



Improved performance and stability of Ag-infiltrated nanocomposite $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}-(\text{Y}_2\text{O}_3)_{0.08}(\text{ZrO}_2)_{0.92}$ oxygen electrode for $\text{H}_2\text{O}/\text{CO}_2$ co-electrolysis



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HIGHLIGHTS

- An Ag-loaded nanocomposite electrode was prepared for $\text{H}_2\text{O}/\text{CO}_2$ co-electrolysis.
- Electrochemical performance and lifetime of the Ag-loaded cell were improved.
- Both ohmic and polarization resistances were decreased with Ag load.
- Effect of varied $\text{H}_2\text{O}/\text{CO}_2$ ratio on cell performance was investigated.

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ABSTRACT

An Ag-infiltrated nanocomposite LSCF ($\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$)-YSZ (yttria stabilized zirconia) oxygen electrode is prepared for co-electrolysis of steam and CO_2 . Scanning electron microscopy (SEM) associated with energy dispersive X-ray spectroscopy (EDS) is employed to verify that nano-Ag particles are formed into the porous LSCF-YSZ electrode. Polarization curves and electrochemical impedance spectra (EIS) of the cell as well as long-term durability are investigated. In comparison with the Ag-free cell, the Ag-loaded cell exhibited improved performance and long-term stability when 45% H_2O , 45% CO_2 , and 10% H_2 is introduced as inlet gas. With infiltration of metal Ag, the ohmic resistance of the cell decreases from $0.14 \Omega \text{ cm}^2$ to $0.11 \Omega \text{ cm}^2$, and the polarization resistance from $0.30 \Omega \text{ cm}^2$ to $0.17 \Omega \text{ cm}^2$ at 800°C . No significant deterioration of the Ag-infiltrated cell is observed when operating for 200 h at 1.3 V and 750°C . With respect to varied $\text{H}_2\text{O}/\text{CO}_2$ ratio (1:2, 1:1, and 2:1) in feed gas, higher H_2O percent content is resulted into higher cell performance, despite the fact that varied inlet gas composition did not dramatically influence the cell performance.

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1. Introduction

Solid oxide electrolysis cells (SOECs) which are the inverse process of solid oxide fuel cells (SOFCs) have attracted considerable interest in the achievement of zero-emission and high-purity hydrogen production [1,2]. At high operation temperature ($\sim 800^\circ\text{C}$), H_2O can be split into H_2 with low electrical energy requirement. High temperature is beneficial to low cell internal resistance and fast electrode kinetic, leading to high electrolysis efficiency. Except for steam electrolysis, SOECs can be also used in $\text{H}_2\text{O}/\text{CO}_2$ co-electrolysis to yield syngas (H_2 and CO), resulted into

synthetic liquid fuels production via Fischer-Tropsch processes [3]. High temperature electrolysis allowing the use of waste heat from power stations or renewable energy (wind energy, solar energy, etc.) provides a possible means of recycling CO_2 into a useful liquid fuel, thereby reducing greenhouse gas emission and increasing energy independence [4].

SOEC technology has been developed based on SOFC research and materials [5]. In SOFCs, yttria stabilized zirconia (YSZ) and nickel-YSZ (Ni-YSZ) are the most commonly used electrolyte and anode materials, respectively. As well as for SOFCs, the YSZ and Ni-YSZ materials can also be employed in SOECs as electrolyte and hydrogen electrode, respectively, exhibiting good electrochemical performance and stability [6,7]. In terms of oxygen electrode materials, $\text{Ba}_{0.9}\text{Co}_{0.7}\text{Fe}_{0.2}\text{Nb}_{0.1}\text{O}_{3-\delta}$ [8], LSM (lanthanum strontium manganite) [9,10], and NiCoO_4 spinel [11] have been reported for

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oxygen reduction and evolution reaction. Nevertheless, electrochemical stability of these materials for SOEC operation was still required to be further demonstrated. Although perovskite LSM has been widely used in SOECs by traditional screen printing method, significant degradation behavior attributed to delamination of the oxygen electrode from the electrolyte has been observed under the electrolysis operation mode [12,13]. An infiltration technology has been developed on fabrication of nano-structured electrode, with reduced polarization resistance and improved cell performance [14]. Mixed ionic electronic conduction (MIEC) oxide LSCF ($\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$) has been studied to simultaneously provide high electronic and ionic conductivity and high oxygen electrochemical reduction/oxidation activity [15,16]. Moreover, it has been reported that Ag nanoparticles were introduced into the oxygen electrodes to improve the electrocatalytic activity of the electrode [17]. Low concentration of Ag nanoparticles attached LSCF electrode was prepared by plasma deposition method to show higher power density than that without Ag introduction [18]. Sun et al. [19] has investigated composite Ag-Sm_{1.8}Ce_{0.2}CuO₄ (SCC) composite electrode prepared by the infiltration method. Compared with the Ag-free SCC electrode, the composite electrode showed enhanced catalytic activity for the oxygen reduction reaction and chemical stability. However, long-term stability investigation will further be required to verify the effect of Ag addition. A study by Sakito et al. [17] has demonstrated that AgNO₃ solution infiltrated into porous LSCF electrode exhibited the enhancement of the cell power density and long-term stability as well for SOFC application. In Ref. [20], it has been proposed that the particle size of the distributed Ag into the scaffold was a key to the catalytic activity of the electrode, influencing the cell performance output. Reducing the Ag particle size and increasing the Ag dispersion improved the cell performance. In addition, a symmetric SOFC was prepared from an SDC (samaria-doped ceria) framework with Ag infiltration [21]. The fabricated Ag-SDC/SDC/Ag-SDC cell exhibited low polarization resistance not only in H₂ atmosphere but also in carbon-containing fuel (CO). Accordingly, the composite Ag-SDC electrode showed both high electrical conductivity and oxygen reduction activity.

Current research papers [22,23] have shown that the Ag-infiltrated composite electrode was efficient in SOFC operation. Nevertheless, few works involving electrochemical performance and long-term durability of the Ag composite electrode were presented in SOEC technology for steam/CO₂ co-electrolysis. In this study, an Ag-LSCF composite electrode was prepared by the infiltration technology for SOFC/SOEC. The electrochemical performance of the Ag-infiltrated LSCF-YSZ oxygen electrode was investigated for steam/CO₂ co-electrolysis, during which an Ag-free electrode was measured for comparison. Furthermore, the long-term stability of the composite Ag-LSCF electrode was evaluated for the co-electrolysis of steam/CO₂, compared with the Ag-free LSCF electrode.

2. Experimental

2.1. Fabrication of nano-structured NiO-YSZ/YSZ/LSCF-YSZ cells

The nano-structured NiO-YSZ/YSZ/LSCF-YSZ cell is composed of a NiO-YSZ hydrogen electrode support, a YSZ electrolyte and an LSCF-YSZ oxygen electrode. The single cell was fabricated by tape casting and infiltration technologies. As mentioned in Ref. [24], NiO-YSZ/YSZ/porous YSZ substrate was firstly prepared by the tape casting process and hot isostatic pressing technique. For fabricating the NiO-YSZ hydrogen electrode, nickel oxide (NiO) powder, YSZ (8mol% yttria stabilized zirconia, Tosoh Company, Japan), and graphite (Furunda Zirconium Material Co. Ltd., China)

porous former were thoroughly mixed at a weight ratio of 50:50:10. Ethanol-butanone solvent, castor oil dispersant, dibutyl phthalate (DBP) plasticizer, and polyvinyl butyral (PVB) binder were then added and ball-milled to form NiO-YSZ slurry. Detailed tape casting process for the NiO-YSZ support was obtained in our previous work [25]. For preparing the YSZ electrolyte, the same tape casting process as in the case of the NiO-YSZ substrate was adopted except for no addition of NiO and graphite. The porous YSZ layer was prepared with 50 wt% pore former to form backbone. The slurries for the NiO-YSZ support, the YSZ electrolyte, and the porous YSZ layer were separately cast via a tape casting machine (DR-150, made in Japan). These resulting tapes were dried at room temperature overnight. The YSZ electrolyte tape was sandwiched between the NiO-YSZ and porous YSZ layer under a vacuum condition, followed by isostatic pressing at 20 MPa for 10 min using a hot isostatic press (30Tm Shanxi, China). The resulting tri-layer tape was punched into circular discs, and then co-sintered in air at 1300 °C for 10 h in order to densify the YSZ electrolyte layer, during which the binder and graphite were removed. As a result, a NiO-YSZ supported tri-layer structure with a porous YSZ network was formed. In order to fabricate the oxygen electrode, the preparation of LSCF precursor solution and the sequential infiltration method have already been described in our previous publications [26]. Stoichiometric amounts of La(N-O₃)₃·6H₂O, Sr(NO₃)₂, Co(NO₃)₂·6H₂O and Fe(NO₃)₃·9H₂O at a molar ratio of 6:4:2:8 were dissolved with chelating agent into deionized water. Ethanol was then added as an effective additive to lower surface tension of the infiltration solution on backbone. Subsequently, the prepared precursor solution was dropwise infiltrated into the pre-treated porous YSZ backbone by using a microsyringe. The infiltrated samples were dried at 70 °C for multiple infiltration operation and then followed by firing at 450 °C for 2 h to decompose the nitrate. The infiltration-firing steps were repeatedly carried out to achieve 30–50 wt% LSCF loading. After reaching the desired loading, the samples were eventually calcined at 850 °C for 5 h to form nano-sized LSCF particles within the YSZ backbone. The nano-structured NiO-YSZ/YSZ/LSCF-YSZ cells were thus obtained with an active electrode area of 0.2 cm².

2.2. Preparation of active oxygen electrode with nano-Ag infiltration

The electrocatalyst Ag was infiltrated into the oxygen electrode in a similar manner to that described in the LSCF infiltration operation. AgNO₃ powder was dissolved into a mixture solution of deionized water and ethanol. The silver nitrate solution was dripped into the porous LSCF-YSZ oxygen electrode by using a microsyringe and dried at room temperature. The infiltration step was repeated for several times, followed by sintering at 450 °C for 2 h to form nano-structured Ag-LSCF-YSZ composite electrode.

2.3. Cell characterization and electrochemical measurement

The prepared NiO-YSZ/YSZ/Ag-LSCF-YSZ cells were sealed to a horizontal alumina tube by using ceramic paste (Aramco-552, USA). Silver paste (Ag ink, Beijing, China) was printed as current collectors onto both electrode surfaces, and then heat treated at 600 °C for 2 h in air. Silver wires were pressed against the oxygen electrode to obtain sufficient electrical contact with both electrodes in four-wire set up. The cell was placed inside a temperature-controlled tube furnace and heated at a heating rate of 2 °C/min. In order to monitor the cell operating temperature, a P-type thermocouple was placed close to the cell. At 700 °C, low flow rate of hydrogen gas was introduced into the hydrogen electrode for in-situ reduction of NiO

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