A NOVEL HIGH-VOLTAGE SINGLE-CHAMBER SOFC WITH SERIES CONNECTED CELLS
I. OPERATING CHARACTERISTICS OF TWO-SEGMENTS CELL

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ABSTRACT

A novel single chamber SOFC for generating elevated voltages is proposed. The system consists of conventional single cells used in single chamber SOFC and interconnections. The single cells are connected in series and placed in the direction of gas flow. This fuel cell was named as single chamber SOFC with series connected cells (abbreviated to SSSC).

In this study, we deal with an analysis of the series connected segments which play an important role in the SSSC. In order to simplify the analysis, a single cell of SOFC is divided into two segments and the effect of the partition on the power generation characteristics is discussed. The result of the calculation for the two-segments cell revealed that the I-V and I-P characteristics of the cell are affected considerably by the ratio of the lengths of the segments. We obtained a maximum output power when the lengths of the segments are equal and hydrogen utilization is 68 %. The maximum power was higher than that of one-segment cell by 2.0 mW. When the utilization increases to 85 %, difference in output power between one-segment and two-segments cells increased to 5.2 mW.

INTRODUCTION

The single chamber fuel cell was proposed by Gool in 1965 (1) and demonstrated in practice by Dyer in 1990 with a hydrated aluminium oxide electrolyte (2). The working mechanism of the single chamber fuel cell is based on the difference in the catalytic activity between the two electrodes for the partial oxidation of the fuel. This idea was applied to SOFC by Hibino and Iwahara in 1993 (3). Recently a single chamber SOFC was demonstrated with a fuel of methane-air mixture at 973-1223 K by Iwahara and Hibino et al. (4, 5).

Since the voltage of a single cell in a conventional SOFC is low, a number of single cells must be series connected to attain a high voltage for practical applications. Furthermore, the complicated structure of a conventional SOFC stack with separators increases the production cost. By using the concept of the single chamber SOFC, it is possible to attain high voltage and power with an internally interconnected series of single cells in a single gas chamber. This single chamber SOFC abbreviated to SSSC is schematically shown in Fig. 1. We expect that this system is easier to produce than that
of conventional SOFC because of the simple stack structure.

In this report, we deal with an analysis of the series connected segments which play an important role in the SSSC. Then a single cell of conventional SOFC is divided into two segments and the effect of the partition on the power generation characteristics is discussed. Further analysis on the series connected segments in a single chamber SOFC is made in the following report.

FORMULATION OF THE SINGLE CELL SYSTEM

The Single Cell Model Used for the Analysis

Prior to the analysis of the divided cells, we formulate a single cell system. The analysis of the operating characteristics of a single cell was conducted on the basis of the calculation model shown in Fig. 2. In order to simplify the calculations, we considered a conventional H₂-O₂ fuel cell and neglected the electric resistance of both electrodes. On the cathode side, oxygen mass balance was considered, and on the anode side, hydrogen and water vapor mass balances were considered.

Mass Balances on the Cathode and the Anode Sides

The oxygen mass balance in a local element of the cathode side area for time Δt can be expressed as Eq. [1] by means of a local cathode current density and Faraday's law.

\[ fC_{O_2}(x)\Delta t = fC_{O_2}(x + \Delta x)\Delta t + \frac{i_c(x)}{nF}W\Delta x\Delta t \]

\( CO_2(x) \) and \( CO_2(x + \Delta x) \) are the oxygen concentration at \( x \) and \( x + \Delta x \), respectively, \( i_c(x) \) is the cathode current density, \( n \) is the number of electrons transferred in the cathode reaction, \( F \) is Faraday constant. The oxygen diffusion process near the electrode was neglected and the electrode reaction was taken into account. From Eq. [1] we have a differential equation of Eq. [2].

\[ \frac{dC_{O_2}(x)}{dx} = -\frac{W}{nF}i_c(x) \]

The mass balances in a local element of the anode side area are also expressed as

\[ \frac{dC_{H_2}(x)}{dx} = -\frac{W}{mF_f}\{i_a(x)\} \]

for the hydrogen mass balance, and

\[ \frac{dC_{H_2O}(x)}{dx} = \frac{W}{mF_f}\{i_a(x)\} \]

for the water vapor mass balance, where \( C_{H_2}(x) \) and \( C_{H_2O}(x) \) are hydrogen and water vapor concentration, respectively, \( i_a(x) \) is the anode current density and \( m \) is the number of electrons transferred in the anode reaction.
Potential Distributions in the Single Cell

We describe the potential distribution in Fig. 3 as a function of $x$. Fig. 3 indicates that the electromotive force $\Delta E_{\text{eq}}$ is divided into the cathode overpotential $\eta_c$, anode overpotential $\eta_a$, the IR drop in the electrolyte, and the cell voltage $V$. The potential balance for the single cell can be expressed as,

$$\Delta E_{\text{eq}} = \eta_c + \eta_a + IR_e + V$$  \[5\]

where $I$ is cell current and $R_e$ is the resistance of electrolyte. When the load current is applied, cathode and anode open circuit potentials $E_{eq,c}$ and $E_{eq,a}$ change to $E_c$ and $E_a$. In this study, we assumed that the electric resistance of the electrodes is low and the oxide ions transfer quickly into the electrolyte, then $E_c$ and $E_a$ are constant in the range of $x$ (6). Consequently, the sum of the activation overpotential $\eta(x)$ and concentration overpotential $\Delta E(x)$ is constant over the $x$ direction on either electrodes. Then we have

$$\eta_c = \eta_c(x) + \Delta E_c(x) = \eta_c(x) + \frac{RT}{nF} \ln \frac{P_{O_2}(x)}{P_{O_2}(0)} = \text{const}$$  \[6\]

for the cathode, and

$$\eta_a = \eta_a(x) + \Delta E_a(x) = \eta_a(x) + \frac{RT}{mF} \ln \frac{P_{H_2O}(x)}{P_{H_2O}(0)} = \text{const}$$  \[7\]

for the anode, where suffixes c and a mean cathode and anode, respectively, $R$ is gas constant, $T$ is temperature, and $P_{O_2}(x)$, $P_{H_2}(x)$ and $P_{H_2O}(x)$ are the partial pressures of respective species at $x$.

Calculation of Fuel Partial Pressure for the Single Cell

Current density at $x$ can be related to activation overpotential at $x$ by the Butler-Volmer equation. We assumed that the dissociation and adsorption of the reactive gases on the electrode surfaces are the rate determining steps, and the exchange current density for the cathode and anode reactions are proportional to $P_{O_2}^{1/4}(x)$ and $P_{H_2}^{1/4}(x)$, respectively (7). Consequently the current density is given by

$$i_c(x) = k_c P_{O_2}^{1/4}(x) [\exp\left\{-\frac{F}{RT} \eta_c(x)\right\} - \exp\left\{-\frac{F}{RT} \eta_a(x)\right\}]$$  \[8\]

for the cathode, and

$$i_a(x) = k_a P_{H_2}^{1/4}(x) [\exp\left\{-\frac{F}{RT} \eta_c(x)\right\} - \exp\left\{-\frac{F}{RT} \eta_a(x)\right\}]$$  \[9\]

for the anode, where $k_c$ and $k_a$ are the rate constants. Applying Eqs. [8] and [9] to Eqs. [2] and [3], and using Eqs. [6] and [7], we can obtain the differential equations related to oxygen, hydrogen, and water vapor partial pressures,

$$\frac{dP_{O_2}(x)}{dx} = -\frac{A k_c P_{O_2}^{1/4}(x)}{n} [\exp\left\{-\frac{F}{RT} (\eta_a - \frac{RT}{nF} \ln \frac{P_{O_2}(x)}{P_{O_2}(0)})\right\}$$

$$- \exp\left\{-\frac{F}{RT} (\eta_a - \frac{RT}{nF} \ln \frac{P_{O_2}(0)}{P_{O_2}(0)})\right\}]$$  \[10\]
for the cathode side, and

\[
\frac{dP_{\text{H}_2\text{O}}(x)}{dx} = - \frac{dP_{\text{H}_2}(x)}{dx} = \frac{AK_{\text{H}_2\text{O}}}{m} \left[ \exp\left\{ - \frac{F}{RT} \left( \frac{\eta_{\text{H}_2\text{O}}}{mF} \ln \frac{P_{\text{H}_2\text{O}}(x)}{P_{\text{H}_2\text{O}}(0)} \right) \right\} - \exp\left\{ - \frac{F}{RT} \left( \frac{\eta_{\text{H}_2}}{mF} \ln \frac{P_{\text{H}_2}(x)}{P_{\text{H}_2}(0)} \right) \right\} \right]
\]

for the anode side, where \( A = \frac{WRT}{0.1013/F} \). If \( P_{\text{O}_2}(0) \) is given, we can obtain the oxygen partial pressure distribution \( P_{\text{O}_2}(x) \) for various \( \eta_{\text{H}_2\text{O}} \) by solving Eq. [10]. And applying this distribution to Eq. [8] with Eq. [6], the cathode current density distribution \( i_c(x) \) can be obtained. On the anode side of the cell, when \( P_{\text{H}_2}(0) \) and \( P_{\text{H}_2\text{O}}(0) \) are given, we can obtain hydrogen and water vapor partial pressure distributions \( P_{\text{H}_2}(x) \) and \( P_{\text{H}_2\text{O}}(x) \) for various \( \eta_{\text{H}_2} \) by solving the simultaneous differential equation of Eq. [11]. And applying these distributions to Eq. [9] with Eq. [7], the anode current density distribution \( i_a(x) \) can be obtained.

Conditions for the calculation are listed in Table I. Considering the report by Wang and Hibino (5), the values of 0.12 atm, 0.057 atm and 0.025 atm were selected as the most proper values for \( P_{\text{O}_2}(0) \), \( P_{\text{H}_2}(0) \) and \( P_{\text{H}_2\text{O}}(0) \) for the single chamber SOFC with LSGM electrolyte. Electromotive force \( \Delta E_{\text{eq}} \) can be calculated by applying these values to the Nernst equation expressed as,

\[
\Delta E_{\text{eq}} = \Delta E_{\text{eq}}^{\circ} + \frac{RT}{2F} \ln \frac{P_{\text{H}_2}(0)P_{\text{O}_2}(x)^{1/2}(0)}{P_{\text{H}_2\text{O}}(0)}
\]

where \( \Delta E_{\text{eq}}^{\circ} \) is the standard electromotive force at \( T \) K. The electromotive force \( \Delta E_{\text{eq}} \) at 1223 K is obtained as 0.920 V. This value reasonably agrees with the experimental value of the OCV (0.85 V) reported by Hibino et al. (5).

We solved the differential equations, Eqs. [10] and [11], using the "NDSolve" command of Mathematica. The partial pressure distributions calculated for various \( \eta_{\text{H}_2\text{O}} \) and \( \eta_{\text{H}_2} \) are shown in Fig. 4.

**Calculation of Total Cell Current**

Current density distributions \( i_c(x) \) and \( i_a(x) \) were obtained using Eqs. [8], [9], and \( \eta_{\text{H}_2\text{O}}(x) \), \( \eta_{\text{H}_2}(x) \), \( \eta_{\text{O}_2}(x) \) and \( \eta_{\text{H}_2\text{O}}(x) \) in Eqs. [8] and [9] were obtained by Eqs. [6] and [7] with \( \eta_{\text{H}_2\text{O}} \), \( \eta_{\text{H}_2} \) and the partial pressures. Then the total cathode current \( I_c \) and total anode current \( I_a \) were calculated using Eqs. [13] and [14].

\[
I_c = \int_0^L i_c(x) dx \tag{13}
\]
\[
I_a = -\int_0^L i_a(x) dx \tag{14}
\]

The total cell current \( I \) is equal to \( I_c \) and \( I_a \). Figure 5 shows \( I_c \) and \( I_a \) as a function of the \( \eta_{\text{H}_2\text{O}} \) and \( \eta_{\text{H}_2} \) for various rate constants. We can refer to Fig. 5 to estimate \( \eta_{\text{H}_2\text{O}} \) and \( \eta_{\text{H}_2} \) at a given cell current for various rate constants.
Calculation Process of I-V and I-P Curves

The electrolyte resistance $R_e$ can be expressed as

$$R_e = \frac{\rho d}{WL} \quad \text{[15]}$$

We use the value of the electrolyte resistivity as 3.98 $\Omega$ cm at 1223K which was reported by Ishihara (8). Now we can calculate the cell voltage $V$ as a function of the cell current $I$, for given values of $k_c$ and $k_a$. First, distributions of the partial pressures $P_{O_2}(x)$, $P_{H_2}(x)$ and $P_{H_2}O(x)$ were calculated using Eqs. [10] and [11], with the boundary conditions of $P_{O_2}(0)$, $P_{H_2}(0)$ and $P_{H_2}O(0)$, and for various values of cathode overpotential $\eta_c$ and anode overpotential $\eta_a$. The calculations were made with different values of $k_c$ and $k_a$ as 0.1, 0.05 and 0.02 A cm$^{-2}$. The cell current $I$ was calculated using Eqs. [8], [9], [13] and [14]. Then we can obtain the relations between $I$ and $\eta_c$, and $\eta_a$, for various $k_c$ and $k_a$. The results are plotted in Fig. 5. We calculate $\Delta E_{eq}$ by Eq. [12] with the values of $P_{O_2}(0)$, $P_{H_2}(0)$ and $P_{H_2}O(0)$. Then by using Eq. [5], we can obtain the value of $V$ because $IR_e$ is known.

The calculated I-V and I-P curves are compared in Fig. 6 with the experimental results reported by Hibino (5). Good agreement was obtained between the experimental data and the calculated curves when $k_c$ is equal to $k_a$ and the value is 0.1 A cm$^{-2}$. We use this value for the rate constants in the following analysis of the operating characteristics of the two-segments cell. Partial pressures at the end of the single cell, $P_{O_2}(L)$, $P_{H_2}(L)$ and $P_{H_2}O(L)$, are shown in Fig. 7 as a function of cell current.

Operating Characteristics of Two-Segments Cell

Modeling of Two-Segments Cell

Schematic diagram of the two-segments cell was described in Fig. 8. The front segment and rear segment are placed in the direction of the gas flow. The effects of the mixing of the cathode and anode side gases in the gap between the segments were neglected. First the values of $P_{O_2}(L_1)$, $P_{H_2}(L_1)$ and $P_{H_2}O(L_1)$ are obtained from Fig. 7 for a given current $I$. Then the I-V curve for the rear segment can be calculated by the above mentioned procedure. The total voltage and power for the two-segments cell were obtained as the sum of the values of front and rear segments. The lengths of the segments are listed in Table II for the two-segments cells of Cell II-VI. Cell I denotes a single cell without division.

I-V and I-P Characteristics of Two Segments Cell

Hydrogen utilization $U_{H_2}$ for the one-segment and two-segments cells are expressed as

$$U_{H_2} = \frac{I}{mFC_{H_2}(0)} \times 100 \quad \text{[16]}$$

and
respectively. The cell current $I$ passing through the two-segments cell is half of the one-segment cell (Cell I) for the same value of $U_{H_2}$.

Figure 9 shows the results of the calculation of I-V and I-P curves for Cells I-VI. It is obvious from the result that the output power of these cells increases in the following order, Cell II, VI < Cell III, V < Cell I (one-segment cell) < Cell IV. We obtained a maximum output power when the lengths of the segments are equal and hydrogen utilization is 68%. The maximum power was higher than that of one-segment cell by 2.0 mW. When the utilization increases to 85%, difference in output power between one segment cell and Cell IV increases to 5.2 mW. The output powers of the one-segment cell and two segments cells at $U_{H_2}$=68 and 85% are summarized in Table III.

**Energy Losses in One-Segment Cell and Two-Segments Cell**

The energy loss caused by electrode overpotential at given cell current for the one segment cell and the two-segments cells are expressed as

$$P_{t,one}(op) = \left( \eta_{c} + \eta_{h,1} \right) \cdot I$$

and

$$P_{t,two}(op) = \left( \eta_{c,1} + \eta_{c,2} + \eta_{h,1} + \eta_{h,2} \right) \cdot I$$

respectively. Where suffixes 1 and 2 mean front segment and rear segment, respectively.

Figure 10 shows the energy loss caused by electrode overpotential. It is obvious from this figure that when $U_{H_2}$ is 68%, $P_{t,two}(op)$ of Cell IV is lower than $P_{t,one}(op)$ by 12 mW. When $U_{H_2}$ increases to 85% the difference increases to 21 mW. This reduction means the lowering of the energy loss arising from the Nernst loss. When the cell area of the segment decreases, the current density in the segment increases. $P_{t,two}(op)$ of Cell II, III, V and VI are larger than that of Cell IV because the electrode overpotential in the smaller segment increases due to the larger current densities.

The energy loss caused by IR drop in the electrolyte at a given cell current for the one-segment cell and the two-segments cells are expressed as

$$P_{t,one}(IR) = \frac{\rho d}{W} \cdot \frac{1}{L} \cdot I^2$$

and

$$P_{t,two}(IR) = \frac{\rho d}{W} \left( \frac{1}{L_1} + \frac{1}{L_2} \right) \cdot I^2$$

respectively. Figure 10 also shows the energy loss caused by the IR drops in the electrolytes of the cells. $P_{t,two}(IR)$ of Cell IV is equal to $P_{t,one}(IR)$. $P_{t,two}(IR)$ of Cell II, III, V and VI are considerably larger than $P_{t,one}(IR)$. This is caused by the large IR drops due to the large current densities in the smaller segments which are the front cells of Cell II and III, and the rear cells of Cell V and VI.
It is observed in Fig. 10 that dividing the single cell leads to a reduction of the energy loss related to the electrode overpotential. This effect is remarkable when $U/H_2$ is high and $L_1$ is equal to $L_2$. In the case that $L_1$ is not equal to $L_2$, the energy loss related to the IR drop in the electrolyte increases when the cell is divided into segments. This effect is also remarkable when $U/H_2$ is high. When $U/H_2$ is high, the difference in the energy loss related to IR drop between one-segment cell and two-segments cells ($P_{1\text{seg}(IR)}-P_{2\text{seg}(IR)}$) is larger than that related to the electrode overpotential ($P_{\text{cell}(op)}-P_{\text{cell}(op)}$). Therefore, when we consider the total loss value, the two-segments cell with equal segment lengths shows the minimum loss value. These relations in the energy losses are summarized in Table III.

**CONCLUSIONS**

In the conventional fuel cell systems, cells must be stacked to increase the output voltage. We propose a single chamber SOFC in which cells are connected to produce a high voltage. Separators and gas distribution devices are not required in this system. The production cost of this system may be lower because of the simple structure. Numerical analysis of a simple model of the two-segments cell revealed that this fuel cell system can generate a high voltage and output power. This is because the energy loss related to the electrode overpotential can be reduced by dividing the single cell. Therefore by increasing the number of the segments, this system could generate high voltages necessary for portable electronics devices.

**LIST OF SYMBOLS**

- $d$: electrolyte thickness (cm)
- $W$: width of a cell (cm)
- $f$: flow rate of inlet gas (cm$^3$ s$^{-1}$)
- $L$: length of a cell (cm)
- $C_i(x)$: concentration of species i (mol cm$^{-3}$)
- $i(x)$: current density (A cm$^{-2}$)
- $n$: number of electrons transferred in cathode reaction
- $F$: Faraday constant
- $m$: number of electrons transferred in anode reaction
- $\Delta E_{eq}$: electromotive force (V)
- $\eta$: electrode overpotential (V)
- $V$: cell voltage (V)
- $I$: cell current (A)
- $R_e$: electrolyte resistance ($\Omega$)
- $E_{eq}$: open circuit potential (V)
- $E$: electrode potential (V)
- $\eta(x)$: activation overpotential (V)
- $\Delta E(x)$: concentration overpotential (V)
- $R_{\text{load}}$: external resistance ($\Omega$)
- $R$: gas constant
- $T$: operating temperature (K)
- $P_i(x)$: partial pressure of species i (atm)
- $k$: rate constant (A cm$^{-2}$)
- $\Delta E_{eq}^*$: standard electromotive force (V)
- $\rho$: electrolyte resistivity ($\Omega$ cm)
- $U/H_2$: hydrogen utilization (%)
- $P_{\text{cell}(op)}$: energy loss caused by electrode overpotential (W)
- $P_{\text{IR}}$: energy loss caused by IR drop in the electrolyte (W)

Subscripts:
- $c$: cathode
- $a$: anode
- 1: front segment
- 2: rear segment
- one: one-segment cell
- two: two-segments cell
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Table I. Values used for the calculation.

| $T$ [K] | $P_{O_2}(0)$ [atm] | $P_{H_2}(0)$ [atm] | $P_{H_2O}(0)$ [atm] | $d$ [cm] | $W$ [cm] | $f$ [cm$^3$ s$^{-1}$] | $L$ [cm] | $n$ [-] | $m$ [-] | $\rho$ [Ω cm] |
|--------|------------------|-------------------|---------------------|--------|--------|----------------|--------|--------|--------|----------------|
| 1223   | 0.12             | 0.057             | 0.025               | 0.05   | 1.0    | 5              | 0.5    | 4      | 2      | 3.98           |

Table II. Length of the segment cells in the two-segments cell shown in Fig. 8.

|   | L$_1$ [cm] | L$_2$ [cm] |
|---|------------|------------|
| Cell II | 0.2 | 0.8 |
| Cell III | 0.3 | 0.7 |
| Cell IV | 0.5 | 0.5 |
| Cell V | 0.7 | 0.3 |
| Cell VI | 0.8 | 0.2 |

Cell I : without division ($L=1.0$ cm).

Table III. Comparison of output power, energy losses by electrode overpotential and by IR drop in the electrolyte.

| $U_H$ [%] | type of cell | output power [mW] | overpotential loss [mW] | IR loss [mW] |
|----------|--------------|-------------------|------------------------|-------------|
| 68       | I            | 133               | 180                    | 27          |
|          | II, VI       | 102               | 186                    | 43          |
|          | III, V       | 123               | 175                    | 32          |
|          | IV           | 135               | 168                    | 27          |
| 85       | I            | 122               | 263                    | 43          |
|          | II, VI       | 81                | 264                    | 67          |
|          | III, V       | 110               | 251                    | 51          |
|          | IV           | 127               | 242                    | 43          |
**Figure 1.** Schematic diagram of a single chamber SOFC with series connected cells, which is abbreviated to SSSC.

**Figure 2.** Calculation model for the analysis of the operating characteristics of a single cell.

**Figure 3.** Schematic diagram of the potential distribution in the single cell.

**Figure 4.** Distributions of the partial pressures in the single cell for $k_c=k_a=0.1$ A cm$^{-2}$. (a) $P_{O_2}(x)$ for various $\eta_{c,\alpha}$ and (b) $P_{H_2}(x)$ and $P_{H_2O}(x)$ for various $\eta_{a,\alpha}$.

**Figure 5.** Cell current of the single cell described in Fig. 2 as a function of $\eta_{c,\alpha}$ and $\eta_{a,\alpha}$ at 1223 K for various rate constants $k_c$ and $k_a$. 

Electrochemical Society Proceedings Volume 2001-16
Figure 6. Comparison of the calculated and experimental I-V (a) and I-P (b) characteristics for a single cell of SOFC using a LSGM electrolyte at 1223 K: (■) Experimental values (5), (–) Calculated curves for $f_{c}=0.1$ A cm$^{-2}$ and $f_{a}=0.1$ A cm$^{-2}$.

Figure 7. Partial pressures of the reactive species at the end of the cell ($x=L$).

Figure 8. Two-segments cell model used for the calculation of operating characteristics.

Figure 9. I-V and I-P characteristics calculated for the one segment (Cell I) and two segments cells (Cell II-VI) at 1223 K.

Figure 10. Energy losses caused by electrode overpotential and caused by IR drop in the electrolyte.