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Electrochemical and chemical stability performance improvement of $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{0.91}\text{Al}_{0.09}\text{O}_{3-\delta}$ cathode for IT-SOFC through the introduction of a GDC interlayer

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ABSTRACT

In order to enhance the electrochemical performance of solid oxide fuel cells (SOFCs), a $\text{Gd}_{0.2}\text{Ce}_{0.8}\text{O}_{2-\delta}$ (GDC) interlayer was introduced to $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{0.91}\text{Al}_{0.09}\text{O}_{3-\delta}$ (BSFA) cathode. The chemical stability and microstructure of the samples were characterized by X-Ray diffraction (XRD) and scanning electron microscopy (SEM). The electrochemical performance was tested by impedance spectroscopy. The results show that BSFA cathode is chemically and mechanically stable with the existence of a GDC interlayer. The polarization resistance of BSFA cathode with a GDC interlayer was $0.3009 \Omega \text{ cm}^2$ and the maximum power density (P_{max}) of an anode supported single cell reached 1321 mW cm^{-2} at $750 \text{ }^\circ\text{C}$. Furthermore, a BSFA-GDC composite cathode was prepared and presented excellent performance with the P_{max} of 1957 mW cm^{-2} at $750 \text{ }^\circ\text{C}$, providing a feasible choice for IT-SOFC cathodes.

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Introduction

Fuel cell is an electrochemical device that converts chemical energy into electrical energy with high efficiency and low pollutant emissions [1]. Various types of fuel cells have been developed and solid oxide fuel cells (SOFCs) have been shown to be the most efficient fuel cells and expected to be early commercialization [2–9]. So far, many studies have been

focused on the development of new electrode and electrolyte materials [10–19]. A key challenge is to develop suitable cathode materials for catalyzing the oxygen reduction reaction below $800 \text{ }^\circ\text{C}$ (intermediate temperature, IT) [8,20]. Several requirements for the cathode for SOFC to meet include high electronic conductivity, chemical stability and thermal expansion match with electrolyte [21]. At lower operating temperatures, however, the cathodic polarization of some cathodes is substantial because of the poor ionic conductivity. Apart from

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that, other issues like thermal expansion coefficients (TEC) mismatch also limit the choice of cathode materials [22].

In the search for new SOFC cathode materials, several materials with different structure appear to have great potential. $\text{Sr}_2\text{Fe}_{1.5}\text{Mo}_{0.5}\text{O}_6$ (SFM) series, with the traditional perovskite structure, shows adequate electrical conductivity in both air and hydrogen environments, excellent redox stability, and promising performance as electrodes in symmetrical SOFCs [23]. $\text{La}_2\text{NiO}_{4+x}$ (LNO), with the K_2NiF_4 structure, presents high oxygen ionic mobility, strong electrocatalytic activity towards the oxygen reduction, and proper thermal expansion coefficients [24]. Although $\text{La}_2\text{NiO}_{4+x}$ and related compounds have most of the intrinsic properties needed for a good cathode, the performance reported to date falls below expectations [21]. $\text{GdBaCo}_2\text{O}_5$ (GBCO), with the ordered double perovskites structure, attracts attention for application as cathode materials by two features: high electronic conductivity above the metal-insulator transition temperature (around 350 °C) and excellent oxide ion conductivity [25,26]. Improvement for such materials should focus on chemical and long-term stability.

Among many cathode materials, Fe-based perovskite oxides are promising materials due to the less flexible redox behavior of iron [27]. Zhao et al. [28], Efimov et al. [29], and Qian et al. [30] reported that $\text{Ba}_x\text{Sr}_{1-x}\text{FeO}_{3-\delta}$ oxide substituted with lower valence cations Zn^{2+} , Cu^{2+} or Ni^{2+} for the B-site of the perovskite reveals a high oxygen permeation flux accompanied with exceedingly high phase stability. $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{0.8}\text{Zn}_{0.2}\text{O}_{3-\delta}$ and $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{0.8}\text{Cu}_{0.2}\text{O}_{3-\delta}$ were also employed as cathode materials for IT-SOFCs, yielding good electrochemical performances.

Furthermore, our previous work [31] for the first time has demonstrated that the novel perovskite oxides $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{1-x}\text{Al}_x\text{O}_{3-\delta}$ ($x = 0-0.2$) as possible cathode materials based on an Y_2O_3 -stabilized ZrO_2 (YSZ) electrolyte for IT-SOFCs. In particular, the $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{0.91}\text{Al}_{0.09}\text{O}_{3-\delta}$ (BSFA) cathode exhibited the optimal electrochemical performances among BSFAs with different Fe ratio. The maximum power density (P_{max}) of 512 mW cm^{-2} was achieved as low as 750 °C for an anode-supported single cell with BSFA cathode, which implies that BSFA should be a potential material as IT-SOFC cathode with high electrocatalytic activity for the oxygen reduction reaction (ORR).

Given the above considerations, we focus on the further examination of chemical compatibility, TEC matching between cathode and electrolyte material, along with the influence of electrode microstructure. This work presents the performance

optimization of BSFA cathode with a $\text{Gd}_{0.2}\text{Ce}_{0.8}\text{O}_{2-\delta}$ (GDC) interlayer and BSFA-GDC composite cathode, as well as giving a brief study of the function of the interlayer. The introduction of a GDC interlayer and preparation of BSFA-GDC composite cathode solved the problem of low chemical stability between BSFA and YSZ. As a result, the electrochemical performance of BSFA cathode was obviously improved and even further improved by BSFA-GDC composite cathode.

Experimental

BSFA oxide was synthesized by a sol–gel method combined with a zeotropic-distillation process. The gel prepared combined with appropriate n-butyl alcohol was thoroughly stirred. The compound was distilled and calcined at different temperatures (800 °C, 850 °C, 900 °C, 950 °C, or 1000 °C) for 10 h.

The chemical compatibility of BSFA and YSZ (8 mol% Y_2O_3 - ZrO_2 , Tosoh Company) was examined by co-firing of the BSFA and YSZ powders at 1100 °C for 3 h at a 1:1 weight ratio. In addition, the chemical compatibility of BSFA with $\text{Gd}_{0.2}\text{Ce}_{0.8}\text{O}_{2-\delta}$ (GDC, Ningbo Institute of Materials Technology & Engineering) was tested in the same way after co-firing at 1050 °C, 1100 °C, and 1150 °C for 3 h, respectively.

X-Ray diffraction (XRD) (X' Pert PRO MPD) (Cu K α radiation, operated at 40 kV, 40 mA) was used to confirm the crystal structure of BSFA powder and determine the presence of any impurity phases in BSFA-YSZ and BSFA-GDC composites. For TEC measurement, the as-synthesized BSFA oxide was first pressed into rectangular bars with nominal dimensions of 25 mm \times 5 mm \times 5 mm under 100 MPa pressure. Then these bars were sintered at 1150 °C for 6 h before the TEC test by a dilatometer (Netsch DIL 402C/4) over the 40 °C–1000 °C temperature range.

Three-electrode setup was used to measure the electrochemical performance of BSFA cathode. YSZ electrolyte pellets (15 mm diameter \times 0.8 mm thickness) were prepared and fired at 1500 °C for 6 h. The GDC slurry was screen-printed (5 μm) onto the YSZ electrolyte, and fired at 1400 °C for 6 h. BSFA powders were made into a slurry by being mixed with 1 wt% ethyl cellulose, 18 wt% starch and organic solvent (terpineol), and screen-printed onto the GDC film, and then sintered at 1050 °C, 1100 °C and 1150 °C for 3 h in air, respectively. The commercial Pt paste was painted as a reference electrode on the YSZ electrolyte surface about 1 mm away from the cathode, and painted as the counter electrode to the

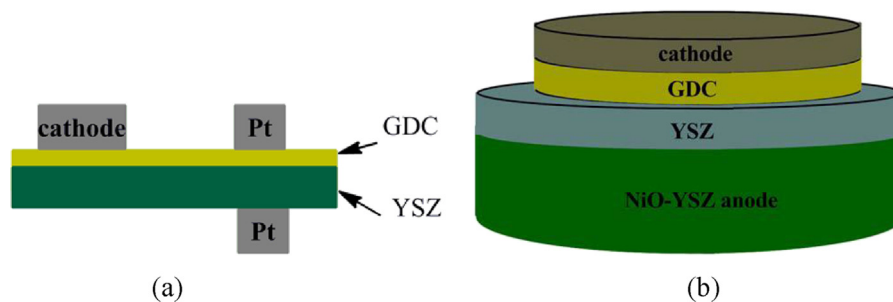


Fig. 1 – (a) Schematic diagram of three-electrode setup with GDC interlayer. (b) Schematic diagram of an anode-supported single cell with GDC interlayer.

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