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Technical Communication

Performance of double-perovskite $\text{YBa}_{0.5}\text{Sr}_{0.5}\text{Co}_2\text{O}_{5+\delta}$ as cathode material for intermediate-temperature solid oxide fuel cells

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ABSTRACT

Double-perovskite $\text{YBa}_{0.5}\text{Sr}_{0.5}\text{Co}_2\text{O}_{5+\delta}$ (YBSC) was investigated as potential cathode material for intermediate-temperature solid oxide fuel cells (IT-SOFCs). YBSC material exhibited a good chemical compatibility with the $\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.8}\text{Mg}_{0.115}\text{Co}_{0.085}\text{O}_{2.85}$ (LSGMC) electrolyte up to 950 °C for 2 h. The substitution of Sr for Ba significantly enhanced the electrical conductivity of the YBSC sample compared to undoped $\text{YBaCo}_2\text{O}_{5+\delta}$, and also slightly increased the thermal expansion coefficient. At 325 °C a semiconductor–metal transition was observed and the maximum electrical conductivity of YBSC was 668 S cm⁻¹. The maximum power densities of the electrolyte-supported single cell with YBSC cathode achieved 650 and 468 mW cm⁻² at 850 and 800 °C, respectively. Preliminary results suggested that YBSC could be considered as a candidate cathode material for application in IT-SOFCs. Copyright © 2011, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.

1. Introduction

In recent years, research on new cathode systems, the oxygen-deficient double-perovskites $\text{LnBaCo}_2\text{O}_{5+\delta}$ (Ln = La, Pr, Nd, Sm, Gd and Y), has received increasing attention for their potential applications in intermediate-temperature solid oxide fuel cells (IT-SOFCs) [1–8]. These double-perovskites possess an ordered structure which consists of alternating lanthanide and alkali–earth planes along the *c*-axis as described in the following stacking sequence $[\text{LnO}_8]\text{--}[\text{CoO}_2]\text{--}[\text{BaO}]\text{--}[\text{CoO}_2]$, whereas the oxygen vacancies are primarily located in the rare–earth planes $[\text{LnO}_8]$ [9–11]. Such structure reduces the oxygen bonding strength and provides disorder-

free channels for oxygen ion motion. This significantly improves the oxygen transport properties [9,10,12], which appears to be favorable for the reduction reaction of oxygen in SOFC cathode.

Similar to other cobalt-based perovskite cathodes, the double-perovskite $\text{LnBaCo}_2\text{O}_{5+\delta}$ cathodes also had high thermal expansion coefficients (TECs) due to the low-spin to high-spin transition of Co [6,13]. The thermal expansion mismatch between the cathode and electrolyte can cause the thermal stress in SOFCs. This resulted in the delamination of the cathode layer from the electrolyte or the cracking of the electrolyte and hence a poor long-term durability of the SOFCs [14]. Therefore, the TECs of the cathode and electrolyte should

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be as similar as possible to minimize thermal stress in SOFCs. The Ln = Y sample showed the lowest TEC in the LnBaCo₂O_{5+δ} systems but suffered from interfacial reaction with the La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{2.8} (LSGM) electrolyte after calcining at 1000 °C for 3 h, which limited its further use as a cathode in SOFCs [6]. Our latest research showed that, by using the cobalt-containing electrolyte material La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.115}Co_{0.085}O_{2.85} (LSGMC) instead of LSGM and lowering the calcining temperature of cathodes on electrolyte, the interfacial reaction between cathode and electrolyte can be avoided effectively [15].

Recently, Kim et al. reported the performance of LnBa_{0.5}Sr_{0.5}Co₂O_{5+δ} (Ln = Pr, Sm and Gd) as a possible cathode material for IT-SOFCs, and the SmBa_{0.5}Sr_{0.5}Co₂O_{5+δ} has showed an excellent electrochemical performance [16]. In addition, the substitution of Sr for Ba not only improved the chemical compatibility of the GdBa_{1-x}Sr_xCo₂O_{5+δ} cathodes with the above two electrolytes, but also enhanced the catalytic activity for oxygen reduction reaction of the GdBa_{1-x}Sr_xCo₂O_{5+δ} cathodes with a tetragonal structure [17]. Moreover, the subsequent studies also demonstrated that the double-perovskites LnBa_{0.5}Sr_{0.5}Co₂O_{5+δ} (Ln = Pr, Nd, Sm and Gd) could be potential cathode materials for applications in IT-SOFCs on the oxide ion-conducting and proton-conducting electrolytes [18–22]. More recently, Aurelio et al have reported that the substitution of Sr for Ba improved the stability of Y(Ba_{1-x}Sr_x)Co₂O_{5.5} materials [23]. With an aim to develop a novel cathode material, the double-perovskite YBa_{0.5}Sr_{0.5}Co₂O_{5+δ} was investigated as potential cathode material for IT-SOFCs. The phase composition, chemical compatibility, electrical conductivity, TEC and cell performance were presented.

2. Experimental

2.1. Sample preparation

The double-perovskite YBa_{0.5}Sr_{0.5}Co₂O_{5+δ} (YBSC) was synthesized by a solid-state reaction. Stoichiometric amounts of commercial powders Y₂O₃ (99.9%), BaCO₃ (99.5%), Co₃O₄ (99%) and SrCO₃ (99%) were mixed and ground in an agate mortar and pestle. The well-mixed powders were pressed into pellets and calcined at 1000 °C for 12 h in air. The calcined samples were reground, pressed into pellets (∅13 mm × 1 mm) and cylinders (∅6 mm × 5–6 mm) at 220 MPa for the electrical conductivity and thermal expansion measurements. They were finally sintered at 1100 °C for 20 h in air, and cooled to room temperature at a rate of 100 °C per hour. The LSGMC, Ce_{0.8}Sm_{0.2}O_{1.9} (SDC) and NiO powders were synthesized by the glycine-nitrate process [24,25]. The dense LSGMC electrolyte pellets were obtained by sintering at 1450 °C for 10 h. The composite anode was prepared by mixing NiO and SDC powers in a 65:35 weight ratio.

2.2. Characterization

The X-ray diffraction (XRD) pattern of the sample was collected in a diffractometer using Cu radiation (Rigaku-D-Max γA, λ = 0.15418 nm) at room temperature. An angle size of 0.02° and scanning range of 20–80° were applied. The

electrical conductivity of the YBSC sample was measured in air by the van der Pauw method between 25 and 850 °C in both heating and cooling cycles and their average values were determined. The data were collected after equilibration at each temperature for 20 min. The TEC of the sample was measured using a push-rod dilatometer (Netzsch DIL 402C) between 30 and 850 °C with a heating rate of 5 °C min⁻¹ in air.

The cell performance was tested using an electrochemical analyzer (CHI604C) in an electrolyte-supported cell configuration NiO-SDC/SDC/LSGMC/YBSC. An SDC interlayer was first screen-printed onto the LSGMC electrolyte pellets, and subsequently sintered at 1300 °C for 1 h in air. The NiO-SDC composite anode was screen-printed onto the interlayer, and sintered at 1250 °C for 4 h in air. Similarly, the YBSC cathode was screen-printed onto the opposite side of the LSGMC electrolyte pellets and sintered at 950 °C for 2 h in air.

3. Results and discussion

3.1. XRD analysis

Fig. 1(b) shows the XRD pattern of the sample YBSC sintered at 1100 °C for 20 h in air. For comparison, the XRD patterns of the YBaCo₂O_{5+δ} (JCPDS#47-0735) and LSGMC samples were also included. The YBSC composition was crystallized in a single phase with double-perovskite structure. This result is in a good agreement with the data of JCPDS#47-0735. There is no detection of any additional diffraction peaks attributable to impurities. The XRD peaks of the YBSC sample shift slightly to the high-angle direction compared to undoped YBaCo₂O_{5+δ}, suggesting that the lattice parameters decrease due to substitution of a smaller Sr²⁺ for Ba²⁺ (the ionic radius values are 0.144 nm for Sr²⁺ and 0.161 nm for Ba²⁺, respectively, in 12-coordination [26]). To investigate the interface reactivity between the cathode and electrolyte, the mixtures of the two powders in a 1:1 weight ratio were calcined at 950 °C for 2 h and the phase composition was analyzed by the XRD

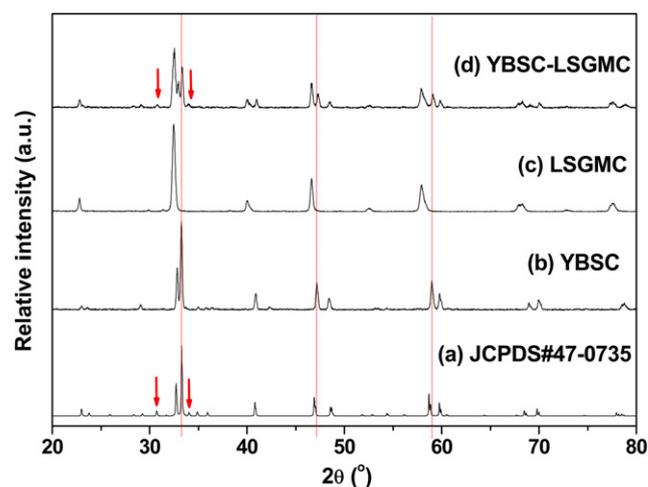


Fig. 1 – XRD patterns of the samples: (a) JCPDS#47-0735, (b) YBSC sintered at 1100 °C for 20 h in air, (c) LSGMC sintered at 1450 °C for 10 h in air, and (d) YBCF-LSGMC mixtures calcined at 950 °C for 2 h in air.

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