



Performance improvement of inert-substrate-supported tubular single cells via microstructure modification



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HIGHLIGHTS

- Inert-substrate-supported tubular single cells are prepared.
- The microstructure and electrochemical performance of the cells are investigated.
- Increasing anode current collector thickness renders a lowered ohmic loss.
- Modifying cathode microstructure allows a reduced polarization loss.
- An improvement of MPD up to 555 mW cm⁻² is gained at 800 °C in hydrogen.

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ABSTRACT

Inert-substrate-supported tubular single cells, with a configuration of porous yttria-stabilized zirconia (YSZ) supporter/Ni anode current collector/Ni–Ce_{0.8}Sm_{0.2}O_{1.9} anode/YSZ/Ce_{0.8}Sm_{0.2}O_{1.9} bi-layer electrolyte/La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-δ} cathode, have been fabricated. The Ni anode current collector layer is prepared by a dip-coating process, while the cathode layer is produced by drop-coating and dip-coating processes, respectively. The electrochemical performances of the single cells are examined with respect to the microstructures of these two layers. The results indicate a significant effect of the Ni layer thickness on its sheet resistance and the ohmic resistance of the single cells. The preferred dip-coating time of the Ni layer is determined to be four. At 800 °C in hydrogen fuel, the single cell with a Ni layer dip-coated for four times and a drop-coated cathode layer exhibits a maximum power density of 500 mW cm⁻². The electrochemical performance of the single cell is further enhanced by modifying its cathode microstructure. Preparing the cathode layer by dip-coating for four times allows a decrease of the overall electrode polarization by 25% and an increase of the maximum power density to 555 mW cm⁻².

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1. Introduction

Solid oxide fuel cells (SOFCs) are energy conversion devices with high efficiency offering environmental advantages. Tubular SOFCs show merits of reduced sealing area and high mechanical strength. They can endure rapid start up and shut down cycles, which is a significant practical advantage over their planar counter-parts [1–3]. In the past decade, Ni-containing anode supported tubular SOFCs have become a research focus because of their low ohmic and polarization losses together with high maximum power densities (MPD) [4–6]. However, the Ni-containing anode supporters

suffer from poor mechanical stabilities under redox cycling conditions, arising from the reduction and oxidation of the nickel. This problem leads to the expansion of anode supporters, occurrence of cracks in the electrolyte layers and ultimately performance degradation or even failure of the single cells [7–10].

To improve the redox cycling stability of Ni-containing anode supported cells, the mechanism and kinetics of the nickel oxidation and reduction processes in the anode have been extensively studied [8,11,12]. High porosity and large pore size have been found to be favorable for the structure stability of the anode in redox cycles. Moreover, the effect of nickel particle size on the redox cycling performance has been evaluated [9,13]. It has been demonstrated that coarse nickel particles are beneficial for the redox stability of the anode supporters. In general, the redox cycling performances of Ni-containing anode supported single cells remain far from

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satisfactory in view of the requirements for commercial application of SOFC technology.

Various attempts have been made to improve the redox cycling stability of Ni-containing anode supported single cells. Among them, one effort is to modify their configurations by using the mechanical modeling method [8,14,15]. An inert-substrate-supported configuration is expected to give high redox cycling stability on account of good mechanical stability of the inert-substrate supporter. Recently, our preliminary results have experimentally validated this expectation, with an inert-substrate-supported tubular single cell, using a porous yttria-stabilized zirconia (YSZ) supporter and Ni anode current collector. The single cell displays a good redox cycling performance within seven redox cycles at 800 °C [16].

Nonetheless, the electrochemical performance (MPD = 337 mW cm⁻² at 800 °C) of the inert-substrate-supported tubular single cell is unsatisfactory, as compared with that (MPD = 925 mW cm⁻² at 800 °C) of the anode-supported coin-type single cell employing identical cell materials (anode, electrolyte and cathode) [16,17]. This could be ascribed to the differences in configuration between the two cells. A long current path and thin current collector layer at the anode side would be responsible for the large ohmic resistance of the inert-substrate-supported tubular single cell [18]. Then optimizing the microstructure of the Ni anode current collector layer should be helpful for improving the performance of the tubular single cell. On the other hand, cathodic polarization, as widely recognized, accounts for a dominant part of the total electrode polarization of a single cell [19–21]. As such, a modified cathode microstructure should be contributive to reduction of the total electrode polarization loss. In this context, it is anticipated that the electrochemical performance of the inert-substrate-supported tubular single cell could be enhanced by modifying the microstructures of these two layers.

In this work, we have produced inert-substrate-supported tubular single cells with a configuration of porous YSZ supporter/Ni anode current collector/Ni–Ce_{0.8}Sm_{0.2}O_{1.9} (SDC) anode/YSZ/SDC bi-layer electrolyte/La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-δ} (LSCF) cathode. The microstructures of the anode current collector layer and the cathode layer have been tuned by controlling their fabrication processes and conditions. The electrochemical performances of the single cells have been evaluated in relation to the microstructures of the two layers.

2. Experimental

2.1. Powder preparation

SDC powder and NiO-SDC powder containing 61.3 wt.% NiO were synthesized by urea combustion. LSCF powder was synthesized by glycine-nitrate combustion. The details of the powder syntheses have been reported previously [22–24].

2.2. Fabrication of the single cell

The porous YSZ supporter was fabricated by an extrusion process. Poly(methyl methacrylate) (S50, Sunjin Chemical) pore former was mixed with the YSZ (TZ-8Y, TOSOH Co., 99.9%) powder to make the porous YSZ supporter. The poly(methyl methacrylate) pore former amount was determined to be 25 wt.% based on the results of our previous research [16]. 16 wt.% NiO and 1.8 wt.% poly(methyl methacrylate) powders were mixed with a lab-made organic carrier (I) to form the slurry for the anode current collector. The organic carrier (I) was composed of 48 wt.% 2-butanone (Samchun Chemicals), 48 wt.% ethanol

(Hayman Specialty Products), 0.5 wt.% triethanolamine (Samchun Chemicals), 1.0 wt.% dibutyl phthalate (Aldrich), 1.0 wt.% polyethylene glycol 300 (Showa Chemical), and 1.5 wt.% Butvar B-98 (Sigma). The NiO layer was deposited on the outer surface of the YSZ supporter by dip-coating. Repetitive coatings were carried out from one to five times to tailor the thickness of the layer. To prevent the formation of cracks and pin-holes during sintering due to the evaporation of the organics, each layer was dried in air for 12 h and then calcined at 600 °C for 1 h before the coating of the next layer.

16 wt.% NiO-SDC and 1.8 wt.% poly(methyl methacrylate) powders were mixed with the organic carrier (I) to form the slurry for the anode. The NiO-SDC was coated onto the surface of the NiO layer via the same dip-coating process. The samples were heated at 1000 °C to adjust the shrinkage behavior in the sintering process.

17 wt.% YSZ powder was mixed with lab-made organic slurry (II) by ball-milling for 24 h. The organic slurry (II) was comprised of 40 wt.% α -terpineol (Kanto Chemical Co. Inc), 30 wt.% B73210 organic binder (Ferro Electronics Materials), and 30 wt.% ethanol. The YSZ layer was deposited on the surface of the NiO-SDC layer by dip-coating for one time. The SDC layer was fabricated on the surface of the YSZ layer using the same process. The samples were then co-sintered at 1400 °C for 4 h in air. The inner and outer diameters of the tube were 2.3 and 4.0 mm, respectively.

Finally, the LSCF cathode layer with a surface area of 1.00 cm² was coated onto the surface of the SDC electrolyte layer by drop-coating and dip-coating, respectively. For the drop-coating process, 14 wt.% LSCF powder was mixed with organic slurry (III) containing 1 wt.% ethyl cellulose (Showa Chemical) and 99 wt.% ethanol by ball-milling for 24 h to obtain a cathode ink. The ink was dropped onto the SDC surface when the tube was rotated at a constant speed by an electric motor. As for the dip-coating process, 10 wt.% LSCF powder was mixed with a lab-made organic carrier (IV) to form a slurry for the cathode. The organic carrier (IV) was composed of 63 wt.% 2-butanone, 31 wt.% ethanol, 0.5 wt.% triethanolamine, 1.0 wt.% dibutyl phthalate, 1.0 wt.% polyethylene glycol 300, and 3.5 wt.% Butvar B-98. The cathode slurry was coated on the SDC surface by dip-coating. Repetitive dip-coatings were carried out from one to five times to change the thickness of the cathode. The dip-coating process of the cathode is similar to that for producing the anode current collector. The cathode was sintered at 950 °C for 2 h in air.

2.3. Structure characterization

The porosity of the YSZ supporter was examined using a porosimeter (AutoPore IV 9500, Micromeritics). The YSZ supporter showed a porosity of 39% after sintering at 1400 °C, which was in accordance with our previous results [16]. The microstructure of the cells was investigated by a scanning electron microscope (SEM, JSM-6400, JEOL). The thicknesses of the Ni and Ni-SDC layers were determined by an energy-dispersive X-ray spectroscopy (EDX, APOLLO X) using the line scan mode. The thicknesses of the LSCF, SDC, and YSZ layers were estimated by an image analysis method using the Image-Pro Plus 6.0 software [25].

2.4. Electrical resistance

A four-probe method was applied to measure the sheet resistance of the Ni layer and LSCF layer, respectively. The sheet resistance of the Ni layer on the porous YSZ supporter was measured in hydrogen, while the sheet resistance of the LSCF layer on the surface of SDC was obtained in air [16].

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