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Enhanced performance of solid oxide fuel cells by introducing a transition layer between nanostructured cathode and electrolyte

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ABSTRACT

A transition layer between $\text{Ce}_{0.8}\text{Sm}_{0.2}\text{O}_{2-\delta}$ (SDC) electrolyte and $\text{Sr}_{0.8}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$ (SCF0.8) nano cathode was introduced to improve electrochemical performances of solid oxide fuel cells (SOFCs). A calcining – stripping method was used to introduce the transition layer on the SDC electrolyte. Then a nano porous cathode was coated on the transition layer. The microstructure of the nano cathode was optimized by adjusting its calcining temperature. It was found that the transition layer played a vital role in improving the electrochemical performances of the nanostructured cathode by enhancing the interface connection between cathode and electrolyte. Good electrochemical performance was obtained as the nano cathode calcined at 700 °C after introducing the transition layer. The polarization resistance of the anode-supported SOFC at 700 °C was significantly reduced from 0.20 $\Omega\text{ cm}^2$ to 0.05 $\Omega\text{ cm}^2$, while the power density of the single cell was increased from 116 to 444 mW cm^{-2} .

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Introduction

Solid oxide fuel cells (SOFCs) have attracted much attention because of their high efficiency, good environmental benefits and high flexibility to various fuels [1–3]. However, the high-temperature (800–1000 °C) operation causes many key technical issues, such as higher systems costs, performance degradation and slow start-up cycles, and these technical difficulties have limited the commercialization of this

transformative technology [4,5]. These drawbacks are easy to be overcome by lowering the operation temperature of SOFCs because the problems related to materials costs of interconnectors, sealing, durability of all the components are no longer difficult to be solved in the intermediate-low (IL) temperature range (600–800 °C) [6–9]. However, a loss of power density is inevitable at lower operation temperatures due to a considerable decrease of ionic conductivity of the electrolyte and catalytic activity of the electrodes. The ohm losses can be

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minimized by reducing the thickness of the electrolyte or/and adopting electrolytes with high ionic conductivity [10,11]. While now the overall performance loss of anode-supported SOFCs at lower operation temperatures is mainly dominated by the activation polarization of the sluggish electrode reactions, i.e., oxygen reduction reactions on the cathode [12,13].

Oxygen reduction reactions take place at the triple phase boundaries (TPBs) of traditional composite cathodes like yttria-stabilized zirconia (YSZ)/ $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_{3-\delta}$, or on the whole surface of mixed ionic-electronic conducting (MIEC) cathodes like $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_{3-\delta}$ [14,15]. To improve SOFCs performances at IL temperature, it is necessary and feasible to reduce the polarization loss of the cathode by developing cathode materials with high catalytic activity or decreasing the grain size of cathode materials down to nano-scale to increase the active area. Nanoparticles with large surface area have long TPBs and more catalytic active sites, and will show high oxygen reduction rate at the gas–solid interfaces. In the recent studies, many efforts are focused on the nanostructured cathode to improve the electrochemical performance of SOFCs at IL temperature [16–22]. Usually, SOFCs cathodes are prepared via a calcining step at high temperatures ($>1000\text{ }^\circ\text{C}$) to enhance the connection between the cathode layer and the electrolyte layer. The connection between cathode and electrolyte has strongly influence on the performance of the cells. However, nanoparticles cannot sustain their sizes after calcined at such high temperatures. One feasible approach is to impregnate cathode precursors into porous layer of ionic conductor oxides, for instance SDC, GDC, YSZ, etc, to form nano composite cathodes after heat treated at lower temperature [12]. For example, performance was remarkable enhancement by impregnating a solution into the pre-sintered porous YSZ structure to form a nano-structured $\text{La}_{0.8}\text{Sr}_{0.2}\text{Co}_{0.5}\text{Fe}_{0.5}\text{O}_3 + \text{YSZ}$ composite cathode [23]. Additionally, electrostatic spray deposition method was used to prepare a double layer nano/micro porous cathode, which was efficient to expand the triple phase boundary and improve the catalytic activity of the cathode [15]. Thus, optimization of the cathode microstructure with high porosity, large TPBs and good adhesion between cathode and electrolyte is useful to enhance the electrochemical performance of SOFCs.

The previous investigation on oxygen absorption/desorption kinetics indicated that $\text{Sr}_{0.8}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$ (SCF0.8) has high catalytic activity toward oxygen reduction reactions in the temperature range of $450\text{--}600\text{ }^\circ\text{C}$ [24]. Therefore, we chose SCF0.8 as the cathode material. Here, we report a new method

for the preparation of nanostructured cathodes to enhance the electrochemical performances of anode-supported SOFCs. The nanoparticles of SCF0.8 with size around 238 nm were prepared by high-speed ball milling. A SCF0.8 transition layer was introduced on the SDC electrolyte by high-temperature sintering, followed by stripping porous SCF0.8 from the SDC electrolyte. Performance of the anode-supported SOFCs with nano cathode was effectively improved through the introduction of transition layer. The results demonstrated the vital role of transition layer for the connection of nano cathode and electrolyte.

Experimental

Preparation of nanoparticles

$\text{Sr}_{0.8}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$ (SCF0.8) powder was prepared by a combined Ethylene Diamine Tetra-acetic Acid (EDTA)–citric acid method [25]. Stoichiometric amounts of $\text{Sr}(\text{NO}_3)_2$ ($\geq 99.5\%$), $\text{Co}(\text{N-O}_3)_2 \cdot 6\text{H}_2\text{O}$ ($\geq 99.0\%$), $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ($\geq 98.5\%$) were dissolved in deionized water to form an aqueous solution. Then citric acid and EDTA acid were added in the above solution with a mole ratio of citric acid, EDTA and total metal ions of 1.5:1:1. pH value of the solution was adjusted to about 7 by concentrated ammonia. After water was evaporated on a hot plate, the resultant purple gel was combusted on an electric oven. Then, the ash was calcined in air for 5 h at $950\text{ }^\circ\text{C}$ to remove the carbon residues. To obtain the nano particles, the SCF0.8 powder was repeatedly ball-milled in ethyl alcohol, and then the resultant suspension was centrifuged at 3000 rpm for 3 min to remove the big particles and obtain the nano-size particles (denoted as SCF0.8nano).

Preparation of single cells

The electrochemical performances of the SCF0.8nano as cathodes of SOFCs were characterized on anode-supported $\text{Ce}_{0.8}\text{Sm}_{0.2}\text{O}_{2-\delta}$ (SDC) electrolyte. A highly fluffy SDC powder was used to prepare the electrolyte membranes. The powder was synthesized via the combined EDTA–citric acid complexing route and calcined in air at $800\text{ }^\circ\text{C}$ for 5 h. The following shows the preparation procedure of an anode-supported electrolyte disc (see Fig. 1), (1) a mixed powder containing NiO, SDC and graphite with weight ratio of 45:45:10 was firstly pressed at $\sim 60\text{ MPa}$ to produce a substrate; (2) the fluffy SDC powder was then uniformly added on the substrate followed by co-pressing at $\sim 210\text{ MPa}$ for several minutes; (3) the shaped green discs were sintered at $1490\text{ }^\circ\text{C}$ for 200 min. The resultant anode-supported electrolyte discs were about 18 mm in diameter and $540\text{ }\mu\text{m}$ in

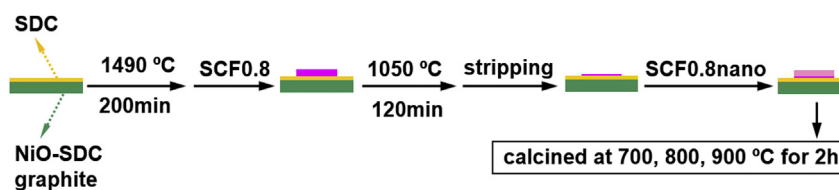


Fig. 1 – Schematic figure for the preparation of anode-supported solid oxide fuel cells.

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