



# Solid oxide fuel cells supported on cathodes with large straight open pores and catalyst-decorated surfaces



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## ABSTRACT

The  $\text{La}_{0.6}\text{Sr}_{0.4}\text{FeO}_{3.8}\text{-Gd}_{0.1}\text{Ce}_{0.9}\text{O}_{1.95.8}$  (LSF-GDC) support with large straight open pores was formed by the phase inversion tape method, and used as a cathode for solid oxide fuel cells (SOFCs) comprising GDC electrolyte and Ni-GDC anode.  $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_{3.8}$  (LSC) nano-particles were deposited deeply inside the cathode using the impregnation method. The single cell with the LSC-modified cathode exhibited a maximum power density of  $726 \text{ mW cm}^{-2}$  at  $750 \text{ }^\circ\text{C}$ , much higher than the one with the un-modified cathode ( $298 \text{ mW cm}^{-2}$ ). The polarization resistance determined with the impedance spectroscopy was  $0.025 \Omega \text{ cm}^2$  for the former, which was one order of magnitude smaller than that for the latter. It is concluded that the presence of large straight open pores in the phase inversion-derived cathode allows for easy impregnation of catalytically active nano-particles, enlarging the triple-phase-boundaries for oxygen reduction reaction and hence enhancing the electrochemical performance of the cathode-supported SOFCs.

## 1. Introduction

Solid oxide fuel cells (SOFCs) can directly convert chemical energy of a fuel gas to electrical energy, and have attracted much attention due to their high energy conversion efficiency, fuel flexibility and low emission [1]. Currently, the research and development focuses on SOFCs supported on thick porous electrodes (anode or cathode). Compared to the anode-supported SOFCs, the cathode-supported SOFCs with thin anodes exhibit better durability and reliability due to substantially reduced deleterious impact from the volume contraction and expansion within the anode redox cycles [2,3]. The lanthanum strontium manganite (LSM) cathode-supported SOFCs have been reported to achieve a long operation time [4]. However, the electrochemical performance of the cathode-supported SOFCs is lower than that of the anode-supported ones, because the high temperature required for preparation of the cathode support and electrolyte reduced the electrocatalytic activity of the cathode [5,6]. One way to cope with this problem is to deposit catalytically active nano-particles on the surface of the cathode skeleton [7,8]. It has been reported that impregnating the cathode with oxide-ionic, electronic, or mixed ionic and electronic conducting electrocatalysts led to a substantially increased power density of the single cell and reduced polarization loss of the cathode

[9–11].  $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_{3.8}$  (LSC) is known to possess high electrocatalytic activity, good electronic and ionic conductivity, suitable for modification of the cathode [12,13].

The effectiveness of the surface modification via impregnation depends on the microstructure, especially the pore structure of the cathode [14,15]. The pores in the cathode are usually introduced using the pyrolyzable substances such as starch and graphite [16,17]. The as-formed pores have irregular shape, and the pore paths are tortuous, which might reduce the effectiveness of the impregnation [18]. Recently, it has been reported that electrodes with large straight open pores can be prepared using the phase inversion tape casting method [19,20]. The SOFC supported on the phase inversion-derived electrode exhibited improved electrochemical performance due to the reduced gas transport resistance in the electrode and increased accessible triple-phase-boundaries (TPBs). The unique pore structure of the phase inversion-derived electrode may also make it easy to impregnate the interior surface of the electrode, especially in the electrode/electrolyte interface region.

$\text{La}_{1-x}\text{Sr}_x\text{FeO}_{3.8}$  has been explored for use in intermediate temperature SOFCs as cathode due to its sufficient electronic conductivity ( $324 \text{ Scm}^{-1}$  at  $650\text{--}750 \text{ }^\circ\text{C}$  for  $\text{La}_{0.6}\text{Sr}_{0.4}\text{FeO}_{3.8}$ ) and appreciable oxygen ionic conductivity, matching thermal expansion coefficient and

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chemical compatibility with dope ceria electrolyte [21,22]. In the present study,  $\text{La}_{0.6}\text{Sr}_{0.4}\text{FeO}_{3.8}$  (LSF) in composite with  $\text{Gd}_{0.1}\text{Ce}_{0.9}\text{O}_{1.95-8}$  (GDC) was therefore used as cathode for fuel cells comprising GDC electrolyte and Ni-GDC anode. The LSF-GDC support with large straight open pores was prepared using the phase inversion tape casting method, and its inner surface was modified with LSC nano-particles via the impregnation method. The electrochemical properties of the fuel cells were measured, and discussed in relation to the microstructure and surface modification of the cathode.

## 2. Experimental

### 2.1. Preparation of LSF-GDC cathode

The  $\text{La}_{0.6}\text{Sr}_{0.4}\text{FeO}_{3.8}$  powder used for preparation of the cathode was synthesized by a solid state reaction method.  $\text{La}_2\text{O}_3$  (CP, Sinopharm Chemical Reagent Co.),  $\text{SrCO}_3$  (Sinopharm Chemical Reagent Co.) and  $\text{Fe}_2\text{O}_3$  (CP, Sinopharm Chemical Reagent Co.) were mixed in their stoichiometric ratio in the nominal compositions. The mixtures were calcined at  $1100^\circ\text{C}$  for 10 h and  $1150^\circ\text{C}$  for 10 h with an intermediate ball milling. The as-prepared LSF powder was mixed with GDC (Ningbo SOFCMAN Energy Technology Co.) in a weight ratio of 6:4 in ethanol by ball milling for 6 h, and dried in the oven.

The LSF-GDC cathode was prepared using the phase inversion tape casting method described in Ref. [23]. Two slurries were used for preparation of the cathode. One slurry was composed of LSF-GDC (63.0 wt%), graphite (7 wt%) (Shanshan technology Co.), Polyethersulfone (PESf) (4.5 wt%) (Radel A-100, Solvay Advanced Polymers), *N*-methyl-2-pyrrolidone (NMP) (24.4 wt%) (CP, Sinopharm Chemical Reagent Co.) and Polyvinylpyrrolidone (PVP) (1.1 wt%) (K30, Sinopharm Chemical Reagent Co.). The other slurry consisted of graphite (60 wt%), PESf (5.5 wt%), NMP (33.1 wt%) and PVP (1.4 wt%). The slurries were degassed in vacuum until no bubble was observed. The two slurries were then co-tape casted, and immersed in a water bath for solidification via a phase inversion process. The as-formed LSF-GDC green tape was punched into disks with a diameter of 15 mm and dried at  $85^\circ\text{C}$ . The green disks were then pre-treated at  $1100^\circ\text{C}$  for 2 h.

### 2.2. Preparation and surface modification of cells

Cathode-supported fuel cells with a configuration of LSF-GDC/GDC/Ni-GDC were fabricated. The GDC slurry was applied to the LSF-GDC cathode by dip coating method, followed by sintering at  $1500^\circ\text{C}$  for 10 h to form a dense GDC electrolyte film. The Ni-GDC anode (with a weight ratio of 6:4) was deposited to the GDC film using the screen printing method, and sintered at  $1300^\circ\text{C}$  for 3 h. The NiO powder used here were prepared by the nitrate-citrate combustion method [24].

To investigate the superior of the cathode with large straight pores, symmetric cathode supported cells with a configuration of LSF-GDC/GDC/LSF-GDC were fabricated as well. A GDC green tape prepared using conventional tape casting method was laminated on the both sides by the phase inversion-derived LSF-GDC green tape, and co-sintered at  $1500^\circ\text{C}$  for 10 h [25].

The sintered cells were modified by LSC nano-particles using the impregnation method. The stoichiometric  $\text{La}(\text{NO}_3)_3$  (Analytical Reagent, Sinopharm Chemical Reagent Co.),  $\text{Sr}(\text{NO}_3)_2$  (Analytical Reagent, Sinopharm Chemical Reagent Co.) and  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  (Analytical Reagent, Sinopharm Chemical Reagent Co.) were dissolved in deionized water with ethanol and citric acid to prepare a 0.5 M  $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_{3.8}$  (LSC) precursor solution. The solution was injected to the LSF-GDC electrode of the sintered cell, and calcined at  $350^\circ\text{C}$  for 1 h. After repeating the impregnation/calcination step several times, the cathode was subjected to heat treatment at  $750^\circ\text{C}$  for 1 h to decompose the nitrate and form perovskite oxide LSC. The LSC loading was calculated by measuring the weights of the cells before and after impregnation.

### 2.3. Characterization of cathode and cells

The phase composition of the cathode was analyzed with X-ray diffractometer (XRD, TTR-III, Rigaku, Tokyo, Japan), and the microstructure of the cathode and single cells was examined by scanning electron microscopy (SEM, JSM-6390LA, JEOL, Japan).

The cathode-supported fuel cells were sealed to a ceramic tube using ceramic adhesive (Aremco, Ceramabond 552). Silver (Ag) paste (Sino-platinum Metals Co., Ltd., Kunming, China) and Ag wires were used as the current collectors and lead wires, respectively. During the cell measurement, humidified  $\text{H}_2$  (with 3 vol%  $\text{H}_2\text{O}$ ) with a flow rate of 30 sccm was used as fuel, while the cathode was exposed to the ambient air. The current density-voltage curves (*i*-*V*) and the electrochemical impedance spectra (EIS) of the cathode-supported fuel cell and symmetric cathode cell were measured by the electrochemical system (Zahner IM6e electrochemical workstation, Kronach, Germany) at the operating temperature ranging from  $650$  to  $750^\circ\text{C}$  with an interval of  $50^\circ\text{C}$ . The EIS were collected under open circuit voltage (OCV) condition with an AC amplitude of 10 mV in the frequency range of 0.1 to  $10^6$  Hz.

## 3. Results

### 3.1. Phase composition and morphology of the cathode

XRD analysis revealed that the sintered cathode was composed of fluorite-structured GDC and perovskite-structured LSF (Fig. 1). No peaks except those for LSF and GDC were observed, indicating that no chemical reactions occurred between the two oxides.

The cathode consisted of a finger-like layer of thickness  $390\ \mu\text{m}$  underneath a skin layer of thickness  $\sim 17\ \mu\text{m}$  (Fig. 2a). The finger-like layer contained fairly uniform honeycomb pores with  $40\text{--}70\ \mu\text{m}$  in diameter (Fig. 2b). These large pores were fully open to the external surface and straightly aligned along the thickness direction of the cathode. The sintered cathode possessed well-grained structure (Fig. 2c). After impregnation (5 wt% loading), LSC nano-particles were deposited deeply inside the cathode (Fig. 2d). Most of the LSC nano-particles were connected to each other especially in the grain boundary region, while some were discretely distributed on the surface of the LSF-GDC grains.

### 3.2. Electrochemical performance of cells

Fig. 3 shows the overall cross-sectional SEM image of the fuel cell

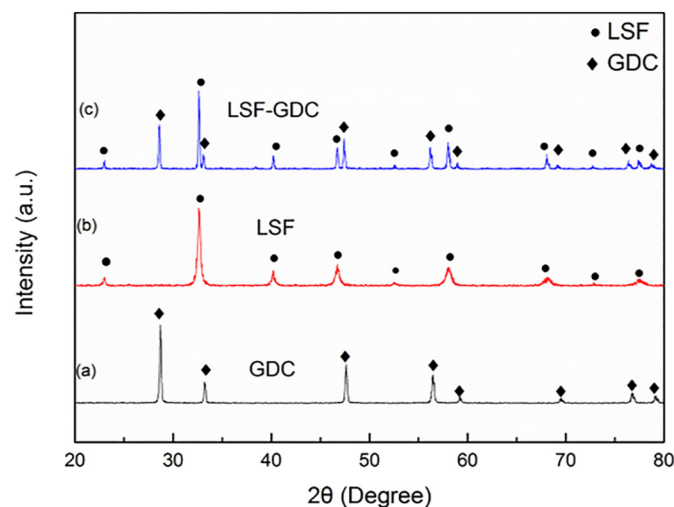


Fig. 1. The XRD patterns of (a) commercial GDC powder, (b) LSF powder and (c) the LSF-GDC composite cathode after sintering at  $1500^\circ\text{C}$  for 10 h.

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