A Finite Element Analysis of the Gas Diffusion Layer of a Polymer Electrolyte Fuel Cell

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Multi-component gas transport phenomena in a cathode electrode layer of Polymer Electrolyte Fuel Cells (PEFC) are investigated by using a two-dimensional computational model developed by Yi and Nguyen. In the past research, we encountered a problem that the solution did not converge when the same settings for physical parameters with them were used in our steady simulations. In the present study, we extend our simulation from steady to unsteady simulation to avoid the problem. Also, based on the results, the effect of changing permeability in the simulations is discussed with reporting possible errors regarding the value of permeability shown in the reference at the same time.

List of symbols

\( C_{O_2} \) : concentration of oxygen mol/cm\(^3\)
\( C_{O_2,ref} \) : oxygen reference concentration for the oxygen reaction mol/cm\(^3\)
\( \Delta t \) : time step s
\( e^- \) : electron
\( F \) : Faraday constant, 96,487 C/equiv
\( H_2O \) : water
\( H^+ \) : hydrogen ion
\( I \) : current density A/cm\(^2\)
\( I_o \) : exchange current density for the oxygen reaction A/cm\(^2\)
\( L \) : width of electrode cm
\( M \) : molecular weight g/mol
\( O_2 \) : oxygen
\( P \) : gas pressure atm
\( R \) : gas constant, 82.08 cm\(^3\) atm/mol K or 8,314 J/mol K
\( t \) : time s
\( v \) : velocity of gas or liquid cm/s
\( w \) : mass fraction
\(x\) : direction across the channel and shoulder cm
\(y\) : direction through the electrode cm

**Greek**
\(\rho\) : density of gas g/cm\(^3\)

**Subscripts**
\(O_2\) : oxygen
\(H_2O\) : water
\(N_2\) : nitrogen

1. **Introduction**

Fuel cells are expected to be a promising key technology in the future because they are ecologically friendly using chemical combination of the molecules of a fuel and oxidizer without burning in the process, and advantageous in saving energy. In particular, numerous researches have been conducted on Polymer Electrolyte Fuel Cells (PEFC) used for the power source of fuel-cell vehicles and for an electric power source in the household because of its favorable characteristics such as low operating temperature or high energy density.

The characteristics of its electric power generation depend on the structure, the material used inside the cell and the operating condition. In particular, to prevent the major causes of poor performance of PEFC such as cathode electrode flooding, understanding the electrochemical state inside an operating fuel cell is necessary in addition to optimization of its shape. However, it is not always easy to measure the state by experiments. Therefore, component transport phenomena in fuel cells have been analyzed by numerical computations.

There have been numerous studies regarding the computation of fuel cells. However, only a few studies have already been carried out on interdigitated gas distributors which are expected to be effective for the cathode electrode flooding\(^1\), and its electrochemical state is not thoroughly understood.

In previous studies, Yi and Nguyen\(^2\) conducted two-dimensional analysis of a gas diffusion layer by using finite-difference algorithm, and Berning, et al.\(^3\) conducted three-dimensional analysis of a Proton Exchange Membrane (PEM) fuel cell sheet. In addition, Lum and McGuirk\(^4\) studied the effects of various operating parameters on the performance comparing their results with Yi and Nguyen’s\(^2\). Lin and Nguyen\(^5\) developed two-dimensional two-phase model of a PEM fuel cell.

In the present study, multi-component gas transport phenomena in a cathode electrode layer of PEFC are investigated by using a two-dimensional computational model developed by Yi and Nguyen\(^2\) and our finite element scheme. In the past research\(^6\), we encountered a problem that the solution did not converge when the same settings for physical parameters with them were used in our steady
simulations. In the present study, we extend our simulation from steady to unsteady simulation to avoid the problem. Also, based on the results, the effect of changing permeability in the simulations is discussed with reporting possible errors regarding the value of permeability shown in Ref. 2 at the same time.

2. Modelling of a gas diffusion layer\(^2, 7\)

2.1 Computational domain

By forcing the reactant gases to flow into the electrodes in order to exit as compared to flowing over the surface of the electrodes in conventional parallel-channel gas distributors, the interdigitated gas distributors in effect have converted the transport of the reactant/product gases to/from the catalyst layers from a diffusion mechanism to a forced convection mechanism\(^7\). Computational domain is the part of a cathode gas diffusion layer, i.e., an electrode shown in Fig. 1 with total width of the domain of 0.3 cm, and the value of parameters are shown in Table 1. In the computation, mass conservation law,

\[ \frac{\partial \rho}{\partial t} = -\nabla \cdot (\rho \mathbf{v}), \]  

Darcy's law,

\[ \mathbf{v} = \frac{\kappa}{\mu} \nabla P, \]  

chemical conservation equation where \(i\) represents oxygen or water,

\[ \frac{\partial \rho w_i}{\partial t} = D_i \nabla \cdot (\rho \nabla w_i) - \nabla \cdot (\rho w_i \mathbf{v}), \]  

ideal gas law,

\[ \rho = \frac{MP}{RT}, \]  

relational expression of average molecular weight,

\[ \frac{1}{M} = \frac{w_{O_2}}{M_{O_2}} + \frac{w_{H_2O}}{M_{H_2O}} + \frac{w_{N_2}}{M_{N_2}}, \]  

and relational expression of mass fraction,

\[ w_{O_2} + w_{H_2O} + w_{N_2} = 1 \]  

are assumed. Using these laws, a simultaneous equation is derived in order to obtain unknown functions of gas pressure \(P\), mass fraction of oxygen \(w_{O_2}\) and mass fraction of water \(w_{H_2O}\).

By substituting Eqs. (2) and (4) into Eq. (1), and Eqs. (2), (4), (5) and (6) into Eq. (3), the simultaneous partial differential equations for the unknown functions \(P, w_{O_2}, w_{H_2O}\) can be yielded.
\[ \frac{\partial MP}{\partial t} = \frac{\kappa}{\mu} \nabla \cdot (MP \nabla P) \]  \hspace{1cm} (7)

\[ \frac{\partial MP_{W_3}}{\partial t} = D_{H_2} \nabla \cdot (MP \nabla W_{H_2}) + \frac{\kappa}{\mu} \nabla \cdot (MP \nabla P) \]  \hspace{1cm} (8)

\[ \frac{\partial MP_{H_2,O}}{\partial t} = D_{H_2,O} \nabla \cdot (MP \nabla W_{H_2,O}) + \frac{\kappa}{\mu} \nabla \cdot (MP \nabla P) \]  \hspace{1cm} (9)

where \( M \) is given by

\[ M = \frac{1}{\frac{w_{O_2}}{M_{O_2}} + \frac{1-w_{O_2}}{w_{H_2,O}} M_{H_2,O}} \]  \hspace{1cm} (10)

2.2 Boundary conditions

At the inlet \( \Gamma_1 \), both gas pressure and mass fraction are given.

\[ P = P^m, \hspace{1cm} W_{O_2} = W_{O_2}^m, \hspace{1cm} W_{H_2,O} = W_{H_2,O}^m. \]  \hspace{1cm} (11)

At the supporting section of the separator \( \Gamma_2 \), it is assumed that there is no inflow and outflow in the normal direction of the wall because it touches the solid wall.
\[ \frac{\partial P}{\partial n} = 0, \quad \frac{\partial \omega_{i_2}}{\partial n} = 0, \quad \frac{\partial \omega_{H_i(\ell)}}{\partial n} = 0. \] \tag{12}

At the outlet \( \Gamma_3 \), it is assumed that the pressure is given, and there is no gradient in mass fractions.

\[ P = P^{\infty}, \quad \frac{\partial \omega_{i_2}}{\partial n} = 0, \quad \frac{\partial \omega_{H_i(\ell)}}{\partial n} = 0. \] \tag{13}

At \( \Gamma_4 \) and \( \Gamma_6 \) which are the center points of the channels, it is assumed that there is no flow in the \( x \)-direction and no concentration change.

\[ \frac{\partial P}{\partial n} = 0, \quad \frac{\partial \omega_{i_2}}{\partial n} = 0, \quad \frac{\partial \omega_{H_i(\ell)}}{\partial n} = 0. \] \tag{14}

It is assumed that the following chemical reaction occurs in the catalyst layer \( \Gamma_5 \).

\[ 2H^+ + \frac{1}{2} O_2 + 2e^- \rightarrow H_2O. \] \tag{15}

The relationship between mass fluxes of oxygen and water generated along with electrode reaction and current density can be defined using Faraday's law as follows;

\[ n_{i_2} = \frac{M_{i_2} I}{4F}, \] \tag{16}

\[ n_{H_i(\ell)} = -\frac{M_{H_i(\ell)} (1 + 2 \alpha) I}{2F}. \] \tag{17}

The relational expressions of the mass fluxes in the catalyst layer are given as follows;

\[ n_{i_2} = -\varepsilon \rho \mathcal{D}_{i_2} \frac{\partial \omega_{i_2}}{\partial n} + \omega_{i_2} \left( n_{i_2} + n_{H_i(\ell)} \right) \] \tag{18}

\[ n_{H_i(\ell)} = -\varepsilon \rho \mathcal{D}_{H_i(\ell)} \frac{\partial \omega_{H_i(\ell)}}{\partial n} + \omega_{H_i(\ell)} \left( n_{i_2} + n_{H_i(\ell)} \right) \] \tag{19}

By substituting Eqs. (16) and (17) into Eqs. (18) and (19), boundary conditions of \( \omega_{i_2} \) and \( \omega_{H_i(\ell)} \) in the catalyst layer can be obtained.

\[ \frac{\partial \omega_{i_2}}{\partial n} = \left[ \frac{M_{i_2} - 2(1 + 2 \alpha)M_{H_i(\ell)} \omega_{i_2} - M_{i_2} \omega_{H_i(\ell)}}{4F \varepsilon \rho \mathcal{D}_{i_2}} \right] I, \] \tag{20}

\[ \frac{\partial \omega_{H_i(\ell)}}{\partial n} = \left[ \frac{M_{i_2} - 2(1 + 2 \alpha)M_{H_i(\ell)} \omega_{H_i(\ell)} + 2(1 + 2 \alpha)M_{H_i(\ell)}}{4F \varepsilon \rho \mathcal{D}_{H_i(\ell)}} \right] I. \] \tag{21}

Next, the relationship between current density and oxygen concentration in the catalyst layer is given using the Tafel equation.

\[ I = I_0 \frac{C_{i_2}}{C_{i_2,ref}} \exp \left( \frac{4kF}{RT} \eta \right). \] \tag{22}

The relational expression of oxygen concentration and mass fraction is as follows;

\[ C_{i_2} = \frac{\rho \omega_{i_2}}{M_{i_2}}. \] \tag{23}

Then, by substituting what is obtained by substituting Eq. (23) into Eq. (22), into Eqs. (20) and (21), boundary conditions of \( \omega_{i_2} \) and \( \omega_{H_i(\ell)} \) in the final catalyst layer \( \Gamma_5 \) are given.
\[
\frac{\partial w_{i_{1},l}}{\partial n} \bigg|_{y=h} = \left\{ \frac{M_{i_{1}} - 2(1+2\alpha)M_{H_{2}O}}{4M_{i_{1}},e} \right\} w_{i_{1},l} - M_{i_{1}} \left( \frac{a}{D_{i_{1},l},F} \right) \exp \left( \frac{4kF}{RT} \eta \right).
\]  

(24)

\[
\frac{\partial w_{H_{2}O},l}{\partial n} \bigg|_{y=h} = \left\{ \frac{M_{i_{1}} - 2(1+2\alpha)M_{H_{2}O}}{4M_{i_{1}},e} \right\} w_{H_{2}O},l + 2 \left\{ (1+2\alpha)M_{H_{2}O} \right\} w_{i_{1},l} \left( \frac{a}{D_{i_{1},l},F} \right) \exp \left( \frac{4kF}{RT} \eta \right).
\]  

(25)

where \( a = \frac{h}{\sqrt{\nu_{i_{1}},n_{i_{1}}}} \) is assumed. The relational equation between mass fluxes of oxygen and water and normal velocity is

\[
\varepsilon \rho n_{i_{1}} = n_{i_{1}} + n_{H_{2}O}.
\]  

(26)

Boundary conditions of \( P \) in the catalyst layer can be obtained by substituting Eqs. (2), (16), (17), (22), and (23) into Eq. (26).

\[
\frac{\partial p}{\partial n} \bigg|_{y=h} = \left\{ \frac{M_{i_{1}} - 2(1+2\alpha)M_{H_{2}O}}{4M_{i_{1}},e} \right\} w_{i_{1}},l \left( \frac{a\mu}{kF} \right) \exp \left( \frac{4kF}{RT} \eta \right).
\]  

(27)

Concerning the mathematical formulation showed in this chapter, we basically follow the work of Yi and Nguyen's\(^2\). In their work, it is known that there are some errors regarding the inclusion of an extra porosity term in the terms of the right hand side of Eqs. (18)-(21), (24)-(27). However, it is confirmed that the error doesn't change the present results and discussions.

3. A finite element scheme

Basic equations (7), (8), and (9) and boundary conditions (11), (12), (13), (14), (24), (25) and (27) obtained in the modeling also with initial conditions described below are solved by a numerical scheme, where three stages are included in one time step. The present computations solve the unsteady equations by the time-marching method to obtain the steady solution. This is an intrinsic difference in the numerical methods between the present study, and Yi and Nguyen's\(^2\). Using the \( (n) \)th-step solution, the \( (n+1) \)th-step solution will be derived. The superscript represents the number of the time step. In the time difference term, the lumped mass approximation is used.

\[
\frac{M^{n+1}(\rho^{n+1}) - \rho^{n}}{\Delta t} = \nabla \cdot \left( M^{n}(\rho^{n}) \nabla \rho^{n+1} \right)
\]  

(28)

\[
\frac{M^{n+1}(\mu^{n+1}) - \mu^{n}}{\Delta t} = \nabla \cdot \left( M^{n}(\mu^{n}) \nabla \mu^{n+1} \right) + \sum_{\mu} \nabla \cdot \left( M^{n}(\mu^{n}) \nabla \mu^{n+1} \nabla \mu^{n+1} \right)
\]  

(29)

\[
\frac{M^{n+1}(\nu_{i_{1},l}^{n+1}) - \nu_{i_{1},l}^{n}}{\Delta t} = \nabla \cdot \left( M^{n}(\nu_{i_{1},l}^{n}) \nabla \nu_{i_{1},l}^{n+1} \right) + \sum_{\mu} \nabla \cdot \left( M^{n}(\nu_{i_{1},l}^{n}) \nabla \nu_{i_{1},l}^{n+1} \nabla \nu_{i_{1},l}^{n+1} \right)
\]  

(30)

where
\[ M^{(n)} = \frac{1}{\frac{w^{(n)}_{\Omega_1}}{M_{\Omega_1}} + \frac{w^{(n)}_{H_G,\Omega}}{M_{H_G,\Omega}} + \frac{1 - w^{(n)}_{\Omega_1} - w^{(n)}_{H_G,\Omega}}{M_{N_1}}} \]

and

\[ M^{(n)^*} = \frac{1}{\frac{w^{(n+1)}_{\Omega_1}}{M_{\Omega_1}} + \frac{w^{(n+1)}_{H_G,\Omega}}{M_{H_G,\Omega}} + \frac{1 - w^{(n+1)}_{\Omega_1} - w^{(n+1)}_{H_G,\Omega}}{M_{N_1}}} \]

Eq. (28) is a diffusion equation while Eqs. (29) and (30) are convection-diffusion equations. Each equation is discretized in space by the Finite Element Method (FEM) using the triangular piecewise linear element. The present FEM approach is especially advantageous in treating complex passage shapes in fuel cells. Convergence to the stationary solution is judged by the rates of variation \( \| \tilde{x}^{(n+1)} - \tilde{x}^{(n)} \|_2 / \| \tilde{x}^{(n)} \|_2 \). Here, \( \tilde{x}^{(n)} \) denotes a vector that consists of \( \tilde{P}^{(n)}, \tilde{\omega}^{(n)}_{\Omega_1}, \) and \( \tilde{\omega}^{(n)}_{H_G,\Omega} \), i.e.,

\[ \tilde{x}^{(n)} = (\tilde{P}^{(n)}, \tilde{\omega}^{(n)}_{\Omega_1}, \tilde{\omega}^{(n)}_{H_G,\Omega})^T. \]

(31)

and \( \| \|_2 \) represents the Euclidean norm. When it becomes less than \( 10^{-5} \), the computation is assumed to reach a steady state and is terminated.

To solve the linear simultaneous equations, the asymmetric skyline Gaussian elimination is used. In the computations, the same parameter setting used in the steady computation\(^6\) is adopted. The number of elements and the number of nodes are 1,200 and 671, respectively. Mesh independency of our results has already been confirmed. The time increment of the computation is \( \Delta t = 0.01 \text{ s} \), and the time when the computation is stopped considering that the solution reaches the steady state is 1.52 s. Initial conditions for the variables are set as

\[ P = 1.04 \times 10^5 \text{ atm}, \quad w^{(n)}_{\Omega_1} = 0.21, \quad w^{(n)}_{H_G,\Omega} = 0.00. \]

(32)

The computation was performed on a personal computer equipped with a Pentium4 3.2GHz processor. The time required for each computation was approximately twenty minutes.

4. Results and discussions

Firstly, the permeability of the electrode \( \kappa \) is set to be \( 1.25 \times 10^{-5} \text{ cm}^2 \). This is the minimum value with which we can obtain a solution by the steady computation\(^6\). As shown in Fig. 2, the rates of variation of pressure, oxygen and water decrease with time. Figs. 3, 4 and 5 show distribution of pressure, mass fraction of oxygen and mass fraction of water in the gas diffusion layer respectively comparing the present results with Yi and Nguyen's\(^2\). Concerning the pressure distribution, the variation in the y direction defined in Fig. 1 is small and air flow in the x direction is rather dominant compared to that in the y direction. From Fig. 4, oxygen is consumed due to the reaction in the catalyst
layer as air moves from the inlet to the outlet. From Fig. 5, water is generated due to the reaction. These results agree with those of the steady computation. Also, they are similar to those obtained when \( \kappa \) is \( 9.9 \times 10^{-10} \) cm\(^2\) in the results of Ref. 2.

In the next, the permeability of the electrode \( \kappa \) is set to be \( 9.9 \times 10^{-10} \) cm\(^2\). Figs. 6, 7 and 8 show distribution of pressure, mass fraction of oxygen and mass fraction of water in the gas diffusion layer respectively. From Figs. 4, 5, 7 and 8, the gradients of the distributions of oxygen and water become slightly milder than those when the permeability of the electrode \( \kappa \) is \( 1.25 \times 10^{-5} \) cm\(^2\). On the other hand, the effect of varying the permeability has a great influence on the pressure distribution as is found from the comparison between Figs. 3 and 6. As the permeability becomes smaller, pressure in the catalyst layer becomes larger. In the computation, the same permeability with Ref. 2 is used. However, the resultant pressure becomes clearly different.

<table>
<thead>
<tr>
<th>Table 1. Values of parameters</th>
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<tbody>
<tr>
<td>Inlet channel width ((L_1))</td>
</tr>
<tr>
<td>Shoulder width ((L_2 - L_1))</td>
</tr>
<tr>
<td>Outlet channel width ((L - L_2))</td>
</tr>
<tr>
<td>Electrode height ((h))</td>
</tr>
<tr>
<td>Maximum side length of the triangular mesh</td>
</tr>
<tr>
<td>Permeability ((\kappa))</td>
</tr>
<tr>
<td>Porosity of the electrode ((\varepsilon))</td>
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<tr>
<td>Inlet weight fraction of oxygen ((w^o_{in}))</td>
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<tr>
<td>Inlet weight fraction of water ((w^w_{in}))</td>
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<tr>
<td>Inlet weight fraction of nitrogen ((w^n_{in}))</td>
</tr>
<tr>
<td>Inlet channel pressure ((P^m))</td>
</tr>
<tr>
<td>Outlet channel pressure ((P^{out}))</td>
</tr>
<tr>
<td>Temperature ((T))</td>
</tr>
<tr>
<td>Viscosity of the gas ((\mu))</td>
</tr>
<tr>
<td>Effective diffusion coefficients of oxygen ((D^o_{in}))</td>
</tr>
<tr>
<td>Effective diffusion coefficients of water ((D^w_{in}))</td>
</tr>
<tr>
<td>Constant for current expression ((a))</td>
</tr>
<tr>
<td>Transfer coefficient of oxygen reduction reaction ((k))</td>
</tr>
<tr>
<td>Overpotential ((\eta))</td>
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<tr>
<td>Net water transport coefficient of the membrane ((\alpha))</td>
</tr>
</tbody>
</table>
In Fig. 9, velocity vectors for the permeabilities of $1.25 \times 10^{-5}$ cm$^2$ and $9.9 \times 10^{-10}$ cm$^2$ are compared. In the latter case, an unphysical result where air flows out from the inlet is obtained. The distributions of current density along the electrode width for both permeabilities are compared with that of Ref. 2 as shown in Fig. 10. The result with the permeability of $1.25 \times 10^{-5}$ cm$^2$ also gives a similar result with Ref. 2.

Based on these results, the present authors think that there are possible errors regarding the value of permeability shown in Ref. 2.

5. Conclusion

Multi-component gas transport phenomena in a cathode electrode layer of PEFC are investigated by using a two-dimensional computational model developed by Yi and Nguyen$^2$ and our numerical scheme. In the present study, we have conducted unsteady simulations to avoid the convergence problem we encountered in the past research$^6$. Based on the results, the effects of changing permeability in the simulations are discussed.

The effects on the distribution of oxygen and water are found to be small. However, in the present computations with the same permeability reported in Ref. 2, pressure gradient becomes too large and results are considered to be invalid from a physical point of view. These results show possible errors regarding the value of permeability shown in Ref. 2.

As future works, we are going to extent our method to three-dimensional flows and also two phase flows.

Fig. 2 The rates of variation ($\kappa = 1.25 \times 10^{-5}$ cm$^2$)
Fig. 3 Distribution of pressure
(left: the present result ($\kappa=1.25\times10^{-5}$ cm$^2$), right: results by Yi and Nguyen ($\kappa=9.9\times10^{-10}$ cm$^2$))

Fig. 4 Distribution of oxygen mass fraction
(left: the present result ($\kappa=1.25\times10^{-5}$ cm$^2$), right: results by Yi and Nguyen ($\kappa=9.9\times10^{-10}$ cm$^2$))

Fig. 5 Distribution of water mass fraction
(left: the present result ($\kappa=1.25\times10^{-5}$ cm$^2$), right: results by Yi and Nguyen ($\kappa=9.9\times10^{-10}$ cm$^2$))
Fig. 6 Distribution of pressure ($\kappa=9.9 \times 10^{-10}$ cm$^2$)

Fig. 7 Distribution of oxygen mass fraction ($\kappa=9.9 \times 10^{-10}$ cm$^2$)

Fig. 8 Distribution of water mass fraction ($\kappa=9.9 \times 10^{-10}$ cm$^2$)
Fig. 9 Velocity vectors (left: \( \kappa=1.25 \times 10^{-5} \text{ cm}^2 \), right: \( \kappa=9.9 \times 10^{-10} \text{ cm}^2 \))

Fig. 10 Distribution of current density
(left: the present result; ①: \( \kappa=1.25 \times 10^{-5} \text{ cm}^2 \), ②: \( \kappa=9.9 \times 10^{-10} \text{ cm}^2 \), right: results by Yi and Nguyen (\( \kappa=9.9 \times 10^{-10} \text{ cm}^2 \))
References


