



Nanostructuring of $\text{SmBa}_{0.5}\text{Sr}_{0.5}\text{Co}_2\text{O}_{5+\delta}$ cathodes for reduced-temperature solid oxide fuel cells

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HIGHLIGHTS

- $\text{SmBa}_{0.5}\text{Sr}_{0.5}\text{Co}_2\text{O}_{5+\delta}$ /LSGM cathode of nanostructure is fabricated by impregnation method.
- The lowest ARS values of $\text{SmBa}_{0.5}\text{Sr}_{0.5}\text{Co}_2\text{O}_{5+\delta}$ /LSGM cathode is $0.035 \Omega \text{ cm}^2$ at 550°C and $0.12 \Omega \text{ cm}^2$ at 500°C .
- Power densities of 1.5 W cm^{-2} at 600°C and 0.70 W cm^{-2} at 500°C were obtained.

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ABSTRACT

Here we report the fabrication of composite cathodes for reduced-temperature solid oxide fuel cells by impregnating aqueous solutions corresponding to $\text{SmBa}_{0.5}\text{Sr}_{0.5}\text{Co}_2\text{O}_5$ (SBSCO) into the porous $\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{3-\delta}$ (LSGM) backbones. Examination of X-Ray diffraction patterns indicates that phase-pure SBSCO layered perovskite oxides can be only achieved at calcination temperatures $\geq 900^\circ\text{C}$. Based upon impedance measurement of symmetric cells, the SBSCO–LSGM composites calcinated at 850°C show a trade-off between the SBSCO phase purity and catalyst size, and thereby exhibit minimal cathode polarization resistances with respect to the infiltrate calcination temperature, e.g., $0.035 \Omega \text{ cm}^2$ at 550°C and $0.12 \Omega \text{ cm}^2$ at 500°C at the loadings of 21 wt%. Analysis of impedance spectra under varied oxygen partial pressures suggests that oxygen reduction reactions on the nano-scale SBSCO–LSGM composite are largely dominated by ionization of adsorbed oxygen atoms on the SBSCO surfaces. Thin LSGM electrolyte fuel cells with impregnated Ni anodes and SBSCO cathodes show high power densities of 1.5 W cm^{-2} at 600°C and 0.70 W cm^{-2} at 500°C .

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1. Introduction

Fuel cells have been considered as one of the most important power generation technologies to efficiently convert fuels into electricity in an environmentally friendly manner. Among various types of fuel cells, solid oxide fuel cells (SOFCs) operate at elevated temperature, and exhibit some unique advantages such as high energy efficiencies, wide fuel flexibilities and rapid electrode kinetics without the use of noble metals as the catalysts [1–3]. Extensive efforts have been made to reduce the SOFC operating temperature from the conventional $700\text{--}900^\circ\text{C}$ down to $500\text{--}600^\circ\text{C}$, which allows broad choice of the electrode and interconnecting materials, reduces the system costs and increases the

performance durability [4,5]. Nevertheless, the SOFC power density would drop dramatically at low temperatures due to exponentially increased ohmic resistance from the electrolyte and interfacial polarizations from the electrodes. The large ohmic resistance can be decreased by reducing the electrolyte thickness and/or using alternative electrolyte materials such as doped ceria or doped lanthanum gallate that have much higher oxide-ion conductivities than the standard yttrium-stabilized zirconia (YSZ). Considering the dominant contribution of cathode to the overall interfacial polarizations, especially for the common anode-supported thin electrolyte SOFCs, development of novel cathode materials and microstructures with superior electrocatalytic activity and long-term stability are critically important to achieve high enough power density at reduced temperatures [6,7].

Perovskite oxides such as $\text{La}_{0.8}\text{Sr}_{0.2}\text{FeO}_{3-\delta}$, $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$ and $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_{3-\delta}$ exhibit both electronic and ionic conductivities, and have been extensively investigated as the cathode materials for reduced-temperature SOFCs due to extension of the electrochemically

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reactive region from the traditionally triple-phase boundaries to the entire surface [8,9]. Recently, layered perovskite oxides with the $\text{LnBaCo}_2\text{O}_5$ formula have gained great interest as novel cathode materials because of their high oxygen surface exchange coefficients (k) and self-diffusion coefficients (D), e.g., $D = 2.8 \times 10^{-10} \text{ cm}^2 \text{ s}^{-1}$ and $k = 7.5 \times 10^{-8} \text{ cm s}^{-1}$ for $\text{GdBaCo}_2\text{O}_{5+x}$ at 500°C [10,11], or $D = 3.6 \times 10^{-7} \text{ cm}^2 \text{ s}^{-1}$ and $k = 6.9 \times 10^{-5} \text{ cm s}^{-1}$ for $\text{PrBaCo}_2\text{O}_{5+x}$ at 500°C [12,13]. These values are substantially larger than what observed for most of common perovskite oxides, e.g., $D = 3.9 \times 10^{-7} \text{ cm}^2 \text{ s}^{-1}$ and $k = 1.5 \times 10^{-10} \text{ cm s}^{-1}$ for $\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$ at 500°C [14]. Tarancón explored the potential of $\text{GdBaCo}_2\text{O}_{5+x}$ as new cathode materials and reported a polarization resistance ($R_{p,c}$) value of $0.25 \Omega \text{ cm}^2$ on $\text{Gd}_{0.1}\text{Ce}_{0.9}\text{O}_2$ (GDC) electrolytes at 650°C [15]. GdBaCoFeO_{5+x} (GBCF) was also investigated as cathode for intermediate-temperature solid oxide fuel cells and showed interfacial polarization resistances of 0.42, 0.18 and $0.11 \Omega \text{ cm}^2$ at 550, 600 and 650°C , respectively [16]. Recently, Zhou et al. showed that $\text{SmBaCo}_2\text{O}_{5+x}$ (SBSCO) displayed good catalytic activity for oxygen reduction reactions [17], which can be further enhanced by partial substitution of Sr for Ba. In particular, $\text{SmBa}_{0.5}\text{Sr}_{0.5}\text{Co}_2\text{O}_5$ (SBSCO) exhibited interfacial polarization resistances of 1.01, 0.38, 0.16 and $0.06 \Omega \text{ cm}^2$ at 500, 550, 600 and 650°C [18]. Composites of SBSCO and GDC in a weight ratio of 1:1 showed even lower polarization resistances of $0.10 \Omega \text{ cm}^2$ at 600°C and $0.013 \Omega \text{ cm}^2$ at 700°C [19].

Reducing the feature size of cathode catalysts from the common micron scale down to the nano scale can accelerate oxygen reduction kinetics by providing enlarged number of active sites for surface oxygen exchange [20–22]. A cost-effective approach for obtaining nano-scale cathodes is to incorporate nanoparticles into the pre-formed porous backbones by impregnating appropriate solutions of soluble metal salts with the subsequent calcinations. Simple perovskite oxides such as doped LaCoO_3 , SmCoO_3 and LaFeO_3 have been extensively used as the infiltration catalysts and have shown excellent performance as the cathodes for reduced-temperature SOFCs [23–25]. For example, the $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$ (SSC) infiltrated GDC composites show polarization resistances as low as $0.1 \Omega \text{ cm}^2$ at 550°C [24]. Even better performances, e.g., $0.075 \Omega \text{ cm}^2$ at 550°C and $0.13 \Omega \text{ cm}^2$ at 500°C , were achieved by co-impregnating $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ (SDC) in addition to SSC into the porous LSGM backbones [26]. Note that the $\text{LnBaCo}_2\text{O}_5$ layered perovskite system have rarely been used as the infiltration catalysts. In this work, nano-scale catalysts of $\text{SmBa}_{0.5}\text{Sr}_{0.5}\text{Co}_2\text{O}_5$ (SBSCO) were coated onto the pore walls of LSGM backbone and were evaluated as potential cathodes for reduced temperature SOFCs operating over the regime of $450\text{--}600^\circ\text{C}$.

2. Experimental

Electrochemical properties of nano-scale SBSCO oxides as the active cathode catalysts were evaluated using symmetrical cathode fuel cells, which were based upon tri-layer structures of porous | dense | porous LSGM that were produced by laminating one dense LSGM tape and two porous LSGM tapes on both sides with subsequent co-firing at 1450°C . Fig. 1 showed a typical fabrication process of both symmetrical cathode fuel cells and function fuel cells which used in this paper. To avoid the damage of electrolyte when hot pressing asymmetric function fuel cells, the pressure was set to be 800PSI, much lower than 3000PSI when fabricating symmetrical cathode fuel cells. The porous layers were filled with 40 wt% starch as the fugitive materials, yielding porosities of $\approx 40\%$ as measured using the Archimedes method. The SBSCO catalysts were added into the porous LSGM backbones by impregnating an appropriate precursor solution followed by calcinations at temperatures of $800\text{--}900^\circ\text{C}$. The symmetrical cathode fuel cells were weighed before and after each impregnation–calcination cycle to estimate the loadings of impregnated SBSCO catalysts, which were expressed as the weight ratio of the impregnated oxides to the porous LSGM backbones plus impregnated oxides. A single impregnation/calcination cycle produced a SBSCO loading of 1.5–2 wt% in the porous LSGM backbone, so multiple impregnation/calcinations cycles were performed to increase the catalyst loadings. The precursor solutions were prepared as follows: stoichiometric amounts of the nitrate salts – $\text{Sm}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, $\text{Ba}(\text{NO}_3)_2$, $\text{Sr}(\text{NO}_3)_2$ and $\text{Co}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ —were dissolved in distilled water. Citric acid was then added to the solution with the molar ratio of the citric acid to the total metal ions at 1.5:1. Ammonia was added to the above solution to adjust the PH value at ≈ 4 in order to ensure full chelation of citric acid with metal ions. The precursor solution was subsequently heated on a hot plate until a molar concentration of 0.5 mol L^{-1} was obtained. Electrochemical properties of impregnated SBSCO–LSGM composites were also evaluated on functional fuel cells, where only one side of porous LSGM backbones was coated with SBSCO catalysts while the other was impregnated with nickel nitrate solution with subsequent calcinations at 700°C to form NiO. The typical NiO loadings were 21 wt%.

The phase compositions of the impregnated SBSCO–LSGM composites were examined using X-ray diffraction (XRD, Rigaku D/Max2500). XRD patterns were collected at room temperature with a step size of 0.02° in 2θ over the scanning range of $15\text{--}80^\circ$. The microstructure of the SBSCO–LSGM composites was examined using the field emission scanning electron microscope (FESEM–

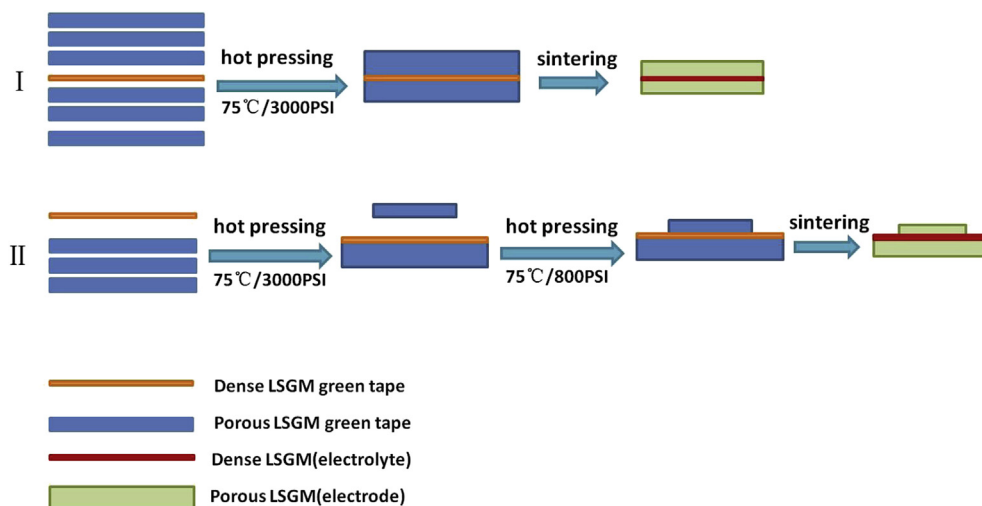


Fig. 1. A typical fabrication process of both (I) symmetrical cathode fuel cells and (II) function fuel cells which used in this paper.

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