



# La<sub>0.8</sub>Sr<sub>0.2</sub>Co<sub>0.8</sub>Ni<sub>0.2</sub>O<sub>3-δ</sub> impregnated oxygen electrode for H<sub>2</sub>O/CO<sub>2</sub> co-electrolysis in solid oxide electrolysis cells

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## HIGHLIGHTS

- LSCN impregnated GDC-LSM is studied as oxygen electrode for H<sub>2</sub>O/CO<sub>2</sub> co-electrolysis.
- The cell has current density of 1.60 A cm<sup>-2</sup> at 1.5 V at 800 °C with H<sub>2</sub>O/CO<sub>2</sub> ratio of 2/1.
- The cell shows a relative high long-term stability performance for 115 h.
- The degradation behavior under co-electrolysis process is analyzed.

## ARTICLE INFO

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## ABSTRACT

High-temperature H<sub>2</sub>O/CO<sub>2</sub> co-electrolysis through reversible solid oxide electrolysis cell (SOEC) provides potentially a feasible and eco-friendly way to convert electrical energy into chemicals stored in syngas. In this work, La<sub>0.8</sub>Sr<sub>0.2</sub>Co<sub>0.8</sub>Ni<sub>0.2</sub>O<sub>3-δ</sub> (LSCN) impregnated Gd<sub>0.1</sub>Ce<sub>0.9</sub>O<sub>1.95</sub> (GDC)-(La<sub>0.8</sub>Sr<sub>0.2</sub>)<sub>0.95</sub>MnO<sub>3-δ</sub> (LSM) composite oxygen electrode is studied as high-performance electrode for H<sub>2</sub>O/CO<sub>2</sub> co-electrolysis. The LSCN impregnated cell exhibits competitive performance with the peak power density of 1057 mW cm<sup>-2</sup> at 800 °C in solid oxide fuel cell (SOFC) mode; in co-electrolysis mode, the current density can reach 1.60 A cm<sup>-2</sup> at 1.5 V at 800 °C with H<sub>2</sub>O/CO<sub>2</sub> ratio of 2/1. With LSCN nanoparticles dispersed on the surface of GDC-LSM to maximize the reaction active sites, the LSCN impregnated cell shows significant enhanced electrochemical performance at both SOEC and SOFC modes. The influence of feed gas composition (H<sub>2</sub>O-H<sub>2</sub>-CO<sub>2</sub>) and operating voltages on the performance of co-electrolysis are discussed in detail. The cell shows a very stable performance without obvious degradation for more than 100 h. Post-test characterization is analyzed in detail by multiple measurements.

## 1. Introduction

In recent years, due to the fossil fuel resource shortage, as well as environmental issue, there have been increasing demands for renewable and clean energy. Many new energy technologies, such as solar, wind, hydro and biomass, have been widely utilized and developed to produce primary electricity. However, the tremendous challenge is that the intermittent nature of these new energy sources is limited by many external conditions (geographical location, temperature, humidity, etc.), which makes it was not as convenient as traditional fossil fuels. Consequently, large-scale energy conversion devices are essential for the conversion and storage of those renewable energy sources [1]. Solid oxide electrolysis cell (SOEC) can effectively use these intermittent energy sources to produce hydrogen or syngas, which can serve as an alternative energy carrier. Hydrogen is considered as an ideal energy

carrier because of its eco-friendly combustion. However, the major problem of hydrogen as an alternative fuel is its safe storage and transport [2,3], while syngas can substitute for hydrogen as a suitable alternative energy carrier.

SOEC is an energy conversion device that converts electrical energy into chemical energy (H<sub>2</sub>, CO or syngas) by electrolyzing H<sub>2</sub>O, CO<sub>2</sub> or H<sub>2</sub>O/CO<sub>2</sub> mixtures [4–6]. In addition, O<sub>2</sub> can be obtained as by-product in this process. Syngas can be used as feedstock through the Fischer-Tropsch (F-T) process to produce synthetic liquid fuel [7], and then the synfuel can be used as alternative fuels in many industrial sectors without changing existing infrastructures [8]. Moreover, electrolysis cells can also be operated reversely as solid oxide full cell (SOFC) to convert chemical energy into electrical energy when the energy supply is insufficient [3]. Therefore, it is necessary to develop SOEC technology as an energy conversion for large-scale application of clean

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energies. Comparing with the sole H<sub>2</sub>O or CO<sub>2</sub> electrolysis, there are several significant advantages of simultaneous electrolysis (co-electrolysis) of H<sub>2</sub>O and CO<sub>2</sub>. The performance of co-electrolysis reaction is much more close to the electrolysis of sole H<sub>2</sub>O, which shows a lower overpotential and polarization resistance in comparison with sole CO<sub>2</sub> electrolysis that needs a much higher activation energy for its slow kinetics. Besides, CO<sub>2</sub> can be reduced to CO in the co-electrolysis process via the reverse water gas shift (RWGS,  $\text{H}_2 + \text{CO}_2 \xrightleftharpoons[\text{WGS}]{\text{RWGS}} \text{CO} + \text{H}_2\text{O}$ ) reaction, which can significantly reduce the consumption of the total electricity. Moreover, the appropriate proportion of syngas (H<sub>2</sub>/CO = 2/1) can be directly used as feedstock to produce syngas by controlling conditions (operating temperature, inlet gas composition and applied voltage or current) of the co-electrolysis process [9–11]. Therefore, it has a significant advantage for developing of mature and feasible solid oxide co-electrolysis cell.

SOECs are similar to solid oxide fuel cells (SOFCs) just operated in reverse, therefore they have similar material compositions and geometric configurations; however, there still have many differences. Due to the changes of operating mode and environment, SOECs have some special requirements on the materials. The current research shows that the SOEC oxygen electrode is the most rapidly attenuated component. LSM (Sr-doped LaMnO<sub>3</sub>) is widely used as the oxygen electrode in SOFC with expressive electrochemical performance, but it shows a high polarization loss and delamination behavior in electrolysis mode [12–15]. Recently, researchers have studied mixed ionic and electronic conductors (MIEC) as oxygen electrode. These materials possess high electronic and oxygen ionic conductivity, leading to a high oxygen surface exchange coefficient at the gas/oxygen electrode interface. Co-based perovskite material promises to be used as oxygen electrodes in SOEC for its high oxygen permeability and catalytic activity. La<sub>0.8</sub>Sr<sub>0.2</sub>Co<sub>0.8</sub>Ni<sub>0.2</sub>O<sub>3-δ</sub> (LSCN) was reported to be a promising alternative cathode material for SOFCs with high electrocatalytic activity [16,17]. Laguna-Bercero et al. [18] have studied the performance of LSCN as a novel electrode for solid oxide reversible cells (SORCs) and found the cell performs equally in both SOFC and SOEC modes. In our previous study [19], the impregnated LSCN oxygen electrode was investigated for H<sub>2</sub>O electrolysis in SOEC with expressive performance. However, so far little attention has been paid to LSCN as oxygen electrode for SOEC to co-electrolyze H<sub>2</sub>O/CO<sub>2</sub> mixture.

In this study, the cell with LSCN-impregnated oxygen electrode was prepared to investigate the electrochemical performance of H<sub>2</sub>O/CO<sub>2</sub> co-electrolysis to prepare syngas. The cell was tested at both electrolysis and galvanic modes for evaluation the performance of the oxygen electrode. The long-term electrolysis stability and the degradation behavior during co-electrolysis process are also discussed and analyzed in detail.

## 2. Experimental

### 2.1. Preparation of single cell

Hydrogen electrode-supported SOEC was used in this research. The structure of this button cell includes Ni-YSZ (Ytria stabilized Zirconia) as hydrogen electrode, YSZ (TZ-8Y, Tosoh, Japan) as electrolyte and impregnated La<sub>0.8</sub>Sr<sub>0.2</sub>Co<sub>0.8</sub>Ni<sub>0.2</sub>O<sub>3-δ</sub> + GDC-LSM as oxygen electrode. The detail preparation of the cell can be found in Ref. [20]. For the oxygen electrode preparation, (La<sub>0.8</sub>Sr<sub>0.2</sub>)<sub>0.95</sub>MnO<sub>3-δ</sub> (LSM) powders were mixed with Gd<sub>0.1</sub>Ce<sub>0.9</sub>O<sub>1.95</sub> (GDC) at 50:50 weight ratio, and added appropriate amounts of adhesive and carbon powders as a pore former to form slurry. The slurry was screen printed on YSZ electrolyte twice, and then sintered at 1200 °C for 2 h in air to form porous GDC-LSM scaffold. The thickness of the prepared scaffold was approximate 15 μm and the active area was 0.5 cm<sup>2</sup> for calculating area specific resistance (ASR) and current density. 0.5 mol L<sup>-1</sup> of LSCN solution was prepared for impregnation [19]. The GDC-LSM scaffold was then

impregnated with the as-prepared LSCN solution and then sintered at 750 °C for 2 h in air. The loading of LSCN was controlled at about 30 wt % by repeating impregnation-drying cycles [17]. For comparison, a cell with GDC-LSM composite oxygen electrode without impregnated La<sub>0.8</sub>Sr<sub>0.2</sub>Co<sub>0.8</sub>Ni<sub>0.2</sub>O<sub>3-δ</sub> was also prepared.

### 2.2. Characterization and measurements

The microstructure was analyzed using field emission scanning electron microscopy (FE-SEM, Sirion 200). The crystal structure was characterized by X-ray diffraction (XRD, Shimadzu XRD-7000S).

For electrochemical characterizations, the hydrogen electrode side of the as-prepared cell was attached to the end of a tubular alumina and was sealed with ceramic adhesive (Aremco Products, Inc.). Pt paste was printed on the surface of the oxygen electrode and sintered at 850 °C in air for 2 h. Pt meshes were attached on both electrodes to work as current collector, and each Pt mesh connected with two Pt wires as current and voltage probes. The four-probe method was used to eliminate the influence of the ohmic loss of the collector conductor and make the test data more accurate.

For SOFC performance test, the hydrogen electrode side was fed with humidified hydrogen (3 vol % at 25 °C, 100 mL min<sup>-1</sup>), and the oxygen electrode was exposed to ambient air at the operating temperature range from 600 °C to 800 °C. For SOEC test, the hydrogen electrode was fed of a mixture of H<sub>2</sub>O-CO<sub>2</sub>-H<sub>2</sub> with different proportions. 20 standard-state cubic centimeter per minute (sccm) flow rate of hydrogen was mixed in the feed gas to maintain a reducing atmosphere for preventing Ni being oxidized at high-temperature, humidity atmosphere. The oxygen electrode was supplied with air at a flow rate of 100 sccm. Electrochemical impedance spectra (EIS) and polarization curves (I-V curves) were measured by Zahner Zennium electrochemical workstation. For EIS, the measurements were recorded under open-circuit voltage (OCV) or different applied voltages of 1.0, 1.2 and 1.4 V. The signal amplitude was 10 mV with frequency range of 0.1 Hz–100 kHz. The cell was also tested for a short-term performance under constant voltages of 1.2, 1.5 and 1.8 V, respectively. The long-term stability performance of the cell was tested in galvanostatic mode at current density of 0.4 A cm<sup>-2</sup>.

## 3. Results and discussion

### 3.1. Microstructure

Fig. 1 (a) shows the cross-sectional image of LSCN + GDC-LSM/YSZ/Ni-YSZ cell. The cell is composed of three layers, namely, the impregnated LSCN + GDC-LSM oxygen electrode (~15 μm), the dense YSZ electrolyte (~10 μm) and Ni-YSZ hydrogen electrode (~800 μm). The interfaces between each layer exhibit compact contact without delamination or cracks. It also can be found that the impregnated LSCN nanoparticles are scattering and uniformly distributing on the surface of GDC-LSM scaffold (Fig. 1 (b)).

### 3.2. Cell performance at SOFC mode

The performance of LSCN impregnated GDC-LSM oxygen electrode cell and GDC-LSM oxygen electrode cell in SOFC mode had been tested respectively before the co-electrolytic performance testing. I-V-P curves and EIS results at different temperatures are illustrated in Fig. 2. As can be seen, the OCV of LSCN impregnated cell is 1.110, 1.095, 1.085, 1.075 and 1.065 V, corresponding to the temperature from 600 to 800 °C. The OCVs are consistent with the theoretical Nernst potentials (such as 1.133 V at 600 °C, 1.099 V at 800 °C) at different temperatures, indicating that the cell is well prepared without crack and the gastight of the cell can separate the fuel and the oxidant effectively. The peak power density of LSCN impregnated cell is 201, 357, 580, 817 and 1057 mW cm<sup>-2</sup>, respectively, at temperature from 600 to 800 °C (Fig. 2

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