Anode-supported solid oxide fuel cells based on $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ electrolyte fabricated by a phase-inversion and drop-coating process

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Abstract:

The performance of anode-supported solid oxide cells is often limited by gas transport in the thick anode substrate. In this study, anode-supported solid oxide fuel cells with a cell configuration of $\text{Ni-Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ (SDC) | SDC | $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$ (SSC)-SDC are fabricated by a combination of phase-inversion tape-casting, direct drop-coating on the green anode, and co-firing. The porous anode substrate is composed of a thin skin layer, a thick finger-like porous layer, and a thin sponge-like porous layer. The skin layer with fine pores offers extended three-phase boundaries (TPBs) at the electrolyte/anode interface while the finger-like porous layer/support provides pore channels for efficient mass transport. Dense SDC electrolyte membranes with a thickness of $\sim 15\text{ }\mu\text{m}$ are obtained at a relatively low co-firing temperature (1400°C) due to the large sintering shrinkage of the green anodes. Peak power densities of 0.608 Wcm^{-2} at 600°C and 0.789 Wcm^{-2} at 650°C have been demonstrated using humidified (3 vol. % water vapor) H_2 as fuel and ambient air as oxidant, suggesting that the

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triple-layered anode effectively decreases the anode polarization resistance and thus enhances the cell performance.

Keywords: Anode-supported solid oxide fuel cells; phase-inversion; direct drop-coating; $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ electrolyte.

1. Introduction

Solid oxide fuel cells (SOFCs) hold promise to be the most efficient and environmentally benign system for power generation from a wide variety of readily available fuels, including coal gas, natural gas and gasified carbonaceous solid fuels (e.g. municipal solid waste and biomass).^[1-6] Among all cell configurations studied, anode-supported SOFCs have attracted significant attention because it allows thinner electrolyte and hence lower Ohmic resistance.^[7, 8] However, the concentration polarization due to mass transport through the relatively thick anodes may impact the performances of such cells.^[9, 10] Aiming to achieving lower concentration polarization loss, porous anode substrates have been proposed and fabricated by tape-casting^[11, 12] and dry-pressing^[13, 14] with additions of pore formers (e. g., graphite, starch). However, the pores/paths are often tortuous and irregular, easily resulting in high resistance to gas transport.^[15] A phase-inversion method has been studied for a facile, one-step fabrication of triple-layered anodes for micro-tubular SOFCs.^[16-20] The anodes consist of a thin skin layer, a thick finger-like porous layer, and a thin sponge-like porous layer. The thick finger-like porous layer can offer more gas transport channels in the anode substrate, thus decreasing the concentration losses.

Compared with the tubular configuration, the planar SOFCs have gained more

popularity due to several advantages, including shorter current paths, higher power density, and reduced manufacturing cost.^[21, 22] However, only few works focus on the fabrication of the planar anode-supported SOFCs by phase-inversion.^[23, 24] Besides, the fabrication of anode-supported SOFCs usually undergoes multiple-steps,^[18, 25] including preparation and pre-sintering of anode substrate, depositing of an electrolyte layer, co-firing of the anode/electrolyte bi-layer, and final coating and firing of a cathode layer. Each step involves at least one high-temperature firing process, making the cell fabrication time-consuming and costly.^[26]

In this study, a planar triple-layered Ni- $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ (SDC) anode was fabricated by a phase-inversion tape-casting process, **during which certain slurry was tape-casted and then solidified via phase-inversion.** The SDC electrolyte slurry was directly drop coated on the surface of the green anode, followed by drying and co-firing. The process without pre-heating of anode substrates is simpler and more cost-effective. Finally, single cells with a configuration of Ni-SDC|SDC| $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$ (SSC)-SDC were fabricated and evaluated.

2. Experimental

Triple-layered anodes were prepared by a phase-inversion tape-casting method. A mixture of NiO (Alfa Aesar, USA) and SDC (Fuel cell materials, USA) powders with a weight ratio of 65:35 was ball-milled with ethanol for 24 h and then dried. Polyethersulfone (PESf, Radel A-300, Ameco Performance, USA) and polyvinylpyrrolidone (PVP, K30, Aldrich, USA) were dissolved into N-methyl-2-pyrrolidone (NMP, Alfa Aesar, USA) to form stable casting solutions.

Subsequently, the dried NiO-SDC powders were added into the casting solutions and ball-milled for 48 h to get a well-dispersed slurry with a weight ratio of NiO-SDC:NMP:PESf:PVP=100:40:10:2.2. After evacuation under vacuum for 30 min, the slurry was tape-casted, and then transferred into a water bath for solidification via a phase-inversion process. The solidified green tape was punched into pellets with a diameter of 13 mm and dried at room temperature. The SDC electrolyte slurry was directly deposited on the green anode substrate by drop coating ^[27]. After drying, the NiO-SDC anode/SDC electrolyte bi-layer was co-fired at 1400°C for 5 h, with a heating rate of 2°C/min below 600°C and 3°C/min above 600°C. As a comparison, NiO-SDC (65:35 wt. %) anode substrates with 15 wt. % graphite additives as pore formers were fabricated by dry-pressing under 360 MPa, and then pre-sintered at 800°C for 2 h. After drop-coating the SDC electrolyte slurry on the surface of the pre-sintered anode substrates, the anode/electrolyte bi-layers were co-fired at 1500°C for 5 h. Finally, the SSC-SDC cathode slurry with a weight ratio of SSC:SDC=70:30 was screen-printed onto the top of the SDC electrolyte layer, followed by firing at 950°C for 2 h to form the cathode. The effective area of the cathode was 0.31 cm².

The current density-voltage curves and electrochemical impedance spectra (EIS) of the cells were acquired using a multi-channel Versa STAT (Princeton Applied Research) at the operating temperatures ranging from 500 to 650°C. NiO paste and silver paste (both purchased from Heraeus, USA) were employed as current collectors on the anode and the cathode side, respectively. Silver wires were used as the lead wires. During cell measurement, humidified H₂ (with 3 vol. % H₂O) with flow rate of

40 sccm was used as the fuel while the cathode side was exposed to ambient air. The cells were first stabilized at 650°C for 30 min to allow complete reduction of NiO to Ni. The impedance spectra were collected under open circuit condition with an AC amplitude of 10 mV in the frequency range of 0.1 Hz to 1 MHz. The morphology and microstructure of the green anode support and the single cells after testing were characterized using a field emission scanning electron microscope (LEO 1530).

3. Results and discussion

Fig. 1(a) shows a typical cross-sectional view (SEM image) of the triple-layered green anode composed of a thin skin layer, a thick finger-like porous layer, and a thin sponge-like porous layer; all the layers are well bonded. The diameters of the finger-like pores vary along the thickness direction, increasing from several microns (near the skin layer) to ~ 60 μm (away from the skin layer). Such structure is expected to allow rapid transport of fuel gas through the anode support layer to the reaction sites as well as the fast removal of product (e. g., water vapor) from the reaction sites during cell operation, thus minimizing the concentration loss from gas transport through the porous anode. The formation of the finger-like pores can be attributed to the exchange between the solvent (NMP) in the slurry and the non-solvent (water).^{[28,}
^{29]} During the exchange process, the sponge-like layer with finer porous structure is formed due to the steep concentration gradient, and the skin layer is originated from the fast exchange between the solvent and non-solvent ^[30, 31]. It is shown in Fig 1 (b) that the region near skin layer contains some micron-channels with a diameter of 1-5 μm , allowing the SDC electrolyte slurry to penetrate into the green anode by direct

drop-coating. To further exhibit the detailed structure, especially the distribution of Ni and SDC particles, the NiO-SDC anode substrate fired at 1400°C for 5 h is reduced to Ni-SDC under H₂ at 650°C. Shown in Fig. 1(c) is the detailed microstructure of the anode backbone around the finger-like pores. It can be clearly seen that the porous Ni and SDC composite anode has a uniform microstructures, providing pore channels for fast gas transport as well as continuous paths for ionic and electronic current flow. In addition, the sponge-like Ni-SDC layer with a finer porous structure at the bottom of the anode substrate (Fig. 1(d)) may act as an effective current collecting layer during cell operation.

Shown in Fig. 2(a) is a cross-sectional view of the triple-layered anode supported cell after testing. It is observed that the SDC electrolyte membrane with a thickness of ~15 µm bonds well to both porous electrodes. The thickness of cathode and anode are ~20 and ~550 µm, respectively. The cathode shows a homogeneous porous structure consisting of fine particles (Fig. 2(b)). The SDC electrolyte membrane is crack-free, continuous, and quite dense with grain sizes of 0.3-1 µm (Fig. 2(c)). The co-firing temperature here (1400°C) is lower than that of the multiple-step process in this study (1500°C for 5 h) and reported elsewhere (1500°C for 6 h) ^[32]. Densification of SDC electrolyte at lower temperatures is due most likely to the larger sintering shrinkage of the green anode compared with the pre-sintered ones. As shown in Fig. 2(d), the skin layer near electrolyte/anode interface shows a homogeneous and fine porous structure, which is beneficial for expanding the triple phase boundaries (TPBs).

Fig. 3 shows the cross-sectional view of the cells fabricated by a conventional

dry-pressing method. No open pores are observed in the SDC electrolyte membrane. The thicknesses of the electrolyte, cathode and anode are 15, 20, 550 μm , respectively, which are similar to those of the triple-layered anode supported cells. As shown in Fig. 3(b), the anode shows a sponge-like structure with some random and irregular pores, which is the only difference between the cells fabricated by the phase-inversion and the dry-pressing method.

Fig. 4(a) shows the I-V and I-P curves of the triple-layered anode supported cells measured at operating temperatures of 500 to 650°C with humidified H_2 as the fuel and ambient air as the oxidant. Peak power densities of 0.182, 0.359, 0.608, and 0.789 Wcm^{-2} with the open circuit voltage (OCV) values of 0.91, 0.88, 0.85 and 0.81 V were obtained at 500, 550, 600 and 650°C, respectively. The OCV values are similar to those for single cells based on dense SDC electrolyte membranes reported elsewhere,^[32, 33] indicating that the SDC electrolyte membrane is sufficiently dense, consistent with the SEM images in Fig. 2 (b) and (c). Under similar testing conditions, the power densities of the cells fabricated by the phase-inversion method is much higher than those of the cells fabricated by the dry-pressing method (0.521 Wcm^{-2} at 650°C, see Fig. 4(b)). Also, the cell performances are higher than those reported by others for the cells based on SDC electrolyte with the identical cathode and anode material but with a sponge-like porous anode fabricated by dry-pressing (e. g., 0.397 Wcm^{-2} ^[34] at 600°C, 0.549 Wcm^{-2} ^[32] at 650°C).

To provide more detailed information, typical electrochemical impedance spectra of these cells are acquired at different temperatures under open circuit conditions, as

shown in Fig. 5(a). The spectra interception with the real-axis at high frequencies represents the ohmic resistance (R_o) of the cell, and the interception in low frequencies represents the total resistance (R_t) of the cell. The total polarization resistance (R_p) of the cell is obtained from the equation below ^[35] :

$$R_p = \frac{R_t - R_o}{\frac{V_{oc}}{E_n} \left[1 - \frac{R_o}{R_t} \left(1 - \frac{V_{oc}}{E_n} \right) \right]}$$

where E_n is the Nernst potential and V_{oc} is the OCV. The calculated R_p and R_o values are plotted in Fig. 5(b). As the operating temperatures increase from 500 to 650°C, the R_p decreases from 2.185 to 0.173 $\Omega \text{ cm}^2$ while the R_o decreases from 0.438 to 0.139 $\Omega \text{ cm}^2$. This can be attributed to the increased ionic conductivity of the electrolyte and faster electrode kinetics with increasing temperature. ^[23]

Shown in Fig. 6 is a direct comparison of impedance spectra measured at 650°C for cells with different anode architecture. The cell fabricated by the phase-inversion method shows a much lower R_t , while the calculated R_p is only ~65% of that of the cell fabricated by the dry-pressing method (0.265 $\Omega \text{ cm}^2$). Considering the fact that the materials for both the anode and the cathode are identical, and the thickness and microstructure of the cathode are similar in two types of cells, the decrease of R_p may be attributed mainly to the unique anode architecture. The R_p comes mainly from the resistance of gas diffusion, gas conversion, and charge transfer. Among them, the gas diffusion resistance is most likely to play a major role due to the large difference of anode architectures between two types of anode. The gas diffusion resistance ($R_{\text{Diffusion (anode)}}$) can be estimated by the following equation^[9]:

$$R_{\text{Diffusion(anode)}} = \left(\frac{RT}{2F}\right)^2 l_a \frac{1}{D_{\text{H}_2\text{O}, \text{H}_2}} \frac{\tau_a}{\rho_a} \left(\frac{1}{P_{\text{H}_2}} + \frac{1}{P_{\text{H}_2\text{O}}}\right) \times \left(1.0133 \times 10^5 \frac{\text{Pa}}{\text{atm}}\right)^{-1}$$

where R , T , F , l_a , $D_{\text{H}_2\text{O}, \text{H}_2}$, τ_a and ρ_a represent the universal gas constant, absolute temperature, Faraday's constant, anode thickness, binary diffusion coefficient for a mixture of H_2O and H_2 , the tortuosity factor of the hydrogen electrode, and the volume fraction porosity of the anode, while P_{H_2} and $P_{\text{H}_2\text{O}}$ represent the partial pressure of hydrogen and water vapor in the anode. Compared to the sponge-like porous anode, the tortuosity of the porous anode with finger-like pores is greatly reduced.^[18, 36] Typically, the tortuosity factor for the anode substrate with straight pores structure is about 2,^[37, 38] much lower than that of the anode substrate with sponge-like structure (with a tortuosity factor value of 6-10^[39]). In addition, the average porosity of the triple-layered anode after reduction of NiO to Ni upon sintering in H_2 is measured to be ~41% by Archimedes method, higher than that of the sponge-like anode fabricated by dry-pressing method (~24%). Therefore, the gas diffusion resistance from the anode fabricated by phase-inversion process is much lower. In our previous work, we found that the electrode with straight pores could effectively facilitate not only the gas diffusion but also the ion-infiltration.^[40-42] It is believed that the cell performance can be further enhanced by catalyst infiltration.

4. Conclusions

To decrease the concentration loss from gas transport in anode-supported SOFCs, a phase-inversion tape-casting process was used to fabricate a triple-layered anode composed of a thin skin layer, a thick finger-like porous layer, and a thin sponge-like porous layer. The skin layer with fine microstructures can offer more TPBs at

electrolyte/anode interface. The finger-like pores can facilitate the fuel gas transport to the reaction sites and the water vapor away from the reaction sites. The sponge-like layer with fine pore sizes can act as the current collecting layer during cell operation. The dense SDC electrolyte membranes were fabricated by direct drop-coating SDC slurry on the green anode with a subsequent co-firing at a relatively lower temperature, making the fabrication process simpler and more cost-effective. The single cells demonstrate peak power densities of 0.608 Wcm^{-2} at 600°C and 0.789 Wcm^{-2} at 650°C , higher than those of the cells with sponge-like porous anodes.

Acknowledgement

This work was supported by the US Department of Energy ARPA-E REBELS Program (under award number DE-AR0000502) and the National Natural Science Foundation of China (No. 51472146). Haibin Sun acknowledges a fellowship from the China Scholarship Council (2014-3026).

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Figure captions:

Fig. 1 (a) A cross-sectional view (SEM image) of the NiO-SDC green anode substrate fabricated by phase-inversion tape-casting, and closer views (higher magnification) of (b) **region near** the skin layer of the green anode substrate, (c) the anode backbone around finger-like pores, and (d) the sponge-like layer of the anode substrate after firing and reduction.

Fig. 2 (a) A cross-sectional view (SEM image) of the triple-layered anode supported cell after testing, and closer views (higher magnification) of (b) the cathode, (c) the SDC electrolyte membrane, and (d) the electrolyte/anode interface.

Fig. 3 (a) A cross-sectional view (SEM image) of the cells fabricated by a conventional dry-pressing method, and (b) a closer view (higher-magnification) of the anode.

Fig. 4 (a) I-V and I-P curves of triple-layer anode supported cells measured at different temperatures using hydrogen as fuel and ambient air as oxidant; (b) Comparison of I-V and I-P curves of cells prepared by phase-inversion and dry-pressing method measured at 650°C.

Fig. 5 (a) Impedance spectra of triple-layered anode supported cells measured at different temperatures under open circuit conditions, (b) R_o and calculated R_p of triple-layered anode supported cells measured at different temperatures using hydrogen as fuel and ambient air as oxidant.

Fig. 6 Comparison of impedance spectra measured at 650°C for cells fabricated by a phase-inversion and a dry-pressing method.

Fig. 1(a)

Fig. 1(b)

Fig. 1(c)

Fig. 1(d)

Fig. 2(a)

Fig. 2(b)

Fig. 2(c)

Fig. 2(d)

Fig. 3(a)

Fig. 3(b)

Fig. 4(a)

Fig. 4(b)

Fig. 5(a)

Fig. 5(b)

Fig. 6