



# Novel quasi-symmetric solid oxide fuel cells with enhanced electrochemical performance



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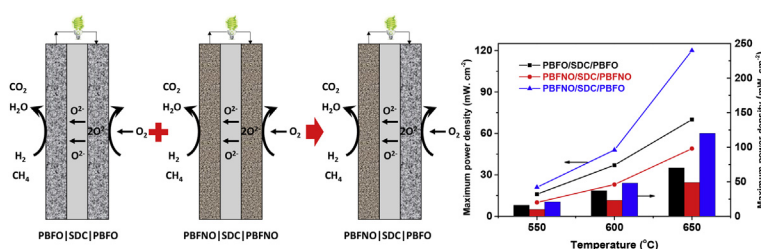
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## HIGHLIGHTS

- Novel quasi-symmetrical solid oxide fuel cells (Q-SSOFCs) are proposed.
- $\text{PrBaFe}_2\text{O}_{5+\delta}$  and  $\text{PrBaFe}_{1.6}\text{Ni}_{0.4}\text{O}_{5+\delta}$  are developed as new SSOFC electrodes.
- Q-SSOFC demonstrates superior performance by comparison with other SSOFC cells.
- Q-SSOFC offers more prospects at lower temperatures.
- Q-SSOFC opens doors for improvement of electrochemical performance in SSOFC.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Symmetrical solid oxide fuel cell (SSOFC) using same materials as both anode and cathode simultaneously has gained extensively attentions, which can simplify fabrication process, minimize inter-diffusion between components, enhance sulfur and coking tolerance by operating the anode as the cathode in turn. With keeping the SSOFC's advantages, a novel quasi-symmetrical solid oxide fuel cell (Q-SSOFC) is proposed to further improve the performance, which optimally combines two different SSOFC electrode materials as both anode and cathode simultaneously.  $\text{PrBaFe}_2\text{O}_{5+\delta}$  (PBFO) and  $\text{PrBaFe}_{1.6}\text{Ni}_{0.4}\text{O}_{5+\delta}$  (PBFNO, Fe is partially substituted by Ni) are prepared and applied as both cathode and anode for SSOFC, which exhibit desirable chemical and thermal compatibility with  $\text{Sm}_{0.8}\text{Ce}_{0.2}\text{O}_{1.9}$  (SDC) electrolyte. PBFO cathode exhibits higher oxygen reduction reaction (ORR) activity than PBFNO cathode in air, whereas PBFNO anode exhibits higher hydrogen oxidation reaction (HOR) activity than PBFO anode in  $\text{H}_2$ . The as-designed Q-SSOFC of PBFNO/SDC/PBFO exhibits higher electrochemical performance than the conventional SSOFCs of both PBFO/SDC/PBFO and PBFNO/SDC/PBFNO. The superior performance of Q-SSOFC is attributed to the lowest polarization resistance ( $R_p$ ). The newly developed Q-SSOFCs open doors for further improvement of electrochemical performance in SSOFC, which hold more promise for various potential applications.

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## 1. Introduction

Solid oxide fuel cell (SOFC) has been developed for over 150

years due to high energy efficiency, environmental friendly and fuel flexibility [1–3]. Traditional SOFC consists of a dense electrolyte asymmetrically sandwiched between porous cermet anode and porous oxide cathode, which are respectively subjected to different atmospheres [4–7]. With an eye to cost and long-term performance stability, symmetrical solid oxide fuel cell (SSOFC) using identical material as both anode and cathode has gained extensively attentions. By employing the same electrode composition, the fabrication procedure of cell can be controlled just in one step. Additionally, chemical and thermal compatibility with other components will also be enhanced, as only one type of interface exists. More importantly, the anode degradation towards sulfur poisoning and carbon deposition can be recovered by simply switching the anode to the cathode. Actually, the regeneration of anode mainly depends on periodic oxidation with air or steam by reversing the gas flows. In very different anode and cathode environments, the suitable SSOFC electrode materials should meet many rather restrictive requirements in both reducing and oxidizing environments, including sufficient conductivity and thermo-mechanical stability as well as reasonably dual electrocatalytic activity for both oxygen reduction reaction (ORR) and hydrogen oxidation reaction (HOR) [8].

Due to the rather restrictive requirements [9], only a limited number of redox-stable perovskite oxides can be effectively applied as both cathode and anode for SSOFC, such as  $\text{La}_{0.7}\text{Ca}_{0.3}\text{CrO}_{3-\delta}$  [10–12],  $\text{La}_{0.75}\text{Sr}_{0.25}\text{Cr}_{0.5}\text{Mn}_{0.5}\text{O}_{3-\delta}$  [13–16],  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Sc}_{0.2}\text{Mn}_{0.8}\text{O}_{3-\delta}$  [17], Fe-substituted (La, Sr) $\text{TiO}_3$  [18,19],  $\text{SrFe}_{0.75}\text{Mo}_{0.25}\text{O}_{3-\delta}$  [20–24],  $\text{La}_{0.6}\text{Ca}_{0.4}\text{Fe}_{0.8}\text{Ni}_{0.2}\text{O}_{3-\delta}$  [25–31],  $\text{Pr}_{0.4}\text{Sr}_{0.6}\text{Co}_{0.2}\text{Fe}_{0.7}\text{Nb}_{0.1}\text{O}_{3-\delta}$  [32–34], and  $\text{La}_2\text{NiO}_4$  [35]. On the other hand, majority of the reported SSOFCs using (La, Sr)(Ga, Mg) $\text{O}_3$  (LSGM) or yttria-stabilized zirconia (YSZ) electrolytes were tested under 800 °C (even higher operating temperature), which greatly restricts the selection of electrode materials and the utilization of cheaper metal interconnects. As a result, these problems restrict the performance of SSOFCs, which cannot meet the required level of commercialization.

With keeping the SSOFC's advantages, a novel quasi-symmetrical solid oxide fuel cell (Q-SSOFC) is proposed to further improve the performance, which optimally combines two different SSOFC electrode materials as both anode and cathode simultaneously. Cobalt-free  $\text{PrBaFe}_2\text{O}_{5+\delta}$  (PBFO) and  $\text{PrBaFe}_{1.6}\text{Ni}_{0.4}\text{O}_{5+\delta}$  (PBFNO, Fe is partially substituted by Ni.) were prepared by a modified Pechini method and applied as new electrode materials for SSOFC, which exhibited desirable chemical and thermal compatibility with  $\text{Sm}_{0.8}\text{Ce}_{0.2}\text{O}_{1.9}$  (SDC) electrolyte [36]. The phase behavior, thermal expansion behavior, electrical conductivity and electrochemical performance of PBFO and PBFNO in both air and  $\text{H}_2$  were studied, and the output performance of SSOFCs and Q-SSOFC were investigated.

## 2. Experimental

### 2.1. Synthesis

Single phase PBFO, PBFNO and SDC powders were all prepared by a modified Pechini method [37]. First, reagent grade  $\text{Pr}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ ,  $\text{Ba}(\text{NO}_3)_2$ ,  $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  as raw materials were mixed evenly with deionized water at stoichiometric ratio, then added citric acid as complexing agent into aqueous solution, a 2:1 mol ratio of citric acid to the metallic cations content was used. pH was adjusted to 7–8 by the addition of ammonia in the ventilation cabinet. Stirred solution was constantly heated on a hot plate until a black viscous gel appeared and auto-ignition occurred. The resulting powders were subsequently calcined at different temperatures for 3 h, to achieve pure phase PBFO powders. Similarly,

the PBFNO and SDC powders were synthesized by same procedure above, then calcined at different temperatures for 3 h and 700 °C for 3 h, respectively.

### 2.2. Sample preparation

The single phase PBFO and PBFNO powders were respectively pressed into bars under a uniaxial pressure of 200 MPa, followed by sintering at 1150 °C for 5 h in air for measurements of conductivity and thermal expansion behavior. The symmetrical half-cells PBFO/SDC/PBFO and PBFNO/SDC/PBFNO were prepared for the anodic and cathodic performance evaluations. The as-synthesized SDC electrolyte powders ball-milled for 24 h was initially pressed into pellets under a pressure of 250 MPa and pre-calcined at 1400 °C for 5 h in air to obtain sufficient strength. Next, PBFO and PBFNO powders were blended thoroughly with ethyl cellulose (binder, 6%) and terpineol (solvent, 36.37%), respectively. Two kinds of homogeneously electrode slurries were coated onto both surfaces of the SDC substrates by screen printing and then co-fired in air at 1000 °C for 3 h. A silver paste was painted on the PBFO and PBFNO layers at the both sides of the pellets to serve as current collectors.

The details of fabrication of the electrolyte-supported cells containing PBFO/SDC/PBFO, PBFNO/SDC/PBFNO and PBFNO/SDC/PBFNO were presented in the half-cells preparation except for the effective electrode surface area of 20%, then the cell was sealed on an alumina tube for performance testing.

### 2.3. Characterization

The phases identification of the PBFO and PBFNO powders calcined at different temperatures were carried out using X-ray diffractometer (XRD) with  $\text{Cu K}\alpha$  radiation source, the diffraction angle range ( $2\theta$ ) 20°–80°. The chemical compatibility between electrolyte and electrode materials calcined at 1000 °C for 5 h were also analysis by XRD. For the measurement of electrical conductivity, a DC four-probe method was used for sintered PBFO and PBFNO bars in different gas atmospheres at descending temperature ranging from 800 °C to 350 °C, with an interval of 50 °C. Thermal expansion data was collected using a NETZSCH DIL 402PC dilatometer in air with a heating rate of 5 °C  $\text{min}^{-1}$  between 100 and 1000 °C. To analyze the electrochemical impedance spectroscopy (EIS) of the symmetric cells, Zahner IM6 electrochemical workstation was applied for 500–650 °C polarization testing. The impedance spectra were recorded at open circuit voltages (OCVs) with a signal amplitude of 5 mV over frequency response range of 0.1 Hz– $10^5$  Hz for reducing atmosphere and 0.05 Hz to  $8 \times 10^6$  Hz for oxidizing atmosphere, respectively. A scanning electron microscope (SEM, Model KYKY EM-3200) was employed to detect the microstructure of the symmetrical cells. Single-cell performance tests were conducted under humidified hydrogen (~3%  $\text{H}_2\text{O}$ ) as the fuel and ambient air as an oxidant at 550–650 °C using an electronic load (IT8500), the impedance was also measured at OCVs using the same operation by adjusting frequency to 0.05– $10^6$  Hz.

## 3. Results and discussion

### 3.1. Phase analysis

Fig. 1(a), (b) shows the XRD patterns of the synthesized PBFO, PBFNO powders after calcination at different temperatures, respectively. It should be noted that the characteristic diffraction peaks of the splitting in double perovskite dot not appear while synthesized in air. This may be attributed to the presence of oxidation state of  $\text{Fe}^{3+}/\text{Fe}^{4+}$  generated by  $\text{Fe}^{2+} \rightarrow \text{Fe}^{3+}$  and  $\text{Fe}^{3+} \rightarrow \text{Fe}^{4+}$ , incorporating adequate oxygen under oxidizing

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