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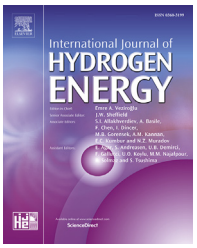

Preparation and properties of $PrBa_{0.5}Sr_{0.5}Co_{1.5}Fe_{0.5}O_{5+\delta}$ as novel oxygen electrode for reversible solid oxide electrochemical cell

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ABSTRACT

In this work, double perovskite-type oxide $PrBa_{0.5}Sr_{0.5}Co_{1.5}Fe_{0.5}O_{5+\delta}$ (PBSCF) is synthesized by the conventional wet chemical method and firstly characterized as the oxygen electrode for reversible solid oxide electrochemical cells (RSOCs). The microstructure and electrochemical performance of RSOCs based on this oxygen electrode are investigated. The maximum power density of the cell reaches 986 mW/cm^2 at 800 °C and the cell has good stability in short-term test in fuel cell (SOFC) mode. In electrolysis cell (SOEC) mode, it displays an electrolysis current density as high as 1.3 A/cm^2 when the temperature, absolute humidity (AH) and applied voltage are 800 °C, 50 vol % and 1.3 V, respectively. The cells also exhibit excellent durability of 120 h in SOEC mode and present good reversibility. The results suggest that the RSOCs based on this oxygen electrode has a very promising prospect.

Introduction

Energy crisis and global warming are the major challenges in modern society during the recent years [1,2]. Reversible solid oxide electrochemical cells (RSOCs), an advanced electrochemical device reversibly operating in SOFC mode and SOEC mode, has attracted more and more attention to alleviate these problems [3–5]. Many kinds of fuels, including H_2, CO and other hydrocarbons, are converted effectively to electricity and heat in SOFC mode [6–8]. And renewable energy can also be reconverted to chemical fuel by the electrolysis of various feedstocks, such as H_2O, CO_2 or CO_2+H_2O in SOEC mode [9–11]. Hence, RSOCs is a promising critical technology for efficient energy conversion and storage.

The traditional RSOCs includes dense yttria-stabilized zirconia (YSZ) electrolyte, nickel-YSZ composite (Ni-YSZ) hydrogen electrode and $La_{0.8}Sr_{0.2}MnO_{3-\delta}$ (LSM) oxygen electrode [12]. Recently, many researchers find out that the oxygen electrode is the key challenge to develop a high performance and durable RSOCs [13,14]. Under the operation condition of RSOCs, the materials of oxygen electrode must

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exhibit good stability as well as high catalytic activity in SOEC mode and SOFC mode. Developing new materials and modifying the oxygen electrode structures are effective ways to improve the electrochemical performance of RSOCs. Chrzan et al. prepared oxygen electrode by infiltrating YSZ backbone with $LaNi_{1-x}Co_xO_{3-\delta}$ catalyst and achieved superior performance [15]. Zhou et al. used $La_{0.5}Sr_{0.5}Fe_{0.8}Cu_{0.15}Nb_{0.05}O_{3-\delta}$ (LSFCN) as robust oxygen electrode and achieved relatively high electrochemical performance and good stability [3]. Ai et al. adopted $Er_{0.4}Bi_{1.6}O_3$ decorated $La_{0.76}Sr_{0.19}MnO_{3+\delta}$ as the oxygen electrode, which displayed extremely high performance and excellent durability [16].

Double perovskite-type $PrBa_{0.5}Sr_{0.5}Co_{1.5}Fe_{0.5}O_{5+\delta}$ (PBSCF) has excellent oxygen surface exchange coefficient, high oxygen ion conductivity and good structural stability, which has been extensively used as the cathode of SOFC [17–21]. PBSCF exhibits remarkably high power densities (2020 mW/cm^2 and 1310 mW/cm^2 at 600 °C and 550 °C, respectively) and low area specific resistance (ASR) (0.056 Ω·cm^2 at 600 °C) [22]. In our previous work, PBSCF has achieved good performance as the oxygen electrode of the SOEC [23]. In this paper, RSOCs based on this oxygen electrode is examined. The electrochemical performances including the power density and polarization resistance in SOFC mode, current density and hydrogen production rate in SOEC mode, the reversibility and stability of a single cell are examined at different operating temperatures and humidity.

Experiment

Materials synthesis

$PrBa_{0.5}Sr_{0.5}Co_{1.5}Fe_{0.5}O_{5+\delta}$ (PBSCF) powders were synthesized by the conventional sol-gel method [22]. Stoichiometric nitrates were dissolved in deionized water; citric acid (CA) and EDTA were then added in proportion ([Me]:[CA]:[EDTA] = 1:1.5:1). The above solutions were mixed together and kept at 80 °C with continuously stirred. The obtained gel was dried at 300 °C for 10 h followed by 5 h heat treatment at 1050 °C.

Single cell fabrication

The hydrogen electrode supported substrate, Ni-YSZ active layer and YSZ electrolyte were prepared by tape casting and screen-printing as described in our previous work [24,25]. The thickness of Ni-YSZ support is about 900 μm. The $Gd_{0.1}Ce_{0.9}O1_{.95}$ (GDC) barrier layer was utilized to prevent the chemical reaction between PBSCF and YSZ. GDC paste was prepared by mixing GDC powder with the binder (weight ratio of 60:40) and then manually ground. The GDC barrier layer was fabricated by coated GDC paste onto the surface of the YSZ electrolyte using screen-printing method and calcined at 1350 °C for 2 h. The pure PBSCF paste was prepared by the same method and coated on the surface of the GDC barrier layer by screen-printing method too, followed by fired in air for 2 h at 1050 °C. The effective area was 0.5 cm^2. Pt paste was coated onto the surface of the oxygen electrode as the current collector and fired at 850 °C for 2 h.

Characterization and measurement

The crystal structure of PBSCF powders and microstructure of the single cell were characterized by X-ray diffraction (XRD, Shimadzu XRD-7000S) and scanning electron microscope (SEM, Sirion 200), respectively. In SOFC mode, the hydrogen electrode and the oxygen electrode were fed the wet hydrogen (3 vol% AH) and ambient air, respectively. In SOEC mode, the hydrogen flowed through a thermostatic water tank in which the humidity was controlled by water temperature. In addition, the gas pipe should be heated to avoid the condensation of steam. The flow rates of hydrogen and air were controlled at 100 sccm. The current density–voltage–power density curve (I–V–P) and electrochemical impedance spectra (EIS) in SOFC mode and the current density–voltage (I–V) and EIS in SOEC mode were recorded by four-probe method with an electrochemical station (IM6, Zahner). The frequency range of the EIS test is 10^5 Hz–0.1 Hz with 10 mV amplitude.

Results and discussion

Crystal structure

The XRD pattern of the pristine PBSCF is presented in Fig. 1. The XRD peaks clearly indicate a single phase without detectible impurity after calcination in air at 1050 °C for 5 h. It possesses the double perovskite structure corresponding to the JCPDS card 53-0131 of $PrBaCo_2O_{5.68}$.

Microstructure

The microstructure of the Ni-YSZ/YSZ/GDC/PBSCF single cell is shown in Fig. 2(a). The thicknesses of YSZ electrolyte, GDC barrier layer and PBSCF oxygen electrode are 10 μm, 7 μm and 20 μm, respectively. It can be clearly found that the YSZ electrolyte is flawless and very dense, it also adheres fairly well to the GDC barrier layer and Ni-YSZ hydrogen electrode.

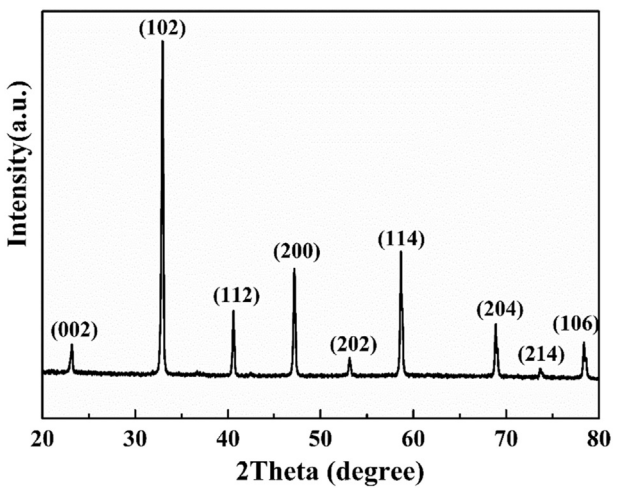

Fig. 1 – XRD patterns of PBSCF powders fired in air at 1050 °C.

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