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# Oxygen reduction mechanism at $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ cathode for solid oxide fuel cell

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ABSTRACT

Perovskite structure $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (BSCF) and $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ (LSGM) powders have been successfully synthesized by glycine–nitrate combustion process. A porous and crack-free BSCF cathode is obtained by spraying the slurry of BSCF powders and terpineol onto LSGM pellet. The oxygen reduction reaction mechanism has been investigated by AC impedance spectroscopy and cyclic voltammetry method. AC impedance spectroscopy analysis shows that there are two different processes in the cathode reaction which are related to oxygen dissociation/adsorption and bulk oxygen diffusion. And the molecular oxygen is involved in the rate-determining step. The polarization resistance decreases with an increase of temperature and the oxygen partial pressure. With an increase of the applied DC bias, the logarithm of the polarization resistance decreases linearly due to additional oxygen vacancies and the lowered chemical potential of oxygen at the BSCF/LSGM interface by the applied voltage. The exchange current density reaches to 182 mA $cm^{-2}$ at 700 °C, suggesting that the ORR kinetics at the BSCF/LSGM interface is high due to the excellent mixed ionic and electronic conductivity of BSCF.

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## 1. Introduction

Solid oxide fuel cell (SOFC) is considered to be a promising energy conversion device which can convert chemical energy of fuel gas directly into electrical energy with advantages of high electrical efficiency and low pollutant emissions. Apart from these general merits as a fuel cell, the most striking advantage of the SOFC lies in its fuel flexibility [1]. Not only hydrogen but also various kinds of hydrocarbon can be used as fuel [2]. The traditional SOFC operated at high temperature up to 1000 °C presents some problems related to the cost of the materials and fabrication. Accordingly, intermediate-to-low temperature solid oxide fuel cell, operating in the range of 500–800 °C, has attracted much attention in recent years [3]. While improvements in intermediate-to-low temperature SOFC have been achieved, their performance is still typically limited by cathode overpotential [4,5]. The reduction of operating temperature will lead to a significant decrease in the cathode activity [6]. As a result, the conventional SOFC cathode, strontium doped lanthanum manganite (LSM), is not suitable for intermediate-to-low temperature SOFC due to its low activity below 800 °C [7]. Therefore, developing new cathode materials with good performance at intermediate-to-low temperature range is the key to low down the operating temperature. Recently, Shao et al. [8] have reported $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (BSCF) as a novel cathode material for

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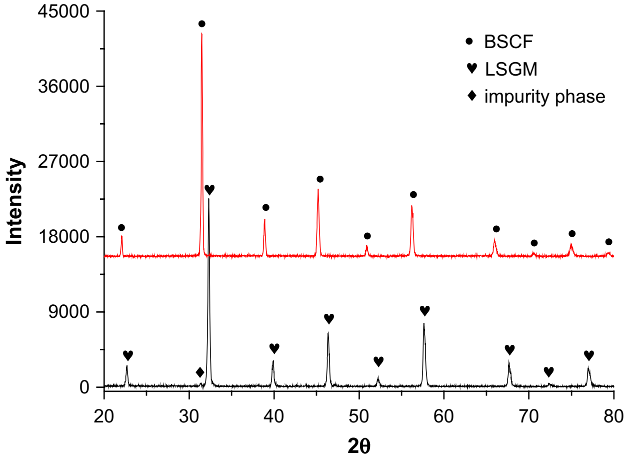

**Fig. 1 – The XRD patterns of BSCF and LSGM powders after calcination.**

the next generation of SOFC. It exhibits satisfactory electrochemical performances in both dual-chamber and single-chamber SOFC. To the best of our knowledge, the oxygen reduction reaction (ORR) processes and mechanisms at BSCF cathode are still unknown. It is well established that the performance of SOFC is limited by oxygen reduction reaction at the cathode. So the understanding of ORR processes and mechanisms is of great importance.

AC impedance spectroscopy technique has been extensively used to obtain information about oxygen reduction mechanisms. This technique has the advantage of making it possible to separate different rate processes in the studied frequency domain, so it is an efficient tool to determine the main processes involved in the reaction [9]. In the present paper, the mechanism of oxygen reduction reaction of BSCF cathode on $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ (LSGM) electrolyte will be investigated by AC impedance spectroscopy technique.

## 2. Experimental

### 2.1. Preparation

The $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ powders were synthesized by glycine–nitrate combustion process, using glycine as both fuel and complex. $Ba(NO_3)_2$, $Sr(NO_3)_2$, $Fe(NO_3)_3 \cdot 9H_2O$, $Co(NO_3)_2 \cdot 6H_2O$ and glycine ($H_2NCH_2COOH$), all of which were analytical reagent grade, were used as starting materials. These starting materials with the required molar ratios were mixed in deionized water to obtain transparent solutions. The solutions were heated to obtain viscous liquids on a hot plate at about 80 °C. Then the viscous liquids were transferred into a baker which was heated to 200–300 °C. The water was evaporated quickly and the solutions got swelled and ignited resulting in porous, foamy and fragile materials. The as-prepared materials were calcined at 850 °C for 2 h in air to obtain pure, well-crystallized powders. Powders of $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ were synthesized by glycine–nitrate combustion process from the following initial materials: $Ga_2O_3$ (99.999%), $La(NO_3)_3 \cdot 6H_2O$ (AR), $Mg(NO_3)_2$ (AR), $Sr(NO_3)_2$ (AR), and glycine ($H_2NCH_2COOH$, AR). $Ga_2O_3$ was first dissolved into strong $HNO_3$ and heated to 200–250 °C until the colorless $Ga(NO_3)_3$ aqueous solution was obtained. The next experimental processes were similar to that of synthesizing BSCF powders. The as-prepared powders were calcined at 1000 °C for 2 h in air to obtain pure, well-crystallized powders.

### 2.2. *Symmetrical fuel cell fabrication*

The calcined LSGM powders were sintered for electrolyte pellet at 1250 °C under a pressure of 40 MPa by Spark Plasma Sintering (SPS). Both sides of the pellet were roughed with 240# SiC grit paper and then cleaned by ultrasonic. The calcined BSCF powders were ball milled overnight to break up the agglomerated particles. 60% ball-milled BSCF powders were dispersed homogeneously into 40% terpineol by ultrasonic to obtain cathode slurry. Then, the slurry was sprayed onto both sides of the LSGM pellet. The symmetrical cell was first heated up to 500 °C to eliminate the organic binders, and then to 1000 °C for 2 h.

### 2.3. *Characterization*

X-ray diffraction analysis (XRD) was carried on to identify phase formation using Cu–Kα radiation at a scanning speed of 0.02° per step on a Rigaku Dmax-RB X-ray diffractometer. Morphology observation of the cathode after sintering was performed in a SUPRA-55 FESEM with an accelerating voltage of 10 keV. Two Pt plates were used as current collectors attached to two Pt wires for symmetrical cell experiments. AC

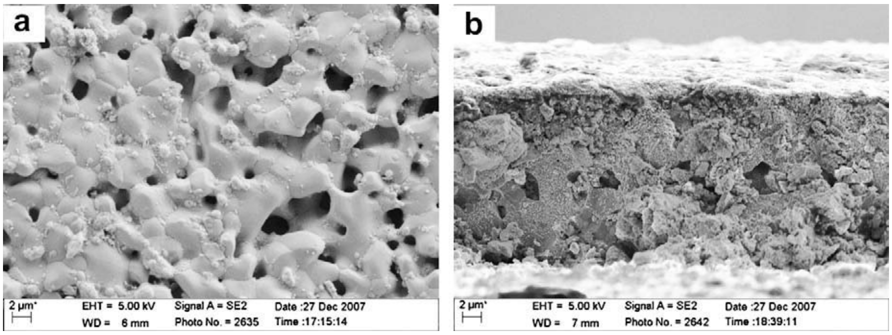

**Fig. 2 – (a) FESEM surface morphology of BSCF cathode on LSGM electrolyte, (b) FESEM image of cross-section at the porous BSCF/LSGM interface.**

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