IMPEDANCE MEASUREMENT AND SIMULATION ON A DISC-TYPE SOFC UNDER POWER GENERATION

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ABSTRACT

AC impedance characteristics under practical loading conditions were numerically calculated and the measurements under DC current conditions were carried out using actual size planar disc-type solid oxide fuel cells (SOFCs) employing doped lanthanum gallate as a solid electrolyte. Under the conditions of relatively high fuel utilization, the change in composition of the gases caused by the electrochemical fuel cell reaction leads to the appearance of gas conversion impedance, and considerable variation exists in Nernst potential and current density along the gas flow axis. A semi-circular behavior that can be represented by an RC parallel circuit was also predicted by the simulation. Dependency of R and C on fuel utilization was calculated, and experimentally measured behavior was compared with the simulated results and discussed.

INTRODUCTION

AIST has started the work on “the development of the method to measure the efficiency of SOFC with sufficient accuracy” since FY 2001 under the project METI. In this work, with the collaboration of KEPCO and MMC, we have started electrochemical measurements using practical scale disc-type planar SOFC developed by Oita University, KEPCO and MMC using doped LaGaO₃ as a solid electrolyte.

Although the understanding of the dynamics of SOFC in practical conditions is very important in pushing SOFC into practical use, relatively few AC impedance measurements have been reported using large-scale SOFC under large current conditions. Presumably, this is because of the inherent difficulties in the experimental measurement and in the interpretation of the impedance data.

In this study, using a relatively large-scale cell that has a simple geometrical structure for the simulation, we have measured the AC impedance under large current and practical fuel utilization conditions. Numerical simulations of the impedance were also conducted and the results compared and discussed.
EXPERIMENTAL

The experimental cell setup is shown in Figure 1. The disc-type cells using La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.15}Co_{0.05}O_3 as solid electrolyte were used as described elsewhere (1). The thickness of the electrolyte was ca. 0.2 mm, and its diameter was 100 mm. Ni/Ce_{0.8}Sm_{0.2}O_{1.9} cermet was used as anode and Sm_{0.5}Sr_{0.5}CoO_3 as cathode. The effective area of the cell is 56.8 cm² (85 mm dia.). Silver paste was painted on the outer surface of the cathode side of the electrolyte, used as a reference electrode. The temperature of the cell was measured by chromel-alumel thermocouples (0.5 mm dia.), which were inserted into each current collector about 10 mm deep toward the center of the disc.

The AC impedance measurements were carried out by superimposing sinusoidal current from 10 kHz to 10 MHz onto DC current using a potentiogalvanostat (Solartron 1287) and a frequency response analyzer (Solartron 1255). AC current was detected through a clamp sensor (Hioki 3275). Voltages between anode and cathode, anode and reference electrode, and cathode and reference electrode were simultaneously recorded digitally (Yokogawa DL708E) and used to calculate the impedance between each electrode by Fourier analysis. It was confirmed that identical results were obtained by the two methods. The impedance of the reference electrode was confirmed to be sufficiently small.

Dry hydrogen and air were supplied to the cell. The flow rate of air was controlled to be five times that of hydrogen. The temperature of the cell was kept at 750°C.

SIMULATION MODEL

Assumptions of the Model

The schematic diagram of a disc-type SOFC as a simulation model (2) is shown in Figure 2. The single cell consists of a composite disc made of a solid electrolyte and electrodes, structured current collectors, and metal end plates for positive and negative electrodes. Hydrogen as fuel and air as oxidant are supplied through orifices at the center of each end plate, and the structured current collectors enabled uniformly distributed axially symmetric gas flow. It is assumed that the solid electrolyte and the current collectors are homogeneous with uniform thickness. Although cell reactions take place on the electrolyte surface, it is assumed that the concentrations of chemical species are constant along the z direction due to perfect mixing by diffusion. It is also assumed that the pressure drop in the channel is small and the total pressure therefore constant. For further simplicity, the temperature is assumed to be uniform. This assumption can be approximately realized by externally heating the end plates during the experiment. The electrical conductivities of current collectors are high enough, and hence the cell potential, \( V_{cell} \), between cathode and anode, is constant everywhere in the cell. And finally, the local cell impedance is assumed to be constant everywhere.

In general, the equivalent circuit of the SOFC is dependent on AC frequency range, cell and electrode materials, and their constructions. A set of representative equivalent circuits is shown in Figure 3. In this study, a set of calculations is carried out using the simplest
equivalent circuit, which consists of a pure resistance, $R_{cell}$, and an electromotive force, $E_{emf}$, as shown in the figure (a). The solid electrolyte such as doped LaGaO$_3$ has electronic conductance as well as ionic conductance. So that variations of the equivalent circuit shown in Figure 3 (b) are also examined, considering reaction resistance $R_r$, double layer capacitance $C_d$, electronic resistance $R_e$, ionic resistance $R_i$ and series ohmic resistance $R_s$.

**Governing Equations**

Basic equations governing the system are as follows. The electromotive force, $E_{emf}$ can be obtained from

$$E_{emf} = -\frac{\Delta G_{H_2O}}{2F} + \frac{RT}{2F} \ln \frac{p_{H_2}p_{O_2}^{0.5}}{p_{H_2O}}$$  \[1\]

where $\Delta G$ = Gibbs free energy, $F$ = Faraday constant, $R$ = gas constant, $T$ = temperature, $p_{H_2}$ = hydrogen partial pressure, $p_{O_2}$ = oxygen partial pressure, and $p_{H_2O}$ = water partial pressure. The electric circuit equation for the equivalent circuit depicted in Figure 3(a) is

$$J_r = \frac{E_{emf} - V_{cell}}{R_{cell}}$$  \[2\]

where $J_r$ = current density with cell reaction and $R_{cell}$ = area specific cell. The equation for the equivalent circuit in Figure 3 (b) is omitted here because of its complexity. However, the relationship between $J_r$, $E_{emf}$ and $V_{cell}$ can easily be obtained for a given circuit. In addition, the cell voltage $V_{cell}$ is given by

$$V_{cell} = V_{dc} + V_{ac} \sin(2\pi ft)$$  \[3\]

where $V_{dc}$ = DC cell voltage, $V_{ac}$ = amplitude of applied ac voltage, $f$ = frequency, and $t$ = time.

Transport equations for molar concentrations of hydrogen, water and oxygen considering the diffusion in the flow direction are

$$\frac{\partial c_{H_2}}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} \left( v_r c_{H_2} - D_{H_2} \frac{\partial c_{H_2}}{\partial r} \right) = -\frac{J_r}{2Fh_f}$$  \[4\]

$$\frac{\partial c_{H_2O}}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} \left( v_r c_{H_2O} - D_{H_2O} \frac{\partial c_{H_2O}}{\partial r} \right) = \frac{J_r}{2Fh_f}$$  \[5\]

$$\frac{\partial c_{O_2}}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} \left( v_a c_{O_2} - D_{O_2} \frac{\partial c_{O_2}}{\partial r} \right) = -\frac{J_r}{4Fh_a}$$  \[6\]

where $c$ = molar concentration, $v$ = flow velocity, $D$ = diffusion coefficient, $r$ = radius, and $h$ = effective channel height. Suffixes $H_2$, $H_2O$, $O_2$, $f$, and $a$ = hydrogen, water, oxygen, fuel, and air, respectively. Furthermore, the continuity may be described as:
\[ \frac{1}{r} \frac{\partial}{\partial r} (rv_r) = 0 \quad [7] \]

\[ \frac{1}{r} \frac{\partial}{\partial r} (rv_a) = \frac{RT}{p} \frac{J_r}{4Fh_a} \quad [8] \]

where \( p \) = total pressure. Finally, the relationship between partial pressure \( p_M \) and molar concentration \( c_M \) of chemical species M is given as follows:

\[ p_M = c_M RT \quad [9] \]

**Numerical Method and Conditions in Calculation**

The above-mentioned partial differential equations were solved numerically using the implicit finite difference method. For the simplification of the calculation, a no-flux boundary condition was assumed for the inlet and outlet of the channels. The stationary solution for the DC case was calculated by assuming the open circuit as an initial condition. Then the result was used as an initial condition for the case where AC current superimposed on DC current. The load current was calculated by integrating the current distribution over the whole cell area as a function of time. The Fourier transforms of the load current and the applied AC voltage were used to calculate the total cell impedance.

The numerical setup of the standard case is shown in Table 1. The condition is based on the experimental setup for the impedance measurement. Additional parameters are AC voltage, load current, and fuel utilization. Because these parameters are not independent, fuel utilization factor \( U_f \) defined below will be mainly used in the following.

\[ U_f = \frac{I_L}{2Fm_{H2}} \quad [10] \]

where \( I_L \) = load current and \( m_{H2} \) = molar flow rate of supplied hydrogen. The calculation was carried out with an applied AC voltage of amplitude of 1 mV for a range of frequencies

**Table 1. Parameters Used for Calculating the Standard Case.**

| parameter                      | standard case |
|--------------------------------|---------------|
| \( D_{eff} \)                  | 8.5 cm        |
| Channel effective height \( h_i \) | 0.1 cm        |
| \( h_a \)                      | 0.1 cm        |
| Fuel flow rate \( G_f \)       | 3 Ncc/min/cm² |
| Air flow rate \( G_a \)        | 15 Ncc/min/cm²|
| Temperature \( T \)            | 1023.15 K     |
| Total pressure \( p \)         | 0.1013 Mp     |
| Fuel utilization \( U_f \)     | 80%           |
| Local cell impedance \( R_{cell} \) | 0.5 Ω cm²    |
| Diffusion coefficient \( D_f \) | 7.06 cm²/s    |
| \( D_a \) fuel                 | 1.65 cm²/s    |
| Inlet \( H_2 \) mole fraction in fuel | 99%          |
| Inlet \( O_2 \) mole fraction in air | 20.95%      |
| Inlet \( N_2 \) mole fraction in air | 79.05%      |

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between 100 Hz and 0.01 Hz. The cell was meshed with 100 elements and time step $\Delta t$ was chosen between 0.1 ms and 5 ms depending on the frequency. The diffusion coefficient was evaluated using the Chapman and Enskog equation as a function of temperature and pressure (3).

RESULTS AND DISCUSSION

The distribution of $E_{emf}$, current density, $p_{H2}$, $p_{H2O}$ and $p_{H2}$ were calculated along the r axis and plotted in Figure 4. In the calculation the electronic conductance of the electrolyte was neglected and fuel utilization was set at 80%. In the figure two conditions were considered, one without the diffusion of gases along the flow axis and one with the diffusion. It can be seen that the current is concentrated at the central region of the disc. The concentration of the current was brought about by the drop of $E_{emf}$ toward the outer edge of the disc. The effect of the gas diffusion is quite noticeable in the anode gas concentrations. Steam, which is the product of anode reaction, tends to diffuse back into the center of the disc, while hydrogen accelerates and flows rapidly to the outside of the cell. These phenomena make $E_{emf}$ smaller throughout the cell, which in turn makes cell performance lower. Because the flow rate of air was assumed to be five times that of fuel, the variation of $p_{O2}$ is not so remarkable compared with the gaseous species in the fuel channel.

AC impedance behavior at the standard condition was calculated and plotted in Figure 5 at various $U_f$ conditions. Although pure resistance was assumed and capacitive elements were excluded for the local cell impedance for the standard condition, the semicircular behaviors were predicted. The behaviors can be represented by an RC parallel circuit and are shown by the solid lines in the figure.

Primdahl et al. (4) have demonstrated the appearance of such a semicircular behavior caused by the shift of Nernst potential via electrochemical reactions in SOFC anode when using a very small cell over which perfect gas mixing was assumed. The semicircular behavior they called gas conversion impedance is represented by an RC parallel circuit where $R$ and $C$ can be expressed as follows.

\[
R_s = \left( \frac{RT}{2F} \right)^2 \left( \frac{1}{p_{H2O}} + \frac{1}{p_{H2}} \right) \frac{1}{N} \tag{11}
\]

\[
C_s = \left( \frac{2F}{RT} \right)^2 \frac{V}{1 + \frac{1}{p_{H2O}} + \frac{1}{p_{H2}}} \tag{12}
\]

where $N$ represents the gas flow rate, $V$ is the volume of the gas over anode related to the anode reaction, and others have their usual meanings.

Although the concentration of current density to the center of the disc increases as $U_f$ becomes larger, as can be expected by plotting the same type of figure as Figure 4 at various $U_f$, the high frequency side interception to the real axis does not change with $U_f$. As expected from Eq. [11], the calculated semicircles in Figure 5 show apparent dependency

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on \( U_f \), i.e., at low \( U_f \), \( pH_2O \) takes a small value and at high \( U_f \), \( pH_2 \) takes a small value making \( R_g \) in Eq. [11] large, while in the middle \( U_f \) region the diameter of the semicircle becomes minimal. Although Eq. [11] and [12] were derived by assuming an ideally small electrode, the same type of relation was found to be predictable in our standard condition, that is, with the cell having extensive electrode area and large in-plane gas distributions.

The time constant of the semi-circle is related to the time required for the gases to pass through the volume over the electrode and can be expressed as follows:

\[
t = \frac{V}{N}
\]

Calculated time constant for the current cell is 0.27 sec, which is close to the value (0.4 sec) obtained from the characteristic frequency (\( Z'' = \text{min.} \)) in Figure 5. The characteristic frequency in Figure 5 remains almost the same by the change in \( U_f \), which is discussed later.

The same type of dependency as in Figure 5 was calculated in the case where lateral diffusion was not considered, and was plotted in Figure 6. The comparison of these figures clearly indicates the negative effect of the lateral diffusion, which was also expected by the calculated \( E_{\text{emf}} \) distribution (Figure 4). The calculated output voltage in the case of Figure 4 was 0.787 V without diffusion and 0.757 V with diffusion.

It seems from Eq. [11] that the fuel utilization at which the diameter of the semicircle becomes minimum is 50%, i.e., \( pH_2O = pH_2 = 0.5 \), although the minimum \( U_f \) points appearing in Figures 5 and 6 are slightly shifted from 50%. But it has to be kept in mind that Eq. [11] was derived assuming ideal gas mixing, and no change of gas concentration was assumed substantially, while in this calculation the composition of the inlet and outlet gas are not the same because of the DC current at a finite cell size. Hence the situation cannot be treated so simply. As a whole, it can be said that the tendency is in fairly good agreement despite the difference of the model. The time constant (0.25 sec) calculated from the characteristic frequency in Figure 6 is slightly less than that obtained from Figure 5 but is in good agreement with 0.27 sec obtained from Eq. [13].

The comparison of \( E_{\text{emf}}, J_f, pH_2, pH_2O, \) and \( pO_2 \) distributions with and without electronic conduction in the electrolyte is plotted with \( r \) in Figure 7. Because the electrolyte used for our experiment is doped LaGaO\(_3\) with a small amount of electronic conduction, the investigation of the effect of electronic conduction on cell performance is of great importance. The comparative analysis was carried out with the standard condition and condition d of Table 2; that is, the transference number of electronic conduction equals 0.024.

The effect of the electronic conduction on \( E_{\text{emf}} \) at the entrance region is only trifle while it is extremely remarkable on \( J_f \) throughout the whole cell. Since the plotted \( J_f \) here is the one including internal short circuit current \( J_s \) as can be depicted in the equivalent circuit of Fig.3(b), the increase of \( J_f \) as compared to the one without electronic conduction is almost due to the superimposition of internal short circuit current. Because of the internal short circuit, hydrogen is consumed more rapidly, and hence more steam is generated. At the exit region hydrogen is almost exhausted, and, accordingly, the contribution of electronic short
Table 2. R and C Values Used for Calculating Figure 8.

| Case | a | b | c | d | e |
|------|---|---|---|---|---|
| Equivalent circuit in Fig 8 | (a) | (b) | (b) | (b) |
| $R_{\text{cell}}$ ($\Omega \text{cm}^2$) | 0.5 | 1 | - | - | - |
| $R_i$ ($\Omega \text{cm}^2$) | - | - | 0.5 | 0.25 | 0.25 |
| $R_r$ ($\Omega \text{cm}^2$) | - | - | 0 | 0 | 0.1 |
| $C_d$ (F/cm$^2$) | - | - | 0 | 0 | 0.16 |
| $R_g$ ($\Omega \text{cm}^2$) | - | - | 12.5 | 10 | 10 |
| $R_e$ ($\Omega \text{cm}^2$) | - | - | 0 | 0.25 | 0.15 |

circuit current in the region to the total current density is quite large. $E_{\text{emf}}$ in the region shows a considerable decrease, and the output voltage is lower. The calculated output voltage is 0.675 V, considerably lower than the standard condition, indicating the significance of electronic conduction in practical applications. The effect of electronic conduction on $p_{O_2}$ is not so remarkable in this case because of the low air utilization assumed for the calculation.

AC impedance behaviors at $U_f = 80\%$ were calculated, changing various parameters in the equivalent circuits listed in Table 2 and plotted in Figure 8. In case b, where only $R_{\text{cell}}$ is doubled compared with the standard case a, only a parallel shift of the curve is observed. In case c where parallel electronic conduction is additionally considered, the high-frequency side interception shifts slightly toward zero, and most of all the diameter of the semicircle becomes considerably larger. The behavior seems to be more exaggerated in case d, where less resistance was assumed for electronic conduction.

In case e, a parallel circuit of an electrical double-layer capacitance and an electrochemical reaction resistance was assumed and added to the equivalent circuit. Because a rather large capacitance value was assumed for the electrical double layer due to the frequency range used for the calculation, only one additional bump appeared at the high-frequency side. The figure would spread into two separate semicircles if more realistic capacitance were to be assumed for the double-layer.

The time constant obtained for each condition differs slightly. The difference in time constant and diameter of the semicircle can be explained by plotting them against effective fuel utilization instead of apparent $U_f$. In the case where electronic short-circuiting occurs, substantial fuel utilization is thought to be higher than the apparent one. Effective fuel utilization ($U_{f,e}$) is then defined by replacing $I_f$ with $I_e$ in Eq. [10], $I_e$ being the integrated $I_f$ over the whole cell. For plotting, $E_{\text{emf}}$ in Figure 3 were replaced by an $R_g$ and $C_g$ parallel circuit, and the calculated semicircles were fitted using properly chosen $R_g$ and $C_g$ values of a corresponding equivalent circuit. Figure 9 shows the dependency of thus calculated $R_g$ and $\tau_g (= R_g C_g)$ on $U_{f,e}$. The general dependency of $R_g$ on $U_{f,e}$ can be predicted from Eq. [11], while the dependency of $\tau_g$ on $U_{f,e}$ could not be predicted from Eq. [13], which indicates the invariability of $\tau_g$ on $U_f$. Anyway, the significant difference of $R_g$ and $\tau_g$ at each condition depicted in Figure 8 was attributed to the difference of $U_{f,e}$ except for case b, where minor deviations were observed. In general, the results confirm the independent behavior of the gas conversion impedance from the local cell impedance.
Experimental results corresponding to the simulated case depicted in Figure 5 are shown in Figure 10. The measurement was conducted by flowing a constant amount of hydrogen (3 ml/min/cm²) and air (15 ml/min/cm²) and adjusting the DC current so the fuel utilization given by Eq. [10] takes each noted value. In the high-frequency side of each curve there appears to be additional impedance behavior that is thought to originate from an electrochemical reaction at the electrodes. Even though the impedance behaviors plotted in Figure 10 are obtained by measuring the cell impedance as a whole, the simultaneous measurement using a reference electrode has revealed that the cathode impedance is insignificant and measured impedance behavior almost reflects that of anode behavior.

As expected, impedance shows apparent dependency on \( U_f \), and the diameter of the semicircle \( (R_g) \) seems to be minimum around \( U_f = 23.3\% \). The difference of minimum \( U_f \) predicted from Figure 5 and seen here is attributed to electronic conduction in the real cell. Effective fuel utilization in our cell is estimated as quite a bit larger than the apparent one.

Another interesting observation in Figure 10 is the high-frequency side interception shifting toward the high-impedance side as \( U_f \) becomes larger. The concentration of current to the center of the disc at higher fuel utilization does not bring this sort of phenomenon. From the results of another set of experiments where, by altering the flow rate of hydrogen and changing the current density at each measurement so the fuel utilization was kept constant, the shift was shown to be brought about by the difference in gas concentration distribution and not by the difference in current density. The high-frequency side interception in this case is thought to include the resistance of the electrolyte and electrode and contact resistance between electrode and current collector. None of the resistance seems to explain the shift. A possible explanation is that the impedance obtained here is not settled at the high-frequency region used in this experiment and that the high-frequency interception still includes impedances originating from electrode reaction. The time constants obtained from Figure 10 also shows minimum at the middle \( U_f \) range as predicted by calculations.

The impedance behavior at various \( U_f \) obtained by keeping the current density at 0.3 A/cm² and changing the hydrogen flow rate is also shown in Figure 11. The high-frequency side interception shifts the same way described above. Because the gas conversion impedance expressed by Eq. [11] is inversely proportional to the gas flow rate, and because the increase in gas flow rate makes the effect of lateral gas diffusion less effective, interpreting the results of Figure 11 can be rather complex. Although the inductive behavior at the low-frequency limit appearing on some curves in Figures 10 and 11 indicates another mechanism for the appearance of low-frequency semicircles, as a whole, gas conversion impedance explains the low-frequency behavior quite well in practical cell operation.

Experiments using larger cells are being done where gas composition is controlled by controlling hydrogen and steam flow rates separately. The effect of gas composition on impedance behavior will be investigated more systematically in the future.

**CONCLUSIONS**

AC impedance behavior of practical-size SOFCs under DC current conditions was numerically simulated. The change in Nernst potential caused by the SOFC electro-
chemical reactions was predicted to be responsible for the semicircular behavior in the complex impedance plot, including the case in which the composition of the inlet and outlet gases differs considerably. $R$ and $C$ values, by which the impedance behavior can be represented, depend on fuel utilization; dependency was estimated by the calculations. These values were found to be treated systematically by plotting them against effective fuel utilization instead of the apparent one. Experimental results also show that the semicircular impedance predicted by the simulation is predominant in the practical SOFC applications. In the experimentally obtained impedance plot, the high-frequency side interception was found to shift toward high impedance values as fuel utilization became larger, which was attributed to the change in gaseous species’ concentration distribution in the cell.

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Figure 1. Schematic of experimental cell configuration.
Figure 2. Calculation model of the SOFC.

Figure 3. Equivalent circuit used for the calculation.
Figure 4. Distribution of $E_{emf}$, $J_n$, $p_{H_2}$, $p_{H_2O}$ and $p_{O_2}$ with and without considering lateral gas diffusion at the standard condition.

Figure 5. Calculated impedance behavior against fuel utilization for the standard case listed in Table 1.
Figure 6. Calculated impedance behavior against various $U_f$ at the standard case only without lateral diffusion.

Figure 7. Distribution of $E_{emf}$, $J_r$, $p_{H2}$, $p_{H2O}$ and $p_{O2}$ with and without considering electronic conduction.
Figure 8. Calculated impedance behaviors using parameters listed in Table 2.

Figure 9. Dependency of $R_g$ and $\tau_g$ on effective fuel utilization.
Figure 10. Experimentally observed impedance variation against $U_f$ at 750°C. (a) $U_f=4.1\%$, (b) 23.3\%, (c) 46.5\%, (d) 69.8\%, (e) 80.0\%

Figure 11. Impedance behaviors obtained with $J_r=0.3$ A/cm$^2$ for various flow rates.