

Properties of Pulsed Laser Deposited $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_{3-\delta}$ - $\text{Ce}_{0.9}\text{Gd}_{0.1}\text{O}_{2-\delta}$ Nano-Composite Cathodes Depending on the Ratio of LSC:GDC

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Lanthanum strontium cobaltite (LSC) is a high performance cathode material of solid oxide fuel cells (SOFCs) at lower operating temperatures. However, successful application of LSC as a SOFC cathode is challenging because it has a high thermal expansion coefficient (TEC) mismatch with general electrolyte materials of the SOFC.[1] To improve this, composite materials which consist of cathode materials and electrolyte materials are used as the electrode.

Several studies on SOFC cathodes have highlighted the benefit of the composite cathode over a single phase cathode. First, composite cathodes are compatible both chemically and physically with electrolytes, thus there would be minimized reaction and defect generation at the electrolyte and the cathode interface. In addition, TEC of the cathode can be controlled, depending on the mixing ratio of the composite material. When the thermal expansion mismatch is minimized, the adhesion of the cathode to the electrolyte would be improved. Moreover, cell performance can be improved by reducing the resistance by expanding the area of contact of the electrolyte and the cathode and reducing the diffusion path within cathode.[2-4]

The benefit of using composite cathodes applies to thin-film-processed cathodes as well. In the previous study, fabricating a LSC-gadolinia-doped ceria (GDC) (5:5 volume ratios) nano-composite thin-film cathode by pulsed laser deposition (PLD) was successfully implemented.[5] The defects due to the TEC mismatch were suppressed, like composite cathodes fabricated by the powder process. There is, however, still lack of the research on the properties of the nano-composite thin-film cathodes in terms of the composition. Therefore, in this study, in-depth analyses of the thin-film-processed composite cathode were attempted. LSC-GDC nano-composite cathodes were fabricated at various mixing ratios (3:7, 5:5, 7:3) by using PLD. The physical properties and the electrochemical properties were investigated as a function of the mixing ratio.

LSC-GDC targets of various mixing ratios were fabricated by sintering pellets at 1200 °C for 5 hrs. Commercial powders of $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_{3-\delta}$ (K-ceracell, Korea) and $\text{Ce}_{0.9}\text{Gd}_{0.1}\text{O}_{2-\delta}$ (Rhodia, France) were used. LSC and GDC were mixed at 3:7, 5:5, 7:3 volume ratios by dry ball milling. A KrF excimer laser (COMPEX Pro 201F, Coherent, $\lambda = 248$ nm) was used as an ablation source and the laser fluence was about 3 J/cm² on the target surface and the target to substrate distance was kept as 5 cm. Each film was deposited at 700 °C and 200 mTorr oxygen atmosphere.

Figure 1 shows XRD patterns of each composition of the target and thin film. In the XRD patterns of target, diffraction peaks of secondary phases were not observed, and only diffraction peaks of LSC and GDC were obtained. It is also shown that the intensity ratio of the peaks was changed by the mixing ratio of LSC and GDC.

In the XRD patterns of films, it was also shown that diffraction peaks by LSC and GDC were only obtained, except a peak by the substrate (Sapphire c-cut wafer, $2\theta = 41.6^\circ$). Peak intensity ratios, however, were different between the target and the film. It seems that the deposited film is slightly textured, unlike the randomly orientated polycrystalline target.

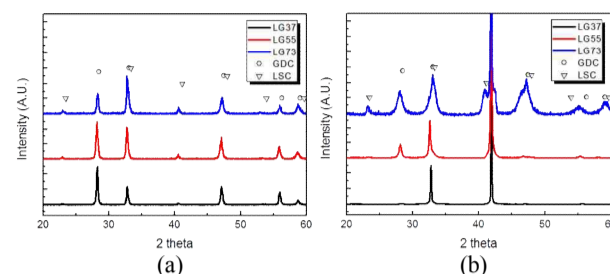


Fig. 1. (a) XRD pattern of target. (b) XRD pattern of thin film.

From the broad width of the peaks, the particle size of LSC and GDC in the films is thought to be on the nanometer scale. The particle size of LSC and GDC of each composition were estimated by the Scherrer equation. The average particle size in the thin film was calculated as few nanometer. The result indicates that nano-composite thin-film cathodes are successfully fabricated.

Arrhenius plots of polarization area specific resistances (ASR_{pol}) from the half-cell measurement for each composition are shown in Figure 2. The activation energies of ASR_{pol} of the LSC:GDC = 5:5 (LG55) and LSC:GDC = 7:3 (LG73) cells are similar. LSC:GDC = 3:7 (LG37) cell is slightly different. It appears that the activation energy of composite cathode is changed when the fraction of GDC in the composite cathode is greater than a certain amount.

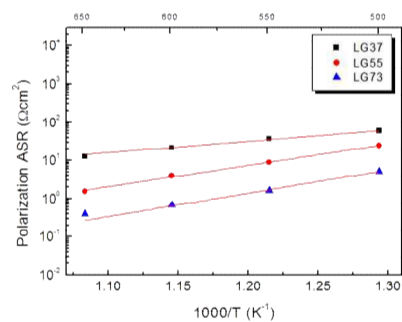


Fig. 2. Arrhenius plots of ASR_{pol} of the half cell.

The ASR_{pol} value decreased as the LSC content increased. In general, powder-processed LSC-GDC composite cathodes show the highest performance at the 5:5 mixing ratio.[4] In thin-film processed nanostructure composite cathodes, this does not seem to apply. It is thought that, when the nanosize particles are homogeneously mixed, the amount of the electrode material is the major factor to affect the composite cathode performance. Further analysis to elucidate the origin of properly differences will be performed.

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