distribution of charge sites, a constant proton concentration in the membrane is used. The momentum equation takes the form of the generalized Darcy relation. The potential equation is derived from Ohm’s Law. Thus, the system equations assuming isothermal condition for membrane are described as follows:

\[
\frac{\partial c_{H,0}^m}{\partial t} + \nabla \cdot N_{H,0}^m = 0,
\]

\[
u_m = -\varepsilon \frac{K_m}{\mu_m} (\nabla p - p g),
\]
Net water molar flux in membrane is given by the Nernst-Planck equation along with the Nernst-Einstein relationship:

\[
N_{H_2O}^m = \frac{n_d}{F} i_m - D_{H_2O}^m \nabla c_{H_2O}^m + c_{H_2O}^m u
\]  

where \( n_d \) is the drag coefficient, and \( D_{H_2O}^m \) is the water self-diffusion coefficient. The detailed expressions for them are given in Appendix A. The proton concentration \( c_{H^+}^m \) is considered constant due to negligible changes.

**Quasi-1D channel model**

The governing equations for the gas channel flow are developed from the mass and momentum conservation laws. The variables in the flow field change only in the flow direction (stream-wise). Taking a general one-dimensional control cell with volume \( dv \), inlet area \( A_i \), outlet area \( A_o \), and wall area \( A_{wall} \), and taking \( Q \) as the vector of dependent variables, \( F \) as the fluxes through the flow-area \( A_i \) and \( A_o \), \( S \) as the source-terms, i.e., the fluxes due to boundary and interface conditions, then the governing equations for the multi-component, non-reactive gas mixture in an integral form can be written as

\[
\frac{d}{dt} \int_{dv} [Q dv + F A_i - F A_o] = S, \tag{19}
\]

where the vectors \( Q \), \( F \) and \( S \) are listed as:

\[
Q = \begin{bmatrix} \rho Y_k \\ \rho \\ \rho u \end{bmatrix}, \quad F = \begin{bmatrix} \rho Y_k u - \rho D_k \nabla Y_k \\ \rho u \\ \rho u^2 + p - \mu \nabla u \end{bmatrix}, \quad S = \begin{bmatrix} \sum_{k=1}^{k=NS} \tilde{m}_k dA \ (k \neq 1, NS - 1) \\ \sum_{k=1}^{k=NS} \tilde{m}_k dA \ (k = 1, NS - 1) \\ f_{wall} \end{bmatrix}, \tag{20}
\]

In Eq. [20], the effects of the species mass transport and the channel viscous friction are taken into account. Since the velocity at the porous channel wall is very small, the effect of mass transfer through the channel wall on the momentum balance is neglected.

Boundary conditions for the bipolar solid wall and the porous GDL interface are implemented. For the bipolar solid wall, a non-slip boundary condition is employed, and no species mass transport is allowed.

The viscous friction is assumed in the flow direction and given by
where the friction coefficient $C_f$ is associated with the friction factor $f$ through the relation:

$$C_f = \frac{f}{4}. \tag{22}$$

The flow inside the gas channel is assumed to be fully developed laminar flow, and thus the friction factor is independent of the relative roughness of the wall, and only depends on the flow Reynolds number. Then, the friction factor is given as (15)

$$f = \frac{64}{Re}, \quad \text{for} \ Re \leq 2300, \tag{23}$$

and the geometry factor $\phi$ is 1.16, fitted from the empirical data corresponding to that of a channel with square cross-section (15). The Reynolds number $Re$ is calculated as:

$$Re = \frac{\rho u D}{\mu}, \tag{24}$$

where the wall area $A_{wall}$ is computed as:

$$A_{wall} = W_p ds. \tag{25}$$

The wetted perimeter $W_p$ is computed through the channel diameter $D$ or the hydraulic diameter (for non-circular cross-section tubes) and the channel cross-section area $A$ as

$$\frac{D}{4} = \frac{A}{W_p}. \tag{26}$$

There exist additional pressure drops at the channel corner due to the flow momentum change and they have been taken into account by the convection and pressure gradient terms in the momentum equation.

**Boundary conditions and interface conditions**

The inlet values of velocity, temperature and species mass fractions at the anode and the cathode inlet are specified. The inlet velocity is a function of the desired current density and the stoichiometric flow ratio for a given fuel cell geometry:

$$u_{in} = \xi \frac{i_{in}}{nF} \frac{RT_{in}}{x_{in}P_{in}} \frac{A_{in}}{A_{ch}} \tag{27}$$
where \( \zeta \) is the stoichiometric flow ratio, \( n = 4 \) for oxygen at the cathode side and \( n = 2 \) for hydrogen at the anode side. In the above equation, \( x_m \) is the molar fraction of oxygen and hydrogen at inlets and \( p_m \) is the inlet static pressure, and \( A_m \) and \( A_{ch} \) are the membrane active area and the gas channel inlet area, respectively.

At the outlets of the gas channel, only the backpressure is specified, and an extrapolated boundary condition is employed for other variables. No-slip boundary condition is specified at all external surfaces.

The interface conditions between the GDL and the gas channel are implemented as the following. The GDL side provides the mass flux, including both the mass convection and diffusion through the interface between the GDL and the gas channel. On the other hand, the gas channel provides to the GDL the values of the dependent variables including the pressure and species concentrations at the interface. The viscous friction at the interface is calculated the same way as the solid wall because the interface velocity is very close to zero and much smaller than the mean channel velocity. Thus, the gas channel governing equations can be readily solved with both mass and momentum flux available.

The interface conditions between the GDL and the membrane layer are implemented as the following. For the GDL, the \( k \)th species mass flux \( f_k \) including both the mass convection and diffusion through the interface is given as:

\[
(\rho u Y_k - D_{eff} \nabla Y_k) |_{GDL} = f_k,
\]

At the anode side, the hydrogen consumption flux based on the local current density is given as:

\[
f_{H_2} = -\frac{M_{H_2}}{2F} \cdot i_a,
\]

where \( i_a \) is the anode catalyst local current density, which is given by the membrane layer by solving the potential equation after the anode and cathode activation overpotentials were given. Water flux at the interface is given as Eq. [18].

At the cathode, oxygen flux due to reaction is given as:

\[
f_{O_2} = -\frac{M_{O_2}}{4F} \cdot i_c,
\]

where the local current density \( i_c \) is calculated through the cathode agglomerate model. Besides the water transfer due to electro-osmotic drag, water created due to electrochemical reaction at the catalyst layer is given as:

\[
S_{H_2O} = \frac{M_{H_2O}}{2F} \cdot i_c.
\]
The total water flux at cathode catalyst layer interface is then given as:

\[ f_{H_2O} = S_{H_2O} + N_{H_2O}^w. \]  \[32\]

The Eqs. [8-10] are solved for the species mass fractions and pressure after obtaining the species mass flux and the total mass flux. The GDL governing equations can be readily solved with given flux and values of dependent variables at the interface boundary.

It is assumed that the water in the GDL is in equilibrium with that in the membrane layer at the interface, which implies that the water activity is same for both membrane and GDL at the interface. The water activity at GDL is calculated as:

\[ a = \frac{RT}{P_{sat}(T)} c_{H_2O}^g \]  \[33\]

where \( P_{sat} \) is the saturation pressure of the GDL, and \( c_{H_2O}^g \) is the water vapor concentration given by GDL side.

The membrane potential equation is solved by obtaining the potential flux (current density) at the cathode side together with the potential value at the anode side. The cathode current density is calculated through the agglomerate model by obtaining cathode activation overpotential, while the potential at the anode side was set as the anode activation overpotential. In many previous studies, the activation overpotentials of anode and cathode are specified as constants. But in this paper, they are computed and updated iteratively, which results in more accurate prediction of the local current density.

First of all, the reversible potential is calculated as:

\[ E_{rev} = 1.23 - 0.9 \times 10^{-3} (T - 298.15) + \frac{RT}{2F} \left( \ln P_{H_2} + \frac{1}{2} \ln P_{O_2} \right), \]  \[34\]

where \( P_{H_2} \) and \( P_{O_2} \) are the partial pressure of hydrogen at the anode inlet and the partial pressure of oxygen at the cathode inlet. After the initialization of the cathode activation overpotential, an initial cathode current density is obtained. Further, the anode activation overpotential is updated according to Eq. [7]. The membrane potential equation is solved to obtain the membrane ohmic overpotential \( \eta_{ohm} \), and finally the cathode overpotential is updated as:

\[ \eta_c = E_{cell} - E_{rev} - \eta_a - \eta_{ohm} \]  \[35\]

where \( E_{cell} \) is the specified cell potential.

This algorithm is capable of predicting the cell current density based on a desired cell potential and stoichiometric flow ratio. The data flowchart among the related modules is shown in Figure 1.
RESULTS AND ANALYSIS

A multi-channel serpentine flow pattern is used for both the cathode and anode sides with co-flow arrangement. Figure 2 shows the gas channel grid, generated using the parametric grid generation code developed in house. The channel width, height and number of inlet and channel turns are used as parameters for the specific grid generations, which can facilitate the simulation-aided channel design optimizations. The grid size for the cathode GDL, the membrane layer and the anode GDL are 10x100x100. The active area of the fuel cell is set as 10 cm x 10 cm and the width and height of flow channels are 1 mm, with total of 10 parallel serpentine channels in parallel. The physical parameters of GDL and membrane layer (Nafion 115) used in this simulation are listed in Table III and Table IV, respectively. The simulations were conducted for different operating pressures,
different cathode air stoichiometric flow ratios and different cathode gas channel inlet humidity, while the anode gas channel inlet was fixed at 100% relative humidity. The operating conditions are listed in Table V.

Table III. Physical parameters of GDL

| Symbol | Parameter         | Values       |
|--------|-------------------|--------------|
| ε      | GDL porosity      | 0.5          |
| K      | GDL permeability  | $1.0 \times 10^{-11} \text{m}^2$ |
| δ      | GDL Thickness     | 0.25 mm      |

Table IV. Physical parameters of membrane layer

| Symbol | Parameter                        | Values                   |
|--------|----------------------------------|--------------------------|
| $\varepsilon_m$ | Membrane porosity | 0.5                      |
| $K_m$  | Membrane permeability           | $1.8 \times 10^{-18} \text{m}^2$ |
| $\mu_m$ | Effective viscosity             | 8.91 kg m$^{-1}$ s$^{-1}$ |
| $\rho_{dry}$ | Dry membrane density         | 2000 kg m$^{-3}$         |
| $M_m$  | Membrane molecular mass         | 1100 kg kmol$^{-1}$      |
| $c_p$  | Proton concentration            | 1.2 kmol m$^{-3}$        |
| $\delta_m$ | Membrane thickness             | 0.127 mm                 |

Table V. Fuel cell operating conditions

| Parameters                  | Values                  |
|-----------------------------|-------------------------|
| Cathode air inlet pressure  | 1, 1.5, 2 atm           |
| Anode inlet pressure        | 1, 1.5, 2 atm           |
| Cathode air inlet humidity  | 50%, 100%               |
| Anode inlet humidity        | 100%                    |
| Air stoichiometric ratio    | 2, 5, 10                |
| Hydrogen stoichiometric ratio | 2                     |
| Cathode air inlet temperature | 353K                  |
| Anode inlet temperature     | 353K                    |
Figures 3-6 show the results of the case with both anode and cathode gas channel inlet pressure at 1.0 atm, and 100% relative humidity. Both the air and hydrogen stoichiometric flow ratios are 2 and the cell potential is 0.5 V. Figure 3 depicts the current density, activation overpotential and species mass fraction distribution at the interface between the cathode catalyst layer and the GDL. It can be seen that there is a strong correlation between the oxygen concentration and current distributions, shown in Figure 3(a, c). Due to the production of water at the cathode catalyst layer and electro-osmotic water flux from the anode side, the water concentration gradually increases along the channel downstream direction, as shown in Figure 3(d). From Eq. [35], since the anode catalyst activation overpotential maintains a small value compared to the cathode catalyst activation overpotential, and the magnitude of membrane resistance is basically proportional to the current density, the magnitude of cathode catalyst activation overpotential basically increases as the current density decreases along the channel downstream direction. The variation percentage based on the average overpotential changes from -3.27% to +5.63% in this case. As we all know that the cathode overpotential exponentially affect the current generation, the variation of cathode activation overpotential thus cannot be neglected in order to accurately predict the current density.

Figure 4(a) shows the cathode gas channel pressure distribution. By employing multi-channel serpentine flow patterns compared to a single serpentine channel, the pressure drop can be decreased due to the reduced flow rate per flow channel. Figures 4(b, c) show the oxygen mass fractions and water mass fraction, respectively. As expected, it shows a gradual depletion of oxygen mass fraction along the channel downstream direction. Water is not only chemically produced at the cathode catalyst layer, but also transported from membrane due to the electro-osmotic drag. Thereafter, water flows through the
GDL into the gas channels, and finally exits at the outlet. Consequently, water concentration gradually increases along the channel downstream direction.

Figure 3. Distributions at the cathode side catalyst layer GDL interface with cell potential specified at 0.5 V: (a) current density; (b) cathode activation overpotential; (c) oxygen mass fraction; and (d) water mass fraction.

Figure 5 shows the current density, activation overpotential and species mass fraction distribution at the interface between anode catalyst layer and GDL. As it was known, the effect of electro-osmotic drag from the anode side to the cathode reduces the water content at anode side, while the back diffusion from the cathode to the anode side mitigates this effect, and the net water flux is a sum of the overall effects of electro-osmotic drag, back diffusion and the convection due to pressure gradient. At the beginning, the hydrogen is largely consumed correlating to a large current density, and the water begins to be discharged shown in Figure 5(c, d). Here, the water transport is dominated by the effect of electro-osmotic drag, which results in a reduced water fraction, and consequently, the hydrogen returns to a higher level, because the summation of water and hydrogen mass fraction must be unity. As the current density decreases, the back diffusion is sufficient to counteract the subsided drag, and the water fraction recovers, and the hydrogen fraction decreases. From the Butler-Volmer equation, the activation overpotential increases as the current density reduces, so that there is a strong correlation between the anode catalyst activation overpotential and current distributions, as shown in Figures 5(a, b). But the values of the anode overpotential are much smaller than those of
the cathode side. Figure 6 shows the anode gas channel pressure and species mass fraction distribution. Compared to the cathode gas channel, the pressure drop is lower resulted from the facts that the gas density is smaller at the anode side and the inlet velocity is much lower than the cathode side. As expected, the species mass fraction distributions are similar to those at the catalyst layer GDL interface.

Figure 4. 3D distributions inside the cathode side gas channel with the cell potential specified at 0.5 V: (a) pressure; (b) oxygen mass fraction; (c) water mass fraction.
Figure 5. Distributions at the anode side catalyst layer GDL interface with cell potential specified at 0.5 V: (a) current density; (b) anode activation overpotential; (c) hydrogen mass fraction; (d) water mass fraction.
The most widely used fuel cell performance evaluation criterion is the polarization curve or I-V curve. Figure 7 shows the simulated polarization curves and various overpotentials under the operating conditions described in Table V. Changes in gas channel inlet pressure or air humidity can result in the changes of inlet gas compositions. Since the saturation pressure for water is only a function of temperature, and remains constant for different inlet pressure, changes in gas channel inlet pressure or humidity result in water and oxygen molar fraction changes. A higher inlet pressure at cathode side leads to a higher oxygen fraction with same inlet humidity, while a lower humidity leads to a higher oxygen fraction with same inlet pressure. At the anode side, the inlet hydrogen fraction increases with inlet pressure increase when the inlet humidity remains same. It can be seen from Figure 7(a) that the higher cathode gas inlet pressure leads to a higher maximum average current density. This increase is significant when the pressure is increased from 1 atm up to 2 atm. Due to the thinness of the catalyst layer and high gas
phase oxygen diffusivity, the oxygen saturation concentration inside the Nafion solution is set as the gas phase oxygen concentration, and the increase in cathode gas inlet pressure can enhance the chemical reaction in the cathode catalyst layer. Shown in Figure 7(c), the magnitude of cathode activation overpotential is decreased when cathode gas inlet pressure is increased. But the benefit from increase in anode gas inlet pressure is limited, as shown in Figure 7(b). Shown in Figure 7(d), the magnitude of anode activation overpotential, though trivial, decreases as inlet pressure increases because the inlet hydrogen fraction increases. However, the membrane resistance increases greatly as the anode inlet pressure increases at larger current density, as shown in Figure 7(e). It is believed that a higher cathode inlet pressure together with a lower anode inlet pressure can facilitate the water transport from the cathode to the anode and thus potentially avoid flooding. Though the two-phase effect is not included in the current study, some insight can be achieved by examining the gas phase water distributions.

In the following, the water distributions at the anode catalyst interface, the cathode catalyst interface and inside the membrane are compared. Figures 8(a, b, c) show the results for the case with the operating condition: anode inlet pressure 1 atm, hydrogen stoichiometric flow ratio 2, relative humidity 100%; cathode inlet pressure 2 atm, relative humidity 50%.
hydrogen stoichiometric flow ratio 2, relative humidity 100%. Figures 8(d, e, f) show the results for the case with only a higher anode inlet pressure 2 atm. It can be seen that a higher water mass fraction is obtained for a lower anode inlet pressure at both the anode and the cathode catalyst interfaces, and a higher liquid water content is obtained in membrane layer as well. Shown in Figure 9, though a lower cathode air relative humidity of 50% is employed, the same results are obtained. It can be concluded that a lower anode inlet pressure together with a higher cathode inlet pressure can help sustain a wet membrane layer.

Figure 8. Water distributions with the cell potential set as 0.5 V, and both cathode air and anode hydrogen relative humidity set as 100%: (a-c) is for the operating condition: $p_a = 1$ atm, $p_c = 2$ atm; (d-e) is for the operating condition: $p_a = 2$ atm, and $p_c = 2$ atm. (a, d) water mass fraction at the anode catalyst interface; (b, e) water mass fraction at the cathode catalyst interface; and (c, f) water content inside the membrane.
Figure 9. Water distributions with the cell potential set as 0.5 V, and cathode air relative humidity set as 50%, anode hydrogen relative humidity set as 100%: (a-c) is for the operating condition: $p_a = 1 \text{ atm}$, $p_c = 2 \text{ atm}$; (d-e) is for the operating condition: $p_a = 2 \text{ atm}$, and $p_c = 2 \text{ atm}$. (a, d) water mass fraction at the anode catalyst interface; (b, e) water mass fraction at the cathode catalyst interface; and (c, f) water content inside the membrane.
Figure 10. Comparison of different cathode air relative humidity. Hydrogen stoichiometric flow ratio set as 2 for all cases, air stoichiometric flow ratio set as 10 for (a, c, e); and 2 for (b, d, f). (a, b) Polarization curves; (c, d) cathode activation overpotential; and (e, f) membrane ohmic overpotential.

Figure 10 shows the polarization curves, cathode activation overpotential and membrane resistances for different cathode air humidity and stoichiometric flow ratios. Since the decrease in the inlet air humidity increases the oxygen mass fraction, and thus it has the same effect as the increase in inlet air pressure. As shown in Figures 10(c, d), the magnitude of cathode activation overpotential decreases as the cathode air humidity
decreases. However, a higher air humidity input can help sustain a wet membrane through back diffusion, and reduce the membrane resistance. As shown in Figure 10(e, f), the magnitude of membrane resistance decreases as the cathode air humidity increases. Particularly, at low current density, higher cathode air humidity results in a better performance. However, at large current density, the water produced at the cathode catalyst layer will dilute oxygen as the air humidity increases, and this can be aggravated by the water dragged from the anode side, and therefore, low cathode air humidity exhibits better performance, as shown in Figure 10(b). This trend is valid as long as the fuel cell is not starved of oxygen. Nevertheless, a higher stoichiometric flow ratio entails a higher cost, and has to be considered carefully when designing the fuel cell system. For lower stoichiometric flow ratio case, at lower current density, increase in air humidity can decrease the membrane resistance. When further increase the air humidity, the membrane resistance no longer decreases, and is eventually counteracted by the effect of oxygen dilution. At higher current density, lower inlet air humidity with higher inlet oxygen mass fraction yields better performance, as shown in Figure 10(a).

CONCLUSION

The multi-resolution fuel cell simulation approach is employed in this study. And a new algorithm for coupling local current density and activation overpotential has been developed, which allows more accurate prediction of the membrane layer resistance. An industrial size of fuel cell has been simulated with different operating pressure and cathode air humidity. It can be concluded from the simulated results, that increase in the cathode air inlet pressure can increase the average current density; but the benefit from increase in anode fuel inlet pressure is trivial. The increase in the cathode inlet relative humidity has an adverse effect on the fuel cells performance at large current densities but a favorable effect at small current density as long as oxygen is sufficient. However, for lower air stoichiometric flow ratio case, at lower current densities, increase in the air humidity can increase cell performance, but further increase of air humidity will induce an adverse affect; at higher current densities, lower inlet air humidity with higher inlet oxygen mass fraction produces a favorable effect on cell performance.

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APPENDIX A

This appendix details the equations for water drag coefficient, water self-diffusion coefficient, water activity and conductivity inside Nafion membrane. Water drag coefficient is given as (1):
The water content $\lambda_{\text{H}_2\text{O}}$ is calculated as:

$$ n_s = 2.5 \frac{\lambda_{\text{H}_2\text{O}}}{\text{SO}_3} = 22. \tag{A.1} $$

$$ \lambda_{\text{H}_2\text{O}} = \frac{c_{\text{H}_2\text{O}}^m}{\rho_{\text{dry}}^m - 0.0126c_{\text{H}_2\text{O}}^m} \tag{A.2} $$

where $\rho_{\text{dry}}^m$ is the dry membrane density, and $M^m$ is the membrane molecular mass. Water self-diffusion coefficient is given in reference (1) as:

$$ D_{\text{H}_2\text{O}} = D'\left\{\exp\left[2416\left(\frac{1}{303} - \frac{1}{T}\right)\lambda_{\text{H}_2\text{O}}\right]^{1.781 - 78.9a + 108a^2}\right\} \tag{A.3} $$

where $a$ is the water activity and $D'$ is the diffusion coefficient measured at constant temperature (30°C) and in coordinates moving with the swelling of the membrane. $D'$ is written as:

$$ D' = \begin{cases} 
2.642276 \times 10^{-13} \frac{\lambda_{\text{H}_2\text{O}}}{\text{SO}_3} & \lambda_{\text{H}_2\text{O}} \leq 1.23 \\
7.75 \times 10^{-11} \frac{\lambda_{\text{H}_2\text{O}}}{\text{SO}_3} - 9.5 \times 10^{-11} & 1.23 \leq \lambda_{\text{H}_2\text{O}} \leq 6 \\
2.5625 \times 10^{-11} \frac{\lambda_{\text{H}_2\text{O}}}{\text{SO}_3} + 2.1625 \times 10^{-10} & 6 \leq \lambda_{\text{H}_2\text{O}} \leq 14
\end{cases} \tag{A.4} $$

And water activity within membrane is given by:

$$ a = \frac{1/2160b - 134183/2160}{b + 797/2160} \quad c_1 = -41956e4 \\
+ 797/2160 \quad c_2 = 139968e3 \\
\frac{b}{c_3} = 382482e7 \\
+ 216(c_1-c_2\lambda_{\text{H}_2\text{O}}/\text{SO}_3) \quad c_4 = 251739e7 \quad \lambda_{\text{H}_2\text{O}}/\text{SO}_3 \leq 14 \\
+ c_4\lambda_{\text{H}_2\text{O}}/\text{SO}_3^{3/2} \quad c_5 = 419904e6 \quad 14 \leq \lambda_{\text{H}_2\text{O}}/\text{SO}_3 \leq 16.8 \\
+ 0.7143\lambda_{\text{H}_2\text{O}}/\text{SO}_3 - 9.0021 \quad 16.8 \leq \lambda_{\text{H}_2\text{O}}/\text{SO}_3 
\quad b = (c_1 + c_2\lambda_{\text{H}_2\text{O}}/\text{SO}_3) \quad \lambda_{\text{H}_2\text{O}}/\text{SO}_3 \leq 14 \\
+ 216(c_1-c_2\lambda_{\text{H}_2\text{O}}/\text{SO}_3) \quad 14 \leq \lambda_{\text{H}_2\text{O}}/\text{SO}_3 \leq 16.8 \\
+ c_4\lambda_{\text{H}_2\text{O}}/\text{SO}_3^{3/2} \quad 16.8 \leq \lambda_{\text{H}_2\text{O}}/\text{SO}_3 \\
+ c_5 \quad \lambda_{\text{H}_2\text{O}}/\text{SO}_3
\tag{A.5} \quad c_3 = -3.2 \times 10^{-3}
\]$$

Membrane conductivity $\sigma_m$ is given by:

$$ \sigma_m = \sigma_{m303} \exp\left[1268\left(\frac{1}{303} - \frac{1}{T}\right)\right] \tag{A.6} $$

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The conductivity of the membrane at 303K, $\sigma_{303}$, is expressed by the following correlation as a function of water content:

$$\sigma_{303} = 0.5139 \lambda_{H_2O} - 0.326, \quad [A.7]$$

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