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## Research on a low temperature reversible solid oxide cell

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### ABSTRACT

In this study, two-electrode-supported solid oxide cells with the structure of “Ni-Sm doped CeO<sub>2</sub> infiltrated porous La<sub>0.9</sub>Sr<sub>0.1</sub>Ga<sub>0.8</sub>Mg<sub>0.2</sub>O<sub>3</sub> (LSGM)| dense LSGM | SmBa<sub>0.5</sub>Sr<sub>0.5</sub>Co<sub>2</sub>O<sub>5</sub> infiltrated porous LSGM” are fabricated. Fuel cell and steam electrolysis performance at different operating temperature (450–600 °C) for different steam content are tested, and reaction mechanism discussed. In the fuel-cell mode, the maximum power density reaches 1.817, 1.345, 0.801 and 0.342 W cm<sup>-2</sup> at 600, 550, 500, and 450 °C, respectively. In electrolysis mode, current density at 1.3 V in 70% H<sub>2</sub>O – 30% H<sub>2</sub> stream is –2.472, –1.616, and –0.727 A cm<sup>-2</sup> at 600, 550, and 500 °C, respectively. The durability of the cell is tested for over 351 h.

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### Introduction

Reversible solid oxide cells (SOC) [1] can be operated in both electrolysis mode (SOEC: to store electricity energy with chemical fuels) and fuel-cell mode (SOFC: to convert fuels into electricity), which is considered as a promising energy storage technology [2–5] to balance the supply-demand mismatch for electricity from renewable energy such as solar and wind energy. Conventional SOFC using Yttria stabilized Zirconia (YSZ) as the electrolyte and Sr-doped LaMnO<sub>3</sub> (LSM) as the air electrode is called as high temperature SOFC, because the YSZ can only meet sufficient ionic conductivity, 0.1 S/cm, at around 1000 °C. So high an operating temperature severely limited the selection of component materials and hindered the commercialization of this technology [6].

Recently, there is a trend to reduce the operating temperature of SOCs into the so-called low temperature (LT) range (lower than 650 °C) for the following reason: at first, if the operating temperature could be decreased down to 500–600 °C, many waste heat sources in industry such as steel manufacturing or those from various combustors could be used for SOEC [7]; secondly, Fischer-Tropsch synthesis reaction [8,9] for directly synthesis of CH<sub>4</sub> [3] can be possible when stable electrolysis of CO<sub>2</sub> [10–12] can be realized on LT-SOCs; third, cheaper cost of components such as separators of SOC at low temperature would reduce the system cost [13]. For this purpose, however, the ohmic resistance and polarizations should be reduced by fine design of the cells and careful selection of materials.

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$\text{La}_{1-x}\text{Sr}_x\text{Ga}_{1-y}\text{Mg}_y\text{O}_{3-0.5(x+y)}$  (LSGM) has sufficient ionic conductivity at reduced temperatures; it is stable and has no electronic conductivity in both oxidizing and reducing atmospheres. Because of such advantages, LSGM has been known as a very promising electrolyte material for intermediate temperature solid oxide fuel cells (IT-SOFC) [14–16] and solid oxide electrolysis cells (IT-SOEC) [17]. In contrast, Sm-doped ceria (SDC) or Gd-doped ceria (GDC) is not a good electrolyte for its partial reduction in reducing atmospheres, although it has comparable ionic conductivity as LSGM and is widely used as a barrier layer or a component in electrode materials [18]. In the temperature range lower than 650 °C, the thickness of LSGM film is expected to be reduced, and then electrode supported LSGM film, especially which supported from both sides, is the ideal choice. In this case, however, the thickness of electrodes increases from normal scale, and the risk of concentration polarization becomes the main concern; there are many model simulation works in the literature which reveal the importance of controlling the diffusion resistance [19–21].

Ni electrode has excellent activity and is a good catalyst for the  $\text{H}_2$  formation during water splitting [22]. To avoid its reaction with LSGM and sintering at high manufacturing temperature, Ni is expected to be infiltrated into pre-sintered porous LSGM skeleton. The obtained nano-particles of Ni have high activity, but its sintering is the main reason of degradation. In our previous study, it has been confirmed that Ni-SDC infiltrated LSGM electrode is more stable and catalytically more active than Ni infiltrated LSGM electrode [23].

$\text{SmBa}_{0.5}\text{Sr}_{0.5}\text{Co}_2\text{O}_5$  is demonstrated a good cathode material, it has high conductivity and shows small area specific resistance (ASR) at intermediate temperatures [24]. And the recent studies show that infiltrated  $\text{SmBa}_{0.5}\text{Sr}_{0.5}\text{Co}_2\text{O}_5$  has even higher catalytic activity and lower ASR [25].

Above materials have been widely studied in literature, but the assembly of them was mainly studied in the SOFC mode; the performance and its stability in SOEC mode is interesting but not well studied. In the present work, two-electrode-supported SOCs with the structure of “Ni-SDC infiltrated porous  $\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_3$  (LSGM) | dense LSGM |  $\text{SmBa}_{0.5}\text{Sr}_{0.5}\text{Co}_2\text{O}_5$  infiltrated porous LSGM” are fabricated. The cells have the dense LSGM film as thin as 16  $\mu\text{m}$  and are expected to have small ohmic resistance; infiltrated electrodes would also have low polarizations. The performance of the SOCs is tested and the stability discussed.

## Experimental

### Cell fabrications and characterization

The scaffold of the symmetrical cells with tri-layer structures of “porous LSGM | dense LSGM | porous LSGM” was fabricated by laminating one dense LSGM green tape and two porous LSGM green tapes on both sides with subsequent co-firing at 1380 °C for 4 h. The green tape for LSGM dense layer was prepared by tape casting technology. For this purpose, commercial LSGM powder was weighted and ball milled for 12 h, in the mixture of xylene and butyl acetate, with triethanolamine as dispersant. Then polyvinyl butyral (PVB) as binder, polyethylene glycol and dibutyl phthalate as plasticizer were

added. The mixture was ball milled for 8 h to get the slurry, which was degassed in vacuum for 5 min, and tape casted on PET tape, following with drying at 50 °C, to get the green tape for the dense LSGM electrolyte film. As for the porous LSGM tape, similar procedure was adopted, but graphite as pole former was added to LSGM powder with the weight ratio being 1:5. During tape casting, the height of blade was 75  $\mu\text{m}$  for the dense layer but 300  $\mu\text{m}$  for the porous layer. During laminating of the green tapes, they were pressed under 3000 psi at 75 °C, and the hold time was 20 min. The green tri-layer tape of “porous LSGM | dense LSGM | porous LSGM” was cut to plates of 14 mm in diameter, and then sintered in air at 1380 °C for 4 h to get the LSGM skeleton of the symmetrical cells with ~20  $\mu\text{m}$  thick dense LSGM electrolyte film in the middle.

For the preparation of  $\text{SmBa}_{0.5}\text{Sr}_{0.5}\text{Co}_2\text{O}_5$  (SBSC) electrode, solution for infiltration was prepared by weighting commercial  $\text{Sm}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ ,  $\text{Ba}(\text{NO}_3)_2$ ,  $\text{Sr}(\text{NO}_3)_2$ ,  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  (all of them are AR grade in purity) and dissolving them into deionized water. Citric acid was added and its molar ratio to the total metal ion was 1.2:1, the concentration of the solution was controlled to be around 0.5 mol  $\text{L}^{-1}$ . The solution was infiltrated into the porous LSGM layer, following by heat treatment at 500 °C for decomposition of the nitrates. This process was repeated several times until the final SBSC phase would be 20w% in the porous electrode. The previous work in our group shows that 21w% SBSC infiltrated LSGM skeleton gives the lowest polarization as a SOFC cathode [25]. The final heat treating temperature was selected as 900 °C (for 2 h), because samples treated independently at this temperature showed pure SBSC phase, while those treated at temperatures bellowing 850 °C showed XRD pattern of  $\text{SrCoO}_3$  as impurity.

For the preparation of Ni-SDC electrode, similar procedure was adopted, but the start materials were  $\text{Sm}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ ,  $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ , and  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  (also AR grade), the calculated Ni ratio in Ni/SDC mixture was 70w%, the weight ratio of infiltrated active substance in the final electrode was controlled to be 20 w%, and the final treating temperature of the infiltrated Ni/SDC mixture was selected as 850 °C.

The morphologies of the cell configuration and infiltrated particles were characterized by scanning electron microscope (SEM, Hitachi S-4800-2).

### Performance characterization

Initial performance of the cell was tested in both fuel-cell mode (FC mode) and electrolysis cell mode (EC mode). DC polarization (I–V) curves and Electrochemical impedance spectra (EIS) were measured at 450, 500, 550 and 600 °C, while a stream of  $\text{H}_2/\text{H}_2\text{O}$  mixture with different ratio (with the total flow rate being 100 sccm) was supplied to the Ni-SDC-LSGM hydrogen electrode and air supplied to the SBSC-LSGM oxygen electrode (flow rate 100 sccm).

Electrochemical impedance spectra (EIS) testing was performed using an IM6 Electrochemical Workstation (ZAHNER, Germany) between  $10^5$  and  $10^{-1}$  Hz, with a 20 mV perturbation under open-circuits conditions. The ohmic resistance ( $R_o$ ) was obtained from the value of the high frequency intercept. The electrode polarization resistances ( $R_p$ ) were measured by the differences between the high and low frequency intersections of the impedance spectrum on the real axis (Z real axis).

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