High Temperature Neutron Diffraction Study of (La0.6Sr0.4)(Co0.8Fe0.2)O3-d Perovskite

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Recently, (La,Sr)(Co,Fe)O3-d perovskites have been investigated as the cathode materials of solid oxide fuel cells, because of the high electronic and high oxide-ion conductivity [1]. However, the detailed crystal structures under cell operation conditions, i.e., at elevated temperatures under oxygen partial pressures, P(O2) <= 0.21atm, have not been reported yet. In this study, we have measured high temperature neutron diffraction data of 40 mol% Sr- and 20 mol% Fe-doped LaCoO3, (La0.6Sr0.4)(Co0.8Fe0.2)O3-d (LSCF), at elevated temperatures, 299<=T<=1273K, under 0.002<=P(O2)<=0.21atm, to investigate the detailed crystal structures and the oxide-ion conduction path.

Neutron diffraction measurements were performed with a 150-detector system, HERMES [2], installed at the JRR-3M reactor in JAEA (Tokai). The neutron wave length was 1.8204(5)A and the diffraction data were collected in the 2 θ range from 20 to 153 deg. in step interval of 0.1 deg. A furnace with Pt-Rh heaters installed in a vacuum chamber was placed on the sample stage, and used for neutron-diffraction measurements at high temperatures. The diffraction data obtained were analyzed using the Rietveld program RIETAN-2000 [3]. The diffraction data of LSCF could be indexed assuming a trigonal symmetry (R-3c, No.167) at 299, 475, and 671K in P(O2) = 0.21atm, or a cubic one (Pm-3m, No.221) between 873 and 1273K under 0.002<=P(O2) <=0.21atm. The Rietveld refinement result of LSFC measured at 1273K in P(O2) = 0.002atm was as follows: Z = 1, a = 3.9333(2)A, α = 90 deg., V = 60.854(6)A^3. The reliability factors were as follows: Rwp = 6.48%, Re = 3.43%, S = Rwp/Re = 1.89.

Figure 1 shows the refined crystal structure of LSFC at 1273K in P(O2) = 0.002atm. Oxide ions showed larger atomic displacement parameters than those of La, Sr, Co, and Fe ions. Oxide ions also indicated larger thermal motion perpendicular to (Co, Fe)-O bonds. These results suggest the oxide-ion conduction in LSCF with an arc shape path [4].

References

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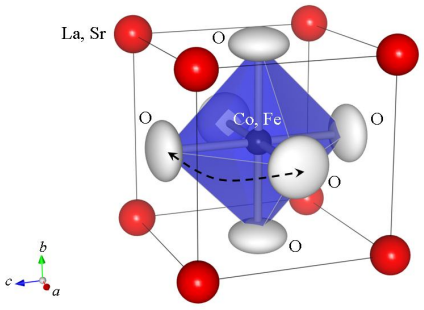

Fig. 1. Refined crystal structure of (La0.6Sr0.4)(Co0.8Fe0.2)O3-d at 1273K under P(O2) = 0.002 atm. The dotted curve with arrows denotes possible conduction path of oxide ions.