



Evaluation of performances of solid oxide fuel cells with symmetrical electrode material



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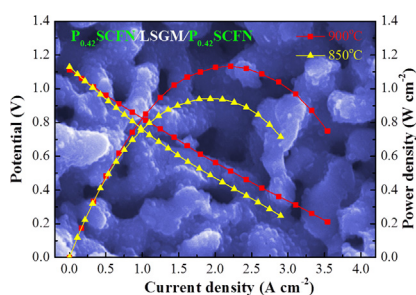
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HIGHLIGHTS

- P_xSCFN oxide is developed as symmetrical electrode material for SOFC.
- Pr element content in P_xSCFN has great effect on SOFC performance.
- Nano-sized Co-enriched particles are obviously formed in anode side.
- Pr³⁺/Pr⁴⁺, Co²⁺/Co³⁺ and Fe²⁺/Fe³⁺/Fe⁴⁺ coexist in P_xSCFN.

GRAPHICAL ABSTRACT



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ABSTRACT

Pr_xSr_{0.6}Co_{0.2}Fe_{0.7}Nb_{0.1}O_{3-σ} (P_xSCFN, $x = 0.38, 0.40, 0.42$ and 0.44) oxide has been developed as symmetrical electrode material for solid oxide fuel cell (SOFC) in our group. In this study, the effect of Pr element contents on the cell performance of the P_xSCFN symmetrical electrode SOFC is investigated in details. Electrochemical impedance spectra (EIS) technique, scanning electron microscopy (SEM), and energy dispersive X-ray spectroscopy (EDX) are used to characterize the electrodes after the performance test. It is found that the nano-sized Co-enriched particles are obviously formed in anode side, which could contribute to the high catalytic activity of the P_xSCFN symmetrical electrode materials. An optimum x value, i.e., 0.42, is found for the P_xSCFN symmetrical electrode materials. As-prepared single cell shows an average slow degradation rate of 0.5 mV h^{-1} during 100 h stability test, indicating that the symmetrical SOFC with PSCFN as electrode has the acceptable stabilization. X-ray photoelectron spectroscopy (XPS) analysis indicated that the content of Sr²⁺ species on the surface decreases to some extent in the case of A-site Pr element slight excess, which could result in the promoting of the catalytic properties of PSCFN material.

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1. Introduction

Solid oxide fuel cells (SOFCs) are electrochemical devices that have advantages of high efficiency, low pollution, fuel flexibility and long term stability and other features which make them attractive for energy conversion [1,2]. Hydrogen is considered the

best fuel for SOFC, but the issues related to its generation, storage and transportation still hinder its large-scale utilization in SOFC. Instead, hydrocarbon fuels become good alternatives for the commercialize utilization of SOFC [3]. To date, Ni-YSZ is a widely-used anode material in SOFC; however, hydrocarbon fuels are easy to be catalytically cracked on Ni, leading to carbon deposited on the surface of the anode and decrease the cell performance. On the other hand, sulfur, even at the magnitude of part per million (ppm) levels in the fuel, could be adsorbed on Ni sites and block its activity for the catalytic oxidation of fuel [4]. In order to solve these problems, Symmetrical electrode SOFC, in which the same material is used in both anode and cathode, was proposed [4–6]. In such symmetrical electrode SOFCs, the possible coke formation and/or sulfur poisoning on the surface of the anode can be potentially eliminated and thereby the anode is regenerated by operating the anode as the cathode in turn; where carbon or sulfur species absorbed on the electrode could be burnt out by oxidant. Simultaneously, the durability of cathode could also be improved in the low O₂ partial pressure at triple phase boundary region of cathode side [5]. In addition, the process for the preparation of the cell becomes more easily since the electrodes may be fired using the same thermal process. This would significantly save fabrication time and reduce the cost.

It is a key issue to develop a symmetrical electrode material with high catalytic activity for commercial utilization of such SOFCs, especially for those SOFCs work in hydrocarbon fuels containing H₂S. Bastidas et al. [4] considered La_{0.75}Sr_{0.25}Cr_{0.5}Mn_{0.5}O₃ (LSCM) as the symmetrical electrode material, and found that the prepared LSCM/YSZ/LSCM cell showed the maximum power density of 0.3 W cm⁻² at 900 °C. Ruiz-Morales et al. [5] optimized microstructure of LSCM and achieved a maximum power density of 0.5 W cm⁻² at 950 °C by using wet H₂ as fuel. Zhu et al. [6] added Sm-doped CeO₂ (SDC) or Ni into LSCM symmetrical electrode and found that addition of 6 wt.% Ni to the anode resulted in the highest power density value reached 0.559 W cm⁻² at 900 °C, which was better than those cells with the addition of SDC. Liu et al. [7] used Sr₂Fe_{1.5}Mo_{0.5}O_{6-σ} (SFM) as the symmetrical electrode material, and the obtained SFM/LSGM/SFM symmetrical cell reached the maximum power densities of 0.835 and 0.23 W cm⁻² at 900 °C in wet H₂ and CH₄, respectively. They investigated the redox stability upon a 5 redox cycle test at 800 °C, and found that there was no any adverse effect on the maximum power output after 5 redox cycles. However, the cell performance was lower than the conventional Ni-based SOFC. Martinez-Coronado et al. [8] used La_{0.5}Sr_{0.5}Co_{0.5}Ti_{0.5}O_{3-σ} (LSCT) as the symmetrical electrode material, and obtained a maximum power density of 0.11 W cm⁻². Chen et al. [9] used Sr-rich La_{0.3}Sr_{0.7}Fe_{1-x}Cr_xO_{3-σ} (LSFC_x, x = 0–0.3) as the symmetrical electrode material, and obtained the maximum power density of 0.3 W cm⁻² at 800 °C in wet H₂ for LSFC_{0.3} sample. All these reported symmetrical electrode material showed relatively low power density performance.

In our previous study [10], Pr_xSr_{0.6}Co_{0.2}Fe_{0.7}Nb_{0.1}O_{3-σ} (P_xSCFN, x = 0.38–0.44) was synthesized and used as the symmetrical electrode. It is found that the excess of A-site Pr element in P_xSCFN, i.e., in the cases of x = 0.42 and 0.44, resulted in a decrease in grain size and the creation of more active sites for oxygen reduction reaction. AC impedance measurements revealed that P_{0.42}SCFN had the best electrochemical catalytic performance in both O₂ and wet H₂ atmospheres. The maximum power densities of the prepared P_{0.42}SCFN/LSGM/P_{0.42}SCFN cell reached as high as 1.13 W cm⁻² in wet H₂ and 0.67 W cm⁻² in wet CH₄ at 900 °C. In this study, the effect of Pr element contents on the cell performances of the P_xSCFN (x = 0.38, 0.40, 0.42 and 0.44) symmetrical electrode SOFC were investigated in more details. Electrochemical impedance

spectra (EIS) technique, scanning electron microscopy (SEM), and energy dispersive X-ray spectroscopy (EDX) were used to characterize the electrodes after the performance test. Furthermore, in order to understand the promoting catalytic properties of P_xSCFN materials, X-ray photoelectron spectroscopy (XPS) analysis was used to investigate the existence states of Pr³⁺/Pr⁴⁺, Co²⁺/Co³⁺, Fe²⁺/Fe³⁺/Fe⁴⁺ and Sr²⁺ species in the PSCFN sample. The obtained results were analyzed and discussed in details.

2. Experimental

2.1. Sample preparation and cell fabrication

P_xSCFN (x = 0.38, 0.40, 0.42 and 0.44) perovskite oxides were prepared by solid-state reaction with the chemicals with high purity: Pr(NO₃)₃·6H₂O, SrCO₃, Co(NO₃)₂·6H₂O, Fe₂O₃ and Nb₂O₅ (99.9% Wako, Japan) [10]. Commercially available powder, La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{3-σ} (LSGM, 99.9% FCM, USA) was used for the preparation of electrolyte. Dense LSGM pellets with 20 mm in diameter were prepared by uniaxial mold pressing LSGM powder at 200 MPa and then sintering at 1450 °C for 10 h in air. After sintering, the electrolyte disk was approximately 20.0 mm in diameter and around 0.265 mm in thickness.

Symmetrical electrode cells of P_xSCFN/LSGM/P_xSCFN for the impedance study were prepared by slurry coating method. That was, P_xSCFN slurry was coated onto both sides of LSGM electrolyte pellet to form the anode and cathode and then, as-prepared cell was sintered in air at 1200 °C for 2 h. The effective area of porous electrode was about 0.38 cm² with a thickness of about 30 μm. For a single cell performance testing, the cell was sealed on an alumina tube using a Pyrex glass ring. Such measurements were carried out in a temperature range of 750–900 °C. For the single cell performance test, the anode was first annealed at 900 °C in pure H₂ atmosphere for 1 h, and then exposed to fuel at a flow rate of 100 cm³ min⁻¹. O₂ was used in the cathode side as the oxidant with a flow rate of 50 cm³ min⁻¹. Electronic contacts were realized by using a Pt mesh with Pt paste. It is reported that Pt has almost no influence on the electrode catalytic activity [11–14]. Electrochemical impedance spectra (EIS) of the single cell after the cell performance test were recorded at open circuit voltage (OCV) over the frequency range of 0.1 Hz–1 MHz with the AC signal amplitude of 10 mV using a frequency response analyzer and a potentiostat (Solartron 1255B and 1287, respectively). The impedance responses were analyzed using the equivalent RC circuit method.

2.2. Material characterization

Morphology and element composition were investigated by scanning electron microscopy (SEM, Hitachi, SU6600, Japan) with an EDX detector. Surface composition and valence of each element were identified for fresh powders by X-ray photoelectron spectroscopy (XPS, AXIS ULTRA DLD, Japan). A monochromatic Al Kα source (hν = 1486.6 eV) was used at a power of 150 W. Binding energy were calibrated with respect to setting the measured binding energy of C1s at 248.8 eV. Resolution of all spectra was performed by XPS Peak Program (XPSPEAK 41).

3. Results and discussion

3.1. Cell performance analysis

Current–potential (*I*–*V*) characteristics of LSGM electrolyte supported symmetrical electrode SOFCs with P_{0.38}SCFN, P_{0.40}SCFN, P_{0.42}SCFN and P_{0.44}SCFN as the electrode are shown in Fig. 1. The

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