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Performance assessment of $Bi_{0.3}Sr_{0.7}Co_{0.3}Fe_{0.7}O_{3-\delta}$-LSCF composite as cathode for intermediate-temperature solid oxide fuel cells with $La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ electrolyte

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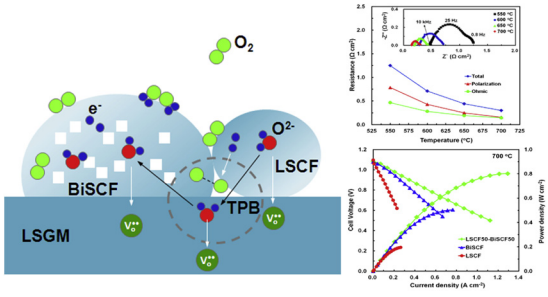
HIGHLIGHTS

- A cubic structure $Bi_{0.3}Sr_{0.7}Co_{0.3}Fe_{0.7}O_{3-\delta}$ is synthesized by solid-state reaction.
- BiSCF3737 is chemically compatible with available LSCF and LSGM electrolyte.
- A low electrode polarization resistance of 0.04 Ω cm^2 is achieved at 700 °C.
- The cell with LSCF50-BiSCF50 shows the highest and maximum power density.
- The LSCF-BiSCF composite are one of promising cathode candidates for IT-SOFCs.

GRAPHICAL ABSTRACT



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ABSTRACT

Perovskite-type $Bi_{0.3}Sr_{0.7}Co_{0.3}Fe_{0.7}O_{3-\delta}$ (BiSCF3737) oxide with perfectly cubic structure based on the P*m*-3*m* space group has been developed and investigated as cathode for intermediate-temperature solid oxide fuel cells (IT-SOFCs). BiSCF3737 is incorporated into $(La_{0.6}Sr_{0.4})_{0.9}Co_{0.2}Fe_{0.8}O_{3\pm\delta}$ (LSCF) to form a composite cathode called LSCF-BiSCF. X-ray diffraction (XRD) results demonstrate that BiSCF3737 has an extremely desirable chemical compatibility with LSCF as well as with $La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ (LSGM) electrolyte. The cells with LSGM electrolyte (0.5 mm thickness) and symmetrical electrodes are fabricated for electrocatalytic activity test. Compared with the pure constituent (LSCF or BiSCF3737), the composite with optimum composition, i.e., LSCF50-BiSCF50, exhibits better electrochemical activity for oxygen reduction. The LSGM electrolyte-supported (~300 μm thickness) cell with LSCF50-BiSCF50 composite cathode exhibits higher power densities of 0.617 and 0.802 W cm^{-2} at 650 and 700 °C, respectively, with humidified H_2 (~3% H_2O) as the fuel and ambient air as the oxidant. Over 78 h stability test at 600 °C indicates that a little performance decrease occurs but no interfacial damage happens, suggesting that LSCF50-BiSCF50 is a potential material for IT-SOFCs.

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1. Introduction

Solid oxide fuel cells (SOFCs) are one important solid-state electrochemical system which can directly convert chemical energy of fuel into electricity in an efficient and clean way, which usually operated at temperatures as high as 1000 °C [1–3]. Reducing the operating temperature down to intermediate range (500–700 °C) with sufficient power output and durability appears to be the challenge of intermediate temperature-SOFC (IT-SOFC). This would have several benefits including reduction of fabrication cost and improvement of the performance for large-scale application with longer operating life [4,5]. However, the electrochemical performance of cathode generally decreases with the decrease in the operating temperature, which directly affects the overall efficiency of cell [6,7]. Thus, the development of novel cathode materials with high electrocatalytic activity and good stability within the reduced temperatures becomes a critical issue for IT-SOFCs research.

Cathode materials normally require those oxides with sufficient mixed oxygen ionic and electronic conductions (MIEC) [8–11]. The most widely investigated MIEC oxides are perovskites and typically those ones based on lanthanum strontium manganite (LSM) [12–14]. However, at reduced temperatures, LSM has low electrocatalytic activity for oxygen reduction due to its negligible oxygen ionic conductivity and low oxygen surface exchange coefficient [15]. Different attempts have been made to improve its performance, including: (i) using composite cathode consisting of LSM and electrolyte material to increase the number of three phase boundary (TPB) and ionic conductivity (for instance, combining LSM with yttria-stabilized zirconia, LSM/YSZ) [13,16], however, cathode delamination and failure sometimes occur since the formation of LSM nanoparticles at the interface minimizes the direct contact of LSM and YSZ and increases the cathode polarization [17]. This suggests that microstructure plays an important role in the cathode polarization; this is particularly important when the composite cathode is used instead of the single composition one to improve the performance. Other attempts, (ii) replacing the B-site Mn with Fe and/or Co to increase the oxygen exchange rate (for instance, $La_{1-x}Sr_xFe_{1-y}Co_yO_{3-\delta}$, LSCF) [18,19], and (iii) replacing the A-site La with various elements to obtain highly active cathode based on manganite perovskites, which are usually more stable in SOFC operation conditions. Recently, Bi replacement of A-site element has been proposed as a novel SOFC cathode material [20–22]. Bi^{3+}, due to its different electronic configuration, specifically, the high polarizability owing to its $6s^2$ lone pair and lower basicity than the previously proposed Ba^{2+} ($Ba_{1-x}Sr_xFe_{1-y}Co_yO_{3-\delta}$, BSCF) [23–25] as well as La^{2+} based materials (LSCF), is expected to increase the structural stability and offer higher concentration and mobility of oxygen vacancies at the reduced temperatures. In addition, previous work has shown that bismuth oxides have positively catalytic effects on oxygen dissociation process [26,27], which is often the rate limiting step in oxygen reduction reaction at the SOFC cathode. However, to the best our knowledge, its electrochemical properties as SOFC cathode have not extensively evaluated.

Since discovered by Isihara et al. [28], Feng and Goodenough [29], and Huang and Petric [30], Sr and Mg doped $LaGaO_3$ (LSGM) oxides have received increasing attention and is currently considered as one of the most promising electrolyte materials for IT-SOFC. It has significantly high oxygen-ion conductivity and stable performance as compared with the conventional yttria-stabilized zirconia (YSZ) (600–800 °C) and negligible electronic conductivity over a wide range of oxygen partial pressures employed under SOFC operating conditions. However, one of the important issues concerning SOFC employing LSGM electrolyte is the choice of appropriate oxide electrocatalysts for the cathode so that it meets certain requirements, for instance, chemical compatibility with the electrolyte and no destructive phase formation within the operating temperature range. Previously, LSCF cathode has been reported and displayed high electrochemical performance as a good MIEC material at the intermediate temperatures [31]. However, it suffers from some thermochemical reactivity with the LSGM electrolyte [32]. This will directly affect the overall performance of the cell system. On the other hand, as mentioned above, LSGM has been considered as one of the most promising electrolyte materials for IT-SOFCs. Therefore, it is necessary to develop alternative cathode materials with good chemical stability and compatibility with LSGM electrolyte, high activity for oxygen reduction, and high conductivity for both oxide ions and electrons specifically at the reduced temperatures.

Meanwhile, it is widely accepted that the composite cathode always shows some improved electrochemical properties, since it can expand its active sites from TPB to the entire zone of the cathode [33,34]. Moreover, it is critical to develop new cathode material (including compositing with the available LSCF) with proper electrochemical performance and compatibility with the electrolyte. LSGM is usually adopted as an electrolyte due to its high oxide ion conductivity at the reduced temperatures. Therefore, the aim of the present study is to develop a novel bismuth-based cubic perovskite oxide, i.e., $Bi_{0.3}Sr_{0.7}Co_{0.3}Fe_{0.7}O_{3-\delta}$ (BiSCF3737), which is expected to have high activity on O_2 dissociation at the reduced temperatures, and analyse its suitability and compatibility with the available material of $(La_{0.6}Sr_{0.4})_{0.9}Co_{0.2}Fe_{0.8}O_{3\pm\delta}$ (LSCF) as composite cathodes. Subsequently, the composite cathodes $(LSCF)_{1-x}$-$(BiSCF)_x$ ($x = 0, 30, 40, 50, 60, 100$ wt.%) are prepared, and the effects of BiSCF3737 content on the performance of composite cathodes in both the symmetrical cells and fuel cells, are investigated under IT-SOFCs operating conditions.

2. Experimental

2.1. Materials synthesis

The original material of $Bi_{0.3}Sr_{0.7}Co_{0.3}Fe_{0.7}O_{3-\delta}$ (designated as BiSCF3737) was synthesized through solid state reaction from the high-purity starting materials of $Bi(NO_3)_3{\cdot}5H_2O$, $Sr(NO_3)_2$, $Co(NO_3)_2{\cdot}6H_2O$, $Fe(NO_3)_3{\cdot}9H_2O$, and $Ce(NO_3)_3{\cdot}6H_2O$ (99.9%, Wako, Japan). Stoichiometric amounts of these powders were mixed and ball milled at a rotation speed of 300 rpm for 6 h in ethanol media. The resulting precursor solutions were evaporated at 80 °C for 24 h and thoroughly ground in an agate mortar and subsequently calcined at different temperatures from 950 to 1050 °C for 8 h in air to obtain fine BiSCF3737 powders. Similarly, GDC ($Ce_{0.9}Gd_{0.1}O_{1.95}$) as a composite anode material was also prepared by the solid state reaction from the appropriate proportions of Ce and Gd nitrates (99.9%, Wako, Japan) mixed with ethanol by ball milling. Then the mixture was evaporated and calcined at 900 °C for 5 h in air to gain the GDC powder. The commercial NiO (Soekawa Chemicals, Japan), $La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ (LSGM, FCM, USA) and $(La_{0.6}Sr_{0.4})_{0.9}Co_{0.2}Fe_{0.8}O_{3\pm\delta}$ (LSCF, Ningbo SOFCMAN Energy Technology, China) powders were used as received for the preparations of anode, electrolyte and cathode, respectively. The mixture of BiSCF3737 and LSCF powders prepared at the optimum calcination condition was ground to form composites with 0, 30, 40, 50, 60, 100 wt.% BiSCF3737 (marked as LSCF, LSCF70-BiSCF30, LSCF60-BiSCF40, LSCF50-BiSCF50, LSCF40-BiSCF60, and BiSCF3737, respectively). To assess the phase reaction, the chemical compatibility of the BiSCF cathode and LSGM electrolyte was investigated with X-ray diffraction (XRD) analysis by sintering the mixed powders in a