Low Temperature Operated SOFCs Using Ceria Based Electrolyte

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Ceria based electrolyte is one of candidates for low temperature SOFCs operated under 600 °C, since it has reasonably high ionic conductivity, and moderate material cost, in spite of the drawback, lower open circuit voltage. In this study, the effect of fabrication process of the cell on the cell performance was examined using micro tubular, anode-supported cell design. Two samples were prepared by co-sintering anode supported tube and electrolyte (Gd
doped CeO2) using the sintering temperatures of 1260 and 1400 °C, respectively. The cell performance test showed that both samples had advantages depending up on the operating temperatures. The sample sintered at 1260 °C showed better cell performance operated at 450 °C, as high as 0.15 Wcm−2, while the sample sintered at 1400 °C showed the power density of 0.49 Wcm−2 at 550 °C, 1.4 times better than that of the sample sintered at 1260 °C. This behavior is resulted from the difference in the conductivity of the electrolyte and the microstructure of the anode, which were prepared form different sintering temperatures.

Key Words: Ceria, Electrolyte, Micro Tubular, Thin Film, Dip Coating

1 Introduction

Solid oxide fuel cells (SOFCs) produce electricity from electrochemical reactions between fuel and oxidant, with high energy conversion efficiency. The development of SOFCs is, thus, of importance and a key for the future energy strategy.1,3) SOFCs are well investigated from fundamental, basic science to application aspects, and one of the research trends is to decrease operating temperature in order to improve life time of the cell module and to reduce cost by utilizing cheaper materials for module components. Reducing operating temperature of SOFCs can be realized by following manners:

1. modification of cell components4,7)
2. use of new materials for SOFC components8-12)
3. introducing new system concept13,15)
4. use of new cell designs16-18)

In this study, we selected doped ceria, which has reasonably high ionic conductivity at lower temperature and low material cost, as an electrolyte material for micro tubular SOFCs. Use of micro tubular design is also expected to increase the volumetric power density for lowering operating temperature. The effect of co-sintering temperature on the cell performance was examined for the optimization of the fabrication process as well as the operating conditions. Impedance analysis was also employed for the investigation of the cell performance.

2 Experimental

Micro tubular SOFCs were fabricated using following method: Anode tubes were made from NiO powder (Sumitomo metal Co., Ltd.), Gd0.5Ce0.5O2−δ (GDC)(Shin-Etsu Chemical Co., Ltd.), poly methyl methacrylate beads (PMMA) (Sekisui Plastics Co., Ltd.), and cellulose (Yuken Kogyo Co., Ltd.). These powders were mixed for 1 h by a mixer 5 DMV-rr (Dalton Co., Ltd.), and after adding the proper amount of water; it was stirred for 30 min in a vacuumed chamber. The mixture (clay) that was prepared from these powders was left over 15 h for aging. The tubes were extruded from a metal mold by using a piston cylinder type extruder. For the preparation of an electrolyte on the anode tube, dip-coating and co-sintering techniques were used. GDC was selected for the electrolyte. A slurry for dip-coating was prepared by mixing the GDC powder, solvents, binder (poly vinyl butyral), dispersant and plasticizer. Conventional dip coating technique was used for coating the layer on the tubular anode support. After coating process, the tube was sintered at 1260 and 1400 °C for 1 h in air. The anode tubes with the electrolyte were, again, dip-coated using cathode slurry, which was prepared in the same manner using La0.8Sr0.2Co0.8Fe0.2O1.9 (LSCF), the GDC powder, and organic ingredients. After dip-coating, the tubes were dried and sintered at 1050 °C for 1 h in air.

Figure 1 shows the image of these samples. As can be seen, the sample of 1260 °C sintering temperature has

Fig. 1 Image of the micro tubular SOFCs (φ2 mm)
white colored electrolyte, which showed less dense structure of the electrolyte. Note that no porous was observed for both samples from SEM measurement. The cell performance was investigated by using a potentiostat (Solartron 1296). The cell size was 2 mm in diameter and 20 mm in length with cathode length of 9.5 mm, whose effective cell area was 0.6 cm². The Ag wire was used for collecting current from anode and cathode, which were both fixed by Ag paste. The current collecting Ag wire was attached at an edge of the anode tube with Ag paste. For cathode, Ag wire was wrapped in 1 mm pitch on the surface of the cathode and fixed using Ag paste. Hydrogen (humidified by bubbling water at room temperature) was flowed inside of the tubular cell at the rate of 10 mL min⁻¹ with the addition of Nitrogen 15 mL min⁻¹.

3 Results and Discussion

Figure 2 shows the performance of the SOFCs prepared at the sintering temperature (Tₜ) of 1260 and 1400°C at the operating temperature of (a) 450 and (b) 550°C. As can be seen, the open circuit voltage (OCV) of the cell was 0.95 and 0.88 V at 450 and 550°C, respectively for both samples. The OCVs decreased as operating temperature increased, which typically explained by an increase of electronic conductivity in the electrolyte. At 450°C (Fig. 2 (a)), the performance of the cell of Tₜ = 1260°C was 1.5 times higher than that of the other. On the other hand, the maximum power density of the cell of Tₜ = 1400°C at 550°C was 1.4 times higher than that of the cell of Tₜ = 1260°C.

In order to understand the behavior shown in Figs. 2, impedance analysis was employed and the results were shown in Figs. 3. At 450°C operating temperature in Fig. 3 (a), the difference in the cell performance was found to be resulted from the difference in the overpotential resistance, which probably related to gas diffusion in the anode. We have shown that the porosity of the anode tube was strongly affected by the sintering temperature. Therefore, it can be concluded that the anode microstructure has strong influence on the performance at 450°C operating temperature.

On the other hand as shown in Fig. 3 (b), the difference in the overpotential resistance is small, and the ohmic resistance (electrolyte resistance) is improved at the sample with Tₜ = 1400°C. Since the OCV is almost the same for two samples, the sintering temperature of 1260°C is reasonably sufficient from the densification point of view, however, the conductivity of the electrolyte seems not. As a result, the performance of the cell with Tₜ = 1400°C showed higher power density. Thus, it is really needed to optimize the fabrication process depending upon the operation conditions when ceria based material is selected for electrolyte.

4 Conclusion

Two samples were prepared by co-sintering anode supported tube and electrolyte (Gd doped CeOₓ) using the sintering temperatures of 1260 and 1400°C, respectively, and were examined in order to observe the effect
of sintering temperature on the SOFC performance operated at less than 600°C. The cell performance test showed that both samples had advantages depending up on the operating temperatures. The sample sintered at 1260°C showed better cell performance operated at 450°C, as high as 0.15 Wcm⁻², while the sample sintered at 1400°C showed the power density of 0.49 Wcm⁻² at 550°C, 1.4 times better than that of the sample sintered at 1260°C. This behavior is resulted from the difference in the conductivity of the electrolyte and the microstructure of the anode, which were prepared from different sintering temperatures. Two important factors can be noted as follows:

(1) The conductivity of the ceria based electrolyte depends up on the sintering temperature, which is probably related to the grain size of the electrolyte. The effect tended to be larger when the operating temperature increased to 550°C.

(2) Microstructure of the anode (porosity) influences the cell performance at lower operating temperature around 450°C, which lead to the better cell performance for the sample with Tₛ = 1260°C.

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References