

Capillary Pressure-Water Saturation Relations for Gas Diffusion Layers Affecting Water Transport and PEFC Polarization Behaviors

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ABSTRACT: Numerical simulations using a two-phase flow, non-isothermal model with the capillary pressure-water saturation relations considered have been carried out to examine the effects of liquid water transport properties of GDL materials on cell performance. A designed GDL having a different capillary pressure-water saturation relation from the conventional GDL showed better cell performance at high current density, indicating improved mass transport. Numerical simulation indicates the direction to develop an engineered design of the porous structure for the GDL substrate to reduce the concentration overpotential.

KEY WORDS: polymer electrolyte fuel cells, water management, gas diffusion layer, mass transport

1. INTRODUCTION

Water management is essential for improving polymer electrolyte fuel cell (PEFC) performance. In conventional PEFC architecture, functional porous transport layers, so-called gas diffusion layers (GDLs) with microporous layers (MPLs) embedded, are employed with electrochemical reaction layers, *i.e.*, catalyst layers (CLs). Under high current density operation and/or high relative humidity conditions, water produced in the cathode is partially/entirely condensed and plugs pores in these porous layers, resulting in deterioration of reactant transport from flow channels to the reaction site in the CLs. Efficient liquid water removal from the cell is needed, and therefore, fundamental understanding and control of water transport processes in PEFCs are still of great importance.⁽¹⁾

Two-phase behaviors in porous media are affected by the physical and chemical properties of the porous media. The co-existing wetting phase and non-wetting phase in porous media are affected by each other in transport processes mainly driven by fluid pressure and capillary force.

In PEFCs, liquid water transport in CLs, MPLs, and GDLs is dominated by capillary force, while viscous and inertial forces are negligible.⁽²⁾ The capillary pressure, p_c , is defined by the

difference of liquid pressure p_l and gas pressure p_g in these porous media and is modeled by the Leverette function $J(s_l)$ as the following equation,

$$p_c = p_l - p_g = -\sigma_w \cos \theta_K \sqrt{\frac{\varepsilon_K}{\kappa_K}} J(s_l) \quad (1)$$

where σ_w , θ_K , ε_K and κ_K are the surface tension of liquid water, the contact angle, the average porosity and permeability of the porous material, respectively and s_l is the liquid water saturation that is defined as the ratio of the pore volume occupied by liquid water to the entire pore volume in a porous medium. For hydrophobic porous media, $\theta_K > 90^\circ$, the liquid phase is difficult to fill the small pores. Thus, the liquid water does not invade the porous media entirely, lowering the local liquid water. Accordingly, porous media used in PEFCs are generally kept hydrophobic to lower the liquid water saturation and prevent the liquid water from plugging pores. The standard Leverett function for hydrophobic porous media has been widely used in the PEFC community,⁽³⁾

$$J(s_l) = 1.417s_l - 2.120s_l^2 + 1.263s_l^3 \quad (2).$$

The capillary pressure-water saturation, p_c - s_l , relations highly affect liquid water transport, resulting in oxygen transport in the

CLs, MPLs, and GDLs. Recently, novel gas diffusion layers embedded with flow channels have been proposed to enhance liquid water removal and oxygen transport.⁽⁴⁾ The newly developed GDL showed less liquid water accumulation, leading to superior cell performance under high relative humidity conditions.⁽⁵⁻⁷⁾ It is suggested that the cell performance improvement can be attributed to its unique porous structure shown in the literature.⁽⁶⁾

In this study, numerical simulations have been carried out to examine the effects of liquid water transport properties of GDL materials on cell performance using a two-phase flow, non-isothermal model with the capillary pressure-water saturation relations considered.

2. PEFC SIMULATION MODEL

2.1. Governing equations

In the present study, a two-phase flow, non-isothermal model was applied to simulate the performance of the PEFC. The conservation equations of mass, momentum, chemical species, energy, and charges were fully coupled with the electrochemical reactions as follows,

$$\nabla \cdot (\rho \mathbf{u}) = Q_m \quad (3),$$

$$\begin{aligned} \frac{\rho}{\varepsilon_k} \left(\mathbf{u} \cdot \nabla \frac{\mathbf{u}}{\varepsilon_k} \right) = & -\nabla p + \nabla \cdot \left[\frac{\mu}{\varepsilon_k} \left\{ (\nabla \mathbf{u} + (\nabla \mathbf{u})^T) - \frac{2}{3} (\nabla \cdot \mathbf{u}) \mathbf{I} \right\} \right] \\ & - \left(\frac{\mu}{\kappa_K} \right) \mathbf{u} + F \\ (k = & \text{CL, MPL, GDL, CH}) \end{aligned} \quad (4),$$

$$\begin{aligned} \rho \mathbf{u} \cdot \nabla \omega_i - \nabla \cdot \left(\rho \omega_i \sum_k D_{ik} \left[\nabla x_k + (x_k - \omega_i) \frac{\nabla p}{p} \right] \right. \\ \left. + D_i^T \frac{\nabla T}{T} \right) = & M_i R_i \\ (i, k = & \text{H}_2, \text{O}_2, \text{H}_2\text{O}) \end{aligned} \quad (5),$$

$$\nabla \cdot \left(\sum_{i=\text{G,L}} [\rho c_p \mathbf{u}]_i T \right) - \nabla \cdot \left(\sum_{i=\text{G,L,S}} k_i \nabla T \right) = S_T \quad (6),$$

$$\nabla \cdot (-\sigma_s^{\text{eff}} \nabla \phi_s) = -i_{\text{src}} \quad (7),$$

$$\nabla \cdot (-\sigma_m^{\text{eff}} \nabla \phi_m) = i_{\text{src}} \quad (8),$$

where \mathbf{u} is velocity (m/s), p is pressure (Pa), T is temperature (K), ρ is density (kg/m³), and μ is viscosity (Pa·s) of the gaseous mixture. In addition, ω , x , D^T , M , ε_k , and κ_K are mass fraction, mole fraction, thermal diffusion coefficient (kg/(m·s)), molecular weight (kg/mol), porosity, and permeability (m²), respectively.

D_{ik} is multicomponent diffusivity (m²/s). The subscripts i and k denote the gas species. Also, R and Q_m represent the source terms. And σ_s^{eff} and σ_m^{eff} are effective electric and ionic conductivities (S/m) while ϕ_s and ϕ_m are solid and polymer electrolyte phases potentials (V), respectively. i_{src} is volumetric current density source (A/m³) and was modeled by the Butler–Volmer equation with couples of transport resistance taken into account. The details of the model can be found in the literature.⁽⁸⁾

2.2. Capillary pressure-water saturation relations

Liquid water transport properties of the GDLs were modeled by Eq.(1) where the average porosity and permeability of the porous material are used with the modified Leverett function.

$$J(s_l) = a s_l - b s_l^2 + c s_l^3 \quad (9).$$

Identical parameters in the above equation were firstly determined to satisfy the capillary pressure-water saturation relation reported for SGL10BA, Case1, in the literature.^(9,10) We also carried out simulations for a designed GDL, Case2, to compare the effects of the capillary pressure-water saturation relations on cell performance and liquid water distribution in GDLs. The parameters used in the present study are listed in Table 1. The simulated capillary pressure-water saturation relations are shown in Fig. 1.

Table 1 GDL Parameters applied in the simulations.

	ε_K	a	b	c
Case1	0.84	0.7277	1.42	1.463
Case2	0.65	0.65	1.5	1.6

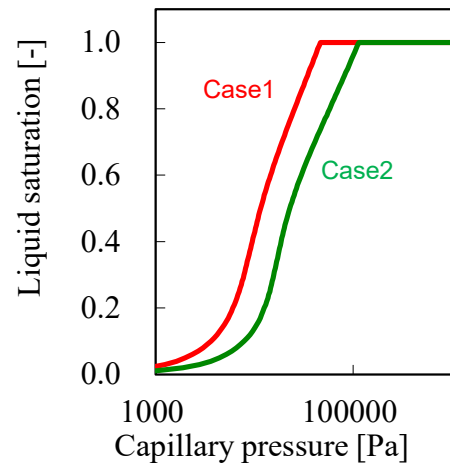


Fig. 1 Capillary pressure-water saturation relation.

2.3. Calculation domain and conditions

The calculation domain with 4 mm long in the flow direction and the flow field calculated in the current study were shown in Fig. 2, where an interdigitated flow field was applied to simulate the cell used for X-ray imaging reported in the literature.⁽⁷⁾ The simulations were carried out at almost 100% RH condition and at room temperature, to examine polarization behaviors as observed in the experiments.⁽⁷⁾

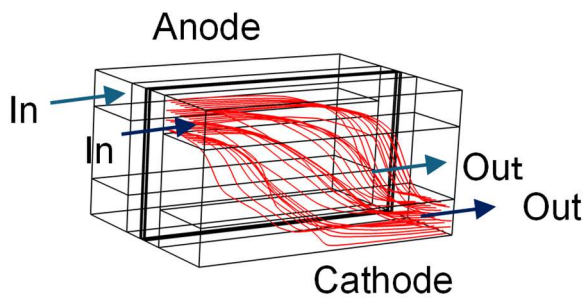


Fig. 2 Calculation domain and flow field.

3. RESULTS AND DISCUSSION

Polarization curves calculated for different GDL properties are shown in Fig. 3. Case2 shows better performance at high current density, indicating improved mass transport. It is noteworthy that simulation results show less deteriorated mass transport in Case2, although the porosity of Case2 is considerably lower compared to Case1.

Fig. 4 shows liquid water saturation distributions of Case1 and Case2. Accumulation of liquid water in the GDL was suppressed in Case2. This is attributed to the capillary pressure-water saturation relation for the GDL at Case2, indicating its ability to maintain lower liquid water saturation in the porous media.

Fig. 5 shows liquid water saturation profiles across the anode GDL to the cathode GDL under the channel center at 2 mm downstream from the inlet. Liquid water saturation in the cathode catalyst layer at Case2 is less than 0.2 while over 0.7 at Case1. Numerical simulation demonstrates that GDL properties highly affect the distribution of liquid water, and engineered design for the GDL substrate further improves liquid water and oxygen transport in PEFCs. This agrees well with experimental observations⁽¹¹⁾ in which X-ray imaging revealed low liquid water content in the GDL with a newly developed porous structure.

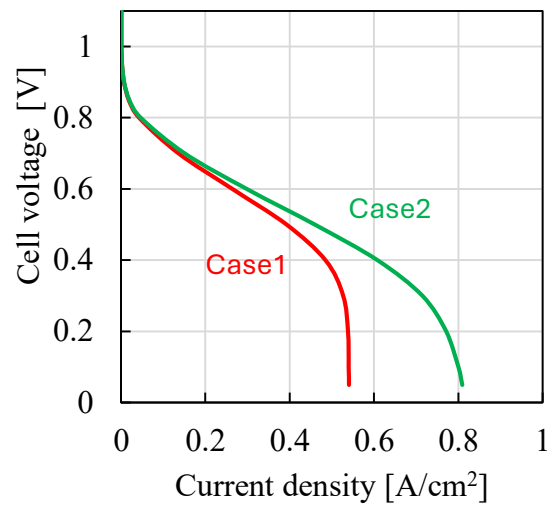
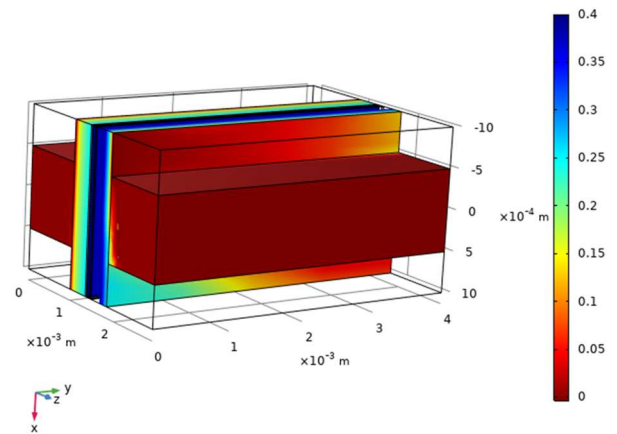
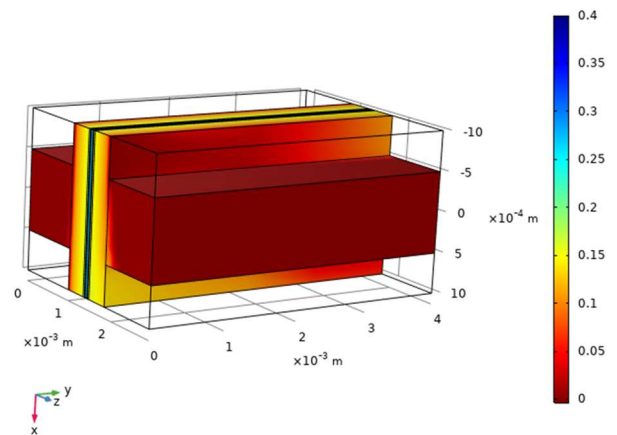


Fig. 3 Calculation results.



(a) Case1



(b) Case2

Fig. 4 Liquid water saturation distributions.

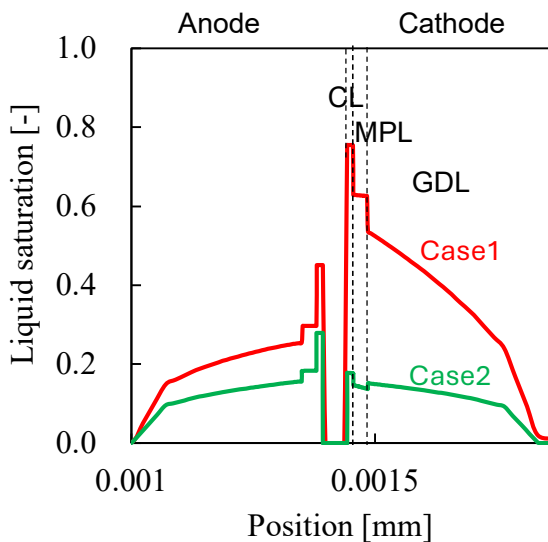


Fig. 5 Liquid water saturation profiles.

4. CONCLUSIONS

Numerical simulations have been carried out to examine the effects of liquid water transport properties of GDL materials on cell performance using a two-phase flow, non-isothermal model with the capillary pressure-water saturation relations considered. A designed GDL having a different capillary pressure-water saturation relation from the conventional GDL was examined, showing better cell performance at high current density, indicating improved mass transport. Numerical simulation indicates the direction to develop an engineered design of the porous structure for the GDL substrate to further reduce the concentration overpotential.

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