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Efficient carbon dioxide electrolysis in a symmetric solid oxide electrolyzer based on nanocatalyst-loaded chromate electrodes

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ABSTRACT

Composite cathode based on redox-stable $\text{La}_{0.75}\text{Sr}_{0.25}\text{Cr}_{0.5}\text{Mn}_{0.5}\text{O}_{3-\delta}$ (LSCM) can be handled to perform for direct CO_2 electrolysis without a flow of reducing gas over the electrode; however, the insufficient electrocatalytic activity of the ceramic composite cathode still limits the electrode performances and current efficiencies. In this case, catalytic-active iron nanocatalyst and iron oxide catalyst were loaded into the LSCM-based composite cathode and anode, respectively, to improve the electrode performances. Then efficient direct CO_2 electrolysis was demonstrated by using the symmetric solid oxide electrolyzer based on LSCM loaded with 2 wt% Fe_2O_3 at 800 °C. The dependences of conductivity of LSCM were studied on temperature and oxygen partial pressure and further correlated to the electrode performance. The loading of nanocatalyst considerably improves the electrode performance and the current efficiency of CO_2 electrolysis was accordingly enhanced by approximately 75% for the impregnated LSCM-based electrode at 800 °C. The synergistic effect of catalyst-active iron nanoparticles and redox-stable LSCM perovskite ceramic leads to the excellent stability and better cathode performance for the direct CO_2 electrolysis at high temperatures.

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Introduction

Solid oxide electrolyzer has attracted a great of interests because it can efficiently and directly convert electrical energy into chemical energy and provide an alternative for producing renewable fuels [1–3]. Significant efforts have been devoted to

the developments of CO produced from CO_2 electrolysis. The technical developments related to its storage and allocations have contributed to the commercialized utilization [4,5]. Therefore, electrochemical reduction of CO_2 into CO has been proposed as a feasible form of energy carrier taking into account that it would be compatible with the existing energy infrastructures. Production of CO fuel or its derivatives from

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CO₂ would be an effective method to access a new form of energy carrier and enable a carbon-neutral cycle [6–8]. High temperature electrolysis shows great promise in the electrolysis of CO₂ using a solid oxide electrolyzer [9]. An oxide-ion-conducting solid oxide electrolyzer is able to electrolyze CO₂ into CO and O₂ by applying external voltages. The O²⁻ moves through the oxide-ion-conducting electrolyte to the anode, where O₂ is formed and released in the oxygen electrode [10]. High temperature electrolysis process is with great promise because the heat partly offers the energy for molecular dissociation which leads to favorable kinetics and thermodynamics because of the high operation temperatures ranging from 700 to 1000 °C even without using expensive noble metals as electrodes [11,12].

Recently, most studies of high temperature electrolysis of CO₂ have been preferentially performed with oxide-ion-conducting solid oxide electrolyzers based on Ni-YSZ composite cathodes [13,14]. However, a significant concentration of CO flowing over the Ni cermet is required to avoid the oxidation of Ni to NiO, which would cause a loss of electronic conductivity and even the failure of the electrode [15]. Irvine et al. have recently proved the efficient electrolysis of CO₂ based on a ceramic LSCM cathode without using a flow of CO, and it has been found that the perovskite ceramic La_{0.75}Sr_{0.25}Cr_{0.5}Mn_{0.5}O_{3-δ} (LSCM) is an active and redox-stable material, which can be used as the efficient cathode material of the solid oxide electrolyzer for that it exhibits comparable cathode performances compared to the traditional Ni-YSZ cermet [16–18]. Especially, LSCM can be used for both anode and cathode because it can work under extreme oxidizing or reducing atmospheres [19,20]. However, the catalytic activity of the ceramic materials may be a constraint on the improvement of their performance, so that the current efficiency is restricted by the insufficient electrocatalytic activity of the LSCM ceramic material. Therefore, it is of great importance to enhance the electrocatalytic activity of the LSCM electrode to improve the performance of the direct CO₂ electrolysis [21].

In this study, active iron metal and iron oxide were loaded onto the composite cathode and anode based on LSCM for direct CO₂ electrolysis, respectively. The electrical properties of the LSCM were investigated and correlated to the electrode performances, and the performance of the direct CO₂ electrolysis with the electrodes based on iron-loaded LSCM cathode and iron oxide loaded anode was systematically investigated at 800 °C.

Experimental

All chemicals utilized in this current investigation were of analytical grade unless otherwise specified. All the powders were acquired from SINOPHARM Chemical Reagent CO., Ltd (China) except specifically stated. The LSCM powders were synthesized via a combustion method with stoichiometric amounts of Cr(NO₃)₃·9H₂O, La₂O₃, SrCO₃, C₄H₆MnO₄·4H₂O and glycine which were mixed and dissolved in nitric acid, then heated the solution until it was combusted, collected the product of the combustion and fired at 1200 °C (2 °C min⁻¹) for 5 h in air [22,23]. The Ce_{0.8}Sm_{0.2}O_{2-δ} (SDC) powders were

prepared by the same method with Sm₂O₃, Ce(NO₃)₃·6H₂O and glycine followed by a heat treatment at 800 °C (2 °C min⁻¹) for 3 h in air [24–27]. The phase formation of the LSCM powders was confirmed using X-ray diffraction (XRD, Cu K_α, 2θ = 3° min⁻¹, D/MAX2500V, Rigaku Corporation, Japan) in 2θ ranging from 10 to 80° and XRD Rietveld refinement was performed using GSAS software. The microstructure of LSCM powders was characterized by high-resolution transmission electron microscopy (HRTEM, JEM-2100F, JEOL Ltd, Japan). The electrode powders with 2 wt% Fe₂O₃ were prepared by milling the LSCM powders with the SDC powders at a 60:40 weight ratio, and then impregnated the powders with stoichiometric concentration of Fe(NO₃)₃ solution followed by a heat treatment at 550 °C (3 °C min⁻¹) for 30 min in air. X-ray photoelectron spectroscopy (XPS, ESCALAB25, Thermo, USA) was performed on a Thermo ESCALAB 250 using monochromatized Al K_α at *hν* = 1486.6 eV to analyze the chemical states of the elements before and after high temperature reduction. The binding energies were calibrated to the C_{1s} peak at 285 eV. Specific surface areas of the bare LSCM powders and the LSCM-Fe₂O₃ powders were measured with the Brunauer Emmett and Teller (BET) method (low temperature adsorption of nitrogen), respectively.

About 2.0 g LSCM powders were pressed into a bar followed by a sintering at 1400 °C (2 °C min⁻¹) for 10 h in air for a conductivity test. The relative density of sample reached 71.02%. The conductivity test was performed in air using the DC four-terminal method with temperature ranging from 300 to 800 °C. Four Ag wires (0.4 mm in diameter) were mounted on the bar by using the conductive adhesive (DAD87, Shanghai Research Institute for Synthetic Resins, Shanghai, China) followed by a heat treatment at 500 °C (3 °C min⁻¹) for 30 min in air. Then the bar was mounted onto a four terminal jig coupled with an oxygen sensor device. The conductivity was recorded versus temperature with an online system at a step 0.5 °C. The dependence of conductivity on the oxygen partial pressure was tested at 800 °C with the oxygen partial pressure ranging from 10⁻² to 10⁻²⁰ atm. The oxygen partial pressure was adjusted by flowing 5% H₂/Ar at the flow rate of 0.5 ml min⁻¹ controlled by a mass flow meter (D08-3F, Sevenstar, China). The oxygen partial pressure and conductivity were recorded using an online sensor (Type 1231, ZrO₂-based oxygen sensor, Noveltech, Australia) and an online multi-meter (Keithley 2000, Digital Multimeter, Keithley Instrument Inc., USA), respectively. The conductivity test of LSCM sample was also performed in 5% H₂/Ar with the temperature ranging from 300 to 800 °C.

A group of 2-mm-thick 8YSZ electrolyte supports were prepared by dry-pressing the 8YSZ powders into green disks with a diameter of ~20 mm followed by sintering at 1500 °C (2 °C min⁻¹) for 10 h in air [28,29]. Two surfaces of the obtained YSZ electrolyte support were mechanically polished with sand paper (1000 mesh), and then ultrasonically cleaned the YSZ electrolytes several times with the electrolytes immersed in ethanol and distilled water. Finally, the disks were dried in air overnight. The composite electrode LSCM-SDC slurry was prepared by directly milling the SDC powders with the LSCM powders at a 40:60 weight ratio in alpha-terpineol in a mortar. In addition, cellulose was added to create porosity in the electrode. The symmetric electrolyzer was prepared by

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