High-resolution synchrotron X-ray powder diffraction study of the orthorhombic-tetragonal phase transition in $La_{0.64}Ti_{0.92}Nb_{0.08}O_3$

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Introduction

Doped $La_{2/3}TiO_3$ compounds have perovskite-type structure and exhibit electrical and dielectrical properties. Thermal expansion and temperature dependence of lattice parameters are important factors in designing the components of solid oxide fuel cells. Conventional X-ray diffractometer produces double wavelengths of $K\alpha_1$+$K\alpha_2$, and broad diffraction peaks with asymmetric shape, which lead inaccuracy in determining the peak positions and thus lattice parameters. Here, we have used synchrotron X-ray powder diffraction technique, having higher angular resolution and simple peak shape (no $K\alpha_2$ peak), compared with conventional X-ray diffractometry, to determine precise lattice parameters of $La_{0.64}Ti_{0.92}Nb_{0.08}O_3$ as a function of temperature and to investigate the phase transition temperature.

Experiments

The synchrotron X-ray diffraction data were collected using beam line BL-3A installed at Photon Factory, KEK, Tsukuba. The wavelength was determined to be λ=1.38010(4) Å after the calibration with NIST CeO_2 sample (a=5.41129 Å). Experimental conditions were: step interval = 0.004˚ and counting time = 10 or 20s. The 004, 020 and 200 reflections were monitored. Peak positions were determined using a profile fitting program *PRO-FIT* (Toraya 1986).

Results and discussion

Peak splitting between orthorhombic 020 and 200 reflections was clearly observed at lower temperatures but it merged at higher temperatures (Fig. 1). With increasing temperature 020 and 200 peaks approached and became single between 355˚ and 388˚C. Although near the transition temperature these two peaks seemed single, but up to 355˚C the profile was able to be decomposed into 020 and 200 peaks successfully.

Figure 2 shows the temperature dependence of lattice parameters of $La_{0.64}Ti_{0.92}Nb_{0.08}O_3$ compound. With increasing temperature, a and c/2 parameters increased considerably compared with b parameter, resulting that a and b parameters coincided above 355˚C, indicating that the orthorhombic phase transformed to tetragonal symmetry. (1) The lattice parameters increased continuously, while b/a ratio decreased continuously with temperature and became unity at the orthorhombic-tetragonal transition point. (2) Good agreement was obtained in the lattice parameter values between heating and cooling. These results of (1) and (2) indicate that the orthorhombic-tetragonal phase transition is continuous.

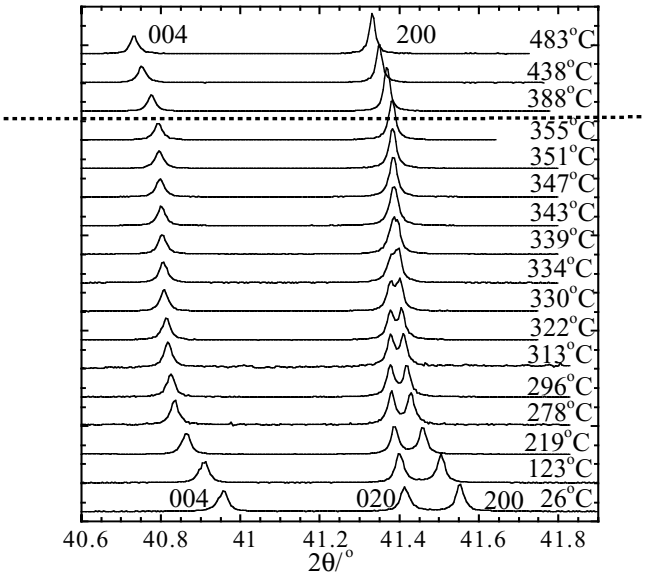

Fig.1. Temperature dependence of 004, 020 and 200 peak profiles of $La_{0.64}Ti_{0.92}Nb_{0.08}O_3$.

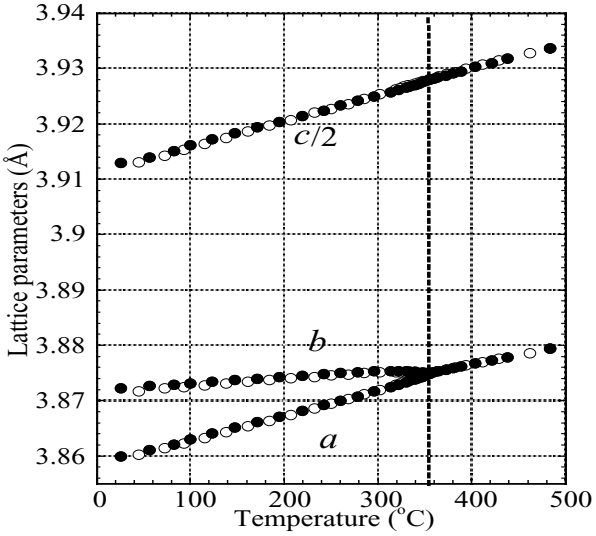

Fig.2. Temperature dependence of lattice parameters of $La_{0.64}Ti_{0.92}Nb_{0.08}O_3$. Filled and open circles denote heating and cooling data, respectively.

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