



Short communication

One-step sintering process of gadolinia-doped ceria interlayer–scandia-stabilized zirconia electrolyte for anode supported microtubular solid oxide fuel cells

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ABSTRACT

A microtubular solid oxide fuel cell (SOFC) consisting of Gd-doped CeO₂ (GDC) interlayer–Sc stabilized zirconia (ScSZ) electrolyte has been prepared on an anode supported tube using one step sintering process at 1350 °C. The cell performance under high fuel utilization condition, with H₂ flow rate as low as 4.3 mL min⁻¹ per 1 cm² electrode area is evaluated. The one-step sintering process results in an improvement of open circuit voltage but an increase of ohmic resistance due to possible high resistive layer and voids observed between GDC and ScSZ layers. The performance of the cell shows power densities of 0.35–0.57 W cm⁻² with corresponding energy efficiencies of 45–20% (LHV), respectively, at 700 °C. Impedance analyses at different voltages have shown that the overpotential due to gas transport is dominant in the low current density region.

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1. Introduction

Solid-oxide fuel cells (SOFCs), which are known to be high efficiency energy devices, have been intensively studied for a variety of applications [1–3]. In order to accelerate the commercialization of SOFCs, lowering the operating temperature of SOFCs is of importance, since it can broaden the selection of materials, and increase the life-time of the cell. So far, many studies related to lowering operating temperatures of SOFCs have been reported, mainly focusing on new electrode and electrolyte materials [4–9]. In addition, many research groups have put considerable efforts toward practical application, for examples, direct use of hydrocarbon fuels by selecting appropriate anode materials [10,11] and giving the stability to the cells under quick-startup/shutdown operation [12–14].

Developing a cost-effective fabrication process is another important issue for commercialization of SOFCs. In our previous study, we have shown that the conventional ceramic processes (extrusion and wet-slurry coating) can be applied to fabricate high performance microtubular type SOFC, using conventional, commercially available raw ceramic materials [15]. The fabrication process includes three successive stages of sintering, to make an electrolyte–anode support at 1350 °C, an interlayer (Gd-doped CeO₂ (GDC)) at 1200 °C, and a cathode at 1100 °C. The GDC

interlayer acts as a protective layer to prevent a reaction between a cathode (especially, cobaltite perovskite type oxide) and a stabilized zirconia electrolyte to form La₂Zr₂O₇ and SrZrO₃ [16]. A shortcoming of the use of GDC is that it may react with the zirconia electrolyte to form (Ce–Zr)O₂ solid solutions at the interface at high sintering temperature above 1300 °C, which have high resistance. Thus, the sintering temperature of the interlayer must be carefully controlled. On the other hand, various different fabrication processes have been evaluated, and successful examples are the use of pulsed laser deposition (PLD) and co-sintering for fabricating dense Sm doped ceria layer on a zirconia electrolyte [17,18].

In this study, to seek for low cost fabrication process, a one step sintering process has been applied to the GDC interlayer–ScSZ electrolyte on the anode supported microtube and evaluated for cell performance. The cell was tested at high fuel utilization, where the cell performance was typically reduced. We then considered the effect of the co-sintering procedure on the energy efficiency of the single cell as a step toward module fabrication. Impedance analysis at different cell voltages was investigated in order to understand the contribution of electrochemical and gas transport processes on the cell performance in open circuit region.

2. Experimental

Cell fabrication process includes following four steps: (1) preparation of a clay mixture for anode tube extrusion and of slurries of an anode functional layer, an electrolyte, an interlayer, and a cathode

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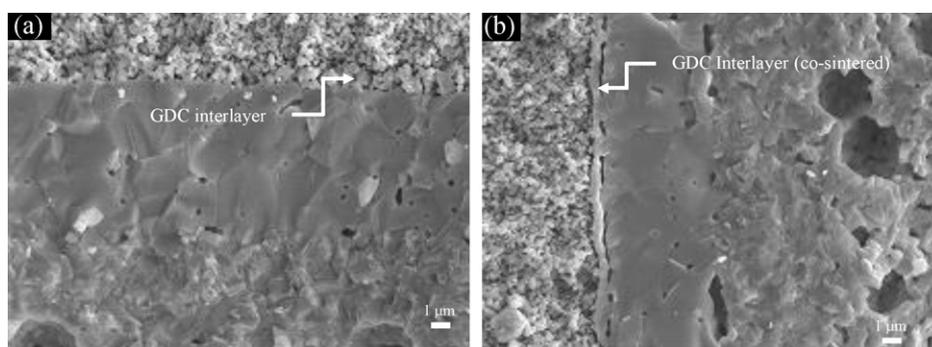


Fig. 1. The cross-sectional fracture SEM image of the microtubular SOFCs prepared using (a) conventional method and (b) one-step sintering process.

for coating process; (2) extrusion of the anode support tubes; (3) the coating process for the anode functional layer, the electrolyte, the interlayer and the cathode; and (4) the sintering process after each coating. The detailed fabrication process and materials for the standard cell were described elsewhere [19].

The microtubular SOFC consisted of an anode (60 wt%: NiO–40 wt%: 8 mol% yttria-stabilized zirconia (YSZ) as a tubular support), an anode functional layer (60 wt%: NiO–40 wt%: 10 mol% scandia-stabilized zirconia (ScSZ)), an electrolyte (ScSZ), an interlayer (10 mol% gadolinia-doped ceria (GDC)), and a cathode (70 wt%: $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-y}$ (LSCF)–30 wt%: GDC).

A one-step sintering process was applied to the ScSZ electrolyte–GDC interlayer. After extruding the anode tube from the clay mixture, the anode tubes were first dipped in the anode functional layer (NiO–ScSZ) slurry and dried in air. Second, they were dipped in the electrolyte (ScSZ) slurry and dried in air. Third, the tube coated with the anode functional layer and electrolyte were dipped in the interlayer (GDC) slurry, and co-sintered at 1350 °C for 1 h in air. Usually, the GDC interlayer coating is applied after the anode tube with the anode functional layer and the electrolyte has been co-sintered at 1350 °C for 1 h in air. The GDC interlayer is then sintered at 1200 °C. Finally, the cathode slurry was dip-coated and sintered at 1100 °C. The dimension of the resulting cell was 1.93 mm in diameter and 3 cm in length with the cathode length of 10 mm, and an effective electrode area of 0.61 cm². Note that this fabrication process is effective for controlling of the microstructure of the anode tube for the improvement of cell performance [20–22].

A scanning electron microscopy (SEM) (JSM6330F, JEOL Ltd, Tokyo, Japan) was utilized for the observation of the cell structure. The performance of the cell was studied using a potentiostat and impedance analyzer (Solartron Analytical, Hampshire, United Kingdom). Ag wire was used for collecting current from the electrodes, which were both fixed using Ag paste. The current collection from the anode side was made from an edge of the anode tube, and the collection from the cathode side was made from the whole cathode area. 20% H₂–Ar balance fuel was allowed to flow inside the anode tube at a flow rate of 13–47 mL min⁻¹ (H₂ flow rate: 4.3–15.4 mL min⁻¹ per 1 cm² electrode area) for power output performance testing. Air was supplied at the cathode side at a flow rate of 100 mL min⁻¹.

3. Results and discussion

Fig. 1 shows SEM images of the microtubular SOFCs prepared using (a) the conventional process (GDC interlayer separately sintered at 1200 °C); and (b) the one-step sintering process. As can be seen, while the GDC interlayer prepared using conventional method included large amount of pores among GDC grains (Fig. 1(a)), the co-sintered GDC interlayer became dense, although

it included voids between the GDC interlayer and the electrolyte (Fig. 1(b)). This has also been observed in the literature [16,23], yet the mechanism of generating voids are not well understood. It is reported that unknown Zr–Sc–Ce–Gd oxide solid solution, which has low ionic conductivity, was detected at the interface and this is probably related to the formation of voids. Thus, an increasing in contact resistance is expected for cells prepared using one-step sintering process.

Fig. 2 shows the performance of the microtubular SOFCs prepared by the one-step sintering process at 700 °C furnace temperature at the fuel (20% H₂ in Ar) flow rate of 13–47 mL min⁻¹. The cell temperature at this furnace temperature was about 692 °C, and stable at each fuel flow rate. As can be seen, the performance of the cell showed 0.35, 0.42, 0.5, 0.55 and 0.57 W cm⁻² maximum power density at fuel flow rates of 13, 17, 25, 34 and 47 mL min⁻¹, respectively. In our previous study [19], a cell prepared using the conventional process showed 0.52–0.88 W cm⁻² at fuel flow rates of 17–47 mL min⁻¹, showing about 19% and 35% reduction from the maximum power density was observed at fuel flow rates of 17 and 47 mL min⁻¹, respectively. It is worth noting that the performance of the cell prepared using one-step sintering process reached a limiting value at high fuel gas flow rates, where the cell would be expected to show increased output power. The high resistance layer between the GDC interlayer and the electrolyte seemed to be the reason for this behavior. The advantage of the cell fabricated using one-step sintering process is seen to be the improvement in the open circuit voltage (OCV) to 1.08 V at the fuel flow rate of 17 mL min⁻¹, compared to 1.01 V for the conventional cell at the same condition. The dense GDC layer (reducing fuel crossover) is expected to improve fuel efficiency. Another possibility was

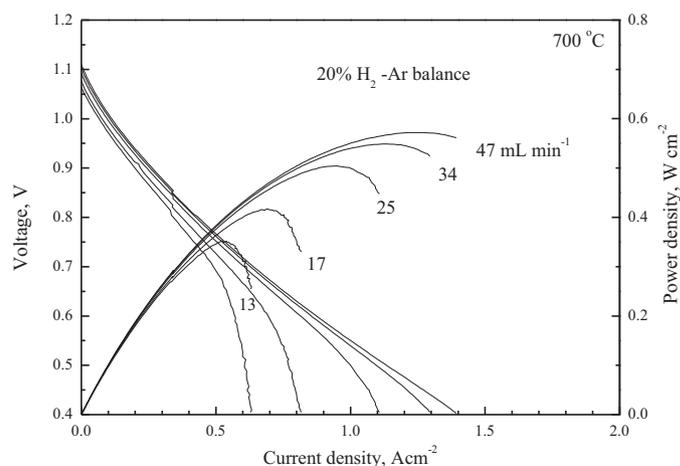


Fig. 2. The IV characterization of the cell prepared using the one-step sintering process at fuel (20% H₂ in Ar) flow rates of 13–47 mL min⁻¹ at 700 °C.

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