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Enhanced performance and stability of interlayer-free $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ - $\text{Ce}_{0.8}\text{Zr}_{0.2}\text{O}_{2-\delta}$ cathode for solid oxide fuel cells

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$\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ cathode

$\text{Ce}_{0.8}\text{Zr}_{0.2}\text{O}_{2-\delta}$ oxide

Interlayer-free

Stabilized zirconia electrolyte

ABSTRACT

Interlayer-free $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ - $\text{Ce}_{0.8}\text{Zr}_{0.2}\text{O}_{2-\delta}$ (LSCF-CZ) composite cathode on YSZ electrolyte has been studied by using X-ray diffraction (XRD), scanning electron microscopy (SEM) and electrochemical impedance spectra (EIS). The addition of CZ suppresses the formation of non-conductive SrZrO_3 at the cathode and YSZ electrolyte interface. As a result, the LSCF-CZ cell performance is greatly elevated. The LSCF-CZ cell exhibits 8.7 times higher current density than the LSCF cell at 0.8 V and 600 °C. The incorporation of oxygen anion from LSCF-CZ cathode into YSZ electrolyte is significantly accelerated. The stability of the LSCF-CZ cathode is remarkably improved as compared with the LSCF cathode at 600 °C.

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Introduction

Perovskite oxides with 3d transition metal Co and/or Fe ions on B-site have excellent catalytic activity for oxygen reduction and superior mixed electronic–ionic conductivity, and are widely studied as cathode for solid oxide fuel cell (SOFC). For instance, $\text{La}_{1-x}\text{Sr}_x\text{Co}_{1-y}\text{Fe}_y\text{O}_{3-\delta}$ (LSCF) cathode displays high electrochemical performance at the intermediate temperature (600–800 °C) [1]. But surface strontium enrichment on LSCF materials causes a bad chemical compatibility with YSZ electrolyte (YSZ = Yttria Stabilized Zirconia) [2–5]. The reaction of LSCF with YSZ to non-conductive SrZrO_3 and/or

$\text{La}_2\text{Zr}_2\text{O}_7$ phases deteriorates the interface of cathode/electrolyte. Many efforts have been given to prevent the reaction at the LSCF/YSZ interface. For instance, LSCF was impregnated into YSZ scaffold and then sintered at low temperature to avoid the interfacial reaction [6]. Gd-doped CeO_2 (GDC) was introduced as a barrier layer to avoid the direct contact between cathode and YSZ electrolyte [7–9]. These methods not only increase the complexity of cell fabrication but also introduce new interface losses between GDC barrier layer and YSZ electrolyte.

It is very important to develop a simple method for improving the LSCF/YSZ interface. A proper modifier can promote the low temperature sintering and improve the

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LSCF/YSZ interface. Bi_2O_3 was added into LSCF cathode to introduce liquid-phase sintering, which yielded a dense LSCF film at the cathode/electrolyte interface and reduced the interface ion transfer resistance [10]. Co_3O_4 as sintering aid in LSCF cathode also optimized the electrode/electrolyte interface. The peak power density of the cell with LSCF- Co_3O_4 cathode reached to 0.26 W cm^{-2} at 600°C [11]. The elemental diffusion or solid state reaction between different phases can be helpful to the improvement of the interface. The solid state reaction between $\text{La}_{1-x}\text{Sr}_x\text{CoO}_{3-\delta}$ (LSC) and MnO_2 greatly restrains the LSC decomposition and stabilizes $\text{La}_{1-x}\text{Sr}_x\text{CoO}_{3-\delta}$ in the presence of chromium species [12].

$\text{Ce}_{1-x}\text{Zr}_x\text{O}_{2-\delta}$ (CZ) oxides are the most used catalytic support because of their outstanding ability to store and release oxygen [13]. Strontium-modification stabilizes CZ oxides, retards the sintering of oxides particles and improves the oxygen storage capacity (OSC) [14]. In this study, $\text{Ce}_{0.8}\text{Zr}_{0.2}\text{O}_{2-\delta}$ - $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ (CZ-LSCF) composite cathode is prepared and applied directly on YSZ electrolyte without CeO_2 -based electrolyte interlayer. The chemical compatibility and reaction between CZ and LSCF are studied by XRD characterization. The performance and stability of CZ-LSCF cathode are investigated based on electrochemical analysis and SEM results.

Experimental

Anode-supported single cells with thin film YSZ ($10 \mu\text{m}$) were fabricated by tape casting method. The home-made $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ (LSCF) powder were synthesized by citric acid ammonium assisted pechini-type method and calcined at 850°C for 4 h. The synthesis details can be seen in the reference [15]. $\text{Ce}_{0.8}\text{Zr}_{0.2}\text{O}_2$ powder was synthesized by a co-precipitation method. The $\text{Ce}(\text{NH}_4)_2(\text{NO}_3)_6$ and $\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$ were used as starting materials. Ammonia water (25wt.%) was applied as precipitator. The precursor solution with $\text{Ce}(\text{NH}_4)_2(\text{NO}_3)_6$ and $\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$ was dropped into ammonia water to yield a yellow cerium–zirconium hydroxides instantly. The hydrous precipitate was filtered and washed with deionized water and ethanol. The dried precipitate was calcined at 400°C for 2 h. The LSCF powder was blended with CZ powder through ultrasonic dispersing in ethanol for 30 min. The cathode slurry, consisting of the dried LSCF-CZ mixtures, organic binders and solvent, was coated on YSZ electrolyte and sintered at 850°C for 2 h.

The current–voltage curves and the corresponding power density (I – V – P) of single cells with LSCF and LSCF-CZ cathode were measured using the two-electrode four-wire method. For comparison, the performance of $(\text{La}_{0.6}\text{Sr}_{0.4})_{0.95}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ with A-site deficiency, denoted as A0.95-LSCF, was also tested at the same condition. Humidified H_2 (100 ml min^{-1}) flow and O_2 (100 ml min^{-1}) flow were supplied as fuel and oxidant, respectively. The electrochemical impedance spectra were measured under open circuit condition using Solartron 1260 frequency response analyzer with Solartron 1287 electrochemical interface with amplitude of 10 mV. The frequency ranged from 10^6 Hz to 0.08 Hz .

The chemical compatibility and reaction among LSCF, CZ and YSZ oxides was characterized by powder X-ray diffraction technique (XRD). XRD patterns were collected in the 2θ range

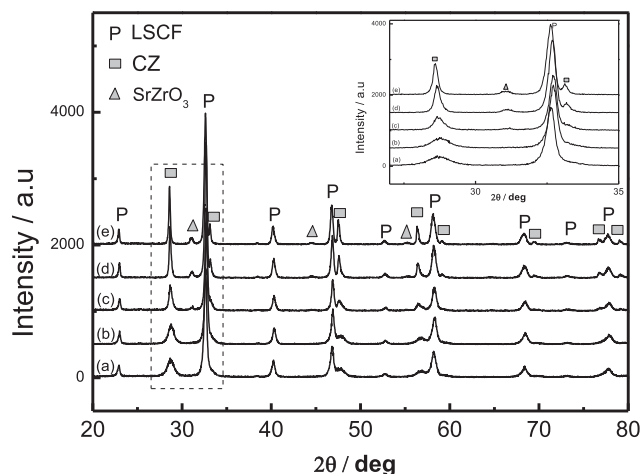


Fig. 1 – XRD patterns of LSCF-CZ mixtures with a weight ratio of 1:1 after sintering at (a) 800°C , (b) 850°C , (c) 900°C , (d) 950°C and (e) 1000°C for 2 h.

of 20 – 80° on a Rigaku D/Max-2500/PC X-ray diffractometer operated at 40 kV and 200 mA using $\text{Cu K}\alpha$ ($\lambda = 1.5406 \text{ \AA}$) radiation. The average crystallite size of the cubic CZ phase was calculated by the Scherrer equation, where the Scherrer constant (particle shape factor) was taken as 0.89. The microstructure of cathode was characterized by scanning electron microscopy (SEM), which was taken on JSM7800F microscope equipped with a field emission gun at 5 kV.

Results and discussion

XRD

XRD technique is used to investigate the chemical compatibility between LSCF oxide and CZ oxide. Fig. 1 shows the XRD patterns of LSCF-CZ samples sintered at 800 – 1000°C . For the

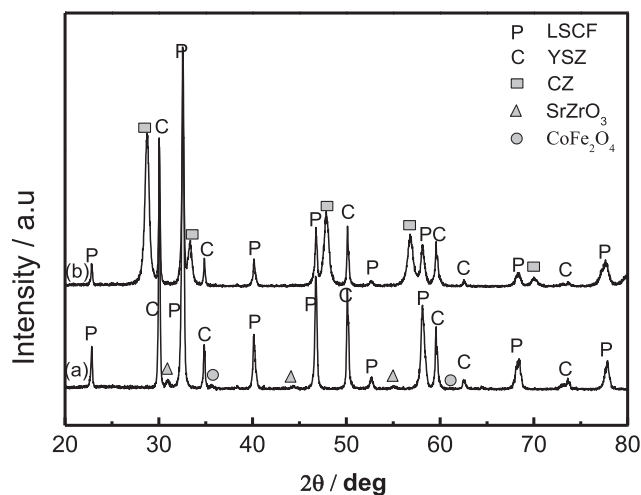


Fig. 2 – XRD patterns of LSCF-YSZ mixtures with a weight ratio of 4:1(a) and LSCF-YSZ-CZ mixtures with a weight ratio of 2:1:1 (b) after sintering at 850°C for 2 h.

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