#### Relation Between Crystal Structure and Ferroelectric Properties of Bi-Si-O Added Sr<sub>1-x</sub>Bi<sub>2+x</sub>Ta<sub>2</sub>O<sub>9</sub>(x=0, 0.2) as a Bulk Ferroelectric Material.

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# **INTRODUCTION**

Ferroelectric  $Sr_{1-x}Bi_{2+x}Ta_2O_9$  (SBT x=0, 0.2) has attention as a promising candidate for FRAM, which has superior a small coercive field (*Ec*) and fatigue-free properties. However, the remanent polarization (*Pr*) of SBT is small<sup>1</sup>. Kijima *et al.* reported that  $Bi_2SiO_5$  added SBT thin film, which prepared by sol-gel method, could be crystallized at lower temperatures<sup>2</sup>. In this paper, we investigated the relation between the crystal structure and ferroelectric properties of Bi-Si-O added SBT(x=0, 0.2) ferroelectric bulk material.

### **EXPERIMENTAL**

Ferroelectric SBT(x=0, 0.2) and paraelectric Bi<sub>2</sub>SiO<sub>5</sub> (BSO2), Bi<sub>4</sub>Si<sub>3</sub>O<sub>12</sub> (BSO4) powder were prepared by conventional solid state reaction from SrCO<sub>3</sub>, Bi<sub>2</sub>O<sub>3</sub>, Ta<sub>2</sub>O<sub>5</sub>, and SiO<sub>2</sub> of 99.9% purity. With regard to the synthesis of Bi-Si-O added SBT, SBT(x=0, 0.2) and BSO2 or BSO4 were used, and wet-mixing was carried out at the prescribed ratio. After wet-mixing, the materials were heated for 2h at 1000~1025°C in air. The phase identification, the metal composition and the surface morphology of the obtained samples were characterized by powder X-ray diffraction, ICP and SEM, respectively. P-E hysteresis loop measurement was carried out using a TF2000FE device (manufactured by aix ACCT) by varying the frequency using the virtual ground mode. The dependence temperature of dielectric constant, the dielectric loss and the Cuie temperature (Tc) were measured by LCR meter. The crystal structure of these samples was determined by neutron powder diffraction using the GPPD of IPNS. The date was refined with the Rietveld analysis by GSAS program. From these results, we investigated the relation between the crystal structure and the ferroelectric properties of these samples.

### **RESULTS AND DISUCUSSION**

From the powder X-ray diffraction patterns, BSO4 added SBT (x=0.2) was confirmed to be a single phase, on the other hands, BSO2 added SBT (x=0.2) was identified as mainly SBT (x=0.2) structure with weak BSO2 (200) peak ( $2\theta$ =11.5°). The metal composition of BSO4 added SBT (x=0.2) was controlled of desired content. Surface morphology, which was observed by SEM, was almost same, that is, the sintering was sufficient of all samples. Figure 1 shows the P-E hysteresis loops of SBT(x=0.2), BSO2 or BSO4 added SBT (x=0.2). From Fig.1, it can be seen that remanent polarization (*Pr*) and coercive field (*Ec*) of a pure SBT (x=0.2) are about  $6.3\mu$ C/cm<sup>2</sup> and 28.0kV/cm. The P-E hysteresis loops significantly changed by added BSO. The *Pr* and the *Ec* of BSO4 added SBT (x=0.2) increased, while the *Pr* decreased and the *Ec* increased of BSO2 added SBT (x=0.2) relative to SBT(x=0.2). Figure 2 shows the temperature dependence of the dielectric constant,  $\varepsilon$ , and the dielectric loss, *tan* $\delta$ , of SBT (x=0.2) and BSO2 or BSO4 added SBT (x=0.2). Curie temperature (*Tc*) changed, and dielectric constant,  $\varepsilon$ , increased significantly by added BSO.

From the crystal structural analysis by Reitveld technique of neutron powder diffraction, the spontaneous polarization and tilting degree, et al of BSO added SBT samples were calculated. The crystal system was orthorhombic, and the space group was A2<sub>1</sub>am of BSO added SBT, and the metal site occupancy was fixed on the basis of the result of ICP. We investigated the relation between these parameters of crystal structure and the ferroelectric property.



## REFERENCES

- 1. A. Machida, N. Nagasawa, T. Ami, and M. Suzuki, *Jpn.J.Appl.Phys.*, **36**, 7267(1997).
- T.Kijima, and H. Ishiwara, *Jpn.J.ApplPhys.*, **41**,716 (2002).