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Performance of $La_{0.1}Sr_{0.9}Co_{0.8}Fe_{0.2}O_{3-\delta}$ and $La_{0.1}Sr_{0.9}Co_{0.8}Fe_{0.2}O_{3-\delta}$ -Ce_{0.9}Gd_{0.1}O₂ oxygen electrodes with Ce_{0.9}Gd_{0.1}O₂ barrier layer in reversible solid oxide fuel cells



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HIGHLIGHTS

- Button cells with LSCF1982-based oxygen electrodes are tested for RSOCs.
- GDC barrier layer prevents reaction between LSCF1982 and YSZ.
- Button cells show stable galvanostatic SOEC operation for 72 h.
- Reversibility is tested by successive SOFC \rightarrow SOEC \rightarrow SOFC operations.
- Possible reasons of irreversibility are discussed.

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ABSTRACT

 $La_{0.1}Sr_{0.9}Co_{0.8}Fe_{0.2}O_{3-\delta} (LSCF1982) \text{ and } La_{0.1}Sr_{0.9}Co_{0.8}Fe_{0.2}O_{3-\delta} - Ce_{0.9}Gd_{0.1}O_2 (LSCF1982 - GDC) \text{ composite}$ oxygen electrodes with a GDC barrier layer are tested in yttria stabilized zirconia (YSZ) electrolyte-based reversible solid oxide cells (RSOCs). Three button cell assemblies (1: NiO-YSZ|YSZ/GDC|LSCF1982; 2: NiO -YSZ/NiO-YSZ/YSZ/GDC|LSCF1982; and 3: NiO-YSZ/NiO-YSZ/GDC|LSCF1982-GDC) are fabricated and their performance in solid oxide fuel cell (SOFC) and solid oxide electrolysis cell (SOEC) modes are studied at different temperatures (600 \leq *T*/°C \leq 800). The use of porous *NiO*–*YSZ* functional layer between hydrogen electrode and electrolyte leads to improvements in SOFC and SOEC (SOFC/SOEC) performance by improving the diffusion of reacting species inside the electrode. The effect of nature of oxygen electrode on SOFC/SOEC performance is studied, which indicates LSCF1982 oxygen electrode gives better performance than LSCF1982–GDC composite oxygen electrode, but LSCF1982–GDC composite oxygen electrode is more durable during reversible SOFC/SOEC operations. Stability of the button cells is studied in galvanostatic SOEC operation for 72 h and in reversible SOFC/SOEC operations. Current -voltage (I-V) tests and Electrochemical Impedance Spectroscopy (EIS) measurements indicate that the button cells show stable operation in SOEC mode. But successive SOFC \rightarrow SOEC \rightarrow SOFC operations indicate that the cells are not completely reversible. The possible reason for poor reversibility is found to be related with the grain growth in LSCF1982-based oxygen electrodes.

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1. Introduction

Reversible solid oxide cells capable of working as SOFC for electricity generation from fuel and as SOEC for fuel generation

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form electrolysis of steam and CO₂ have created increased interest in the field of development of environmentally friendly ways of energy storage and electricity generation [1,2]. A RSOC system has the potential to be used as an efficient electrical energy storage device at the power plants based on intermittent renewable energy sources and could provide effective and flexible power delivery, and its smaller modules, acting as a rechargeable battery, could play an important role in distributed power generation [3,4]. Also, the possibility of utilization of waste heat generated by high temperature industrial



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processes and excess electricity produced in off-peak hours at conventional power plants and renewable sources for steam electrolysis in SOEC has made this technology more attractive [5–7].

Although in principle SOEC is a reverse mode of SOFC as schematically represented in Fig. 1, individual modes presents somewhat different conditions at the electrodes during its operation. Therefore, the selection of suitable electrolyte and electrode materials compatible for stable operation in SOFC mode as well as in SOEC mode is important factor for designing a RSOC. YSZ and Ni-YSZ, respectively, have been widely used electrolyte and hydrogen electrode for SOEC and SOEC [6,8]. Though the composite of lanthanum strontium manganite with YSZ (LSM-YSZ) is widely used as oxygen electrode in SOEC, composites of other perovskite materials, such as lanthanum strontium ferrite (LSF), lanthanum strontium copper ferrite (LSCuF), lanthanum strontium cobalt ferrite (LSCoF), and lanthanum strontium cobaltite (LSCo), with YSZ can be potential material for oxygen electrode [1,9–13]. Since the energy losses due to electrode polarization, especially that of oxygen electrode, and degradation due to the delamination of oxygen electrode, have often been limiting factors in deciding the performance and practicability of SOECs, considerable efforts have been made to improve the performance of oxide electrode [6,14]. Because of their high catalytic activity and oxygen permeability, Co-based perovskite materials could be potential oxygen electrode in SOEC, but during the operation at high temperature they suffer from the drawback of reaction with YSZ electrolyte [15]. By applying a thin layer of another electrolyte, which does not reacts with YSZ at high temperature and, therefore, acts as a barrier layer to prevent the interfacial reaction between YSZ electrolyte and Co-based perovskite oxygen electrode, this drawback could be avoided [16].

In previous works, we have investigated electrochemical properties of LSCF1982 [17–20]. We have employed microwave heat-treated LSCF1982 as a cathode for ceria electrolyte-based intermediate temperature fuel cell and found that, at 650 °C, the fuel cell with microwave heat-treated cathode shows better performance than conventional heat treated cathode [20]. Based on our previous findings in ref. 20, it would be interesting to analyze the performance of LSCF1982 as oxygen electrode in RSOC at the temperatures $600 \le T/^{\circ}C \le 800$. But, like other Co-based perovskite materials, LSCF1982 suffers from the drawback of interfacial reaction between LSCF1982-based electrodes and YSZ electrolyte. Doped ceria-based materials show no reactivity with LSCF1982 and they can be used as electrolyte with LSCF1982-based



Fig. 1. Schematic representation of processes occurring in a RSOC during the operation in SOFC and SOEC modes.

electrodes, but their narrow electrolytic regime, because of partial reduction of Ce^{4+} to Ce^{3+} under reducing conditions, makes them less favorable [16,21,22]. Notwithstanding the above fact, doped ceria, such as GDC, can be used as a barrier layer to avoid interfacial reaction between LSCF1982-based oxygen electrode and YSZ electrolyte.

The optimization of electrode configuration and microstructure has been found to be quite beneficial for the performance of SOFCs. suggesting that such manipulations could be important factor for the RSOCs' performance as well [23]. The introduction of an electrode functional layer between an electrode and an electrolyte has shown to be effective for the improvement of cell performance. Similarly, the use of composite electrodes, having electrolyte as one of its constituents, help improving the stability of cell by enhancing the compatibility of electrolyte and electrode materials at their interface. In previous studies on single RSOCs, SOFC and SOEC modes have shown differences in performance, primarily because of differential behavior of cell components during SOFC/ SOEC modes [1,12,13,24,25]. Ni-YSZ fuel electrodes were found to be less active in SOEC mode than in SOFC mode due to the contribution of a diffusion-limited process during SOEC operation [24], and Ni-YSZ fuel electrodes were reported to be susceptible to aging in high-steam environment, because of coarsening of Ni particles [25]. O'Brien et al. [12] have demonstrated that a single cell with Ni-YSZ fuel electrode and LSM oxygen electrode showed small difference in the values of area specific resistance (ASR) in SOFC and SOEC modes. But a study by Eguchi et al. [13] showed a strong dependence of electrode polarization on the operation condition of SOFC and SOEC, and a cell with Ni-YSZ hydrogen electrode and LSM oxygen electrode resulted in small electrode polarization during SOFC operation whereas a cell with Pt hydrogen electrode and LSCo oxygen electrode showed small electrode polarization in SOEC condition. Also, they studied the reversibility of SOFC and SOEC as a function of the H₂O partial pressure (pH_2O) and H_2 partial pressure (pH_2) in the H_2-H_2O mixture and observed significant concentration overvoltage in the *I–V* characteristics of SOFC and SOEC operating in different *p*H₂O and *p*H₂ atmospheres. Marina et al. [1] compared the inefficiencies associated with electrode reactions during SOFC and SOEC operations using RSOCs with Ni-YSZ or La_{0.35}Sr_{0.65}TiO₃-Ce_{0.5}La_{0.5}O_{2-\delta} composite as hydrogen electrode and LSF, LSCuF, LSCoF or LSM as oxygen electrode.

Therefore, considering the above mentioned factors to improve solid oxide cell performance, in the present work, we design three different single cell configurations, primarily based on YSZ electrolyte with a GDC barrier layer, Ni–YSZ hydrogen electrode and LSCF1982 or LSCF1982–GDC composite oxygen electrode, as given below:

Button cell 1 NiO–YSZ|YSZ/GDC|LSCF1982

Button cell 2 NiO-YSZ/NiO-YSZ| YSZ/GDC|LSCF1982

Button cell 3 NiO-YSZ/NiO-YSZ| YSZ/GDC|LSCF1982-GDC

GDC layer is employed as a barrier to prevent the reaction between YSZ and LSCF1982 at the electrolyte/oxygen electrode interface. The introduction of a porous *NiO*–*YSZ* functional layer between the dense electrolyte and the hydrogen electrode substrate is to improve the diffusion of reactants inside the electrode. The comparative performances of these single cells as intermediate temperature RSOCs in SOFC mode and SOEC mode are analyzed with respect to variations in oxygen electrode composition, and hydrogen electrode configuration. *I*–*V* measurements and EIS are used for the electrochemical characterization of the cells. We investigate the performance stability of cells 2 and 3 during galvanostatic SOEC operation for 72 h. Also, the reversibility of the button cells is tested by successive SOFC \rightarrow SOEC \rightarrow SOFC operations. Download English Version:

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