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Porous $\text{YFe}_{0.5}\text{Co}_{0.5}\text{O}_3$ thin sheets as cathode for intermediate-temperature solid oxide fuel cells

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ABSTRACT

In this work, porous $\text{YFe}_{0.5}\text{Co}_{0.5}\text{O}_3$ (YFC) thin sheets were synthesized by citric acid method. The crystal structure, morphology, thermal expansion, electrical conductivity, and electrochemical properties of YFC were investigated to evaluate it as a possible cathode on $\text{BaZr}_{0.1}\text{Ce}_{0.7}\text{Y}_{0.2}\text{O}_3$ (BZCY) electrolyte for intermediate-temperature solid oxide fuel cells (IT-SOFCs). An orthorhombic perovskite structure was observed in YFC. The conductivity of YFC is 183 S cm^{-1} at 750°C in air. The coefficient of thermal expansion of composite cathode YFC-BZCY is closer to BZCY electrolyte than YFC. The composite cathode represents a relatively low polarization resistance (R_p) of $0.07 \Omega \text{ cm}^2$ at 750°C in air due to the porous thin sheet-like cathode. The oxygen reduction reaction process and the reaction activation energy of cathode were also analyzed. An anode-supported cell of NiO-BZCY|BZCY|YFC-BZCY is fabricated by a simple method of co-pressing. The power density of the cell is 303 mW cm^{-2} at 750°C as the thickness of electrolyte is $400 \mu\text{m}$. The results suggest that YFC is a promising cathode candidate for IT-SOFC.

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Introduction

Solid oxide fuel cells (SOFCs) are considered as energy conversion devices, which can directly convert chemical fuels into electricity with high energy efficiency and low environmental impact [1–4]. Generally, the operating temperature of the current SOFC technology is still high ($800\text{--}1000^\circ\text{C}$) to ensure the high efficient power supply [5–8]. However, such high operating temperatures require the use of expensive materials for fuel cell interconnectors and cause the

compatibility problems between electrode and electrolyte, long start-up time, and large energy input to heat the cell up to the desired temperature [9–13]. The high operating temperature limits the SOFC applications [14]. Therefore, there is a pressing need to develop new materials which are able to generate a reasonable power density at intermediate temperatures (IT, $600\text{--}800^\circ\text{C}$) [15–18].

The current SOFCs are commonly based on oxygen-ion-conducting electrolyte which requires a high activation temperature [19]. In attempt of lowering operating temperatures, the thermo-activation reactions of the oxygen-ion-

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conducting electrolyte in SOFCs are limited. So many studies are carried out to employ the proton conducting electrolytes with higher conductivity at IT as alternative electrolyte materials for SOFCs [20–22]. One of the key problems for this kind of electrolyte is the compromise between high conductivity and chemical stability. For example, doped BaCeO₃ with high ionic conductivity has insufficient chemical stability in an atmosphere of H₂O and CO₂, which is restricted to the applications for SOFC [23]. The new breakthrough of proton conducting electrolyte reported by Liu and co-workers [24–26] showed that Ba(Zr_{0.1}Ce_{0.7}Y_{0.2})O₃ (BZCY) presented high proton conductivity at temperatures below 550 °C. It is significant to develop novel cathode materials with BZCY electrolyte for IT-SOFCs.

Nowadays, the perovskite-type oxides (ABO₃) as cathodes have attracted great attention in the SOFCs application [27]. Among the numerous different perovskites, rare-earth cobalt containing perovskites (LnCoO₃; Ln = Y or lanthanide) have been studied as potential cathodes for SOFCs since they exhibit specific magnetic properties and electrochemical catalytic activity for oxygen reduction reactions (ORR) [28]. In addition, it is generally accepted that Co-based perovskite oxides are suitable cathodes at reduced temperatures [29]. The properties of perovskite such as electrical conductivity, chemical stability and thermal stability are closely related to the nature and oxidation states of the B-site cations, as the cobalt ions change the oxidation state (Co⁴⁺, Co³⁺, Co²⁺) and alter the spin state of the Co³⁺ ion [30–32]. Meanwhile, LnFeO₃ (Ln = Y, Sr, Ba or lanthanide) belonging to ABO₃ type perovskite reflects excellent properties of mixed ionic-electronic conductivity (MIEC) [33,34]. The transition metal Fe has variable oxidation and spin states, which potentially contribute high catalytic activity and strong resistance in cathode environment [35]. Yttrium orthoferrite (YFeO₃) [36], as one of iron-based perovskites, crystallizes in a distorted perovskite structure with an orthorhombic unit cell (Fig. 1(a)). The pure YFeO₃ in n-type conduction is not suitable for the cathode of SOFC, as the electrical conductivity is decreasing with the increasing oxygen partial pressure [37]. So the substitution of B-site cations by transition metal Co with variable valences can enhance the conductivity [33] and maintain the charged oxygen vacancies due to charge imbalance [38,39]. The spin states of Co ions could also be changed by a chemical substitution of Fe, and the existence of cobalt can bring the phase separation of perovskites through the formation of Co–O and Fe–O bonds [40,41].

Herein, the porous YFe_{0.5}Co_{0.5}O₃ (YFC) thin sheet was synthesized by citric acid combustion method as a potential cathode for SOFCs. The nano-structured cathodes like hierarchically nanoporous [42,43] or nanoporous rod-like structure [44] possess the high specific surface which is contributed to gas transport during ORR process and would benefit the adherence between cathode and electrolyte. The properties of porous YFC thin sheets are studied including the crystal structure, morphology, electrical conductivity and electrochemical properties, respectively. The ORR process of cathode is investigated to figure out the rate-limiting step of the reactions for the cell at different temperatures. The performances of the cell are also experimentally examined.

Experimental

Preparation of cathode powder and electrolyte substrate

YFe_{0.5}Co_{0.5}O₃ was synthesized by citric acid method. The stoichiometric ratios of nitrates were used from chemicals of Y(NO₃)₃·6H₂O (99.99%), Fe(NO₃)₃·9H₂O (98.5%), Co(NO₃)₂·6H₂O (99.0%). The starting step was prepared by dissolving the nitrate hydrate in small quantity of distilled water with high-speed stirring. Then the citric acid monohydrate (C₆H₈O₇·H₂O, 99.5%) was added into the solution. Continuous stirring and heating at 80 °C until the gel was obtained. Subsequently, the gel was dried at 110 °C to get a xerogel. Then, the xerogel was processed by the combustion reaction at 500 °C for 2 h (at a heating rate of 2 °C min⁻¹) and was sintered at 950 °C for 10 h (at a heating rate of 5 °C min⁻¹) in air to get the pure phase of YFC.

The electrolyte powder (BaZr_{0.1}Ce_{0.7}Y_{0.2}O₃, BZCY) was obtained by the solid state reaction method. The particular ratio of BaCO₃ (99.0%), ZrO₂ (99.0%), CeO₂ (99.0%), and Y₂O₃ (99.9%) were completely mixed and sintered at 1300 °C for 10 h to get the BZCY powder. The electrolyte pellet was fabricated by the method of dried pressing and sintered at 1500 °C for 10 h in air. The electrolyte disk was 13 mm in diameter and 1.2 mm in thickness.

Fabrication of cell

The ink is fabricated by cathode powders (YFC-BZCY with mass ratio of 7:3 and YFC). The grinded cathode powder was mixed with solvent mixture (terpilenol and turpentine, with volume ratio of 95:5) to get the slurry. Then it was painted on the centre of BZCY disk including both sides through screen-printing technique. Finally, Pt paste was used as current collector by sintering.

The anode-supported SOFC on BZCY was manufactured by the simple method of co-pressing. The anode powder is a mixture of NiO, BZCY and the pore former with mass ratio of 70:30:10. The method of co-pressing and sintering at 1400 °C for 10 h was adopted in order to fabricate the anode-supported BZCY composite dual-layer. Finally, the composite cathode (the mixture of YFC and BZCY at mass ratio of 7:3) was screen-printed on the other side of BZCY disk and sintered to obtain the anode-supported SOFC.

Characterization

The phase structure of YFC powder and chemical compatibility between YFC and BZCY were measured by X-ray diffraction (XRD) in the 2θ range of 10–70° operated at room temperature. The obtained XRD profile of samples was analyzed by the Rietveld method using Reflexing tools in the Material studio 5.5 [45]. Thermogravimetry analysis (TGA) of the synthesized YFC powder was measured from room temperature to 850 °C in air. The specific surface area of YFC powder was figured out by N₂ adsorption–desorption analyses at –196 °C by means of the Brunauer-Emmet-Teller (BET) method, then the pore size distributions of the sample were calculated by Barrett-Jioner-Halenda (BJH) method. In

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