



Short communication

Novel layered perovskite oxide PrBaCuCoO_{5+δ} as a potential cathode for intermediate-temperature solid oxide fuel cells

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ABSTRACT

A novel layered perovskite oxide PrBaCuCoO_{5+δ} (PBCCO) is employed as a potential cathode for intermediate-temperature solid oxide fuel cells (IT-SOFCs). Thermal expansion and electrochemical performance on samarium-doped ceria (SDC) electrolyte are evaluated. The thermal expansion coefficient (TEC) of PrBaCuCoO_{5+δ} (PBCCO) is close to that of SDC electrolyte and electrical conductivity of PrBaCuCoO_{5+δ} (PBCCO) reaches the general required value of cathode material. Symmetrical electrochemical cell with the configuration of PrBaCuCoO_{5+δ} (PBCCO)/SDC/PrBaCuCoO_{5+δ} (PBCCO) applied for the impedance studies, the area specific resistance of PrBaCuCoO_{5+δ} (PBCCO) cathode is as low as 0.047 Ω cm² at 700 °C. A maximum power density of 791 mW cm⁻² is obtained at 700 °C for the single cell consisting of PrBaCuCoO_{5+δ} (PBCCO)/SDC/NiO–SDC. Preliminary results indicate that PrBaCuCoO_{5+δ} (PBCCO) is especially promising as a cathode for IT-SOFCs.

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1. Introduction

As a promising energy conversion device, solid oxide fuel cells (SOFCs) have attracted widespread attention due to its high efficiency, fuel flexibility and environmental benefits, and now it is on the way to practical application and commercialization [1,2]. Nowadays, lowering the operation temperature of SOFCs from high temperature (~1000 °C) to intermediate-temperature (600–700 °C) can not only significantly improve materials' compatibility for the SOFCs system, but also reduce the capital costs of production and application [3]. However, a key obstacle to reduced-temperature operation of SOFCs is the poor activity of traditional cathodes such as La_{1-x}Sr_xMnO₃ (LSM) [4].

Accordingly, the development of alternative cathode with high electrochemical performance is indispensable to make intermediate-temperature SOFCs (IT-SOFCs) technology successful. Recently, Zhang et al. have reported that among the LnBaCo₂O_{5+δ} oxides examined, PrBaCo₂O_{5+δ} (PBCO) has the highest bulk diffusion coefficient and surface exchange coefficient [5], implying it may perform better than others as promising cathodes for IT-SOFCs.

However, to our knowledge, these cathodes, Co occupying the whole of B-sites, often suffer some problems like high thermal expansion coefficients (TECs) in practical long-term applications.

Therefore, in order to reduce disadvantages and improve the performance, much attention should be paid to the B-sites ions. In this work, the ceramic powder with a composition of PrBaCuCoO_{5+δ} (PBCCO) synthesized by auto ignition process was examined as a novel cathode for IT-SOFCs based on samarium-doped ceria (SDC) electrolyte.

2. Experimental

PrBaCuCoO_{5+δ} (PBCCO) powders were prepared by auto ignition process. Pr(NO₃)₃·6H₂O, Ba(NO₃)₂, Cu(NO₃)₂·6H₂O and Co(NO₃)₂·6H₂O were dissolved at the stoichiometric ratio and citric acid was then added, which was used as complexation agent. Molar ratio of citric acid/metal was set at 1.5. After that, this solution was stirring on a heating plate until it changed into black foam and finally ignited to flame, forming the primary powders. The as-synthesized powders were then calcined at 950 °C for 3 h to remove the carbon residue and obtain the desired PrBaCuCoO_{5+δ} (PBCCO) phase measured by X-ray diffraction (PW-1730).

Thermal expansion of the specimen was measured from 30 to 1000 °C using a dilatometer (SHIMADZU50) at a heating rate of 10 °C min⁻¹ in air and Electrical conductivity of PrBaCuCoO_{5+δ} (PBCCO) was studied using the standard DC four-probe technique on H.P. multimeter (Model 34401) from 400 to 800 °C in air.

Symmetrical electrochemical cells with the configuration of PrBaCuCoO_{5+δ} (PBCCO)/SDC (Sm_{0.2}Ce_{0.8}O_{1.9})/PrBaCuCoO_{5+δ} (PBCCO) and PrBaCo₂O_{5+δ} (PBCO)/SDC (Sm_{0.2}Ce_{0.8}O_{1.9})/PrBaCo₂O_{5+δ} (PBCO) applied for the impedance research

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were measured by electrochemical instrument (CHI604C 0.1 Hz–100 kHz) from 600 to 700 °C in air. Morphology and microstructure of symmetrical electrochemical cells after one thermal cycling were observed by scanning electron microscopy (SEM, FEI XL30).

The single cell was fabricated by a dry-pressing process utilizing SDC powders prepared by the glycine–nitrate method and calcined at 600 °C for 2 h [6]. To make a single cell, a mixed powders of NiO+SDC+starch (in a weight ratio of 65:35:20) were pressed at 200 MPa as a substrate (25 mm in diameter and 0.5 mm thick), SDC powders were then added, distributed uniformly, and co-pressed onto the substrate at 250 MPa. The bilayer was subsequently sintered at 1300 °C for 5 h, resulting in an SDC film of about 30 μm thick on the NiO–SDC substrate. Fine PrBaCuCoO_{5+δ} (PBCCO) powders were mixed thoroughly with a 10 wt% ethylcellulose–terpineol binder to prepare the cathode slurry, which was painted on the SDC electrolyte membrane to form a single cell. Afterwards, the cell was fired at 950 °C for 3 h in air. Electrochemical characterizations were performed from 600 to 700 °C under ambient pressure. The single cell performance was tested using an electrochemical instrument (CHI604C) with hydrogen as fuel and ambient air as oxidant.

3. Results and discussion

As shown in Fig. 1a, the prepared powder of PrBaCuCoO_{5+δ} (PBCCO) calcined at 950 °C for 3 h exhibits layered perovskite phase which is consistent with the data reported by Zhou et al. [7], indicating that the layered perovskite structure is well developed after calcination. Fig. 1 also presents the XRD spectra of electrolyte/anode bilayer sintered at 1250 °C for 5 h. It is obvious that there were only peaks corresponding to SDC in the electrolyte membrane (Fig. 1b) and to NiO and SDC in the anode substrate (Fig. 1c) with no peaks attributable to impurities detected.

As shown in Fig. 2, the thermal expansion coefficient of PrBaCuCoO_{5+δ} (PBCCO) and PrBaCo₂O_{5+δ} (PBCO) is $15.2 \times 10^{-6} \text{ K}^{-1}$ and $24.1 \times 10^{-6} \text{ K}^{-1}$, respectively. The mismatch of TEC between the electrolyte and cathode will result in delamination at the cathode/electrolyte interface, or cracking of the electrolyte because of the stress developed upon heating and cooling [8]. It is clear that the TEC of PrBaCuCoO_{5+δ} (PBCCO) is much closer to that of SDC ($\sim 12 \times 10^{-6} \text{ K}^{-1}$ [9]) than PrBaCo₂O_{5+δ} (PBCO) as expected. This implies that PrBaCuCoO_{5+δ} (PBCCO) might be more suitable as a cathode based on SDC electrolyte for IT-SOFCs.

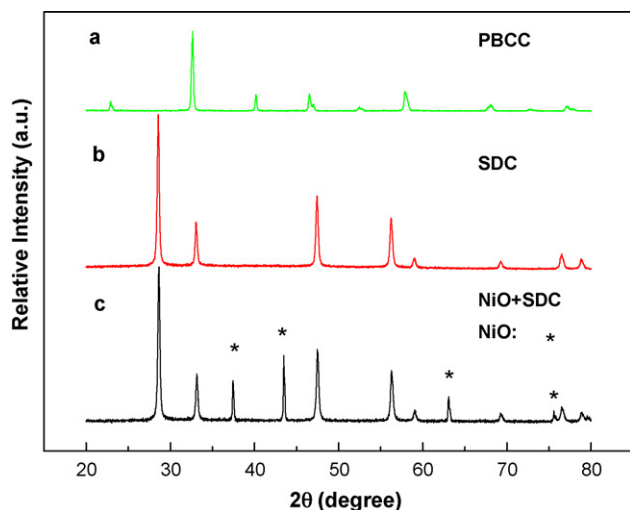


Fig. 1. XRD patterns of (a) PrBaCuCoO_{5+δ} (PBCCO), (b) Sm_{0.2}Ce_{0.8}O_{1.9} (SDC) membrane and (c) NiO–SDC anode substrate (*: NiO).

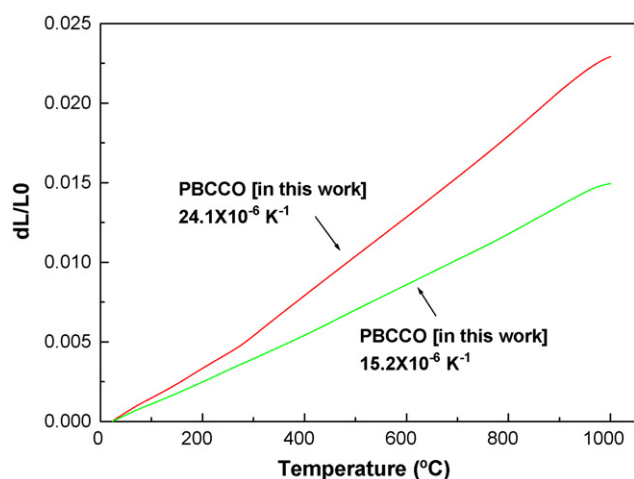


Fig. 2. Thermal expansion of different samples of PrBaCuCoO_{5+δ} (PBCCO) and PrBaCo₂O_{5+δ} (PBCO) in air.

Fig. 3 shows the electrical conductivity behaviors of the PrBaCuCoO_{5+δ} (PBCCO) sample versus the temperature when exposed to an air atmosphere. For cathodes of IT-SOFCs, the general required value for the electrical conductivity is about 100 S cm^{-1} at the operating temperature [10]. The electrical conductivity of PrBaCuCoO_{5+δ} (PBCCO) reaches the above standard absolutely within the 400–800 °C temperature range, though it is slightly lower than PrBaCo₂O_{5+δ} (PBCO) [5]. A similar behavior of LaBaCuCoO_{5+δ} has been reported by Zhou et al. [7]. Therefore, the electrical conductivity of PrBaCuCoO_{5+δ} (PBCCO) as a cathode is acceptable for application in IT-SOFCs.

Symmetric cells were prepared with SDC serving as the electrolyte. Fig. 4 shows the typical impedance spectra of the symmetrical cells from 600 to 700 °C in air. The high frequency intercept corresponds to overall electrolyte resistance of the symmetric cell including ionic resistance of the electrolyte, while the low frequency intercept corresponds to the total resistance of the symmetric cell. Therefore, the difference between the high frequency and low frequency intercepts with the real axis represents the area specific resistance (ASR) of the two interfaces [11]. It is clear that the ASR significantly reduces with the increasing temperature. It is worthy to note that the ASR of the PrBaCuCoO_{5+δ} (PBCCO) cathode is $0.207 \Omega \text{ cm}^2$ at 600 °C, $0.101 \Omega \text{ cm}^2$ at 650 °C and $0.047 \Omega \text{ cm}^2$ at 700 °C, respectively. Compared with the ASR of PrBaCo₂O_{5+δ}

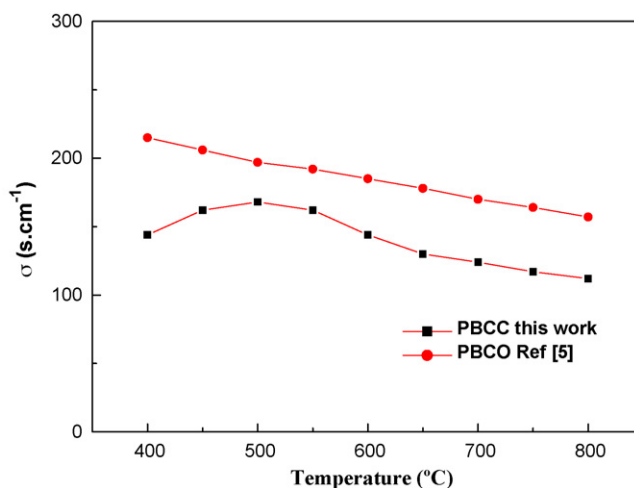


Fig. 3. Temperature dependence of the conductivity for PrBaCuCoO_{5+δ} (PBCCO) sample in air.

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