ANALYSIS OF THE TUBULAR SOFC SYSTEM USING NON-ISOTHERMAL SIMULATION

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ABSTRACT

We have developed a non-isothermal simulation program incorporating inner reforming for a SOFC system using tubular cells. In this paper, the following points are considered, (i) including the radiation heat transfer, (ii) modeling shape of a half-sphere at the bottom of the cell, and (iii) applying the experimental results of electrodes performance of a tubular cell made by the electrochemical vapor deposition. This simulation reveals the details of the temperature and current distributions. From these results, we discuss the reliability of tubular SOFC systems.

INTRODUCTION

The solid oxide fuel cell (SOFC) is considered the next generation fuel cell thanks to its many advantages, including high energy efficiency and fuel adaptability. The tubular SOFC system, it has already demonstrated good reliability and performance stability by verification tests, especially on the 25 kW cogeneration unit test in collaborated by Tokyo Gas, Osaka Gas and Westinghouse (1).

With increase in the size of cell, mechanical reliability becomes even more important. It is time consuming and and costly to investigate all the design and operating conditions experimentally to optimize the system design. In addition, since the SOFC is operated at temperatures as high as 1273K, it is difficult to measure the distributions of current density, temperature of solids and gases, and gas
compositions.

We have developed a computer program for the tubular solid oxide fuel cells incorporating an internal reforming process (2). Using this calculation, we reported a comparison of the power density and the efficiency of a tubular SOFC system with the conventional and the fuel recycling operations. This time, the following points were improved, (i) including the radiation heat transfer, (ii) modeling shape of a half-sphere at the bottom of the cell, and (iii) applying the experimental results of electrodes performance of a tubular cell (3,4). By using this simulation, the critical conditions for cracking should be identified

MODELING AND CALCULATING PROCEDURES

Only the modeling and calculating procedures which were improved are explained here. Others were described in our previous paper (2).

Configuration of the System and the Cell. In this paper, a system shown in Figure 1 was calculated. Figure 2 and Table 1 show schematic diagram and dimensions of a tubular cell used for this calculation, respectively. The cell was assumed to be a 300 mm long tube. Figure 3 shows a configuration of the cell. The tubular cell and alumina tube were cut into 46-50 slices along Z-axis, and a half of the tubular cell was cut into 12-18 element along 0-axis. A hemispheric region was divided to 5 rings along Z-axis. The calculation was conducted for only half of the cell because the cell used for calculation is symmetric to the plane containing both current collectors.

Since the direction of the current in the anode and cathode layer is perpendicular to the Z-axis, a slice of the tubular cell was modeled by a transmission line composed only of resistors and electromotive forces (Figure. 4). In the hemispheric region, the direction of the current in the anode and cathode layers is no longer perpendicular to the Z-axis, because the generated current at the hemispheric region was collected at the top of the nickel felt.

Electrochemical and Chemical Reactions. Fuel gas is assumed to be reformed methane at a steam/carbon (S/C) ratio of 2.5. The chemical species in the fuel gas we have considered were H₂, H₂O, CO, CO₂, and CH₄. As the gas flowed into the cell, the gas composition varies with electrode reactions, reforming and shift reactions.

Since the SOFC is operated at a high temperature around 1273 K, these reactions are assumed to be at equilibrium in the following calculations. Deviations from equilibrium in this present calculation are ignored.
Current Distribution. The polarization at the electrodes mainly consists of activation and concentration polarization. The authors have reported the polarization characteristics of the air and fuel electrodes fabricated by the electrochemical vapor deposition method (EVD) (3,4,5), and we used these results for calculations of the current distribution.

Radiation Heat Transfer. Energy flux of the radiation is given by the Stefan-Boltzmann's equation:

$$E_{\text{rad}} = \varepsilon \sigma T^4$$  \[1\]

where $\varepsilon$ is the emissivity of the cell components and $\sigma$ is the Stefan-Boltzmann constant. In a high temperature, radiation is an the important mode of heat transfer. As shown in Figure 5, radiant heat rays from the cell are reflected and absorbed repeatedly by the cell itself or gas, and weakened gradually. Although the tubular cell is of relatively simple shape, it is difficult to evaluate the transferred radiant heat by the analytical method. Therefore, to consider the contribution of the radiant heat transfer, we adopted the Monte Carlo method, in which a number of imaginary "photons" were radiated from an element of the cell, and their behavior, such as translation, scattering, and absorption, was calculated for each photon. By counting the absorbed energy at each element, and the distribution of the energy radiated from one element is numerically determined.

Solution Algorithm. Figure 6 shows an algorithm to obtain temperature distribution of the cell. First, the distribution of the current density was calculated for a fixed temperature of the cell and gases. Based on the distribution of the current density, temperature and composition of the gases were calculated under adiabatic condition. Then the heat balance in the cell and radiant heat transfer were computed. In the final module, the temperature distribution of the cell was calculated by the finite element method. This iteration was continued until convergence of the temperature distribution.

RESULTS AND DISCUSSION

Operation Parameters. Temperatures of the fuel and air at the gas inlet were set at 1070 K and 1050 K respectively. The fuel gas is reformed methane with steam at $S/C=2.5$ as described above, and the oxidant gas is air. Fuel utilization was 80%, and air utilization was 25%. Total loading current from the cell was set to 32.4 A/cell
Current Distribution. Figure 7 shows the current distribution when methane concentration at the fuel inlet was 0%. According to this calculation, along the Z-axis the current density was rather intense near the fuel inlet. This is due mainly to the depletion of the fuel gas. Along the θ-axis, the current density around 90° was low near the fuel inlet. This is due to the resistance of the electrodes from a electrochemical reaction site to a current collector. When the current density \( j \) increases, the polarization resistance \( \eta/j \) decreases, but the electrode resistance is constant.

Figure 8 shows the distribution of the in-plane current in (a) the anode layer, and (b) the cathode layer at the top of the cell. As expected, intense current was found around the negative cell contact in the anode layer (Figure 8a), while the current was intense around the interconnector in the cathode layer (Figure 8b). In the hemispheric region, it was also seen that the current flowed via the head of the cell from the negative cell contact toward the interconnector. This is due to a lack of cell contacts in this region.

Temperature Distribution. In our previous paper (2), the calculated results were confirmed by comparing with the experimental data. Under the same operating conditions, it shows better matching with the experimental data. Figure 9 shows the temperature distribution. It indicates that the temperature gradient at the hemispheric region is large. This may become the critical position for cracking. There is no nickel felt at hemispheric top. Because nickel felt has large heat conductivity, its presence is a large influence. The top of the cell is the position where reliability is low, because (i) it is the edge of interconnector film which has different thermal expansion characteristics than the other materials, and (ii) there is a plug of the air electrode tube made by the injection forming method. Therefore, it is not desirable to have the maximum temperature gradient of the cell here. In order to improve the reliability against cracking, the heat conduction at the hemispheric top should be reinforced. The heat conduction between a cell and the partitions are located above and below it is not considered in this calculation. When a cell comes in contact with the partitions directly, the temperature gradient at the hemispheric region is reduced.

Figure 10 shows the temperature distributions along the Z-axials for 0-10% methane concentration. Because this reforming reaction of CH₄ is a highly endothermic reaction \( (\Delta H_0 = 229 \text{ kJ mol}^{-1}) \), the temperature at the hemispheric top drops.
Tubular SOFC Systems. For the system shown in Figure 1, it is clear from figure 10 that the application of in-situ reforming lowers both the reliability and the performance of the SOFC system.

We have evaluated the performance of our tubular cell experimentally (5), and reported that the temperature dependence at 1200-1300 K was small, but that the operation below 1173 K lowered the power density considerably. Because fine particle of nickel are in the cell, a difference of few degrees in temperature around 1273 K changes the rate of degradation of the cell. If operations which reduce the temperature distribution become practical, optimizing the operating temperature should promote the higher power density or output voltage and longer life time for the same cell. In order to limit the temperature distribution, the following cooling methods have been suggested, (i) fuel recycling (2), (ii) the natural gas reformers are integrated between the cells (7), and (iii) preheating of fuel using fuel pipes inside the cell chamber (2). Among these, (ii) should be put into practice in 1997 on the 100 kW module by Westinghouse (7).

CONCLUSIONS

We have developed a non-isothermal simulation program incorporating inner reforming and radiant heat transfer for an accurately modeled single tubular cell. It is found that the temperature gradient in the hemispheric region is large. This may become the critical position for cracking. To improve the cell reliability, operating conditions must be designed to reduce its temperature gradient.

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Convection

Radiation

Conduction

Figure 1 A tubular SOFC system.

Fuel

Convection

Radiation

Conduction

Air

Fuel

Cell contact

Anode

Electrolyte

Cathode

Figure 2 Schematic diagram of cross-section of self-supported tubular cell.

Table 1 Cell components and dimension.

| Components          | Material          | Dimension                      |
|---------------------|-------------------|--------------------------------|
| Air electrode tube  | La(Sr)MnO₃        | 9/15mm inner/outer diameter    |
| Electrolyte         | YSZ              | 20μm thick                     |
| Fuel electrode      | Ni-YSZ           | 100μm thick                    |
| Interconnector      | LaCr(Mg)O₃       | 20μm thick, 5mm width          |
| Ni felt             |                  | 2mm thick, 5mm width           |
| Alumina tube        |                  | 4/6mm inner/outer diameter     |
Figure 3 Schematic illustrations of element discretization of tubular cell.

Figure 4 Equivalent circuit for a slice of tubular cell.

Figure 5 Scheme of radiant heat transfer in tubular SOFC.
Radiated photon was scattered in the cell, and absorbed gradually.
Figure 6 An algorithm for calculating temperature distribution of a tubular cell.

Figure 7 Distribution of current density.
Figure 8 Current distribution. (a) in anode layer (b) in cathode layer at top of the tubular cell.

Figure 9 Calculated temperature distribution of the cell.

Figure 10 Calculated temperature distribution of the cell.