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The local structure in *B*-site modified $(Na_{0.5}K_{0.5})NbO_3$-$LiNbO_3$ system

Yumi INAGAKI, Yuya SHINKAI, Isao KAGOMIYA* and Ken-ichi KAKIMOTO
Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya, Aichi 466-8555, Japan

Introduction

Lead-free piezoelectric $(1-x)(Na_{0.5}K_{0.5})NbO_3$-$xLiNbO_3$ (LNKN) system has been attracted interest because the x=0.06 sample shows the maximum piezoelectric property of d_{33}=235pC/N[1]. In LNKN, it is well known that the doped Li is substituted in the *A*-site of perovskite-type structure, and the local structure especially NbO_6 unit structure has been clarified[2]. However, *B*-site modified system has not been investigated. In this study, Ta atom (ion radius: 0.64Å), which size is comparable with that of Nb atom, was doped to substitute for *B*-site. Then the effect of the *B*-site modification on the local structure was investigated using synchrotron radiation powder diffraction, XAFS and Raman spectroscopy measurement.

Experimental

The samples were prepared by a conventional mixed oxide method. The raw materials of high-purity powders of Na_2CO_3, K_2CO_3, Nb_2O_5, Li_2CO_3, and Ta_2O_5 were weighed to obtain the compositions according to the formula of $Li_{0.06}(Na_{0.55}K_{0.45})_{0.94}Nb_{1-x}Ta_xO_3$, where x is 0, 0.03, 0.05, 0.08, 0.10 and 0.15 (LNKNT). The weighed powder was mixed with ball-milling for 24 h in acetone. The dried mixture was calcined at 850°C for 10 h. The powder was subsequently sieved through a sieve with openings 150μm and pressed into disks of 12mm in diameter, followed by cold-isostatic-pressing under 200MPa. The disks were sintered at 1065-1075°C for 3h. A synchrotron radiation powder diffraction of the obtained samples was carried out using by a multiple two theta detector system (MDS) at BL-4B2. The EXAFS measurement was performed by BL-7C. The EXAFS measurement data was analyzed by REX2000 (Rigaku Co. Ltd., Japan) software. The Raman scattering spectra were excited using 514.5nm radiation from Ar^+ laser operating at 10mW and were collected by a microscopic Raman spectrometer (JASCO, NRS-2000).

Result and Discussion

Atomic distance of Nb-O is shown in Fig. 1. Although the ionic radius of Ta atom is as large as Nb atom, the atomic distance of both Nb-O_1 and Nb-O_2 became long when Ta content is over 0.8 mol%. In addition, the Raman mode of A_{1g} was shifted to lower wavenumber with increasing Ta content. These results indicated that the volume of NbO_6 unit in LNKNT increased with increasing Ta content. Particularly, the volume of NbO_6 unit for the case of over 0.8 mol% Ta content was large compared to the LNKNT in the case of Ta content less than 0.8mol%. However, the *c*-axis of lattice constant was decreased by Ta incorporation into *B*-site as shown in Fig. 2. It means that the volume of LNKNT lattice became small with increasing Ta content. Therefore, it is considered that the large NbO_6 unit was tilted in the small lattice when the Ta is substituted in B-site of LNKN perovskite.

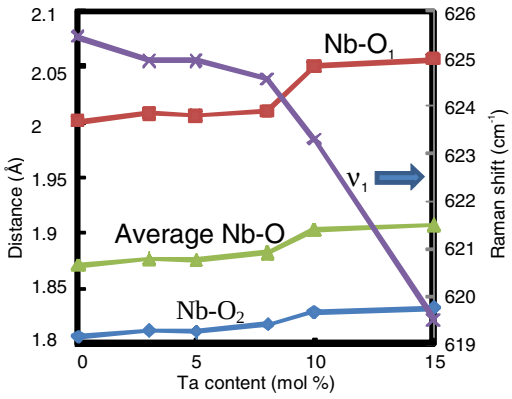

Fig. 1 Atomic distance of Nb-O for LNKNT analyzed by XAFS measurement and A_{1g} Raman mode.

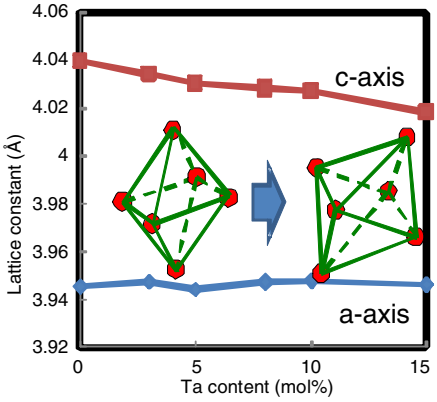

Fig. 2 Lattice constant and the model of NbO_6 unit in LNKNT.

References

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* kagomiya@nitech.ac.jp