Polymer electrolyte fuel cells (PEFCs) have been receiving a great deal of attention in the scientific research community recently due to their potential to reduce our petroleum-based energy use and pollutant emissions associated with petroleum-based energy generation. Phenomena involved in a PEFC are complex and encompass the fundamentals of heat transfer, species transport, multiphase flows, and electrochemical reactions. Multiphase flow originates from water production by the oxygen reduction reaction (ORR) in the cathode of a PEFC. When the local water vapor partial pressure reaches its saturation value, condensation occurs and liquid emerges, leading to the co-existence of gas and liquid and resultant two-phase flow (see Fig. 1). Liquid water must be efficiently removed from the reaction sites in the catalyst layer (CL) where the ORR takes place, through the gas diffusion layer (GDL), and eventually out of the flow channels of a PEFC in order to avoid its adverse impact on the operating PEFC, particularly the through-plane water profiles. High water diffusion in the vapor phase, as expected. Without strong water diffusion in the vapor phase, the liquid-water profiles are found to increase with current density in the cathode GDL but indicate a complex trend in the anode. Effect of varying GDL thermal conductivity on water distribution is also examined.

A multidimensional mathematical model is presented for simulating the coupled phenomena of gaseous fuel/reactant flows, species (including liquid water) transport, heat transfer, hydrogen oxidation, and oxygen reduction reactions in a polymer electrolyte fuel cell (PEFC). The present work focuses on elucidating water distribution in the through-plane direction, in particular across the membrane electrode assembly (MEA) and gas diffusion layer (GDL). Two-dimensional model predictions are computed numerically and compared with available experimental data from neutron radiography or imaging. Using the same set of model parameters, reasonably good agreements are obtained quantitatively between the computed water profile in the MEA–GDL component and the neutron-imaging data reported by two separate research groups, and qualitatively between model prediction and the data from another (third) group. Case-study simulations are carried out for PEFC operation at various temperatures, relative humidities, and current densities. It is found that liquid-water content is lower at higher cell temperatures due to greater water evaporation and stronger water diffusion in the vapor phase, as expected. Without strong water diffusion in the vapor phase, the liquid-water profiles are found to increase with current density in the cathode GDL, but indicate a complex trend in the anode. Effect of varying GDL thermal conductivity on water distribution is also examined.

© 2010 The Electrochemical Society. [DOI: 10.1149/1.3498997] All rights reserved.

Manuscript submitted January 21, 2010; revised manuscript received September 14, 2010. Published October 27, 2010.
Water conservation: Weber and Hickner attributed the large discrepancy to their pseudo-Hickner et al.44 of Sandia National Laboratories and Mukundan and channel effects and incorporate a detailed MEA model. Model validation was performed by direct comparisons of the computed water through-plane profiles with the neutron-imaging data from Hickner et al.44 of Sandia National Laboratories and Mukundan and Borup45 and Borup46 of LANL. The validated model was then applied to elucidate the effects of current density, relative humidity (RH), and temperature on water distribution and PEFC performance. Effect of varying GDL thermal conductivity was also examined. It should be noted, this paper is not intended to focus on modeling new phenomena. Rather, we make one of the first attempts to address an important issue of comparing the predictions from a fuel cell model and the neutron imaging data in terms of water through-plane distribution.

Mathematical Model

Governing equations.— In the present paper, we consider the electrochemical and transport phenomena in the key components of a PEFC, namely, the MEA consisted of the catalyst layers and membrane, GDLs, gas flow channels, and bipolar plates. Two-phase transport is modeled in the regions of the CLs, membrane, and GDLs. Since the current mathematical treatment for the microporous layer (MPL) region, as demonstrated by Weber and Hickner’s model predictions, appears unable to describe the smooth transition. However, in the present work; rather, we account for the MPL effect by adding several source terms. The key aspects regarding the model parameters and source terms. Table I lists the expressions for the electrochemical and transport phenomena that are closely related to the topic of this paper are elaborated below.

<table>
<thead>
<tr>
<th>Surface term</th>
<th>S_a</th>
<th>S_cl</th>
<th>S_w</th>
<th>S_0</th>
<th>S_g</th>
<th>S_diff</th>
<th>S_geo</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bipolar plates</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Gas channels</td>
<td>- ∇  \rho</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>GDL</td>
<td>- \frac{\mu}{k_{Dl}}</td>
<td>j</td>
<td>0</td>
<td>-j</td>
<td>-j</td>
<td>-j</td>
<td>-j</td>
</tr>
<tr>
<td>Anode catalyst layer</td>
<td>- \frac{\mu}{k_{cl}}</td>
<td>j</td>
<td>0</td>
<td>-j</td>
<td>-j</td>
<td>-j</td>
<td>-j</td>
</tr>
<tr>
<td>Cathode catalyst layer</td>
<td>- \frac{\mu}{k_{cl}}</td>
<td>j</td>
<td>0</td>
<td>-j</td>
<td>-j</td>
<td>-j</td>
<td>-j</td>
</tr>
<tr>
<td>Membrane</td>
<td>-</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Charge conservation (protons): \[ 0 = \nabla \cdot (\sigma_{\text{eff}} \nabla \Phi_{\text{m}}) + S_{\text{q(m)}} \]  
[6]

Charge conservation (electrons): \[ 0 = \nabla \cdot (\sigma_{\text{eff}} \nabla \Phi_{\text{e}}) + S_{\text{q(e)}} \]  
[7]

where \( \rho \) the multiphase mixture density, \( \sigma \) the superficial fluid velocity vector, \( p \) the pressure, \( C_{i}/C_{w} \) the molar concentration of reactant/water, \( T \) the temperature, \( \Phi_{\text{m}}/\Phi_{\text{e}} \) the electronic/electrolyte phase potentials. \( S_{\text{diff}} \) includes the diffusion fluxes in gaseous, liquid, and solid electrolyte phases. \( S_{\text{geo}} \) represents the hydraulic permeation water flux through the membrane. The electrochemical and transport processes are coupled together through the model parameters and source terms. Table I lists the expressions for several sources terms. The Butler–Volmer equation and source terms can be found in Ref. 24 and 27. The key aspects regarding the model and electrochemical/transport phenomena that are closely related to the topic of this paper are elaborated below.

Electrochemical kinetics.— The electrochemical reactions take place within the catalyst layers as follows

HOR in the anode: \( H_2 \rightarrow 2H^+ + 2e^- \)  
[8]

ORR in the cathode: \( O_2 + 4e^- + 4H^+ \rightarrow 2H_2O \)  
[9]

The Butler–Volmer equation is customarily adopted to describe the electrochemical reaction rate

\[ j = a_i \left\{ \exp \left( \frac{\alpha_i}{RT} F \cdot \eta \right) - \exp \left( - \frac{\alpha_i}{RT} F \cdot \eta \right) \right\} \]  
[10]

where the surface-to-volume ratio \( \alpha \) evaluate the roughness of porous electrodes. The presence of liquid water in the catalyst layer may reduce the reaction area as follows

\[ \alpha = (1 - \delta)\alpha_{0} \]  
[11]

where the liquid-water saturation \( s \) is defined as the volume fraction of liquid water in the void space.

In PEFCs, HOR is fast, thus yielding a relatively small anode overpotential. Therefore, Eq. 10 for the anode can be adequately simplified to a linear kinetic equation. For ORR, sluggish kinetics results in a large cathode overpotential. Consequently, the Butler–Volmer equation can be approximated by the Tafel kinetics. In addition, the reaction rate of the ORR highly depends on temperature as described by the Arrhenius expression. The HOR and ORR kinetic rate expressions are thus simplified to the following

anode: \[ j_a = a_i^{\text{ref}} \left( \frac{C_{\text{H}_2}}{C_{\text{H}_2}^{\text{ref}}} \right)^{1/2} \left( \frac{\alpha_i + \alpha_{0}}{RT} \cdot F \cdot \eta \right) \]  
[12]
cathode: \( \dot{j}_c = -a \dot{k}_{\text{ref}} \exp \left[ -\frac{E_a}{R} \left( \frac{1}{T} - \frac{1}{353.15} \right) \right] \left( \frac{C_{O_2}}{C_{\text{sat}}} \right) \times \exp \left( -\frac{\alpha F}{RT} \cdot \eta \right) \) \[13\]

where \( E_a \) denotes the activation energy for ORR at the Pt/Nafion electrode and the surface overpotential is defined as

\[ \eta = \Phi^{(0)} - \Phi^{(m)} - U_o \] \[14\]

The equilibrium potential \( U_o \) is a function of temperature.\(^{1,24,27}\)

**Two-phase transport in porous media.**— Two-phase transport in PEFCs originates from water production by the ORR. In a PEFC, both the GDL and catalyst layer are highly porous media, allowing heat and species transport. Liquid may emerge in the pore when the water vapor partial pressure reaches its saturation pressure, resulting in two-phase flow. The two-phase mixture density is defined as\(^{49}\)

\[ \rho = s \rho^{(0)} + (1-s) \rho^{(e)} \] \[15\]

The saturation \( s \) can be obtained from the mixture water concentration \( C_w \)

\[ s = \begin{cases} 0, & C_w < C_{\text{sat}} \\ \frac{C_w - C_{\text{sat}}}{\rho^{(0)} M_w - C_{\text{sat}}}, & C_w > C_{\text{sat}} \end{cases} \] \[16\]

One major interaction between the two-phase flows is described through the relative permeabilities \( k^{(0)} \) and \( k^{(e)} \), defined as the ratio of the intrinsic permeability of liquid and gas phases, respectively, to the total intrinsic permeability of a porous medium. Physically, these parameters describe the extent to which one fluid is hindered by others in pore spaces, and hence can be formulated as a function of liquid saturation. One formula for the relative permeabilities is as follows

\[ k_s^{(0)} = s^3 \quad \text{and} \quad k_s^{(e)} = (1-s)^3 \] \[17\]

In addition, residual liquid may be present as observed by Hickner et al.,\(^{44}\) who probed the through-plane distribution of residual water at vanishing current. Modeling residual liquid impact has been attempted by several studies.\(^{39,40,52}\) In the present work, we exclude the residual water due to the following two considerations: (1) there are no accurate/validated models on residual water for the carbon-fiber-based GDL in PEFCs; (2) the data of in situ residual water are not available for cases with various currents which will be used for model validation. Without the direct data from the in situ experiment, adding residual water (and its spatial distribution) becomes arbitrary and may be unsuitable to be used for elucidating the water profile in an operating fuel cell.

**Fuel, oxidant, and water transport.**— In the gaseous phase, hydrogen, oxygen, and water follow similar transport mechanisms. Modeling their transport can be found in our previous paper,\(^{7}\) therefore not repeated here. To account for the liquid impact, we modify the effective diffusivity by

\[ D^{(e)} = D^{(e)} \left[ \sigma (1-s)^3 \right] \] \[18\]

The convection corrector factor \( \gamma \) is a function of \( s \)

\[ \gamma_s = \frac{s \rho^{(e)}}{\rho^{(0)} (1-s)} \quad \text{and} \quad \gamma_v = \frac{\rho^{(e)} \Lambda^{(e)} C_{\text{sat}}}{M_w} \] \[19\]

where \( \Lambda^{(e)} \) is the relative mobilities of individual phases. The capillary pressure \( P_c \), which may be the primarily driving force for liquid-water transport, is given as follows

\[ P_c^{(e)} - P_c^{(0)} = \sigma \cos(\theta) \left( \frac{\rho^{(e)}}{K} \right)^{1/2} J(s) \] \[20\]

where \( \sigma \) is the surface tension. \( J(s) \) for the hydrophobic diffusion media considered in the present work is given by\(^{7}\)

**Figure 1.** (Color online) The schematic of water in a PEFC.

\[ J(s) = 1.417 s - 2.120 s^2 + 1.263 s^3 \] \[21\]

Once the capillary pressure is calculated, the flux \( j^{(0)} \) in the water equation of Eq. 5 can be obtained through

\[ j^{(0)} = \frac{\lambda^{(0)} \Phi^{(0)}}{v} \left[ \right] \] \[22\]

**Water transport in the Nafion membrane.**— Water in the Nafion membrane is bonded with the sulfonic acid groups in the ionomer. The water content \( \lambda \) is typically adopted to characterize the membrane hydration level and defined as the number of water molecules per sulfonic group. It is related to the equivalent water concentration through the following equation

\[ C_{w}^{(m)} = \alpha \frac{\rho^{(m)} \lambda}{EW} \] \[23\]

where \( \rho^{(m)} \) and \( EW \) are the density and equivalent molecular weight of the membrane, respectively. In the above equation, we use \( \alpha \) to account for several factors that may affect the water content within the membrane, such as membrane swelling due to hydration by water (which reduces the membrane density), compression due to load or force applied at the external wall surfaces (which results in less water than one would expect theoretically), and incomplete disassociation of proton. Two processes may shape the membrane water content profile: one is the water electro-osmotic drag, the other is water back transport including diffusion and hydraulic permeation (see Fig. 1). The coefficient of the electro-osmotic drag \( n_d \) depends on the local water content\(^{54}\)

\[ n_d = \begin{cases} 1.0 & \text{for } \lambda \leq 14 \\ \frac{1.5}{8} (\lambda - 14) + 1.0 & \text{otherwise} \end{cases} \] \[24\]

The water diffusion coefficient in the membrane is also a function of water content and the diffusive flux is given by

\[ G_w^{(m) diff} = -D_w^{(m)} \nabla C_{w}^{(m)} \] \[25\]

where

\[ D_w^{(m)} = \begin{cases} 3.1 \times 10^{-3} \lambda (e^{0.28\lambda} - 1) \cdot e^{[-2436/T]} & \text{for } 0 < \lambda \leq 3 \\ 4.17 \times 10^{-3} \lambda (1 + 161e^{-\lambda}) \cdot e^{[-2436/T]} & \text{otherwise} \end{cases} \] \[26\]
As to the catalyst layers or Gore™ membranes, the diffusivity in the portion of the ionomer is modified by

$$D_{m,eff}^{(i)} = \alpha_{m}^i D_{m}^{(i)}$$  \[27\]

Assuming local microscopic thermodynamic equilibrium between ionomer and surrounding fluid, one can combine the diffusive transport in the void space and ionomer by defining an effective diffusion coefficient

$$D_{m,eff}^{(i)} = e_{m}^i D_{m}^{(i)} + e_{m}^i \rho_{m}^{(i)}RT \frac{d\lambda}{d\lambda_{m}}$$  \[28\]

where $\rho_{m}^{(i)}$ is the density of a dry membrane and the membrane water content is calculated following Spring et al., Luo et al., and Wang\textsuperscript{27}

$$\lambda = \begin{cases} 
0.043 + 17.81a - 39.85a^2 + 36.0a^3 & \text{for } 0 < a \leq 1 \\
14 + 8a & \text{for } 0 < s \leq 1 
\end{cases}$$

$$\alpha_m = \frac{C_s}{C_{sat}} \text{ and } C_{sat}(T) = \frac{P_{sat}(T)}{RT},$$

$$\log_{10} P_{sat} = -2.1794 + 0.02953 \times 10^{-5} (T - 273.15) - 9.1837 \times 10^{-3} (T - 273.15)^3$$  \[29\]

In addition to diffusion, hydraulic permeation may be another mechanism for water transport in the membrane. The permeation flux is determined by the membrane permeability $K_m$ and liquid pressures\textsuperscript{27}

$$\dot{G}_{m,perm} = -\frac{K_m}{M_w} \frac{\partial P}{\partial l}$$  \[30\]

Here, we assume liquid-water pressure in the membrane is linear in the through-plane direction, determined by the difference of $P$ at the membrane surface.

Heat transfer.— Fuel cell produces waste heat during its operation. The major heat generation mechanisms are the reversible/irreversible electrochemical processes, Ohmic resistance, and phase change. The source term $S_T$ in the energy equation can be generalized by

$$S_T = \int [\lambda (I + T) \frac{dU_{m}}{dT} + \rho_{m}^{(i)} \frac{d\rho_{m}^{(i)}}{dt} + \frac{\partial S_T}{\partial T} + S_{fl}]$$  \[31\]

where $S_{fl}$ represents the heat release/absorption due to water condensation/evaporation and the associated phase change rate can be calculated through the water two-phase transport equation. Note that some terms in Eq. 31 may vanish in some fuel cell components, e.g., in the anode GDL, there is no ionic currents, therefore the second term on the right disappears. In the bipolar plates, only the third term on the right is nonzero in the above thermal source.

Boundary conditions.— Equations 1-7 form a complete set of governing equations with eight unknowns: $d$ (with three components), $P$, $C_p$, $C_w$, $T$, $\Phi^{(i)}$, and $\Phi^{(s)}$. Their corresponding boundary conditions are briefly described as follows:

Flow inlet boundaries: The inlet velocity $\vec{u}_m$ in a gas channel is expressed by the respective anode or cathode stoichiometric flow ratio, i.e., $\xi_{\text{an}}$ or $\xi_{\text{ca}}$, defined at the average current density $I$ as

$$(\vec{u}_{\text{an,inlet}})/\vec{u}_{\text{ca,inlet}} = \frac{IA_{\text{an}}}{F} \left( \begin{array}{c} \xi_{\text{an}} \\ \frac{2C_{H2}A_{\text{an}}}{\xi_{\text{an}}} \\ \frac{4C_{O2}A_{\text{ca}}}{\xi_{\text{ca}}} \end{array} \right)$$  \[32\]

where $A_{\text{an}}$, $A_{\text{ca}}$, and $A_{\text{inlet}}$ are the flow cross-sectional areas of the anode and cathode gas channels and the membrane, respectively. The inlet molar concentrations are determined by the inlet pressure and humidity according to the ideal gas law.

Outlet boundaries.— Fully developed or no-flux conditions are applied.

Walls.— No-slip and impermeable velocity condition and no-flux condition are imposed for the mass, momentum, proton, and species conservation equations. The boundary conditions for the electronic potential $\Phi^{(i)}$ and temperature $T$ at the bipolar plate outer surfaces can be expressed as

$$\frac{\partial \Phi^{(i)}}{\partial n} = \frac{IA_{\text{cell}}}{\sigma A_{\text{cell}}},$$

$$\frac{\partial \Phi^{(s)}}{\partial n} = 0$$  \[33\]

where $A_{\text{cell}}$ is the area of the cathode outer surface. At other wall surfaces, symmetry conditions are applied for these two variables. Further details can be found in Ref. 27.

Numerical Procedures

The governing equations, Eq. 1-7, along with its boundary conditions are discretized by the finite volume method and solved in the commercial CFD software package, Fluent (version 6.0.12), by SIMPLE (semi-implicit pressure linked equation) algorithm. The SIMPLE algorithm updates the pressure and velocity fields from the solution of a pressure correction equation, solved by algebraic multigrid method. Following the solution of the flow field, the energy, species, proton, and electron equations are solved. The source terms and physical properties are implemented in a user-defined function and the species/charge transport equations are solved through the software’s user-defined scalars. The mesh of a single-channel 2D PEFC employed for the numerical study is shown in Fig. 2. Totally 50 gridpoints are adopted in the through-direction for the GDLs (both anode and cathode) to precisely capture the through-plane liquid transport. Twelve gridpoints are employed to capture transport in the fuel cell land-channel structure. The geometrical and operating parameters are listed in Table II. In all the simulations to be presented in the next section, the converged values of species imbalance (i.e., $H_2$, $O_2$, and $H_2O$) are all less than 1% and equation residuals are smaller than $10^{-6}$.

Results and Discussion

The water saturation and temperature distributions in both anode and cathode GDLs for three operating temperatures and the current density of 0.75 A/cm$^2$ are displayed in Fig. 3 and 4. From the water-content (in terms of liquid saturation) contours in Fig. 3, it can be seen that part of the cathode GDL is free of liquid water for the 80°C case, whereas it is fully wetted for the other two cases (60 and
This can be explained by the water-vapor-phase diffusion, which is stronger at a higher operating temperature and may be comparable with water production rate. This mechanism drives water vapor from the region under the channel to that under the land, thus enhancing the local saturation under the land at higher operating temperature. This explanation can also be applied to describe the trend in the anode: part of the GDL is in single-phase operation condition of 0.75 A and 0.65 V, which is due to the larger voltage loss arising from the irreversible process.

Figure 5 shows the distributions of the water content in the membrane for the three cases. It can be seen that the water-content contours in the 80°C case is quite different from those in the other two cases: the under-channel membrane region has a lower water content at 80°C. This can be explained with the aid of the water distribution in the anode in the previous figures where at 80°C a large under-channel area in the anode side is dry or free of liquid. For the 40 and 60°C cases, the contours are similar to each other.

In Fig. 6, the through-plane water profiles computed from the present 2D simulation are compared with the neutron-imaging data from Hickner et al. at three operating temperatures. Agreement between model prediction and experimental data is seen to be reasonably good. In the cathode, the liquid volume fraction drops from the catalyst layer to the channel side, indicative of the direction for

<table>
<thead>
<tr>
<th>Table II. Geometrical, physical, and operating parameters.</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Quantity</strong></td>
</tr>
<tr>
<td>Channel depth/width and land width</td>
</tr>
<tr>
<td>Catalyst layer/membrane thickness</td>
</tr>
<tr>
<td>Anode/cathode GDL thickness</td>
</tr>
<tr>
<td>Anode/cathode pressures, P</td>
</tr>
<tr>
<td>Porosity of GDLs/catalyst layers, ε</td>
</tr>
<tr>
<td>Humidification in the anode and cathode</td>
</tr>
<tr>
<td>Water concentration correction factor, α</td>
</tr>
<tr>
<td>Electronic conductivity of GDLs/bipolar plates, σ_eff</td>
</tr>
<tr>
<td>Viscosity of liquid water, µ</td>
</tr>
<tr>
<td>Permeability of GDL/membrane, K_GDL/K_m</td>
</tr>
<tr>
<td>Surface tension, liquid-water-air (80°C), σ</td>
</tr>
<tr>
<td>Thermal conductivity of the membrane/catalyst layer/GDL/bipolar plate</td>
</tr>
<tr>
<td>Contact angle of the GDL, θ_c</td>
</tr>
<tr>
<td>Exchange current density × reaction surface area, aᵢ₀aᵢ₋aᵢ₋ revered</td>
</tr>
<tr>
<td>Exchanger density × reaction surface area, aᵢ₀aᵢ₋aᵢ₋ revered</td>
</tr>
</tbody>
</table>

40°C. This can be explained by the water-vapor-phase diffusion, which is stronger at a higher operating temperature and may be comparable with water production rate. This mechanism drives water vapor from the region under the channel to that under the land, thus enhancing the local saturation under the land at higher operating temperature. This explanation can also be applied to describe the trend in the anode: part of the GDL is in single-phase operation condition of 0.75 A and 0.65 V, which is due to the larger voltage loss arising from the irreversible process.

Figure 5 shows the distributions of the water content in the membrane for the three cases. It can be seen that the water-content contours in the 80°C case is quite different from those in the other two cases: the under-channel membrane region has a lower water content at 80°C. This can be explained with the aid of the water distribution in the anode in the previous figures where at 80°C a large under-channel area in the anode side is dry or free of liquid. For the 40 and 60°C cases, the contours are similar to each other.

In Fig. 6, the through-plane water profiles computed from the present 2D simulation are compared with the neutron-imaging data from Hickner et al. at three operating temperatures. Agreement between model prediction and experimental data is seen to be reasonably good. In the cathode, the liquid volume fraction drops from the catalyst layer to the channel side, indicative of the direction for

![Figure 3](image_url)  **Figure 3.** (Color online) The contours of the liquid water saturation at the operating condition of 0.75 A/cm², RHa/c = 100/100%, and (a) 40°C and 0.59 V, (b) 60°C and 0.65 V, and (c) 80°C and 0.67 V.

![Figure 4](image_url)  **Figure 4.** (Color online) The contours of temperature at the operating condition of 0.75 A/cm², RHa/c = 100/100%, and (a) 40°C and 0.59 V, (b) 60°C and 0.65 V, and (c) 80°C and 0.67 V.
liquid drainage or water removal. The trends of water across the anode/cathode GDL and MEA can be readily explained by the detailed 2D water distributions shown in Fig. 3.

In order to provide a simple direct analysis on the through-plane water distribution in the cathode GDL, one can assume the capillary pressure as the only driving force for liquid movement. A 1D analysis can be performed to equate the capillary flux to the net liquid-water flux

\[
\frac{k^{(j)}}{v^{(j)}} \frac{dP}{dx} = \frac{(1 + 2\alpha)}{2F} \left[ 1 - G_{\text{vapor}} \right] M_w
\]

where \( G_{\text{vapor}} \) represents the portion taken away or added by the water-vapor diffusion. Furthermore, rewriting the above equation leads to

\[
- D \frac{ds}{dx} = \frac{(1 + 2\alpha)}{2F} \left[ 1 - G_{\text{vapor}} \right] M_w
\]

where \( D = -\frac{v}{\mu} \cos(\theta) (K_e) \sqrt{\sigma} (J) \frac{ds}{dx} \). The above simple analysis indicates that the capillary action is similar to a diffusion process, with the “diffusion coefficient” as a function of the liquid saturation \( s \). Note that a number of the factors, including current density \( I \) and water vapor-phase flux \( G_{\text{vapor}} \), may affect the profile of the through-plane water content. For the 80°C case, more water can be transported via the vapor phase, therefore \( G_{\text{vapor}} \) becomes larger (hence the right side becomes smaller) in the diffusive flux formula of Eq. 35, which may contribute to its lower cathode water profile observed in this figure.

In addition, similar analysis can be applied in the anode where the trend is slightly different with that in the cathode as shown in Fig. 6: at or near the anode channel/GDL interface (dimensionless position \( x \)), the liquid volume values for 60 and 80°C are about the same whereas that for 40°C is much higher. Figure 3 shows that a very small portion of anode GDL exhibits single phase at 40°C whereas a large portion for 60 and 80°C, which is the reason for the higher liquid volume for 40°C. However, the strong vapor phase diffusion in the anode at 80°C leads a high liquid saturation under the land as compared with 60°C, which may explain the comparable liquid volume values for these two temperatures.

In addition, protons may dissociate incompletely from the backbone, which can directly affect the water content in the membrane. Other factors such as membrane swelling, compression due to load, or force applied at the external wall surfaces may also affect water content in the membrane, as explained previously. In the present work, an \( \alpha \) value of 0.15 was used and this value renders a reasonably good or satisfactory agreement on the water profile between the model prediction and the experimental data. Weber and Hickner adopted a value of unity for \( \alpha \), which results in a much larger deviation ( > 300%) factor of 4 difference between their pseudo 2D (or 1+1D) prediction and experimental data. A value of \( \alpha \) lower than unity is physically more plausible. We also use the same \( \alpha \) value to compute water profile across the MEA-GDL and compare with the experimental data reported by Mukundan and Borup and Borup of LANL, and the comparison results are shown in Fig. 7. Here, we adopt the model geometry to match the LANL’s experimental setup (e.g., the N117 membrane and GDL). It can be seen that our model prediction agrees reasonably well with their data at two different current densities. In addition, the LANL data show a similar trend on water profile to that from Hickner et al., particularly in the membrane.

It is worthy to mention that application of neutron imaging in fuel cell is relatively new and the probed curves are also affected by several uncertainties such as geometric blur in neutron imaging, detector resolution, fuel cell misalignment, and other mishandlings. Geometric blur is determined by several factors such as the distances between detector and object, \( h \), and between detector and neutron source, \( L \), and the size of the neutron beam source, \( d \), (the geometric resolution = \( d \times h \); for an excellent discussion on this topic, see http://physics.nist.gov/MajResFac/NIF/geometric.html.) Misalignment might introduce a considerable error given the small thicknesses of the GDL and MEA, for an example, a misalignment of 1° (the angle \( \Theta \) between the neutron beam and fuel cell MEA plane) will introduce an error of \( \sim 350 \mu \text{m} \) (\( \sim \sin \Theta \times L_p \)) given that the projected length \( L_p \) of the fuel cell is 2 cm. By comparing with the experimental data from the two separate, reputable groups using one set of model parameters, we can minimize the effect of uncertainties involved in neutron imaging on our model validation.
In addition, despite the reasonable good agreement we also admit that there exists discrepancy between the model prediction and neutron imaging data (at some location the error is close to 20%), which is likely due to the above mentioned uncertainties such as the “geometric blur” and detector resolution, and exclusion of MPLs and anisotropic properties of GDLs/membrane in the model. Moreover, discontinuities of water profiles at the MEA–GDL interface are shown in the model prediction, but not in the neutron imaging curves. A water depth discontinuity likely exists in a real fuel cell as membrane water concentration is several orders of magnitude higher than the gas phase. One possible reason of the smooth curves in neutron imaging data is the “geometric blur,” which may “blur” or smooth the discontinuities. To approximate the effect of the “geometric blur,” we applied the Gaussian blurring (or smoothing) technique56,57 to the model prediction, as was done by Preston et al.38 In this smoothing technique, a Gaussian kernel \( g(x) = \frac{1}{\sqrt{2\pi\sigma^2}} e^{-\frac{x^2}{2\sigma^2}} \) is adopted to transform the value at a point (similar to a pixel in the neutron image) by the weighted average of itself and points in the neighborhood, i.e., \( F(x) = \int_{-\infty}^{\infty} f(x') g(x - x') \, dx' \), where \( \sigma \) is the standard deviation, \( x \) location, \( f(x) \) the curve prior to smoothing, and \( F(x) \) the smoothed curve. The smoothing process or detailed procedure adopted in the present work for a discrete set of points \( \{f_i\} \) in the model prediction is as follow:

1. Create a new set of data \( \{f_i\} \) at uniform gridpoints based on the linear interpolation of a model prediction;
2. Choose a Gaussian kernel (here we used a seven points discrete kernel) \( g_m \) where \( m = -3, -2, -1, 0, 1, 2, 3 \);
3. Calculate a set of new data \( \{F_i\} \) by using \( F_i = \sum_{m=1}^{7} f_i \cdot g_m \);
4. Replace \( \{f_i\} \) by \( \{F_i\} \) and repeat Steps 1–3 till a sufficient degree of smoothing is achieved (in the present work five to ten repeats were carried out).

The final data set \( \{F_i\} \) was then used to plot the smoothed curve, which was also shown in the figure. It can be seen that the discontinuities disappear, better matching the neutron imaging profiles. However, it is worthy to note that capturing such water discontinuity is important for understanding of the liquid-water behavior in a fuel cell. Hopefully, our present work will motivate the experimentalists to obtain more accurate data using neutron imaging, X-ray, or other techniques. We also hope that our present work will motivate modellers to develop higher fidelity models so as to more accurately predict water distribution.

Figure 8 displays the computed current density profiles for the three cell temperatures considered in the present work. As the average current density is set at 0.75 A/cm², variation of the local current is around this average value. Small variations are observed for the three cases with the largest one appearing at 80°C. Its explanation can be aided by Fig. 5: as this operation is controlled by the Ohmic resistance, the lower water content under the channel at 80°C leads to a higher Ohmic polarization, causing a relatively lower local reaction rate. However, the variation is small due to the membrane being highly hydrated for the three cases as shown in Fig. 5.

Figure 9 shows the current density variations at 40°C operating temperature and various RHs. At low RH, the membrane hydration levels may be quite different between the regions under the channel and land; consequently, the Ohmic resistance will differ greatly from location to location. This figure indicates that large current density variations take place for the two cases of dry anode (i.e., the anode RH is 50%). For the other cases where anode is fed with fully humidified gas, the degree of variation is small even for the case in which 50% RH is used in the cathode.

Figure 10 shows the detailed liquid-water distribution within the anode and cathode GDLs for the three dry cases. It can be seen that in the driest case (50% RH on both sides) liquid water still appears in the region is limited in a minute area under the land, while vast portion of the GDL (both anode and cathode) is subjected to dry operation. Comparing with the cases of 50% RH in the anode and cathode (the other side is 100% RH, i.e., RHa/c = 50/100% and 100/50%, respectively), the dry anode indicates a smaller wetted
region in the GDL, particularly in the anode side. For the case that only cathode is dry (i.e., $R_{Ha/c} = 100/50\%$), all the region of the MEA is in the two-phase operation; consequently, a higher water content is secured in the membrane, leading to a small variation of the current density as shown in the previous figure.

Figure 11 shows the water profiles across the MEA–GDL for various RHs (Fig. 11a) and current densities (Fig. 11b). It can be seen from Fig. 11a that the anode water content directly increases with the RH value in the anode whereas in the cathode side, part of the GDL for the case of $R_{Ha/c} = 100/50\%$ is even higher than that at $50/100\%$. Figure 11b displays the water-content profiles at various operating current densities for $R_{Ha/c} = 100/100\%$. It can be seen that the cathode water-content profile increases with the current density; however, the anode side indicates a complex pattern: $0.75\, \text{A/cm}^2$ gives rise to the highest water content in the anode.

Figure 12a shows the detailed 2D water saturation for the case of $1.5\, \text{A/cm}^2$, $R_{Ha/c} = 100/100\%$, and $40^\circ\text{C}$ when using different GDL thermal conductivities: (a) $3.0\, \text{W/m K}$, (b) $1.0\, \text{W/m K}$, and (c) $6.0\, \text{W/m K}$.

Conclusions

A multidimensional model was developed and employed to elucidate the water distribution across the MEA–GDL component in a PEFC. The present model is based on the conservation of mass, momentum, species (including liquid water), energy, and charges.
2D simulation results were computed using the model. We found that liquid-water content may be lower at higher cell temperatures due to greater water evaporation and stronger water diffusion in vapor phase. At 40°C operation where water diffusion in vapor phase is relatively weak, the liquid-water content was found to increase monotonically with current density in the cathode GDL, but a complex trend was observed in the anode. It was also found that changing the RHs in the anode and cathode profoundly affects the liquid-water profile in the PEF. Moreover, reducing the GDL thermal conductivity may shrink the size of two-phase region in the PEF. Results from the present study also show that 2D (through-plane in-plane dimension) fuel cell simulation is required to be able to predict the water distribution across the MEA–GDL. Lastly, predictions computed from the present model were compared with the neutron-imaging data reported by two separate groups in terms of plus in-plane dimension.

References
5. A. Rowe and X. Li, J. Power Sources, 102, 82 (2001).
27. Y. Wang, J. Power Sources, 185, 261 (2008).