High-current-density performance of polymer electrolyte fuel cells (PEFCs) is known to be limited by transport of reactants and products. In addition, at high current densities, excessive amount of water is generated and condenses, filling the pores of electrodes with liquid water, and hence limiting the reactant transport to active catalyst. This phenomenon known as “flooding” is an important limiting factor of PEFC performance. In this work, the governing physics of water transport in both hydrophilic and hydrophobic diffusion media is described along with one-dimensional analytical solutions of related transport processes. It is found that liquid water transport across the gas diffusion layer (GDL) is controlled by capillary forces resulting from the gradient in phase saturation. A one-dimensional analytical solution of liquid water transport across the GDL is derived, and liquid saturation in excess of 10% is predicted for a local current density of 1.4 A/cm². Effect of GDL wettability on liquid water transport is explored in detail for the first time. Furthermore, the effect of flooding on oxygen transport and cell performance is investigated and it is seen that flooding diminishes the cell performance as a result of decreased oxygen transport and surface coverage of active catalyst by liquid water.

The objective of this paper is to provide a fundamental understanding of liquid water transport in hydrophobic GDL and the effect of flooding on the PEFC performance. The paper starts with explaining the two-phase transport in hydrophobic porous GDL. Then, we define the onset of the two-phase mixture and derive a set of analytical equations using a control volume analysis of water balance to determine the threshold current density for two-phase occurrence. Finally, we give basic definitions of two-phase transport and provide one-dimensional solutions of liquid water and oxygen transport in GDL, and investigate the effect of liquid water, GDL wettability, and flooding on the performance of PEFC.

Two-Phase Flow and Transport in Porous GDL

GDL is an essential component of PEFCs. It serves as a support for the polymer electrolyte membrane (PEM), distributes reactant gas over the catalyst layer, and conducts electrons from reaction sites to the outer circuit. Figure 1 shows scanning electron microscopy (SEM) images of commercial carbon-paper and carbon-cloth GDLs. At high current densities, the electrochemical reaction rate is faster than the amount of reactants supplied, specifically oxidizer; therefore, the reaction rate is limited by the transport rate of oxidizer to the reaction sites, which is sandwiched between GDL and PEM. In addition, when the liquid water blocks some pores of GDL, the reactant transport is further limited. As such, PEFC GDL is commonly Teflonized to provide highly hydrophobic surface for easy removal of liquid water.

When water vapor pressure exceeds the saturation level, condensation starts, forming a tree-like liquid water percolation in the porous GDL, as illustrated in Fig. 2. The liquid water reaches the interface of porous GDL and open channel, forming liquid droplets. Inside GDL, liquid water is driven by capillary (wicking) action. This capillary action is a result of capillary pressure distribution, which is defined as the difference between gas and liquid-phase pressures. In hydrophobic GDL, the capillary pressure is negative; hence, the liquid pressure is larger than the gas-phase pressure, whereas in hydrophilic media, the gas-phase pressure is higher than that of the liquid phase.

\[ p_l = p_g - p_t \]
In addition, the liquid pressure increases with the fraction of void spaces occupied by liquid water; therefore, a liquid pressure gradient is formed from higher to lower liquid saturation regions. This pressure gradient becomes the driving force for liquid water flow, as schematically shown in Fig. 3. In PEFCs, the liquid saturation is higher at the catalyst layer, due to water generation and electro-osmotic drag, than the GDL-channel interface. Therefore, the liquid pressure gradient formed in the GDL drives liquid water from the reaction sites toward the open channel. Figure 3 also displays a nearly constant gas pressure profile across the two-phase zone due to much lower gas-phase viscosity. That is, it does not incur much gas pressure drop to drive the gas flow through thin GDLs.

It is interesting to note from Fig. 3 that although the relative magnitude of the liquid to gas pressure is different in hydrophobic GDL than the hydrophilic one, both media provide capillary action to drive liquid water from the inside to its surface. However, the slope of the capillary pressure (i.e., the driving force) is different depending on whether water is the wetting or nonwetting phase. The greater slope of the capillary pressure near the evaporation front (i.e., \( s \approx 0 \)) in hydrophobic GDL is indicative of more effectiveness of this type of medium for water removal.

Figure 3. Schematic illustration of liquid and gas-phase pressure profiles in hydrophilic and hydrophobic porous media.

The shape of the liquid droplets emerging at the channel—GDL interface is governed by the wetting characteristics of the GDL surface. On a hydrophilic surface, which has a contact angle of less than 90°, liquid spreads over it, whereas on a hydrophobic surface, which has a contact angle greater than 90°, the droplet is more of a sphere-like shape, covering less pore entry, as pictured in Fig. 4.

Onset of the two-phase regime.—In this section, an analytical solution predicting the onset of two-phase flow is presented in one-dimension, considering the water transport phenomena in the through-plane direction, i.e., from the anode to cathode. The goal is...
to estimate the threshold local current density under which two-phase conditions start to appear. This onset is defined when the maximum water vapor concentration, occurring at the cathode GDL-membrane interface, becomes equal to the saturation value corresponding to the cell temperature.

Water transport in PEFCs is governed by the following phenomena: generation of water at the cathode due to the oxygen reduction reaction (ORR), forward (i.e., anode to cathode) or backward (i.e., cathode to anode) diffusion of water across the membrane, electro-osmotic drag of water from anode to cathode, and convective removal of water to the gas channel. The considered domain for derivation is sketched in Fig. 5.

The convective mass-transfer coefficient at the channel-GDL interface is calculated using the analogy between heat and mass transfer. It is known that the Nusselt number (i.e., dimensionless heat-transfer coefficient) is constant for laminar flow in ducts and is equal to 4.36 for the constant heat flux condition and 3.66 for the constant surface temperature. Furthermore, using the heat/mass-transfer analogy, the Sherwood number (i.e., dimensionless mass-transfer coefficient) can also be correlated with the Nusselt number with the ratio $Sc/Pr$.

For this part of the work, membrane water content is assumed to be constant across its thickness and is calculated from the arithmetic average of water contents on the anode and cathode sides of the membrane. Water diffusion coefficient in the membrane is calculated by an empirical relation using this average water content. Water vapor diffusion coefficient in the GDL is corrected for tortuosity effects using the Bruggeman correlation.

A control volume analysis of water balance under steady-state conditions at points 1-4 of Fig. 5 provides a set of linear equations for unknowns $C_{1}^{H_{2}O}$, $C_{3}^{H_{2}O}$, $C_{4}^{H_{2}O}$, and $I_{onset}$. These relations are:

Anode channel—GDL interface -1-

$$h_{m,a}(C_{1}^{H_{2}O} - C_{0}^{H_{2}O}) = D_{GDL,a}^{H_{2}O} \frac{C_{1}^{H_{2}O} - C_{2}^{H_{2}O}}{\delta_{GDL}}$$

Anode GDL—membrane interface -2-

$$D_{GDL,a}^{H_{2}O} \frac{C_{2}^{H_{2}O} - C_{1}^{H_{2}O}}{\delta_{GDL}} = D_{mem}^{H_{2}O} \frac{C_{3}^{H_{2}O} - C_{2}^{H_{2}O}}{\delta_{mem}} - \frac{I}{2F}$$

Membrane—cathode GDL interface -3-

$$D_{GDL,c}^{H_{2}O} \frac{C_{3}^{H_{2}O} - C_{4}^{H_{2}O}}{\delta_{GDL}} + D_{mem}^{H_{2}O} \frac{C_{3}^{H_{2}O} - C_{2}^{H_{2}O}}{\delta_{mem}} = (2n_{d} + 1) \frac{I}{2F}$$

Cathode GDL—membrane interface -4-

$$h_{m,c}(C_{4}^{H_{2}O} - C_{5}^{H_{2}O}) = D_{GDL,c}^{H_{2}O} \frac{C_{1}^{H_{2}O} - C_{4}^{H_{2}O}}{\delta_{GDL}}$$

For the solution shown in Fig. 7 assumes that the in-plane gradients...
of water concentration, e.g., along the channel or in the channel-to-land direction, are negligibly small and hence considers only the transport of water in anode to cathode (through-plane) direction. However, in low-stoichiometry cases, there are strong in-plane gradients, thereby requiring multidimensional numerical modeling. This is addressed in a companion paper.21

Basic parameters of two-phase flow in GDL.—Liquid saturation.—Liquid saturation, $s$, is defined as the volume fraction of the total void space of porous media occupied by the liquid phase. Hence, $s$ is defined as

$$s = \frac{V_l}{V}$$  

[8]

Saturation may vary from zero to unity and saturations of all phases add up to unity.

Relative permeability.—In liquid-gas two-phase flow in porous media, the available pore space is shared by liquid and gas; thus, the cross-sectional area available for each fluid is less than the total available pore space. This phenomenon introduces the concept of relative permeability $k_{r, \phi}$, which defines the ratio of intrinsic permeability of a phase $k$ at a given saturation level to the total intrinsic permeability of the porous medium. In this work, GDL is assumed to be a homogeneous porous medium, with relative permeabilities proportional to the cube of phase saturation, i.e.

$$k_{r, \phi} = s^3; \quad k_{r, g} = (1 - s)^3$$  

[9]

Capillary pressure.—Capillary pressure between two phases is expressed as

$$p_c = \sigma \cos \theta \left( \frac{e}{K} \right)^{1/2} J(s)$$  

[10]

where $J(s)$ is the Leverette function and is given by the following relation

$$J(s) = \begin{cases} 
1.417(1 - s)^2 - 2.120(1 - s)^2 + 1.263(1 - s)^3 & \text{if } \theta_c < 90^\circ \\
1.417s^2 - 2.120s^2 + 1.263s^3 & \text{if } \theta_c > 90^\circ 
\end{cases}$$  

[11]

Note that for a hydrophilic medium, the wetting phase is the liquid phase. Therefore the Leverette function is expressed in the gas-phase saturation, whereas in hydrophobic medium, the gas phase becomes the wetting phase and so the liquid-phase saturation is defined as

$$s = 1 - \cos \theta_c$$

[12]

Combining Eq. 13 and 14 yields

$$J(s) = 1.417s - 2.120s^2 + 1.263s^3$$  

[14]

Combining Eq. 13 and 14 yields

$$J(s) = \frac{\sigma \cos \theta_c (e/K)^{1/2}}{v} s^3(1.417 - 4.240s + 3.789s^2)$$  

[15]

Considering the one-dimensional transport of liquid water only in the through-plane direction, the Eq. 15 reduces to an ordinary differential equation, which can be solved analytically. For hydrophobic ($\theta_c > 90^\circ$) GDL,
solution of liquid water transport. Figure 8. Liquid saturation profile across GDL predicted by 1D analytical solution of liquid water transport.

\[
s'(1.417 - 4.240s + 3.789s^2)\frac{ds}{dx} = \frac{I}{2F} M_{H_2O} \frac{v}{\sigma \cos \theta_s (eK)} x^{\frac{5}{2}} \tag{16}
\]

or

\[
s'(0.35425 - 0.8480s + 0.6135s^2) = \frac{I}{2F} M_{H_2O} \frac{v}{\sigma \cos \theta_s (eK)} x + C_1 \tag{17}
\]

Following the same procedure, the following equation is derived for hydrophilic (\(\theta_s < 90^\circ\)) GDL

\[
s'(-0.2415 + 0.6676s - 0.6135s^2) = \frac{I}{2F} M_{H_2O} \frac{v}{\sigma \cos \theta_s (eK)} x + C_1 \tag{18}
\]

In these equations, the integration constant \(C_1\) is governed by the boundary condition, which is the liquid saturation at the GDL-channel interface. Once liquid saturation at this boundary is known, \(C_1\) can be easily calculated. Here, for simplicity, it is assumed that no liquid water is present in the open channel.

For a local current density of 1.4 A/cm², the saturation profiles obtained from this equation for GDLs having contact angles of 80 and 100° are given in Fig. 8. The results show that the liquid saturation level at the GDL-catalyst layer interface can be as high as 10%, even in the case of no liquid water in the gas channel, which may result in severe flooding. Note that in this case the net water transport across the membrane is neglected; however, in real PEFC operation, there may be a significant water transport from anode to cathode due to electro-osmotic drag, especially if the anode stream is well humidified. In addition, it is also seen that the liquid saturation level near the catalyst layer interface is higher for hydrophilic than hydrophobic media, suggesting a higher water removal rate for hydrophobic (\(\theta_s > 90^\circ\)) GDL. This is because the liquid water flow is driven by the liquid pressure gradient, and the latter depends on the first derivative of the liquid pressure with respect to liquid saturation. It is seen from Fig. 3 that the slope of liquid pressure curve is steeper for hydrophobic GDL at lower liquid saturations (i.e., up to 20%); therefore, the liquid water removal rate is higher than hydrophilic GDL. This is one distinctive advantage of using hydrophobic GDL in PEFC.

Figure 9 shows the effect of contact angle of hydrophobic GDL on the liquid water distribution. It is seen that the water distribution has a great dependence on contact angle, and the larger the contact angle than 90°, i.e., the more hydrophobic, the lower the liquid saturation at the catalyst layer-GDL interface.

**Effect of flooding on performance.**—As mentioned earlier, flooding reduces the fuel cell performance in two separate manners: (i) by hindering the oxygen transport from the gas channel to the reaction sites and (ii) by covering electrochemically active sites with liquid water. These phenomena can be studied by a simple analysis if the catalyst layer is assumed infinitely thin and the cathode liquid saturation is uniform. Oxygen is transported from the cathode gas channel to the cathode GDL via convection, and then diffuses through the cathode GDL to the catalyst layer. In this derivation, convective transport of oxygen across porous GDL is neglected. When the catalyst layer is assumed infinitely thin, ORR now occurs at the membrane-GDL interface, and the oxygen consumption rate by electrochemical reaction, ORR, is given by

\[
f_{O_2} = \frac{I}{4F} \tag{19}
\]

per unit reaction surface. Electrochemical kinetics is governed by the Butler-Volmer equation, and because the cathode kinetics is sufficiently sluggish, that can be expressed by Tafel kinetics, i.e.,

\[
I = (1 - s) a_i \frac{C_{O_2}^{ref}}{C_{O_2}^{rex}} \exp \left( \frac{\alpha F \eta}{RT} \right) \tag{20}
\]

In Eq. 20, the factor \((1 - s)\) represents the active area reduction due to liquid water coverage of catalyst particles.

Oxygen is transported from the channel to the reaction zone via convection in the gas channel and molecular diffusion in the GDL. Therefore, the molar flux of oxygen from the gas channel to the reaction site can be expressed as
Figure 10. Cathode overpotential vs. liquid saturation in cathode GDL for different channel oxygen concentrations. Local current density is 2 A/cm².

\[ j^{O_2} = \frac{C^{O_2}_{ch} - C^{O_2}_{rea}}{D^{O_2} [\varepsilon(1-s)]^{1/5} + \frac{1}{h_m}} \]  \[ \text{[21]} \]

In this equation, \( C^{O_2}_{ch} \) and \( C^{O_2}_{rea} \) refers to the oxygen concentration in the gas channel and at the reaction site, respectively, while \( h_m \) represents the convective mass-transfer coefficient at the channel-GDL interface. Convective mass-transfer coefficient is calculated using the same analogy of heat and mass transfer, as explained earlier, and found to be 0.032 m/s for a square gas channel of 1 mm. The molar flux of oxygen is equal to the consumption rate of oxygen. Therefore, using Eq. 20 and 21, oxygen molar concentration at the reaction surface is calculated as

\[ C^{O_2}_{rea} = C^{O_2}_{ch} - \frac{I}{4F} \left[ \frac{\delta^{\text{GDL}}}{D^{O_2} [\varepsilon(1-s)]^{1/5} + \frac{1}{h_m}} \right] \]  \[ \text{[22]} \]

Inserting the expression for oxygen concentration at the reaction site from Eq. 22 to the cathode kinetics equation, Eq. 20, one can obtain the cathode overpotential as a function of oxygen concentration in the channel, liquid saturation in the GDL, and the local current density, as follows

\[ \eta(s,I,C^{O_2}_{ch}) = \frac{RT}{\alpha_e F} \ln \left( \frac{I C^{O_2}_{rea}}{C^{O_2}_{ch}} \right) \times \left[ \frac{1}{D^{O_2} [\varepsilon(1-s)]^{1/5} + \frac{1}{h_m}} \right] \]  \[ \text{[23]} \]

In Fig. 10, the cathode overpotential is plotted vs. liquid saturation in GDL, using Eq. 23, for a local current density of 2 A/cm² and different channel oxygen concentrations. It is seen that the sharp increase in the cathode overpotential, which identifies the mass-transfer limitation of the cathode reaction, may be seen above 75% liquid saturation when channel oxygen concentration is 50 mol/m³, which approximately corresponds to the oxygen concentration near the inlet of a pure oxygen feed PEFC at 80°C and 1.5 atm. However, it is also seen that for a free-stream oxygen concentration of 5 mol/m³, which may correspond to the oxygen concentration near the outlet of an air-feed PEFC operating at a stoichiometric ratio of 2, the mass-transfer limitation starts to appear at as low as 10% liquid saturation.

In Fig. 11, the cathode overpotential vs. local current density is plotted for different liquid saturation values, for an oxygen concentration in the channel of 4 mol/m³. As expected, magnitude of the cathode overpotential increases with increasing local current density. Furthermore, increase in the liquid saturation of GDL results in an increase in cathode overpotential. The increase in the cathode overpotential with increasing liquid saturation is slighter for low current densities and becomes much higher at high current densities. This is attributed to the fact that at higher current densities, the cathode reaction requires more oxygen and the reaction rate is limited by the rate of oxygen transport to the reaction sites. However, in lower current densities the cathode reaction is controlled by activation losses; hence, the change in the cathode overpotential with increasing liquid saturation is not as high as at higher current densities. It is also clearly seen that the limiting current density is controlled by the amount of liquid water present in GDL and catalyst layer.

The analytical expression obtained can be further utilized to separate the two effects of flooding: (i) coverage of active catalyst areas by liquid water and (ii) hindering of oxygen transport by liquid water. In Fig. 12, the hypothetical cases of cathode overpotential are plotted for a local current density of 2 A/cm² near the inlet with a channel oxygen concentration of 10 mol/m³. In the first case, both of the limitations resulting from liquid water are considered. In the second case, it is assumed that liquid water does not cover any active reaction sites; therefore no active area reduction is considered and only the \( O_2 \) transport limitation by liquid water is accounted for. In the last case, it is assumed that the liquid water presence does not interfere with \( O_2 \) transport. Hence, only the effect of coverage of active reaction sites by liquid water is considered. It is seen that the limitation of oxygen transport by liquid water is more dominant over the entire liquid saturation range. Therefore, it is concluded that the primary effect of flooding on the PEFC performance is the reduced effective oxygen diffusivity in GDL due to the filling of pores with liquid water.

The same approach that is used to describe cathode polarization can be used to quantify the anode polarization. In addition, ohmic
losses associated with ionic transport can be determined, assuming that the membrane is fully saturated with liquid water. Under these assumptions, the cell potential can be easily determined by

\[ V_{\text{cell}} = V_{\text{oc}} - \eta_a - \eta_c - IR \]  

[24]

The cell dimensions and operating parameters for these calculations are given in Table I. Because the membrane is assumed to be fully hydrated by liquid water, the membrane ionic conductivity is constant and the ionic losses in membrane and catalyst layers can be approximated as

\[ IR = i \left( \frac{\delta_{\text{mem}}}{k_{\text{mem}}} + 2 \frac{\delta_{\text{cl}}}{k_{\text{cl}}} \right) \]  

[25]

Figure 12. Separate flooding effect of reduced oxygen transport and reduced active area on cathode overpotential. Oxygen concentration in the channel is 10 mol/m³.

In Fig. 13, the local cell potential is plotted for channel concentrations of 20 and 4 mol/m³ of hydrogen and oxygen, respectively, for different liquid saturations in cathode GDL and catalyst layer. The selected values of concentrations correspond to the downstream of an air-feed PEFC operating at 80°C and 1.5 atm. It is clearly seen that the cell performance is limited by the cathode transport and electrochemical kinetics. As expected, the increasing liquid saturation pulls down the polarization curve, reducing the overall performance of the fuel cell.

The analytical derivations show that the presence of liquid water in porous GDL can dramatically decrease the performance of PEFCs, especially in the cases of low O₂ concentrations in the channel and higher current density.

Table I. Electrochemical and transport properties.

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<thead>
<tr>
<th>Description</th>
<th>Unit</th>
<th>Value</th>
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<tr>
<td>Electrochemical kinetics (typical values)</td>
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<tr>
<td>Anode reference exchange current density</td>
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<td>Cathode reference exchange current density</td>
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<td>Faraday constant</td>
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<td>Transport parameters</td>
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<td>O₂ diffusivity in gas at 353 K and 1.5 atm</td>
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<td>H₂O diffusivity in gas at 353 K and 1.5 atm</td>
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<td>H₂O diffusivity in membrane (at λₚₑᵣ = 5.8, estimated)</td>
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<td>Liquid water viscosity</td>
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<td>Material properties</td>
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Conclusions

The governing physics of liquid water transport in hydrophobic GDL has been presented. It is seen that capillary transport is the dominant transport process to remove water from flooded GDLs. An analytical model was developed to estimate the onset of the two-phase regime in GDLs and the associated threshold current density as a function of various anode and cathode relative humidity combinations. In the two-phase regime, a one-dimensional analytical solution was derived for liquid water transport in both hydrophilic and hydrophobic GDL, and the important role of GDL wettability is elucidated. Finally, the effect of flooding on oxygen transport and cell performance was analyzed, and significant decrease in performance is predicted with increased level of flooding, particularly at high current densities, in accordance with experimental observations. Based on the physics described herein for liquid water transport and flooding in a full PEFC is described in the companion paper.

It is expected that the two-phase transport in PEFCs has different transient characteristics than its single-phase counterpart. Hence, efforts are presently underway to study the effects of liquid water transport and flooding on transient and hysteretic behaviors of PEFCs. In addition, there is a lack of fundamental experiments on water transport in GDLs, and no correlations exist for two-phase transport properties directly relevant to PEFC GDLs, such as relative permeability and capillary pressure. These are the areas of ongoing work in our laboratory.

Acknowledgments

Funding for this work from Department of Energy under cooperative agreement no. DE-FC26-01NT41098, ConocoPhillips, GM Foundation, and NSF under grant no. CTS-9733662 is gratefully acknowledged.

The Pennsylvania State University assisted in meeting the publication costs of this article.

List of Symbols

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<th>Symbol</th>
<th>Description</th>
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<td>(a_{ic})</td>
<td>transfer current density (\times) active catalyst area, (A/m^3)</td>
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Greek

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Subscripts

- \(a\) anode
- \(c\) cathode
- \(ch\) channel
- \(cl\) catalyst layer
- \(g\) gas
- \(GDL\) gas diffusion layer
- \(l\) liquid
- \(mem\) membrane
- \(oc\) open circuit
- \(ref\) reference
- \(rxn\) reaction
- \(sat\) saturation

References