Computational Modeling of the Performance Characteristics of Micro Scale Single-Chamber Intermediate Temperature SOFC System

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Abstract
Performance characteristics of micro scale single-chamber intermediate temperature SOFC (IT-SOFC) has been successfully evaluated by computational modeling. The micro scale SOFC system with composite electrodes and ceria based electrolyte was employed for modeling. The polarization curve was obtained through a series of computational process for structural parameters such as distance between anode and cathode and thickness of electrolyte. Also the effect of the inflow direction of fuel-air mixture on the performance of the fuel cell was quantitatively investigated.

Introduction
Solid oxide fuel cell (SOFC) is highly efficient energy conversion system that converts chemical potential to electrical energy. It can utilize hydrocarbon, CH₄, C₂H₆, C₃H₈, etc. and CO, besides H₂, as a fuel source, and no additional fuel reformer system is required due to high operation temperature. SOFC can be applied to electrical power generation station, auxiliary power source of automobile systems and military applications. Research has mainly focused on the fabrication of various forms of SOFC including tubular and planar types for commercial application. On the other hand, experimental attempts for the development of micro scale SOFC (μSOFC) have been knocking the door to the possibilities as power sources of portable equipments (1,2). For small power generating system, intermediate temperature SOFC (IT-SOFC) with ceria-based electrolyte has been proposed (3-5). Materials such as stainless steel for bipolar plate can be employed in IT-SOFC system and, consequently, low processing cost can be attained. For SOFC system, gas sealing has been a vital issue and the gas sealing amorphous materials need to be selected on the basis of degree of the thermal expansion coefficient matching between components and mechanical strength. In this respect, it is interesting to note that gas sealing problem can be largely mitigated in single-chamber SOFC system since no separation between fuel and air gas is required (6-9).

In SOFC system, computer simulation is known to be very efficient to predict materials properties, reaction kinetics, and unit cell performance. Theoretical modeling of SOFC can be divided three parts. Firstly, basic properties such as ionic conductivity of electrolyte materials can be predicted by molecular dynamics or ab initio approaches (10-12). Secondly, the reaction kinetics at three phase boundary (TPB) in the electrode can be simulated by micro modeling (13-15). Lastly, the detail picture of fluid dynamics, mass...
transport and heat conduction/convection in SOFC system can be obtained through macro modeling. The overpotentials of individual SOFC cell can be successfully predicted through a series of macro modeling (16-18).

In this work, fluid motion, diffusion of fuel-air mixture and oxygen ion conduction was considered in the macro modeling scheme utilized by the 3-dimensional finite element method (FEM). Next, ohmic loss, activation loss and concentration loss was obtained through a series of macro modeling and, consequently, the performance characteristics of micro scale single-chamber IT-SOFC were quantitatively investigated.

**CALCULATION METHODS**

For the simulation, Hibino’s B type single-chamber SOFC (8, 9) was employed (Fig. 1), and the dimensions of the model for computer simulation are listed in Table 1.

![Figure 1. Schematic diagram of calculation model, (a) side view, (b) top view.](image)

| Symbol | SOFC components       | Dimension |
|--------|-----------------------|-----------|
| E_L    | Electrolyte length    | 1000 \(\mu\)m |
| E_W    | Electrolyte width     | 160 \(\mu\)m |
| E_T    | Electrolyte thickness | 20 \(\mu\)m |
| A_L    | Anode length          | 900 \(\mu\)m |
| A_W    | Anode width           | 20 \(\mu\)m |
| A_T    | Anode thickness       | 20 \(\mu\)m |
| C_L    | Cathode length        | 900 \(\mu\)m |
| C_W    | Cathode width         | 20 \(\mu\)m |
| C_T    | Cathode thickness     | 20 \(\mu\)m |
| G_H    | Height of hypothetical gas channel | 200 \(\mu\)m |
The fully dense and nonporous electrolyte was assumed and no penetration of gas species into the electrolyte was allowed. Electrodes with uniform 100 nm diameter pores exhibited electronic and ionic conducting properties. It was also assumed that the oxidation of fuel ingredient of gas mixture is only occurred at the anode, and the reduction of oxygen is only occurred at the cathode. Preceded modeling had assumed that TPB where the electrode reaction was occurred was defined as at the interface between electrolyte and electrode, because most electrode reactions were occurred within 50 μm range in the interface region of electrolyte and electrode (18). In this modeling, however, TPB was assumed to be the whole electrode volume because the thickness of electrodes was less than 50 μm for this micro scale SOFC. For the simplicity of calculation, only hydrogen fuel was considered. Corresponding electrode reactions can be represented as Eqns. [1-2]:

\[
\text{Anode : } 2\text{H}_2 + \text{O}_2 = 2\text{H}_2\text{O} + 4e^- \tag{1} \\
\text{Cathode : } \text{O}_2 + 4e^- = \text{O}_2^2^- \tag{2}
\]

Injected gas mixture was composed of nitrogen, oxygen and a small amount of water vapor, and temperature of the system was isothermally fixed at 773 K. Each loss - ohmic loss, activation loss and concentration loss - was obtained from the voltage drop calculation due to ionic conduction, the finding roots of Butler-Volmer equation, and the concentration distribution of each species resulting from the diffusion and convection in the anode, cathode and gas channel. Detailed procedure of performance characteristics modeling from loss calculation was depicted in Fig. 2. Finally, theoretical Open Circuit Voltage (OCV) could be obtained by the Nernst-equation (19).

![Figure 2. Flow chart of performance characteristics analysis.](image-url)
Ohmic Loss

Ohmic loss is the voltage drop caused by the resistance of migrating oxygen ions and electrons in the electrodes. In this work, only ohmic loss from the ionic conduction was considered since the magnitude of ohmic loss originated from the electronic conduction is rather negligible. Using the value of conductivity of oxygen ion in the electrolyte and electrodes at 773 K, Equation [3] was solved, numerically, and the voltage should be obtained.

\[- \nabla \cdot (\sigma \nabla V) = 0\]  

[3]

Activation Loss

Activation loss is due to the overcoming energy barrier when electrons released during the electrochemical reaction. Therefore, activation loss depends on temperature, fuel species, electrode materials, oxygen partial pressure and so on. Activation loss of IT-SOFC becomes more critical compared to high temperature SOFC. Butler-Volmer equation [4] represents the relation between activation loss and current density.

\[ i = i_0 \left[ \exp\left( \frac{\alpha_a F V_{act}}{RT} \right) - \exp\left(- \frac{\alpha_e F V_{act}}{RT} \right) \right], \quad \alpha_a + \alpha_e = 1 \]  

[4]

Exchange current density \( i_0 \), charge transfer coefficient \( \alpha_a \) and \( \alpha_e \) were referred to H. Zhu’s results (16). The numerical solution for the activation loss \( V_{act} \) was obtained by Newton-Raphson method (22).

Concentration Loss

Differences in concentration of each species between gas channel and TPB lead to concentration loss. In previous results (18), the gas velocity in the porous electrodes was set to be zero. However, in this work, convective diffusion was considered by calculating velocity of gas fluid inside both electrodes. Inlet gas mixture was composed of \( \text{H}_2 : \text{N}_2 : \text{O}_2 : \text{H}_2\text{O} = 0.5 : 0.38 : 0.105 : 0.0105 \) in mole fraction and supposed to obey the ideal gas law, and amount of 180sccm (\( \text{H}_2 : 90\text{sccm}, \text{air} : 90\text{sccm} \)) was injected to gas channel. To calculate velocity distribution in the gas channel and porous electrodes, steady-state incompressible Navier-Stokes equation [5] and Darcy’s law [6] were solved numerically.

\[ \rho (u \cdot \nabla u) - \nabla \cdot \eta \left( \nabla u + (\nabla u)^T \right) + \nabla p = 0 \]  

[5]

\[ \nabla \cdot u = 0 \]

[6]

Permeability \( \kappa \) of anode and cathode was assumed to be \( 1.0 \times 10^{-12} \) m\(^2\). Finally, from the velocity distribution, steady-state diffusion and convection equation [7] was solved to obtain the concentration distribution of reactants and products in the gas channel and electrodes.

\[ \nabla \cdot (-D_i \nabla c_i + c_i u) = R_i \]  

[7]
Table 2 shows the value of employed diffusivity ($D_i$) of each species in the channel and porous media. The diffusivity were calculated from Hirschfelder model based on Lennard-Jones potential (23). In particular, when the diffusivity in the porous electrode were calculated, Knudsen diffusion as well as Bruggman model was considered to represent the actual porous media consisting of randomly distributed tortuous pores.

| Table 2. Diffusivity of each species at 773 K, 1 atm. [cm$^2$/s]. |
|-----------------|-----------------|
|                 | Channel         | Porous electrode |
| H$_2$           | 2.813           | 0.252            |
| N$_2$           | 2.366           | 0.081            |
| O$_2$           | 0.559           | 0.059            |
| H$_2$O          | 0.757           | 0.079            |

Volumetric reaction rate ($R_v$) was defined as a mass quantity of reactants and products per unit volume per second, Eqn. [8]:

$$R_v = \pm \frac{v_i}{4F} \times \text{Area of Reaction Site (ARS)} \times \frac{1}{\text{Electrode Volume}} \times M_i$$

$R_v$ is positive if species $i$ is product (H$_2$O), and it is negative if species $i$ is reactant (H$_2$, O$_2$). $v_i$ is the stoichiometric coefficient of reactions i.e. $v_{H_2O} = 2$, $v_{H_2} = v_{O_2} = 1$.

Area of Reaction Site (ARS) could be calculated from the permeability value. From the calculated concentration distribution, the concentration loss was calculated through the Eqns. [9-10] (16).

$$V_{con,a} = \frac{RT}{4F} \left( 2 \ln \frac{[H_2]_{channel}}{[H_2]_{TPB}} - 2 \ln \frac{[H_2O]_{channel}}{[H_2O]_{TPB}} \right)$$

$$V_{con,o} = \frac{RT}{4F} \left( \ln \frac{[O_2]_{channel}}{[O_2]_{TPB}} \right)$$

**RESULTS AND DISCUSSION**

Figure 3(a) shows the calculated loss corresponding to the variation of current density. In the same manner as other type of SOFC, major loss came from the ohmic loss caused by oxygen ion diffusion. On the other hand, concentration loss was minimal due to the reduced concentration difference between inside electrodes and gas channel since whole volume of electrodes served as reaction sites. Corresponding polarization curve, directly
related to the performance of SOFC, was obtained by subtracting all the calculated losses from the theoretical open circuit voltage (1.133 V), Fig. 3(b).

Next, the effect of the distance between anode and cathode on the polarization curve was quantitatively investigated (Fig. 4(a)). As decreasing the distance between anode and cathode, power density of cell was increased due to the reduction of ohmic loss. (Fig. 4(a)). This agrees well with empirical expectation and is consistent with experimental reports (8, 9).

![Figure 3. (a) Calculated losses as the current density variation (b) Calculated polarization curve when the distance between electrodes, thickness of electrolyte were 20 μm and 20 μm, respectively.](image)

Figure 4(b) shows the polarization curve according to the variation of the thickness of electrolyte. In contrast to Hibino's A type single-chamber SOFC (8, 9), interestingly, the single-chamber SOFC adapted in this study (B type) shows higher power density with thicker electrolyte. This was due to decreasing ohmic loss when the thickness of electrolyte was increased. And it can be concluded that in the manufacturing step of B type single chamber SOFC, thicker electrolyte can be employed without deteriorating the performance characteristics, and no additional support materials for the required mechanical strength is required.

Figure 4(c) shows the polarization curve as the variation of the direction of inflow gas mixture. The highest power density was found when gas mixture was injected into the vicinity of cathode in the direction perpendicular to the electrode axis. This observation can be understood by considering the difference of diffusivity. The diffusivity of O₂ in cathode is much lower than that of H₂ and H₂O in anode, therefore, migration of oxygen gas in the cathode was significantly retarded. However, when inflow gas was injected to the cathode side, the velocity of gas fluid in the cathode was higher than that of the gas fluid in anode, this increment of fluid velocity enhanced convective diffusion of oxygen in the cathode and, eventually, lowered the concentration loss. On the other hand, when the inflow gas direction was parallel to the electrode axis, the power density was predicted to be significantly lowered because the concentration difference of the gas species between the inlet and outlet regions of the SOFC system was extensive, originating from the fact that the length of both electrodes was 45 times longer than the width and height.
Figure 4. Polarization curve corresponding to (a) the distance between two electrodes, (b) the electrolyte thickness, (c) the inflow gas direction.

CONCLUSIONS

Computer simulation based on macro modeling of multi-physical phenomena including ionic conduction, fluid dynamics and gas diffusion was successfully performed, and consequently, ohmic loss, polarization loss and concentration loss were quantitatively derived from the simulation results. The performance of micro scale single chamber SOFC system was found to be largely dependent on the structural parameters, the distance between electrodes and the thickness of electrolyte, and the inflow direction of gas mixture. In general, the better performance can be shown with the narrow interdistance between electrodes and with the thicker electrolyte, in the structural viewpoint. And when the inflow direction is perpendicular to the electrode axis, the specific micro scale single-chamber SOFC cell turned out to represent better performance. It is expected that the developed computational scheme in this study, can serve as an important guideline for manufacturing the micro scale single-chamber IT-SOFC system and optimizing the performance characteristics.

NOMENCLATURE

\[ u, v, w : x, y, z \text{-component of velocity in channel [m/s]} \]

\[ u2, v2, w2 : x, y, z \text{-component of velocity in electrodes [m/s]} \]
$P$: pressure in channel [Pa]

$P_2$: pressure in electrodes [Pa]

$c_i$: concentration of species $i$ [kg/m$^3$]

$i_e$: current density [A/m$^2$]

$V$: voltage [V]

$\sigma$: conductivity [S/m]

$i_0$: exchange current density [A/m$^2$]

$\alpha_a$: charge transfer coefficient at anode

$\alpha_c$: charge transfer coefficient at cathode

$F$: Faraday constant [C/mol]

$V_{\text{act}}$: activation loss [V]

$\rho$: density [kg/m$^3$]

$R$: gas constant [J/mol-K]

$T$: temperature [K]

$u$: velocity vector [m/s]

$\eta$: dynamic viscosity [Pa-s]

$\kappa$: permeability [m$^2$]

$D_i$: diffusivity of species $i$ [m$^2$/s]

$R_i$: volumetric reaction rate of species $i$ [kg/m$^3$.s]

$\nu_i$: stoichiometric coefficient of species $i$

$M_i$: molar weight of species $i$ [kg/mol]

$V_{\text{con.a}}$: concentration loss in anode [V]

$V_{\text{con.c}}$: concentration loss in cathode [V]

$[i]_{\text{channel}}$: concentration of species $i$ at the gas channel [mol/m$^3$]

$[i]_{\text{TPB}}$: concentration of species $i$ at three phase boundary [mol/m$^3$]

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