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Short communication

A novel composite cathode for intermediate temperature solid oxide fuel cell



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HIGHLIGHTS

• $Ni_{0.79}Co_{0.2}Zn_{0.01}O$ (NCZ) – SDC composite was firstly evaluated as cathode for IT-SOFC.

• The output power has been further improved by partial substituting Li for Ni at Ni sites.

• At 600 °C, the cell Ni-SDC|SDC|SDC-Li_{0.2}Ni_{0.79}Co_{0.2}Zn_{0.01}O_2 showed the peak power density of 0.98 W cm⁻².

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ABSTRACT

Mixed transition-metal oxide Ni_{0.79}Co_{0.2}Zn_{0.01}O (NCZ) powders have been prepared by the modified Pechini method and studied as cathode for intermediate-temperature solid oxide fuel cells (IT-SOFCs). The Ni-Ce_{0.8}Sm_{0.2}O_{1.9}(SDC)|SDC|SDC-NCZ single cell achieves 326 mW cm⁻² at 600 °C. The output power has been further improved by partial substituting Li for Ni at Ni sites. The Ni-SDC|SDC|SDC-Li_{0.2}Ni_{0.79}Co_{0.2}Zn_{0.01}O₂ (L2NCZ) cell with the SDC film of 30 µm in thickness shows maximum power outputs of 1.32, 0.98 and 0.62 W cm⁻² at 650, 600 and 550 °C, respectively. The impedance measurements at open circuit conditions show that the overall electrode polarization resistances is 0.033, 0.112, 0.351 Ω cm² at 650, 600 and 550 °C, respectively. The primary results indicate a new family of potential cathode material compatible with the SDC electrolyte for IT-SOFCs.

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1. Introduction

Solid oxide fuel cells (SOFCs) are the most efficient devices yet invented for conversion of chemical fuels directly into electrical power [1,2]. Numerous potential opportunities exist for using SOFC system at various power levels, ranging from portable power sources and as auxiliary units in vehicles, to large-scale power stations [3,4]. Recently their potential application as electricity storage devices has also attracted attention [5–7]. The real-world efficiency of SOFC is largely determined by the activation of oxygen reduction reactions (ORR) on the cathodes. However, poor cathodic performance at intermediate temperature has become the main obstacle to the commercial application.

Mixed transition-metal oxides in $(A, B)O_x$ (A, B = Co, Ni, Fe, Zn, Mn, etc.) composition spaces are among the most active ORR catalysts [8,9]. These oxides have been widely applied in energy storage devices, such as lithium-ion batteries [10], electrochemical

transition-metal oxides as cathode materials for SOFCs have not attracted enough research interest. In this communication, as a first study, we investigated the performance of Ni_{0.79}Co_{0.2}Zn_{0.01}O (NCZ)-SDC as cathode for SOFCs. **2. Experimental**

capacitors [11], and metal- O_2 batteries [12], because the presence of mixed valences in these oxides is highly beneficial for ORR by

providing donor-acceptor chemisorption sites for the reversible

adsorption of oxygen [13,14]. Recently, electrodes based on ZnO/

NiO composition were proven to be extremely efficient for low-

temperature SOFCs [15]. In particular, ZnO/NiO has been used as

the cathode and anode material in a symmetrical configuration, and an excellent fuel cell performance, *e.g.* a maximum power

density of 1107 mW cm⁻², has been obtained at 500 °C. Lithiated

transition-metal oxide composite has also been extensively studied

by many research groups as cathode materials for SOFCs with

excellent fuel cell performances [16-19]. However, these mixed

Ni_{0.79}Co_{0.2}Zn_{0.01}O (NCZ) powders were synthesized by the modified Pechini method with citrate and ethylenediamine





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tetraacetic acid (EDTA) as parallel complexing agents. Proper amounts of chemical reagents of Ni (NO₃)₂·6H₂O, Co (NO₃)₂·6H₂O and Zn (NO₃)₂·6H₂O with the stoichiometric cations of Ni:Co:Zn 0.79:0.2:0.01 were dispersed into deionized water under stirring. EDTA and citric acid were then added to the solution at a mole ratio of total metal ions: EDTA: citric acid of 1:1:1.5. NH₄OH was applied to adjust the pH value of the solution to be around 6.0, and the solution became transparent immediately. Evaporation water on a hot plate, a gel was formed, and then combusted to obtain primary powder, which was subsequently, calcined at 900 °C about 2 h to obtain NCZ powders. To prepare Li_{0.2}Ni_{0.79}Co_{0.2}Zn_{0.01}O₂ (L2NCZ) powders, the aforementioned NCZ was reacted with lithium carbonate by solid-state reaction. A stoichiometric ratio of 0.21:1 = Li: NCZ was taken for the reactions. Both powders were mixed in absolute ethanol using mortar and pestle, and the mixture was annealed at 900 °C for 3 h under air. Ce_{0.8}Sm_{0.2}O_{1.9} (SDC) electrolyte material was prepared with commercially available chemicals of CeO₂ and Sm₂O₃. Powders of L2NCZ and SDC with a weight ration of 3:2 were well mixed, pressed into a pellet, and calcined at 1050 °C for 3 h. All the starting chemicals are analytical reagents from Sinopharm Chemical Reagent Co., Ltd.

The anode-supported SDC bi-layer was prepared by a drypressing method [20,21]. Commercial NiO (99.0%, Sinopharm Chemical Reagent Co., Ltd) and SDC (0.6: 0.4 by weight) powders were mixed and ground with alcohol for 2 h. After the alcohol was evaporated; the dried powder was pre-pressed into discs in a stainless-steel die at 200 MPa as the substrate about 1 mm. Then loose SDC powder, calcined at 700 °C for 3 h, was uniformly distributed on to the anode substrate. co-pressed at 300 MPa and sintered subsequently at 1400 °C for 3 h in air to densify the SDC membrane. Fine NCZ or L2NCZ powder, calcined at 900 °C, was mixed thoroughly with a 40 wt.% SDC and an appropriate amount of terpineol binder to prepare the cathode slurry, which was painted on the SDC electrolyte membrane, and fired at 1050 °C for 2 h in air to form a single cell. Surface of the cathode (L2NCZ-SDC) was covered with a layer of L2NCZ powders in order to prevent the evaporation of Li₂O at the high temperatures. The electrode active area was 0.33 cm². Ag paste was applied as a current collector for both the anode and cathode. Electrochemical measurements of the fuel cell were performed in a ceramic tube with an inner diameter of 10 mm placed inside a furnace. Humidified hydrogen (3% H₂O) was fed to the anode chamber at a flow rate of 100 ml min⁻¹, and air flowed over the cathode surface at a rate of 200 ml min⁻¹. Open circuit voltage and *I*–V curves were collected using digital multimeters (XIELI, DPM) based on a two-chamber configuration.

The phase structure of the composite oxides was determined by room-temperature X-ray powder diffraction (XRD) using an X-ray diffractometer (Rigaku Ultima IV) with filtered Cu K α radiation ($\lambda = 1.5418$ Å). Scanning electron microscopy (SEM, JEOL JSM-7500F) was used to characterize the microstructure of the cell after performance test. Electrochemical impedance spectra (EIS) of the fuel cell over the temperature range of 550–700 °C was performed on a PARSTAT 2273 with frequency range of 0.1 Hz–100 kHz, and the AC amplitude was 10 mV.

3. Results and discussion

Shown in Fig. 1 are the XRD patterns of NCZ and a L2NCZ-SDC mixture after calcination at 1050 °C and SDC are also shown for comparison. As can be seen in Fig. 1, a single phase of NCZ is obtained. Lattice constant of NCZ ($Fm\overline{3}m$) is determined to be $a = 4.177 \pm 0.006$ Å in cubic setting by a least square method. The XRD pattern of L2NCZ calcined at 900 °C for 3 h in air shows the formation of a single phase Li_{0.4}Ni_{1.6}O₂ (JCPDS card No. 81-0095). The XRD pattern of L2NCZ can be clearly indexed as the NiO phase



Fig. 1. XRD patterns of NCZ, SDC, and L2NCZ-SDC mixtures after calcination at 1050 $^\circ\text{C}$ for 3 h.

structure; moreover, it is evident that the peaks corresponding to L2NCZ were shifted to right comparing with NCZ, suggesting that Li have doped into NCZ successfully. The replacement of one Ni²⁺ ion by one Li⁺ ion is accompanied by a simultaneous change of valency from Ni²⁺ to Ni³⁺. The ionic radii of octahedral coordinated Ni²⁺, Ni³⁺, and Li⁺ is 0.069, 0.056, and 0.076 nm, respectively [22]. The sum of Li⁺ and Ni³⁺ radii is 0.132 nm, smaller than the sum of two Ni²⁺ (0.138 nm), which leads to the volume of the unit cell in L2NCZ (*Fm*3*m*, *a* = 4.143 ± 0.002 Å) reduced. To investigate the compatibility between L2NCZ and SDC, a mixture of L2NCZ and SDC powders (weight ratio of 3:2) was fired at 1050 °C for 3 h. Fig. 1 also has shown the XRD patterns of a L2NCZ-SDC mixture after calcination at 1050 °C. No additional diffraction peaks are observed, indicating that the L2NCZ is chemically compatible with the SDC electrolyte at 1050 °C for 3 h.

Fig. 2 shows a SEM micrograph of the cross section of the typical cell after the electrochemical test under H_2 . The composite cathode demonstrates good attachment to the electrolyte without any sign of cracking and delamination, indicating good thermal compatibility between the materials. In this case, the thickness of the cathode layer is about 25 μ m whereas the SDC electrolyte film is



Fig. 2. A typical microstructure of a three-layer anode-supported SDC thin membrane fuel cell.

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