

Short communication

# Chemical compatibility and electrochemical property of intermediate-temperature SOFC cathodes under Cr poisoning condition

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## Abstract

We have investigated the relationship between the chemical compatibility and electrochemical properties of  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Fe}_{0.8}\text{Co}_{0.2}\text{O}_3$  (LSCF),  $\text{LaNi}_{0.6}\text{Fe}_{0.4}\text{O}_3$  (LNF), and  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$  (LSM) as a cathode against the Cr poisoning condition. Powder mixtures of LSCF– $\text{Cr}_2\text{O}_3$ , LSM– $\text{Cr}_2\text{O}_3$ , and LNF– $\text{Cr}_2\text{O}_3$  were heated at 1073 K and analyzed by X-ray powder diffraction with the Rietveld refinement. It was found that LNF powder was less reactive with  $\text{Cr}_2\text{O}_3$  than LSCF and LSM powder from the viewpoint of the consumption of  $\text{Cr}_2\text{O}_3$  in the mixtures. From electrochemical measurement, it was found that the cathodic overvoltage was almost unchanged for cells with LNF cathode, either in the presence or absence of a Cr-containing alloy. On the other hand, the cells with LSCF and LSM cathode in the presence of the alloy exhibited a steep increase in the overvoltage curve. These results show that LNF cathode is more stable against Cr poisoning than the other two cathodes. Therefore, we expect LNF to be a long-life cathode with high stability against Cr poisoning in solid oxide fuel cell because of the low reactivity of LNF with  $\text{Cr}_2\text{O}_3$ . © 2007 Elsevier B.V. All rights reserved.

**Keywords:** SOFC; Cathode; Cr;  $\text{LaNi}_{0.6}\text{Fe}_{0.4}\text{O}_3$ ; LNF; Solid oxide fuel cell

## 1. Introduction

Intermediate-temperature solid oxide fuel cells (SOFCs) operated at around 1073 K have recently attracted attention as high-efficiency and low-cost power generation systems because metallic alloys can be used as interconnects. However, when applying the alloys, there is a problem of poisoning of the cathode with chromium. Vapor chromium species was scattered from the alloys to the cathode at SOFC operation temperatures. The Cr species tend to deposit at the three phase boundaries at the cathode/electrolyte/gas interface, resulting in the rapid degradation of cathode activity [1–3]. This phenomenon is called Cr poisoning. The mechanism of Cr poisoning has not been understood yet.

We have examined LNF ( $\text{LaNi}_{0.6}\text{Fe}_{0.4}\text{O}_3$ ) as a cathode material for intermediate-temperature SOFCs [4,5]. LNF is found to have a high electrical conductivity ( $580 \text{ S cm}^{-1}$  at 1073 K) and its thermal expansion coefficient ( $11 \times 10^{-6} \text{ K}^{-1}$ ) is close to that of zirconia-related materials [4]. A cell using LNF cathode with platinum current collectors shows high-performance electric power generation with the maximum power density of  $1.56 \text{ W cm}^{-2}$  at 1073 K [5]. Furthermore, it has been demonstrated that the cells with LNF cathodes are more stable against Cr poisoning than those with LSM cathodes [6]. LNF is less reactive with  $\text{Cr}_2\text{O}_3$  than LSM during heating time 0–1000 h at 1073 K [7]. It was thus demonstrated that the cells with LNF cathodes are more stable against Cr poisoning than those with  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$  (LSM) cathodes.

Therefore, we believe that LNF is a promising material for the intermediate-temperature SOFC cathode. On the other hand  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Fe}_{0.8}\text{Co}_{0.2}\text{O}_3$  (LSCF) is also attracting attention as cathode material of the intermediate-temperature SOFC because it has high mixed electronic and ionic conductivities [8], with high catalytic activity for the  $\text{O}_2$  reduction reaction [9,10].

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Therefore, in this work, we examined the chemical stability of LSCF as well as that of LSM and LNF in the presence of  $\text{Cr}_2\text{O}_3$  at 1073 K for 0–1000 h by means of X-ray diffraction measurements and a Rietveld refinement analysis. We also investigated the influence of Cr poisoning in electrochemical cells using these cathode materials near Cr-containing alloy.

## 2. Experimental

### 2.1. Sample preparation

For the evaluation of the chemical compatibility of cathode materials against  $\text{Cr}_2\text{O}_3$ ,  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Fe}_{0.8}\text{Co}_{0.2}\text{O}_3$  (LSCF, Powlex, Japan) powder,  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$  powder (LSM, Seimi Chemical, Japan), or  $\text{LaNi}_{0.6}\text{Fe}_{0.4}\text{O}_3$  (LNF, Powlex, Japan) powder was mixed with the  $\text{Cr}_2\text{O}_3$  (Kojundo Chemical Laboratory, Japan) powder. The particle size for these kinds of powder was 1.0–1.3  $\mu\text{m}$ . And the powders were thoroughly mixed with the mortar. The weight ratio of cathode material to  $\text{Cr}_2\text{O}_3$  in the mixed powder was 10:1. These mixtures were heated at 1073 K in ambient air for 100, 200, 400, 800, and 1000 h. The heating/cooling rate was 100  $\text{K h}^{-1}$ .

### 2.2. X-ray diffraction measurements and Rietveld refinement analysis

XRD patterns of the heated mixtures were measured with an XRD instrument (RIGAKU, RINT2000) powered at 30 kV and 100 mA using  $\text{Cu K}\alpha$  radiation in air at room temperature. The diffraction intensity was measured stepwise every  $0.02^\circ$  in the diffraction angle  $2\theta$  range between  $10^\circ$  and  $70^\circ$ . Phases were identified from the measured XRD pattern using the International Center for Diffraction Data (ICDD) database. In Rietveld analysis (RIQAS, Materials Data Inc. Company), assuming that there was no product between  $\text{Cr}_2\text{O}_3$  and cathode materials as an initial condition, the fitting of the measurement value and the calculation value on these patterns was done. Then the background, the molar ratio and lattice constant in each phase, etc. were optimized.

### 2.3. Cell fabrication

Electrochemical measurements were carried out using the electrolyte-supported cells. Either 8 mol% yttria-stabilized zirconia (YSZ, Japan Fine Ceramics, Japan) or 10 mol%  $\text{Sc}_2\text{O}_3$ - and 1 mol%  $\text{Al}_2\text{O}_3$ -stabilized  $\text{ZrO}_2$  (SASZ, Tosoh, Japan) were used for the electrolyte. These electrolyte disks are 35 mm in diameter and 0.3-mm thick. A mixture of 60 wt% NiO and 40 wt% SASZ was used for the anode and painted on one side of the electrolyte disks to form porous electrodes with an area of  $0.785\text{ cm}^2$ . After that, the anode was fired at 1573 K for 2 h. As the cathodes, LSCF, LSM or LNF was used. The cathode area was  $0.785\text{ cm}^2$ . It is placed symmetrical to the anode. Cathode paste was printed on the electrolyte surface of the half-cells and sintered at 1273 K for 4 h, 1273 K for 4 h and 1573 K for 2 h for the LSCF, LNF, and LSM cathodes, respectively. When the LSCF cathode was fabricated, a GDC (gadolinium-doped

ceria,  $80\text{CeO}_2\text{--}20\text{GdO}_{1.5}$ ) buffer layer was inserted between it and the electrolyte to prevent unfavorable solid-state reactions at the LSCF/SASZ electrolyte interface [9]. Pt paste was used as the reference electrode and was painted as ring on the cathode side. The gap between the cathode and reference electrodes was about 10 mm.

### 2.4. Electrochemical measurement

Tested cells were operated at 1073 K. The structure of the equipment for the electrochemical measurements has been previously described [6]. Dry air was fed to the cathode, and  $\text{H}_2$  gas to the anode. A Pt mesh and Pt wires were used as current collectors for the cathode, anode, and reference electrode. And dimensions of chamber in which the cells were placed were  $60\text{ mm} \times 60\text{ mm} \times 250\text{ mm}$ . For the investigation of Cr poisoning, a Cr-containing alloy mesh (Inconel 600 mesh) ( $20\text{ mm} \times 20\text{ mm} \times 0.5\text{ mm}$ ) was placed directly on the Pt printed cathode electrode. The current density was kept at a constant value to measure the time dependence of the cathodic overvoltage. We measured the cathode electrode-reference voltage, and estimated the cathodic overvoltage by eliminating the ohmic loss (IR value) from the voltage. The IR value was estimated by the galvanostatic current interruption method. For this measurement, a current pulse generator (Hokuto Denko, HC-113) was used.

### 2.5. Characterization

The Cr distribution in the cathode was observed with an electron probe microanalyzer (EPMA) (JEOL, JXA-8621R).

## 3. Results and discussion

### 3.1. Chemical compatibility against $\text{Cr}_2\text{O}_3$ of LSCF, LNF, LSM

The XRD patterns of LSCF– $\text{Cr}_2\text{O}_3$ , LSM– $\text{Cr}_2\text{O}_3$ , LNF– $\text{Cr}_2\text{O}_3$  mixtures that had been heated at 1073 K in air for 0–1000 h were measured by XRD. From the XRD pattern of the LSCF– $\text{Cr}_2\text{O}_3$  mixture (Fig. 1),  $\text{SrCrO}_4$  (ICDD:35-0743), a spinel phase, and a  $\text{A}_2\text{O}_3$  phase like  $(\text{Fe,Cr})_2\text{O}_3$  (ICDD:02-1357) or  $\text{Fe}_2\text{O}_3$  (ICDD:73-2234) were identified in addition to the raw materials, LSCF (similar  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Fe}_{0.2}\text{Co}_{0.8}\text{O}_3$  (ICDD:48-0124) to  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Fe}_{0.8}\text{Co}_{0.2}\text{O}_3$ ], and  $\text{Cr}_2\text{O}_3$  (ICDD:38-1479). The spinel phase is attributed to  $\text{CoCr}_2\text{O}_4$  (ICDD:78-0711),  $\text{FeCr}_2\text{O}_4$  (ICDD:89-2618), or  $\text{CoFe}_2\text{O}_4$  (ICDD:22-1086), but they cannot be distinguished from one another because their lattice constants are nearly the same. In the case of the LSCF– $\text{Cr}_2\text{O}_3$  mixture,  $\text{CoCr}_2\text{O}_4$  (ICDD:78-0711) was used as a representative spinel phase. For the same reason,  $(\text{Fe,Cr})_2\text{O}_3$  (ICDD:02-1357) was used as a representative  $\text{A}_2\text{O}_3$  phase. From the XRD pattern of LSM– $\text{Cr}_2\text{O}_3$  mixture (Fig. 2), a  $\text{MnCr}_2\text{O}_4$  spinel phase (ICDD:75-1614) was identified in addition to the raw materials, LSM (ICDD:53-0058) and  $\text{Cr}_2\text{O}_3$ . From the XRD pattern of LNF– $\text{Cr}_2\text{O}_3$  mixture (Fig. 3), a  $\text{NiCr}_2\text{O}_4$  spinel phase

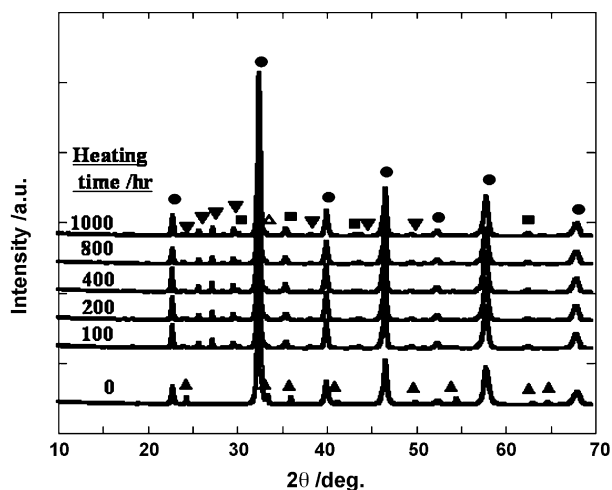


Fig. 1. XRD patterns of LSCF–Cr<sub>2</sub>O<sub>3</sub> mixture powder during heating at 1073 K for 0–1000 h: (●) LSCF, (▲) Cr<sub>2</sub>O<sub>3</sub>, (▼) SrCrO<sub>4</sub>, (■) CoCr<sub>2</sub>O<sub>4</sub> spinel, (△) (Fe,Cr)<sub>2</sub>O<sub>3</sub>.

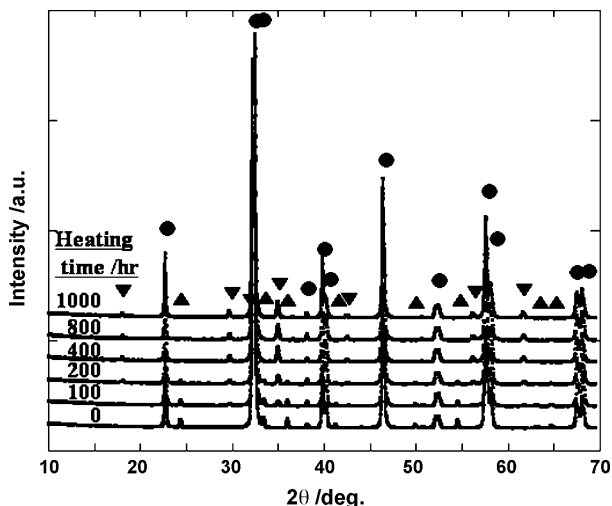


Fig. 2. XRD patterns of LSM–Cr<sub>2</sub>O<sub>3</sub> mixture powder during heating at 1073 K for 0–1000 h: (●) LSM, (▲) Cr<sub>2</sub>O<sub>3</sub>, (▼) MnCr<sub>2</sub>O<sub>4</sub> spinel.

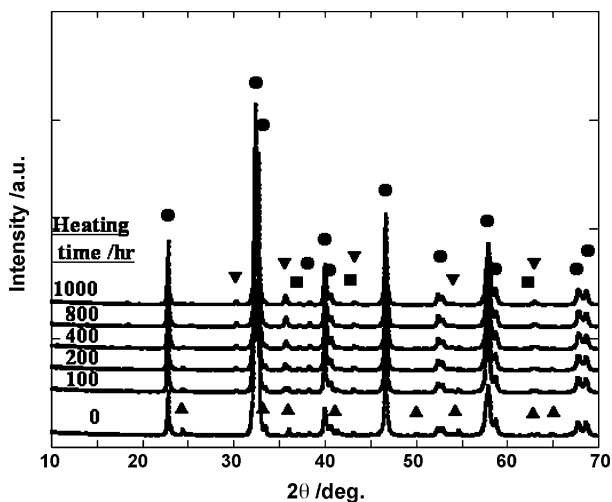


Fig. 3. XRD patterns of LNF–Cr<sub>2</sub>O<sub>3</sub> mixture powder during heating at 1073 K for 0–1000 h: (●) LNF, (■) NiO, (▲) Cr<sub>2</sub>O<sub>3</sub>, (▼) NiCr<sub>2</sub>O<sub>4</sub> spinel.

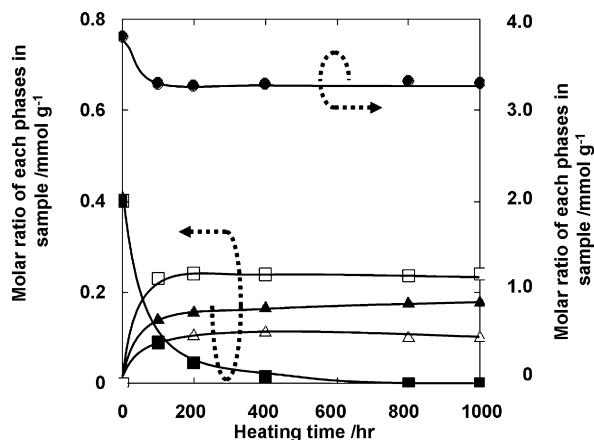


Fig. 4. Molar ratio of phases in LSCF–Cr<sub>2</sub>O<sub>3</sub> mixture during heating at 1073 K for 0–1000 h: (●) LSCF, (■) Cr<sub>2</sub>O<sub>3</sub>, (□) SrCrO<sub>4</sub>, (▲) CoCr<sub>2</sub>O<sub>4</sub> spinel, (△) (Fe,Cr)<sub>2</sub>O<sub>3</sub>.

(ICDD:75-0198) was identified in addition to the raw materials, LNF (ICDD:88-0637), NiO (ICDD:89-7131) (which initially existed in LNF as an impurity) and Cr<sub>2</sub>O<sub>3</sub>.

Assuming that only the identified phases existed in each mixture, the XRD patterns were analyzed by means of the Rietveld method. The Rietveld refinement gave reliability factor  $R_{wp}$  values between 13 and 17 for the LNF–Cr<sub>2</sub>O<sub>3</sub> mixture, 11 and 13 for the LSM–Cr<sub>2</sub>O<sub>3</sub>, 15 and 17 for the LSCF–Cr<sub>2</sub>O<sub>3</sub>. We believe that the  $R_{wp}$  values are acceptable as they are similar to the values obtained by Fálcon et al. in their study of the crystal structure of cathode material by Rietveld refinement [11].

Fig. 4 shows the molar ratio of LSCF, Cr<sub>2</sub>O<sub>3</sub>, SrCr<sub>2</sub>O<sub>4</sub>, CoCr<sub>2</sub>O<sub>4</sub> and (Fe,Cr)<sub>2</sub>O<sub>3</sub> in the LSCF–Cr<sub>2</sub>O<sub>3</sub> mixture as a function of heating time. The molar ratio of the reaction products (SrCr<sub>2</sub>O<sub>4</sub>, CoCr<sub>2</sub>O<sub>4</sub> and (Fe,Cr)<sub>2</sub>O<sub>3</sub>) increased, while that of Cr<sub>2</sub>O<sub>3</sub> decreased, suggesting that the reaction products were caused by the reaction between LSCF and Cr<sub>2</sub>O<sub>3</sub>. It was found that no Cr<sub>2</sub>O<sub>3</sub> existed in the mixture after it had been heated for 600 h.

Fig. 5 shows the molar ratio of LSM, Cr<sub>2</sub>O<sub>3</sub>, and MnCr<sub>2</sub>O<sub>4</sub> in the LSM–Cr<sub>2</sub>O<sub>3</sub> mixture as a function of heating time. The molar ratio of MnCr<sub>2</sub>O<sub>4</sub> increased, while that of Cr<sub>2</sub>O<sub>3</sub> decreased,

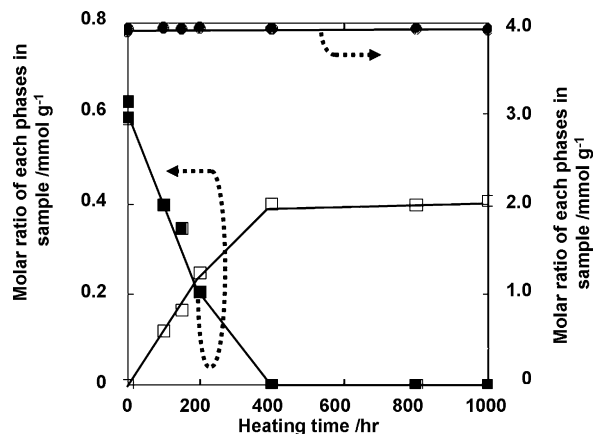


Fig. 5. Molar ratio of phases in LSM–Cr<sub>2</sub>O<sub>3</sub> mixture during heating at 1073 K for 0–1000 h: (●) LSM, (■) Cr<sub>2</sub>O<sub>3</sub>, (□) MnCr<sub>2</sub>O<sub>4</sub> spinel.

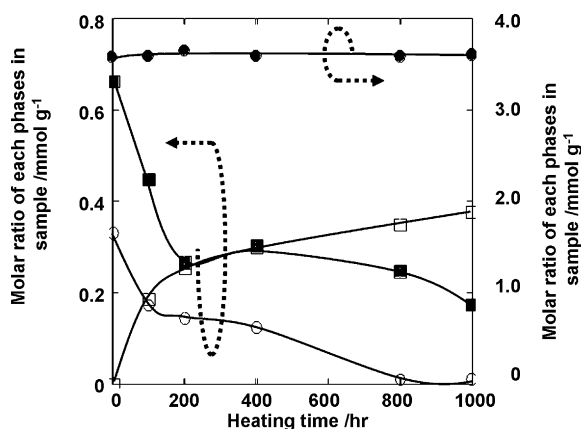


Fig. 6. Molar ratio of phases in LNF–Cr<sub>2</sub>O<sub>3</sub> mixture during heating at 1073 K for 0–1000 h: (●) LNF, (○) NiO, (■) Cr<sub>2</sub>O<sub>3</sub>, (□) NiCr<sub>2</sub>O<sub>4</sub> spinel.

suggesting that MnCr<sub>2</sub>O<sub>4</sub> was caused by the reaction between LSM and Cr<sub>2</sub>O<sub>3</sub>. No Cr<sub>2</sub>O<sub>3</sub> existed in the mixture after it had been heated for 400 h.

Fig. 6 shows the molar ratio of LNF, NiO, Cr<sub>2</sub>O<sub>3</sub>, and NiCr<sub>2</sub>O<sub>4</sub> in the LNF–Cr<sub>2</sub>O<sub>3</sub> mixture as a function of heating time at 1073 K. The molar ratio of LNF nearly unchanged. On the other hand, the molar ratio of NiCr<sub>2</sub>O<sub>4</sub> increased, while that of NiO and Cr<sub>2</sub>O<sub>3</sub> decreased, suggesting that NiCr<sub>2</sub>O<sub>4</sub> was mainly caused by the reaction between NiO and Cr<sub>2</sub>O<sub>3</sub>. It was found that unreacted Cr<sub>2</sub>O<sub>3</sub> still existed in the mixture after the mixture had been heated for 1000 h. From the viewpoint of the consumption of Cr<sub>2</sub>O<sub>3</sub> in the mixtures, the results suggest that LNF powder was less reactive with Cr<sub>2</sub>O<sub>3</sub> than LSCF powder and LSM powder.

The heating time dependence of the refined lattice constants for perovskite phases in each mixture was shown in Fig. 7. The lattice constants of each perovskite phases were nearly unchanged with time.

Here, we discuss the different reactivity of three perovskite compounds with chromia Cr<sub>2</sub>O<sub>3</sub>. LNF differs from LSCF and LSM in two ways: (i) the A site of the perovskite ABO<sub>3</sub> structure is completely filled with lanthanum and there is no strontium and (ii) the B site consists of nickel and iron, which are stable in divalent or trivalent. It appears that LSCF and LNF form oxygen-

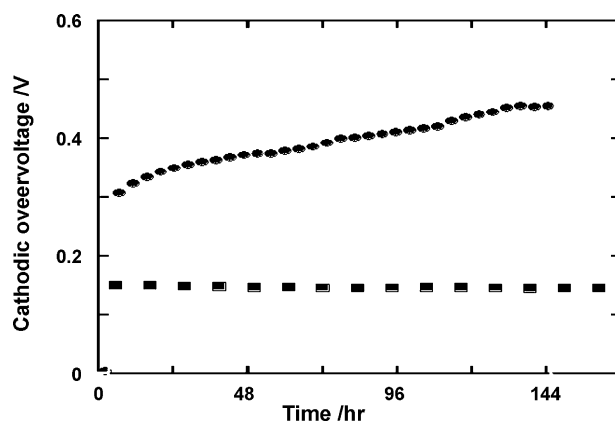


Fig. 8. Cathodic overvoltage (IR corrected) for cells using LSM cathode as a function of time: (●) cell with Inconel 600 at 0.076 A cm<sup>-2</sup> and (■) cell without Inconel 600 at 0.5 A cm<sup>-2</sup>.

deficient compounds with  $d$  in ABO<sub>3-d</sub> being smaller than 3, while LSM has excess oxygen, which would seem to correspond to the facts that Sr<sup>2+</sup> from LSCF, Ni<sup>2+</sup> from LNF, and Mn<sup>4+</sup> from LSM is reactive with chromia. As LNF is relatively tolerant to chromia, it seems that the oxygen deficiency of LNF is small or its strontium-free character contributes to the stability. For further discussion, detailed information is required on the non-stoichiometry of the perovskite compounds, the relative stability of the oxide [12], and the reaction kinetics.

### 3.2. Evaluation of electrochemical properties for cathode under Cr poisoning

We investigated the influence of Cr poisoning on various cells, consisting of LSM, LSCF, or LNF as a cathode. Fig. 8 shows cathodic overvoltage as a function of time for a LSM/YSZ/NiO-YSZ cell with/without Inconel 600. For the sample with Inconel 600, the cathodic voltage increased greatly at a low current density, so that we kept the current density at a low level to measure the time dependence of cathodic overvoltage. The current densities were 0.076 A cm<sup>-2</sup> for the cell with Inconel 600. When tested without Inconel 600, the cathodic overvoltage of this cell did not change greatly with time, com-

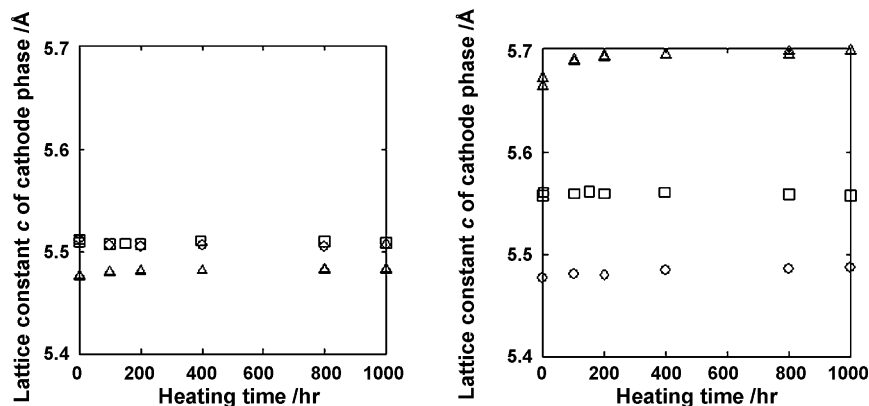


Fig. 7. Lattice constants of the LSCF, LNF, and LSM phases during heating at 1073 K for 0–1000 h: (Δ) LSCF, (○) LNF, (□) LSM.

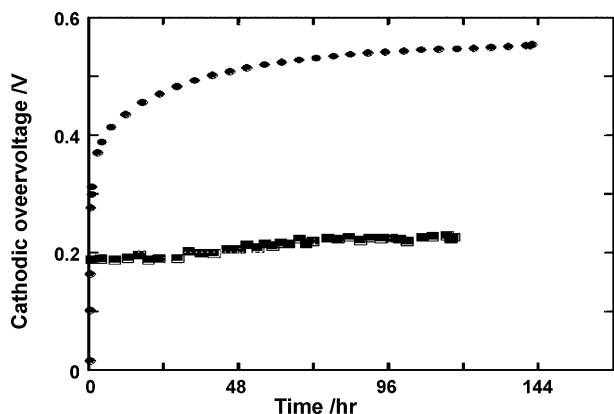


Fig. 9. Cathodic overvoltage (IR corrected) for cells using LSCF cathode as a function of time: (●) cell with Inconel 600 at  $0.7 \text{ A cm}^{-2}$  and (■) cell without Inconel 600 at  $0.7 \text{ A cm}^{-2}$ .

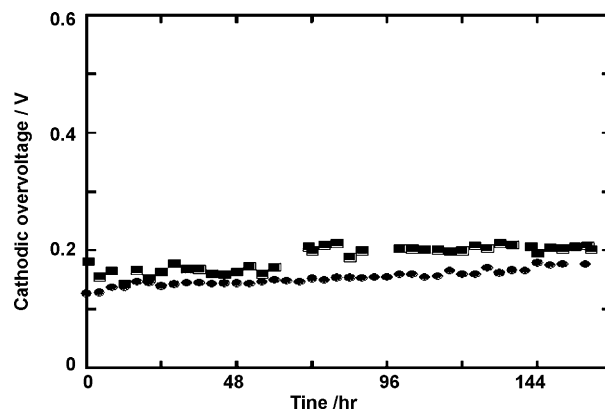


Fig. 10. Cathodic overvoltage (IR corrected) for cells using LNF cathode as a function of time: (●) cell with Inconel 600 at  $0.7 \text{ A cm}^{-2}$  and (■) cell without Inconel 600 at  $0.7 \text{ A cm}^{-2}$ .

pared with that of with Inconel 600. Fig. 9 shows cathodic overvoltage as a function of time for a LSCF/SASZ/NiO-YSZ cell with/without Inconel 600. For both cases, the current densities were fixed at constant values of  $0.7 \text{ A cm}^{-2}$ . However, a different profile of the cathodic overvoltage was obtained for cells using LSCF cathode with/without Inconel 600. When the cell was examined without Inconel 600, the overvoltage was low, about 0.2 V. However, the profile for the cell with Inconel 600 was close to that for the cell using LSM with Inconel 600, though the current density was much larger than that for the LSM cathode. It was thus demonstrated that the LSCF cathode

shows high activity in the absence of chromium species, but it degrades in the presence of it. Fig. 10 shows the cathodic overvoltage as a function of time for a LNF/SASZ/NiO-YSZ cell with/without Inconel 600. The current density was fixed at a constant value of  $0.7 \text{ A cm}^{-2}$ . A similar profile of the cathodic overvoltage was obtained at relatively high current densities for cells using LNF cathode with/without Inconel 600. That means that cathodic overvoltage was almost the same for cells with LNF cathodes, either in the absence or presence of Inconel 600. The above results indicate that the LNF cathodes are more stable against Cr poisoning than the other two cathodes.

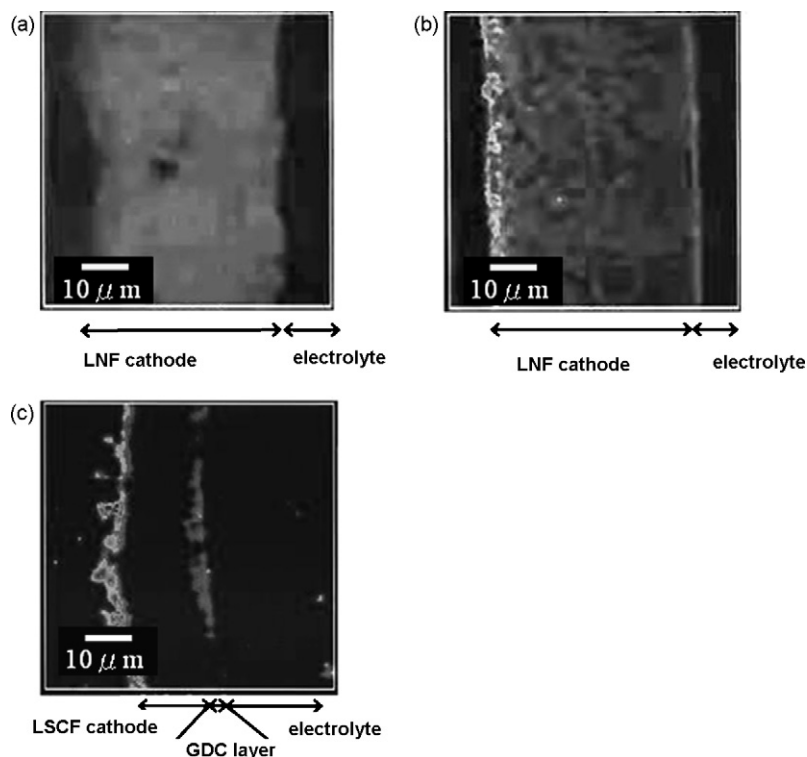


Fig. 11. Chromium distribution determined by EPMA near the interface of the cathode electrode and electrolyte after cell tests with Inconel 600: (a) cell using LNF cathode; (b) cell using LSM cathode; (c) cell using LSCF cathode.



### 3.3. Observation of cathode microstructure by EPMA

Fig. 11 shows the cross-sectional EPMA mapping of Cr around the cathode for the measured cell in the presence of Inconel 600. As shown in Fig. 11(a)–(c), the distribution of Cr could be observed for each cathode layer. As shown in Fig. 11(a), Cr was observed all over in the cathode layer. On the other hand, as shown in Fig. 11(b) and (c), for cells using the LSM and LSCF cathode, Cr was particularly observed near the cathode/electrolyte interface and on the whole cathode layer as well. Fujita et al. have observed chromium concentration near the cathode/electrolyte interface for the cell degraded by Cr poisoning [13]. Assuming that the deposited Cr at around the cathode/electrolyte interface reacts with a part of cathode like an active site, reaction products might play a dominant role in inhibiting electrochemical reaction process such as the dissociative adsorption and movement to the TPB (triple phase boundary) for oxygen molecules or in the loss of the TPB. Therefore, we expect LNF cathode to have a high stability against Cr poisoning due to the low reactivity of LNF with  $\text{Cr}_2\text{O}_3$ .

### 4. Conclusions

We investigated chemical and electrochemical characteristics of cathode materials (LSCF, LSM, LNF) for intermediate-temperature SOFCs. The LNF– $\text{Cr}_2\text{O}_3$ , LSM– $\text{Cr}_2\text{O}_3$ , and LSCF– $\text{Cr}_2\text{O}_3$  mixtures were heated at 1073 K and are analyzed by X-ray powder diffraction with the Rietveld refinement. For the LSCF– $\text{Cr}_2\text{O}_3$  mixture, LSCF reacted with  $\text{Cr}_2\text{O}_3$  to form  $\text{SrCr}_2\text{O}_4$ ,  $\text{CoCr}_2\text{O}_4$  and  $(\text{Fe,Cr})_2\text{O}_3$  and  $\text{Cr}_2\text{O}_3$  was depleted after heating for 600 h. LSM reacted with  $\text{Cr}_2\text{O}_3$  to form  $\text{MnCr}_2\text{O}_4$  to consume all  $\text{Cr}_2\text{O}_3$  in 400 h. LNF had less reactivity with  $\text{Cr}_2\text{O}_3$  and  $\text{Cr}_2\text{O}_3$  still existed in the mixture after heating for 1000 h. From the viewpoint of the consumption of  $\text{Cr}_2\text{O}_3$  in the mixtures, LNF was less reactive with  $\text{Cr}_2\text{O}_3$  than the other two.

We also examined the effect of Cr poisoning on cells consisting of LSCF, LSM, or LNF as a cathode. We found that the

cathodic overvoltage for the cell with LSCF cathode in the presence of Inconel 600 increased. For the cell with LNF cathode either in the presence or absence of Inconel 600, the cathodic overvoltage is almost the same. And for the cell with LSM cathode in the presence of Inconel 600, the cathodic overvoltage increased considerably even at low current density.

From these results, we think that LSCF is a promising material as a high-activity cathode for intermediate-temperature SOFCs. However when metallic alloys are used as their interconnects, the alloy interconnects might be coated to prevent Cr species vaporizing from them. On the other hand, LNF is attractive because in addition to its high electrochemical property as a cathode, the metallic alloy interconnects can be used without coating.

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