



# Degradation factors in (La,Sr)(Co,Fe)O<sub>3-δ</sub> cathode/Sm<sub>2</sub>O<sub>3</sub>–CeO<sub>2</sub> interlayer/Y<sub>2</sub>O<sub>3</sub>–ZrO<sub>2</sub> electrolyte system during operation of solid oxide fuel cells



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## HIGHLIGHTS

- The microstructural change of the LSCF/SDC/YSZ system was analyzed quantitatively.
- The distribution profile of SrZrO<sub>3</sub> in the LSCF/SDC/YSZ system was revealed.
- The TPB length reduced significantly after discharge for 400 h at 1000 °C.
- The agglomeration of SDC and LSCF phases was also confirmed.
- The solid-state reaction between YSZ and SDC proceeded considerably.

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## ABSTRACT

In this study, the microstructural change of the LSCF/SDC/YSZ system upon discharge was analyzed quantitatively by the focused ion beam–scanning electron microscopy as well as the transmission electron microscopy. It is widely recognized that the formation of highly-resistive SrZrO<sub>3</sub> phase degrades the cell performance in this system. In fact, the ohmic loss and cathodic overpotential increased in response to the six-fold increase in the volume of SrZrO<sub>3</sub> phase after 400 h of discharge at 1000 °C. However, the microstructural change proceeded in the whole part of this system, indicating that various degradation factors need to be considered; e.g., 1) the agglomeration of SDC and LSCF phases, 2) the reduction in triple phase boundary length, and 3) the formation of ceria-zirconia solid solution.

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## 1. Introduction

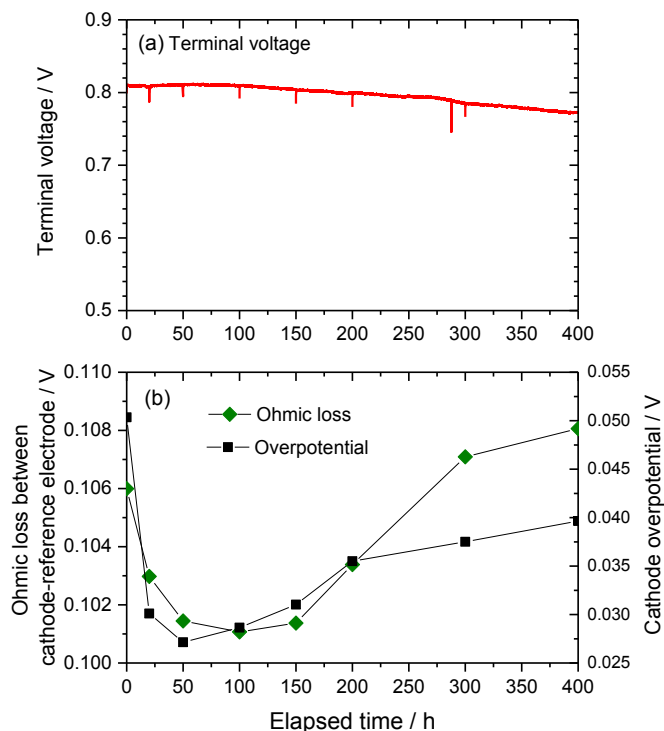
Solid oxide fuel cells (SOFCs) have attracted much attention because of their high energy conversion efficiency, high fuel flexibility, and low environmental load. The residential SOFC cogeneration system has already commercialized in Japan [1–3], but there are still some problems to be settled to ensure the durability and reliability of system.

Lanthanum strontium cobalt iron oxide, (La,Sr)(Co,Fe)O<sub>3-δ</sub> (LSCF), is widely used as a cathode for SOFCs since this material

exhibits high mixed ionic–electronic conductivity. However, LSCF is not chemically compatible with zirconia-based oxides due to the formation of highly-resistive phase of SrZrO<sub>3</sub> and La<sub>2</sub>Zr<sub>2</sub>O<sub>7</sub> [4]. Uchida et al. demonstrated that the insertion of samaria-doped ceria (SDC) interlayer is effective to prevent the solid-state reaction between LSCF cathode and yttria-stabilized zirconia (YSZ) electrolyte [5,6]. Since then, the doped ceria interlayer is widely adopted as a diffusion barrier for the state-of-the-art SOFC cells and stacks [7–14]. However, this procedure does not provide the problem-free solution for this matter. The porosity of barrier layer significantly affects the amount of SrZrO<sub>3</sub> formed as well as the cell performance [15–17]. The solid solution with low ionic conductivity is also produced via the solid-state reaction between doped ceria and zirconia-based oxide, mainly during the sintering step of

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**Fig. 1.** Time courses of (a) terminal voltage and (b) ohmic loss between cathode-reference electrode and cathode overpotential during discharge at  $0.3 \text{ A cm}^{-2}$  for 400 h. Operating temperature:  $1000 \text{ }^\circ\text{C}$ ;  $p(\text{O}_2)$ : 1 atm.

interlayer [18–22]. Although many studies have been conducted to make a strategy for the suppression of these phenomena, the formation mechanism and condition of  $\text{SrZrO}_3$  phase are not fully elucidated [23,24]. Furthermore, the quantitative impact of highly-resistive phases on performance is still under investigation.

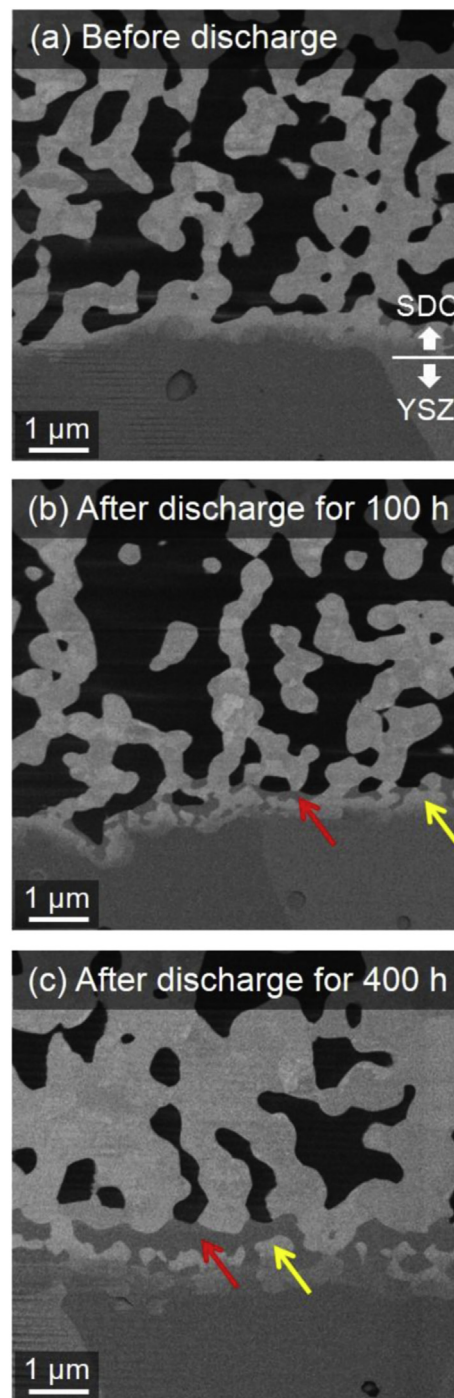
To provide new insights for these phenomena, in this study, the microstructural change of the LSCF/SDC/YSZ system upon discharge was quantified by using the focused ion beam–scanning electron microscope; the discharge operation was conducted at  $1000 \text{ }^\circ\text{C}$  to accelerate the microstructural change. The distribution profile of  $\text{SrZrO}_3$  phase in this cell configuration was visualized. The influence of passed current density on microstructural change as well as performance was also studied. The local structure at the SDC/YSZ interface, especially in the vicinity of  $\text{SrZrO}_3$  formation region, was observed in detail by using a transmission electron microscope.

## 2. Experimental

The electrolyte-supported cells were fabricated as follows. A disk of 8 mol% yttria-stabilized zirconia (abbreviated as YSZ, Tosoh, 24 mm in diameter, 500  $\mu\text{m}$  in thickness) was used as an electrolyte. The Ni–YSZ cermet anode with a volume ratio of 50/50 was prepared from NiO (Wako Pure Chemical Industries) and YSZ powder (8 mol%  $\text{Y}_2\text{O}_3$ – $\text{ZrO}_2$ , Tosoh). The mixture of NiO and YSZ was heat-treated at  $1200 \text{ }^\circ\text{C}$  for 5 h, and mixed with polyethylene glycol to form slurry. The resultant slurry of NiO–YSZ was screen-printed onto the electrolyte and subsequently fired at  $1400 \text{ }^\circ\text{C}$  for 5 h in air. The slurry of  $\text{Ce}_{0.8}\text{Sm}_{0.2}\text{O}_{1.9}$  (SDC, oxalate coprecipitation method [25]) was screen-printed on the other face of electrolyte, and then fired at  $1250 \text{ }^\circ\text{C}$  for 10 h in air. After that,  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_3$  (LSCF, Seimi Chemical) was applied as a cathode; the slurry was screen-printed and subsequently fired at  $1150 \text{ }^\circ\text{C}$  for 5 h in air.

The area of each electrode was  $0.28 \text{ cm}^2$ . The reference electrode of platinum wire was attached to surround the side edge of the YSZ disk and fixed by a platinum paste (N.E. Chemcat Co., U-3402).

The single cell was sandwiched by alumina tubes with Pyrex glass seal as depicted in our previous report [26]. Prior to electrochemical measurements, the anode was reduced under a hydrogen atmosphere at  $1000 \text{ }^\circ\text{C}$ . The gases of  $\text{O}_2$  and 3%  $\text{H}_2$ –97%  $\text{H}_2$  were supplied to the cathode and anode, respectively, with a flow rate of



**Fig. 2.** Backscattered electron images of SDC/YSZ interface (a) before and after discharge at  $0.3 \text{ A cm}^{-2}$  for (b) 100 h and (c) 400 h. Red arrow:  $\text{SrZrO}_3$ ; yellow arrow: SDC. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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