

Dependence of ferroelectric performance and crystal structure on composition and heat treatment of $(\text{Bi},\text{M})_4(\text{Ti},\text{Si})_3\text{O}_{12}$ ($\text{M}=\text{La},\text{Nd}$) ferroelectric oxide

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1. Bi rich (Bi,La)-Ti-Si-O Ferroelectric Material

The La-substituted $(\text{Bi}_{4-x}\text{La}_x)\text{Ti}_3\text{O}_{12}$ is an attractive lead-free material for ferroelectric random access memory (FeRAM) applications because of its relatively large remanent polarization and fatigue-free characteristics. The bulk materials of $\text{Bi}_{3.25+x}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}$ ($x=0, 0.15, 0.25$) [BLT] and $\text{Bi}_4\text{Si}_3\text{O}_{12}$ (BSO) at specific compositional ratio were prepared for ferroelectric materials and some of samples annealed under the vacuum or high oxygen partial pressure (P_{O_2}). We investigated the relationship between the physical properties, crystal structure and ferroelectric performance of the sample before and after various heat treatments and changing composition.

From the results, Pr increases with increasing BSO content, but Pr decreased with increasing Bi content. Moreover, Pr decreased and E_c increased by heat treatment in the most of samples. On the other hand, the dielectric constant (ϵ') at T_c of BLT remarkably increased by adding BSO. From the crystal structure analysis, the bond length and the bond angles of Ti-O are changed. Consequently, the change of distortion in Ti-O₆ octahedra will effect of the change of ferroelectric performance, such as Pr.

2. (Bi,Nd)-Ti-Si-O Ferroelectric Material

We synthesized the new ferroelectric bulk materials of $\text{Bi}_4\text{Si}_3\text{O}_{12}$ (BSO) added $\text{Bi}_{4-x}\text{Nd}_x\text{Ti}_3\text{O}_{12}$ ($x=0.75, 0.85, 1.0, 1.2$). We investigated the dependence of ferroelectric performance and crystal structure on content and heat treatment for $\text{Bi}_4\text{Si}_3\text{O}_{12}$ added $\text{Bi}_{4-x}\text{Nd}_x\text{Ti}_3\text{O}_{12}$.

From the results, the remanent polarization (Pr) of $\text{Bi}_{4-x}\text{Nd}_x\text{Ti}_3\text{O}_{12}$ ($x=0.75, 0.85, 1.2$) samples increased by adding BSO. On the other hand, Pr of $\text{Bi}_{4-x}\text{Nd}_x\text{Ti}_3\text{O}_{12}$ ($x=1.0$) sample shows a maximum, but Pr decreased by adding BSO. Moreover, Pr decreased by heat treatment in the most of samples. The dielectric constant (ϵ') at room temperature and T_c of BSO added $\text{Bi}_{4-x}\text{Nd}_x\text{Ti}_3\text{O}_{12}$ remarkably increased by adding BSO. T_c and ϵ'' at T_c decreased and the dielectric loss ($\tan \delta$) at T_c remarkably decreased by heat treatment. From the results of neutron diffraction analysis, the bond length of Bi(Nd)-O₆ shows the maximum of BSO added $\text{Bi}_{2.8}\text{Nd}_{1.2}\text{Ti}_3\text{O}_{12}$, and it shows the minimum for BSO added $\text{Bi}_3\text{Nd}\text{Ti}_3\text{O}_{12}$. The bond length of Bi(Nd)-O₆ increased with decreasing of Nd content, x , of BSO added $\text{Bi}_{4-x}\text{Nd}_x\text{Ti}_3\text{O}_{12}$ ($x < 1.0$). The total calculated spontaneous ferroelectric polarization (P_s) along a-axis shows a maximum at BSO added $\text{Bi}_3\text{Nd}\text{Ti}_3\text{O}_{12}$. The similar tendency was obtained of no BSO addition sample. It was found that the P_s is related to the Pr. Moreover, the bond length of Bi(Nd)-O₆ changes by heat-treatment and it affects to Pr.