

Characterization of Cathode/Electrolyte Interfacial Impurities in a Solid Oxide Fuel Cell by Soft X-Ray Spectroscopy

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INTRODUCTION

Solid Oxide Fuel Cells (SOFC) have been expected to be clean and high energy conversion power sources [1, 2]. From the viewpoint of SOFC fabrication, control of the electrodes/electrolyte interfaces could be critical because interface impurities generally cause interfacial resistance and decrease the stability of SOFCs. In the interface of $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ (LSM) cathode and ZrO_2 (8-mol% stabilized Y_2O_3) (YSZ) electrolyte, it has been suggested from x-ray diffraction (XRD) measurements that $\text{La}_2\text{Zr}_2\text{O}_7$ may be formed in the annealed LSM/YSZ interface [3]. However, the interfacial impurities have not been clearly identified because XRD provides only information about crystal structure, and none about chemical/electronic states.

Soft x-ray emission/absorption spectroscopy is a powerful tool for determining the chemical/electronic states of materials. It can be used not only for conductive materials but also for insulators, because the x-ray emission/absorption process is generally unaffected by static electricity on the materials. Therefore, soft x-ray emission/absorption spectroscopy has great potential for characterizing SOFC materials. This paper describes the characterization of interfacial impurities in LSM/YSZ by soft x-ray emission/absorption spectroscopy.

EXPERIMENTAL

The interfacial sample was made by the annealing a mixture of LSM and YSZ powder at 1000 °C. Powder samples of LSM, YSZ, and $\text{La}_2\text{Zr}_2\text{O}_7$ were also prepared as references. Soft x-ray spectral measurements of these samples were carried at BL-8.0 [4] in ALS for emission measurements and BL-6.3.2 [5] for absorption measurements with a total electron yield method. We used the OK absorption edge because OK emission/absorption spectra may reflect the environmental chemical states more strongly than ZrL or M spectra.

RESULTS AND DISCUSSION

Before x-ray absorption measurements, OK x-ray emission spectra of the interfacial sample, YSZ, LSM, and $\text{La}_2\text{Zr}_2\text{O}_7$ were measured. Although energy shifts of the emission peaks were clearly observed among these samples, it may be difficult to identify the interfacial species because the individual spectral shapes were broad with FWHM of 3.5 eV.

Figure 1 (a) shows the OK x-ray absorption spectra of the interfacial sample, YSZ, LSM, and $\text{La}_2\text{Zr}_2\text{O}_7$. The spectral shape of LSM (manganese oxide compound) is quite different from those of YSZ and $\text{La}_2\text{Zr}_2\text{O}_7$ (zirconium oxide compounds). Even in these Zr compounds, energy shifts can be observed in the sharp peaks (assigned B, C, D, and E in the figure) between YSZ and $\text{La}_2\text{Zr}_2\text{O}_7$. Therefore, we may analyze the interfacial species from these absorption spectra to see whether $\text{La}_2\text{Zr}_2\text{O}_7$ is formed in the LSM/YSZ interface. Figure 1 (b) shows the absorption spectrum of the interfacial sample minus the LSM fraction: the LSM spectrum was subtracted from the original interfacial sample spectrum up to the disappearance of the 529-eV peak (assigned A) in the original spectrum. Comparing the spectral shape of

YSZ and $\text{La}_2\text{Zr}_2\text{O}_7$, we see that the interfacial sample spectrum with the LSM fraction subtracted looks like a mixture of YSZ and $\text{La}_2\text{Zr}_2\text{O}_7$. To confirm this, we compared the interfacial sample spectrum with the synthesized spectra of YSZ and $\text{La}_2\text{Zr}_2\text{O}_7$ in Figure 2. The fractions of YSZ and $\text{La}_2\text{Zr}_2\text{O}_7$ were varied from 0% to 100 % in a 10% steps in the synthesized spectra. Spectral shape and individual peak energy of the interfacial sample with LSM fraction subtracted agree with those of the synthesized spectra of YSZ: $\text{La}_2\text{Zr}_2\text{O}_7$ = 0.6:0.4. This confirms that $\text{La}_2\text{Zr}_2\text{O}_7$ is formed in the LSM/YSZ interface, which agrees with the XRD analysis [3].

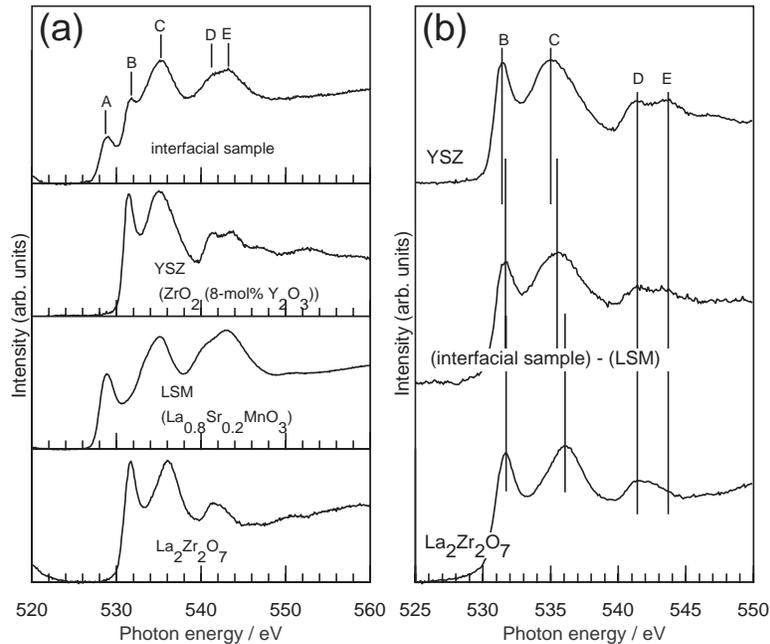


Fig. 1 (a) OK x-ray absorption spectra of LSM/YSZ interfacial sample, YSZ, LSM, and $\text{La}_2\text{Zr}_2\text{O}_7$. (b) Absorption spectrum of the interfacial sample minus the LSM fraction. Spectra of YSZ and $\text{La}_2\text{Zr}_2\text{O}_7$ are also shown as references.

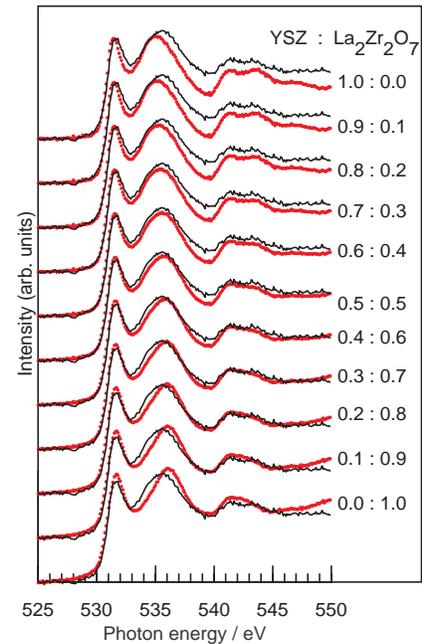


Fig. 2 Comparison of the interfacial sample spectra with the LSM fraction subtracted (solid line) with the synthesized spectra of YSZ and $\text{La}_2\text{Zr}_2\text{O}_7$ (dotted line).

CONCLUSION

Chemical species in a 1000-°C annealed LSM/YSZ interface were analyzed from OK x-ray absorption measurements using $\text{La}_2\text{Zr}_2\text{O}_7$ as a reference sample. Since the spectral shape of the interfacial sample agreed with the mixture of LSM, YSZ, and $\text{La}_2\text{Zr}_2\text{O}_7$, we conclude that $\text{La}_2\text{Zr}_2\text{O}_7$ is formed in the annealed LSM/YSZ interface. This indicates that suppressing the formation of $\text{La}_2\text{Zr}_2\text{O}_7$ in the LSM/YSZ interface is a critical point in advanced SOFC fabrication.

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