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Short communication

Anode-supported micro tubular SOFCs for advanced ceramic reactor system

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Abstract

In this study, micro tubular SOFCs under 1 mm diameter have been fabricated and investigated at 450–550 °C operating temperature with H_2 fuel. The performance of the 0.8 mm diameter tubular SOFC was 110–350 mW cm^{-2} at 450–550 °C operating temperatures. To maximize the performance of the cell as well as to optimize the geometry of tubular cells, a current collecting method used in the experiment was examined. A model was proposed to estimate the loss of performance for single cell due to the current collecting method as functions of anode tube length and thickness. The results showed that the losses of performance were calculated to be 0.8, 2.0, and 4.6% at 450, 500, and 550 °C operating temperatures, respectively, for the 0.8 mm diameter tubular SOFC with the length of 1.2 cm.

Keywords: Tubular SOFCs; Intermediate temperature; Stack; Micro SOFC; Current efficiency

1. Introduction

Development of intermediate temperature (IT) solid oxide fuel cells (SOFCs) has become important in recent years due to its high environmental performance and fuel flexibility. They are expected to decrease material degradation, to prolong stack lifetime, and to reduce cost by utilizing metal materials [1,2]. Recent studies showed outstanding results with high power density of 0.8–2 W cm^{-2} at 600 °C or under using anode-supported planar SOFCs [3–5].

On the other hand, it was shown to be effective to consider the shape of the cells to increase their performance. For example, micro tubular SOFCs had many advantages over conventional planar SOFCs [6–8]. It was shown that micro tubular SOFCs endured thermal stress caused by rapid heating up to operating temperature. It is also possible to design SOFC stacks with larger electrode area in unit volume using micro tubular SOFCs of millimeter to sub-millimeter in diameter.

From this point of view, Advanced Ceramic Reactor Project (2005–2009) supported by NEDO has initiated for realizing such SOFC systems by developing innovative fabrication process technology. So far, applying advanced ceramic processing techniques, we have succeeded in the fabrication of micro tubular SOFCs of millimeters to sub-millimeters in diameter, which are operable at low temperatures between 450 and 600 °C.

In this paper, it is shown the results of *I–V* characterization of single micro tubular SOFC and examination of a current collecting method used in the experiment. A model for this method was proposed to estimate the loss of performance for single cell as functions of anode tube length and thickness for each operating temperature.

2. Experimental

NiO–Gd doped ceria (GDC), GDC, and $La_{0.8}Sr_{0.2}Co_{0.6}Fe_{0.4}O_3$ (LSCF)–GDC were used for anode (tube), electrolyte and cathode, respectively. Fabrication process of the anode tubular SOFC was discussed elsewhere [9]. Fig. 1(a) shows an image of a complete cell and (b) an SEM image of cross-section of the cell. The performance of the cell was investigated using a

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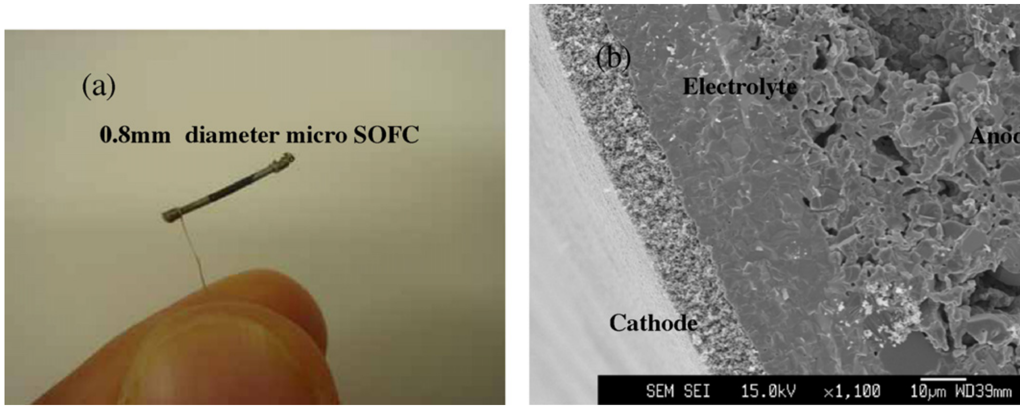

Fig. 1. (a) Picture of a 0.8 mm diameter micro tubular SOFC; (b) cross-sectional SEM images of the 0.8 mm diameter micro tubular SOFC.

Solartron 1260 frequency response analyzer with a 1296 Interface. The Ag wire was used for collecting current from anode and cathode, which were both fixed by Ag paste. Current from anode side was collected from an edge of the anode tube. Twenty percent H_2 (humidified by bubbling water at room temperature) in N_2 was used as a fuel at the flow rate of 25 cm^3 min^{-1}. Cathode side was open to the air without flowing gas. The size of the tubular cell was 0.8 mm in diameter and 1.2 cm long with the cathode length of 8 mm, whose electrode area is 0.2 cm^2.

3. Model

A calculation model as shown in Fig. 2 was considered to describe the current collection method, which was made from the edge of the anode tube. As can be seen, extra anodic resistance (resistance along with the tube) was added to the true cell resistance, which includes ohmic and electrode overpotential resistances due to the current collecting method. Thus, the loss of performance was defined by the ratio of the true cell resistance and actual cell resistance with extra anodic resistance.

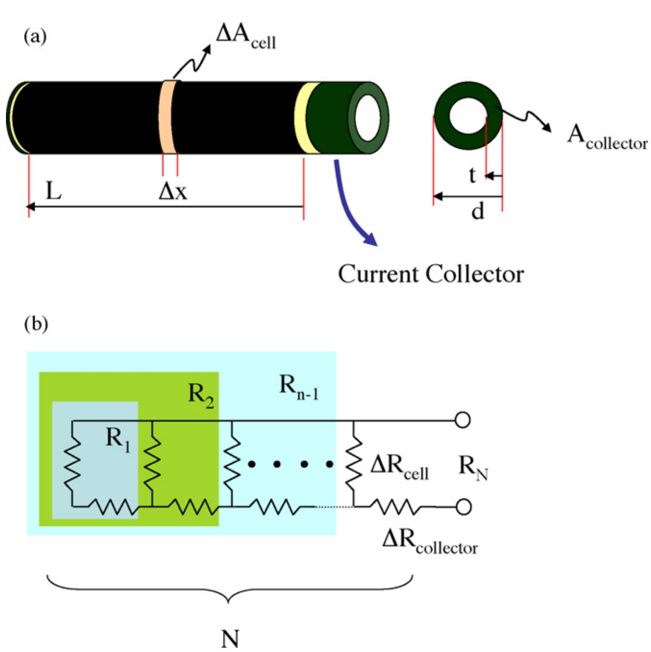

Fig. 2. Proposed model for the anode current collecting method used in this study.

A tubular SOFC with tube diameter, thickness and length of d, t and L, respectively, was divided into N slices. Each slice with the thickness of Δx (=L/N) was assigned to equivalent circuit of $\Delta R_{collector}$ and ΔR_{cell} in Fig. 2(b). $\Delta R_{collector}$ and ΔR_{cell} are the anodic resistance as current collector and the true cell resistance of each slice, respectively. R_1, R_2, ..., R_N shown in Fig. 2(b) were given by following equations:

$$\begin{aligned} R_1 &= \Delta R_{cell} + \Delta R_{collector} \\ R_2 &= \frac{\Delta R_{cell} R_1}{\Delta R_{cell} + R_1} + \Delta R_{collector} \\ &\vdots \\ R_{N-1} &= \frac{\Delta R_{cell} R_{N-2}}{\Delta R_{cell} + R_{N-2}} + \Delta R_{collector} \\ R_N &= \frac{\Delta R_{cell} R_{N-1}}{\Delta R_{cell} + R_{N-1}} + \Delta R_{collector} \end{aligned} \quad (1)$$

where $\Delta R_{collector}$ and ΔR_{cell} are determined from following equations:

$$\Delta R_{collector} = \frac{\Delta x}{A_{collector}\sigma} \quad (2)$$

$$\Delta R_{cell} = \frac{\text{ASR}}{\Delta A_{cell}} \quad (3)$$

where σ and ASR are conductivity of the anode tube in the reducing atmosphere [9] and area specific resistance of the cell (true cell resistance), respectively. ΔA_{cell} and $A_{collector}$ are given as $\Delta A_{cell} = \pi d \Delta x$ and $A_{collector} = \pi t(d-t)$, as shown in Fig. 2(a). Note that the experimental results are equivalent to R_N, which includes the extra anodic resistance. Thus, values of ASR were first estimated from Eqs. (1)–(3) and then, values of ASR were used for further calculation. Since Ag wire and paste were placed on the whole surface of cathode, such performance loss from cathode side due to current collecting method can be negligible. Therefore, current collecting resistance of the cathode part was not directly considered in this calculation. The loss of performance was determined as

$$\text{loss of performance} = \frac{R_N - \text{ASR}/(\pi L d)}{R_N} \quad (4)$$

4. Results and discussion

The performance of the micro tube cell was shown in Fig. 3. As can be seen, the peak power density of 110, 205 and 350 mW cm^{-2} was obtained at 450, 500, 550 °C, respectively. The data points as shown in Fig. 3 for each temperature were used for model calculations.

Using the model, the loss of performance for the tubular SOFC was calculated. In this calculation, appropriate value of N must be selected. For this purpose, the loss of performance was calculated as a function of N using the experimental data set obtained at 550 °C. As shown in Fig. 4, the loss of performance showed convergence for $N > 100$, and therefore, N was set to 1000 for the rest of calculations.

Fig. 5 shows the loss of performance as a function of anode tube length (L) at different operating temperatures (anode tube thickness; $t = 0.2$ mm). As can be seen, the losses of performance were estimated to be 0.8, 2.0, and 4.6% at 450, 500, and 550 °C operating temperatures, respectively. It is also shown that the loss of performance became over 7% for specimen with length of over 1 cm at 550 °C operating temperature. Thus, the length of anode tube should carefully be determined to minimize the loss of performance.

Fig. 6 shows the loss of performance as a function of anode tube thickness (t) at several operating temperatures (anode tube length; $L = 8$ mm). As can be seen, controlling thickness of the anode tube can be very effective to reduce the loss of performance at lower temperature; however, it seems to be difficult to improve by changing anode thickness when it is operated at 550 °C. It appeared that the loss of performance was estimated over 3% at 550 °C operating temperature for actual experimental condition (at $t = 0.2$ mm), while the loss was negligible at 450 °C operating temperature.

After all, to decrease the loss of performance, and to design cell stacks using micro tubular SOFCs, careful consideration

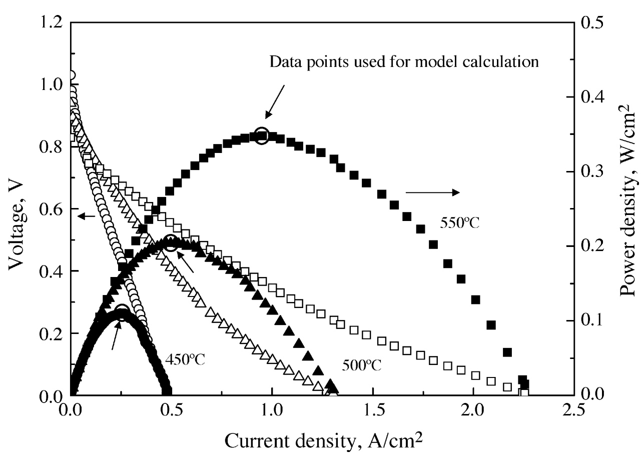

Fig. 3. The performance of the 0.8 mm diameter micro tubular SOFC. Cell voltage and power as functions of current and temperature.

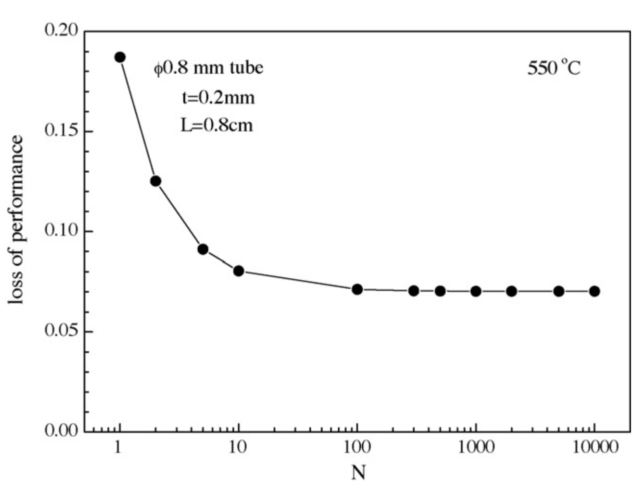

Fig. 4. The loss of performance as a function of number of division (N).

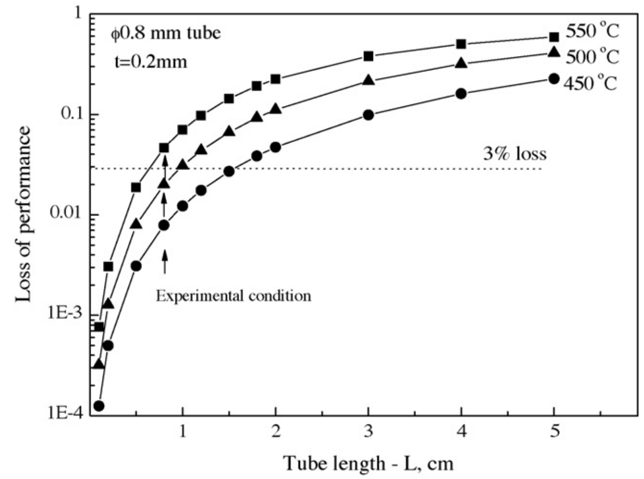

Fig. 5. The loss of performance as a function of anode tube length (L) for different operating temperatures.

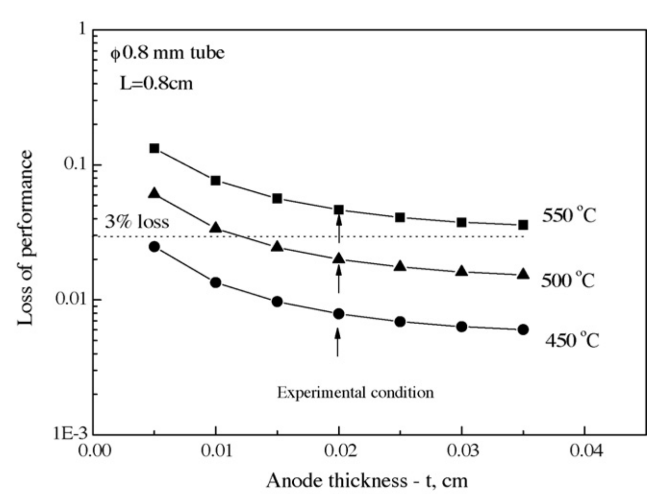

Fig. 6. The loss of performance as a function of anode tube thickness (t) for different operating temperatures.

must be taken and followings can be effective: (i) decrease operating temperature; (ii) increase anode thickness; (iii) increase conductivity of anode; (iv) decrease tube length; (v) use an alternative method for current collection (e.g. current collection from both anode ends).

5. Summary

Micro tubular SOFCs under 1 mm diameter have been successfully fabricated and the performance of the 0.8 mm diameter tubular SOFC was evaluated to be 110–350 mW cm^{-2} at 450–550 °C. Thus, high performance cell stacks with high volumetric power density can be realized by using the micro tubular SOFCs.

A model for the current collecting method was proposed and used to estimate the loss of performance using the experimental data. The losses of performance were estimated to be 0.8, 2.0, and 4.6% at 450, 500, and 550 °C operating temperatures, respectively. In spite of high SOFC performance, the model indicated that the loss of performance due to the current collecting method can be over 7% for the cell with the length of over 1 cm (cathode length). Thus, selection of the length and thickness of the anode tube is crucial to minimize the loss of performance.

Acknowledgement

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