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Enhanced performance of LSCF cathode through surface modification

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ABSTRACT

Mixed ionic-electronic conductors in the family of $\text{La}_x\text{Sr}_{1-x}\text{Co}_y\text{Fe}_{1-y}\text{O}_{3-\delta}$ (LSCF) have been widely studied as cathode materials for solid oxide fuel cells (SOFCs). However, the long-term stability and the limited surface catalytic activity are still a concern. Here we report a new catalyst $\text{La}_{0.4875}\text{Ca}_{0.0125}\text{Ce}_{0.5}\text{O}_{2-\delta}$ (LCC), which can significantly enhance the performance and stability of LSCF cathodes when applied as a thin-film coating on LSCF surface. For example, with $5 \mu\text{L}$ 0.25 mol L^{-1} LCC solution infiltrated into LSCF cathode, the cathodic polarization resistance was reduced by $\sim 60\%$ (to $\sim 0.076 \Omega \text{ cm}^2$) at 750°C , leading to a peak power density of $\sim 1.25 \text{ W/cm}^2$, which is $\sim 18\%$ higher than that for the unmodified LSCF cathode in an anode-supported cell. In addition, stable power output was observed for over 500 h operation at 750°C under a constant voltage of 0.7 V.

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1. Introduction

In recent years, the $\text{La}_x\text{Sr}_{1-x}\text{Co}_y\text{Fe}_{1-y}\text{O}_{3-\delta}$ (LSCF) based cathode for SOFCs has been extensively investigated due to its much higher ionic and electronic conductivity and hence better performance than the conventional LSM cathode [1–3] in the temperature range of $600\text{--}800^\circ\text{C}$. However, the long-term stability of LSCF cathodes is a concern for practical applications [4–6]. Additionally, the oxygen reduction reaction on LSCF cathode is likely limited by the surface exchange process rather than the bulk charge transport process [7–9].

In order to enhance the performance and/or stability of LSCF cathode, several approaches have been studied, including using composite cathode of LSCF-SDC [10,11], LSCF-GDC [2], LSCF-Ag [12] and LSCF-SDC-Ag [13], or using precious metal such as Pt, Pd, and Ag [14–16] to coat the LSCF surface and

enhance its performance. Recently, we proposed a method to improve the performance and stability of a porous LSCF cathode by application of a thin catalytically active coating through infiltration [17]. Improved performance and stability have been demonstrated using thin coating of LSM [9], SSC [18] and SDC [19] on LSCF. Compared to the aforementioned work [2,10–16], this approach can be more readily incorporated in the current fabrication processes at low cost.

Here we report a new catalyst, $\text{La}_{0.4875}\text{Ca}_{0.0125}\text{Ce}_{0.5}\text{O}_{2-\delta}$ (LCC), for LSCF cathode surface modification. Doped ceria compounds have been widely used as catalyst materials due to their high ionic conductivity, favorable surface exchange properties, and oxygen storage capability [20]. Among various dopants studied, La incorporated into the cerium site can significantly enhance the oxygen transfer capability [21]. Although LCC displays relatively poor ionic conductivity,

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the high doping level of ceria by La and Ca (i.e. 50% mol. total substitution) creates high surface oxygen vacancy concentration. When the LSCF cathode is modified by a thin LCC coating, the surface layer with high oxygen vacancy concentration may enhance oxygen molecule dissociation into atomic oxygen [22] and facilitate the oxygen reduction reaction. Even more importantly, the LCC coating can alleviate the enrichment of Sr at the LSCF/SDC interface and LSCF grain surface that is believed to be at least partially responsible for the LSCF degradation [4]. The doped ceria shows excellent solubility of SrO [23], which can reduce the driving force of diffusion of Sr from the LSCF cathode to the YSZ electrolyte and thus help prevent the enrichment of Sr on LSCF grain surface that would otherwise block active sites for oxygen reduction. In this manner, LCC coated LSCF may potentially enhance the stability during operation which would ultimately reduce the cost of SOFC technology.

2. Experimental

To evaluate the polarization resistance of the modified LSCF cathodes, we used symmetric cells with two identical LSCF cathodes on both sides of the YSZ electrolyte: LSCF|SDC|YSZ|SDC|LSCF. Further, the performance enhancement was also validated in anode-supported button cells with a configuration of Ni-YSZ|YSZ|SDC|LSCF.

YSZ electrolyte was used for construction of symmetrical cells in this study so that the LSCF cathodes for symmetrical cells were identical to those for anode-supported YSZ cell. Cylindrical YSZ pellets were prepared by uniaxially pressing a commercially available YSZ powder (Daiichi Kigenso, Japan) followed by sintering at 1450 °C for 5 h to achieve relative density of ~98%. LSCF (from Fuel Cell Materials) green tapes prepared by tape-casting [24] (~50 μm) were then bound onto both sides of a YSZ pellet using a slurry of $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{2-\delta}$ (SDC, 3–4 μm) as a buffer layer, which was subsequently co-fired with SDC on YSZ at 1080 °C for 2 h to form porous LSCF electrodes with an active area of 0.3 cm². The SDC powder was synthesized using a chemical co-precipitation process [25,26]. The SDC powder was then dispersed in acetone with V-006 as binder and ball-milled for 24 h to form a stable SDC slurry.

Aqueous nitrate solutions of LCC precursors with different concentrations (0.05, 0.10, 0.125, 0.25 and 0.5 mol L⁻¹) were prepared by dissolving proper amounts of $\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$, $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ and glycine in distilled water. Glycine was used as a complexing agent to facilitate the phase formation. For each surface modification, 5 μL of LCC solution was infiltrated into a porous LSCF electrode using a micro-liter syringe to control the amount of loading. The infiltrated cell was then fired at 900 °C in air for 1 h to obtain the LCC-infiltrated LSCF. The ASR of cathodes was measured in a two-electrode symmetric cell configuration using two Pt meshes as current collector (without Pt paste to avoid the possible catalytic contribution from Pt). Impedance spectra were acquired using a Solartron 1255 HF frequency response analyzer interfaced with an EG&G PAR potentiostat model 273A with an amplitude of 10 mV in the frequency range from 100 kHz to 0.1 Hz.

For comparison, a mixture of LSCF and LCC with weight ratio of 7:3 was mixed thoroughly with V-006 binder to prepare cathode slurry, and the slurry was then applied to both sides of sintered SDC pellets by brush-painting followed by sintering at 1050 °C to assemble a symmetrical cell. Pure LSCF cathodes and LSCF-SDC (weight ratio of 7:3) composite cathodes were also prepared using the same method for comparison.

To fabricate anode-supported button cells, a tape-cast NiO-YSZ anode-support was first fabricated and pre-fired at 900 °C for 2 h. Then, an active NiO-YSZ layer (~15 μm) and YSZ electrolyte (~15 μm) were sequentially deposited on the anode support by a particle suspension coating process [27,28] followed by co-firing at 1400 °C for 5 h. The LSCF cathode was then applied to the YSZ electrolyte using the same procedures for the fabrication of symmetric cells as described earlier. 5 μL 0.25 mol L⁻¹ LCC solution was infiltrated into cathode followed by firing at 900 °C for 1 h. The whole cell was mounted and sealed on a fuel cell testing fixture, and then tested with humidified hydrogen (3 %H₂O) as fuel and ambient air as oxidant. The cell performances were examined with an Arbin multi-channel electrochemical testing system (MSTAT).

X-ray diffraction (XRD) analysis was used to examine the phase purity of the LCC powders (derived from nitrate solutions) and the LSCF powder as well as the chemical compatibility between LCC and LSCF. The microstructure and morphology of the LCC-infiltrated LSCF cathode was examined using a thermally assisted field emission scanning electron microscope (SEM, LEO 1530). Raman spectroscopy (Renishaw RM 1000, 514 nm excitation) was also performed on both LCC-infiltrated and unmodified cathodes before and after long-term operation of the full cells.

3. Results and discussion

3.1. Structural characterization

Shown in Fig. 1 are the XRD patterns of LCC powders derived from the infiltration solution and subsequently fired at different temperatures. A fluorite phase of LCC, with

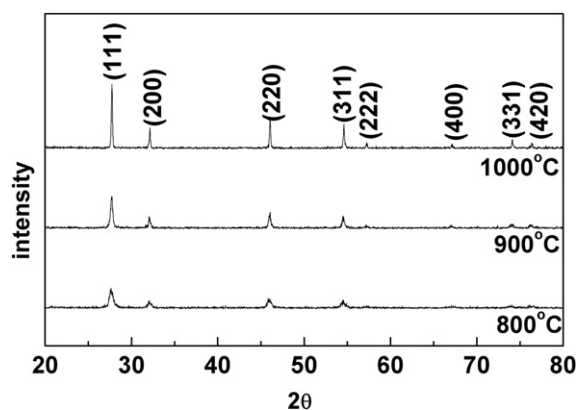


Fig. 1 – X-ray diffraction patterns of LCC powders derived from the infiltration solution and subsequently fired at different temperatures.

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