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Long-term stability of infiltrated $\text{La}_{0.8}\text{Sr}_{0.2}\text{CoO}_{3-\delta}$, $\text{La}_{0.58}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ and $\text{SmBa}_{0.5}\text{Sr}_{0.5}\text{Co}_{2.0}\text{O}_{5+\delta}$ cathodes for low temperature solid oxide fuel cells

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ABSTRACT

Here we report the electrochemical performance and long-term stability of $\text{La}_{0.8}\text{Sr}_{0.2}\text{CoO}_{3-\delta}$ (LSC), $\text{La}_{0.58}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ (LSCF) and $\text{SmBa}_{0.5}\text{Sr}_{0.5}\text{Co}_{2.0}\text{O}_{5+\delta}$ (SBSC) infiltrated (ZrO_2)_{0.89}(Sc_2O_3)_{0.1}(CeO_2)_{0.01} (SSZ) cathodes at low temperatures. At 700 °C, the initial polarization resistance of the infiltrated cathodes increased in following order: SBSC-SSZ (0.054 Ω cm²) < LSC-SSZ (0.084 Ω cm²) < LSCF-SSZ (0.140 Ω cm²). After the heat treatment at 620 °C (820–1400 h), the degradation rate of the polarization resistance of LSC-SSZ, LSCF-SSZ and SBSC-SSZ cathode was 179%, 53.9% and 93.1% kh⁻¹, respectively, while that of the ohmic resistance was 10.6%, 8.86% and 40.9% kh⁻¹, respectively. X-ray diffraction (XRD) and scanning electron microscope (SEM) observation showed that the degradation was mainly caused by the morphological change of the infiltrated particles while the solid reactions between the infiltrated materials and SSZ backbones were not observed.

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Introduction

Solid oxide fuel cells (SOFCs) are energy conversion devices that directly convert chemical energy into electricity with high efficiency and low carbon footprint [1]. Recently, there is a trend to reduce the operation temperature of SOFC to lower temperature (LT) range (<650 °C) in order to reduce system cost, degradation rate as well as start-up and shutdown time

[2]. However, electrochemical impedance of SOFC components will increase with reduced operating temperatures. Infiltration technique is a promising approach for fabricating high performance electrodes with low polarization resistances (R_p) [3–7]. Because the calcination temperature of the infiltrated electrode is much lower than that of the traditional electrode, e.g., 1000–1200 °C for $\text{La}_x\text{Sr}_x\text{Mn}_x\text{O}_x$ (LSM) cathode and 1300–1400 °C for Ni/yttria-stabilized zirconia

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(YSZ) cermet anode, the grain growth is inhibited, resulting in nano-scale particles deposited into the scaffold. Such nano-scale particles increase the effective length of the triple phase boundaries (TPBs) or the number of the active sites for electrochemical reactions and thus achieve high catalytic activities at low temperatures [2]. In addition, due to the low fabrication temperature of the infiltration technique, the solid reaction between electrode and electrolyte materials can be suppressed in some degree. Also, the infiltrated electrode exhibits an improved matching in coefficient of thermal expansion (CTE) with the electrolyte in comparison with the electrode prepared by screen printing and sintering process [3–7].

The infiltration technique was firstly introduced into the SOFC area by Gorte et al. to prepare the Cu–CeO₂ anode with high tolerance to carbon deposition [8–10]. Subsequently, this technique was widely applied by researchers to make high-performance electrodes. For example, the polarization resistance of the Ni infiltrated La_{0.9}Sr_{0.1}Ga_{0.8}Sr_{0.2}O_{3-d} (LSGM) anode was only 0.011 Ω cm² when measured at 550 °C [11]. Sholklauber et al. have demonstrated the performance improvement by direct loading of LSM nano-particles onto a porous YSZ scaffold [12]. Shao et al. showed that the R_p of Sm_{0.2}Sr_{0.8}CoO_{3-δ} infiltrated Sm_{0.2}Ce_{0.8}O_{1.9} (SDC) cathode was only 0.05 Ω cm² at 700 °C [13]. Furthermore, SSC infiltrated LSGM cathode with a low R_p of 0.021 Ω cm² at 650 °C was reported by Han et al. [14].

However, long-term stability of the infiltrated electrode is a critical issue before the successful commercialization of the infiltration technology, even though significant progress has been made in performance enhancement of cell electrodes. Two main reasons will cause the performance degradation of the infiltrated electrodes: firstly, coarsening of infiltrated nano-particles may occur at SOFC operating temperatures, yielding morphological instability [15–19]; secondly, the solid reaction may also occur between the infiltrated material and the scaffold material [13,20]. Tucker et al. reported that the rapid degradation of the cell performance was primarily due to the coarsening of the infiltrated Ni particles [15]. Zhou et al. also showed that coarsening of the nano-scale Ni caused the degradation of the metal-supported fuel cell operating at 650 °C during the 200 h measurement [21]. Han et al. showed that changing infiltrated materials from pure Ni to Ni-SDC mixture produced an improved stability, which was attributed to the restrict growth of Ni by the surrounding SDC [16]. Shao et al. showed that the R_p of Sm_{0.5}Sr_{0.5}CoO_{3-δ} (SSC) infiltrated YSZ cathodes increased from 0.09 to 0.55 Ω cm² after a 300 h test at 700 °C, due to the formation of insulating phase of SrZrO₃ and Sm₂Zr₂O₇ [13,20].

Although stability and degradation mechanism of the cathodes like La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-δ} (LSCF) infiltrated Gd-doped ceria (GDC), LSCF infiltrated YSZ and La_{0.6}Sr_{0.4}CoO_{3-δ} (LSC) infiltrated YSZ have been investigated at the temperature range of 700–750 °C, long-term (>1000 h) stability of such cathodes at a lower temperature (~600 °C) have rarely been reported [20,22,23]. In this paper, we studied the electrochemical properties and long-term (800–1400 h) stabilities of La_{0.8}Sr_{0.2}CoO_{3-δ} (LSC), La_{0.58}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-δ} (LSCF) and SmBa_{0.5}Sr_{0.5}Co_{2.0}O_{5+δ} (SBSC) infiltrated (ZrO₂)_{0.89}(Sc₂O₃)_{0.1}(CeO₂)_{0.01} (SSZ) cathodes at 550–700 °C.

Experimental

Preparation of cathode materials

The precursor solutions of LSC, LSCF and SBSC were prepared by the Pechini method (citrate method). Stoichiometric amounts of nitrate salts were dissolved into distilled water. Citric acid was then added into the solution with the molar ratio of the citric acid to the total metal ions at 1.5:1. Ammonia was added into the solution to adjust the pH value at ~4 in order to ensure full chelation of citric acid with metal ions. The precursor solutions of LSC, LSCF and SBSC were dried and calcined at 600 °C, 700 °C and 800 °C for 2 h to obtain the corresponding powder. Details for the preparation process can be found in our previous work [16].

Fabrication of symmetrical cells

Commercial SSZ powder ((ZrO₂)_{0.89}(Sc₂O₃)_{0.1}(CeO₂)_{0.01}, Daiichi kigenso kagaku kogyo Co., Ltd Japan) and SDC powder (Sm_{0.2}Ce_{0.8}O_{1.9}, Rarechem high-tech Co., China) with appropriate amounts of dispersant, binder, plasticizer, solvent and other additives were used to obtain the slurries for tape casting. The scaffold of symmetrical cells with a tri-layer structure of “porous SSZ | dense SSZ | porous SSZ” was fabricated by laminating one dense SSZ tape and two porous SSZ tapes on both sides with subsequent co-firing at 1300 °C for 4 h. Thicknesses of the porous SSZ layer and dense SSZ layer were ~40 μm and ~200 μm, respectively. The active area of the symmetrical cells was 0.35 cm². The tri-layer structure of “porous SDC | dense SDC | porous SDC” followed the same process described above. Precursor solutions of LSC, LSCF and SBSC were infiltrated into the porous SSZ scaffold and that of LSCF was infiltrated into the porous SDC scaffold, followed by calcinating at 700 °C for 2 h. The infiltration-calcination process was repeated to achieve desired loadings of the cathode materials. A single infiltration/calcination cycle yielded a loading of ≈ 5 wt% and the ultimate loading was controlled at 30 wt%.

Electrochemical measurements

Electrochemical impedance spectra (EIS) testing was performed under open-circuit conditions in ambient air using an IM6 Electrochemical Workstation (ZAHNER, Germany) between 100 kHz and 0.1 Hz with a 20 mV perturbation. The ohmic resistance (R_o) was obtained from the value of the high frequency intercept. The electrode polarization resistance (R_p) was measured by the difference between the high and low frequency intersections of the impedance spectra in the first quadrant on the real axis (Z_{real} axis), and divided by 2 to account for the contributions of two symmetrical electrodes.

Material characterization

The phase structure of synthesized powder was characterized by an X-ray diffraction (XRD, D8 ADVANCE). The diffraction patterns were collected by step scanning in a 2θ range of 20–80° with the scan rate of 10° min⁻¹. To detect potential

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