



# 0.46% unipolar strain in lead-free BNT-BT system modified with Al and Sb



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## ABSTRACT

Lead-free piezoelectric ceramics  $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.935}\text{Ba}_{0.065}\text{Ti}_{1-x}(\text{Al}_{0.5}\text{Sb}_{0.5})_x\text{O}_3$  (BNBT6.5- $x$ AS) were prepared by a conventional solid sintering technique and their structure and electrical properties were systematically investigated. With increasing AS content, BNBT6.5- $x$ AS ceramics underwent a phase transition from ferroelectric to relaxor phase, accompanied with the significant disruption of ferroelectric order. Accordingly, the ferroelectric-relaxor transition temperature  $T_{F-R}$  was shifted down to room temperature, and thus a large unipolar strain of 0.46% (at 80 kV/cm) corresponding to a large signal  $d_{33}^*$  of 573 pm/V is obtained in 1.5 mol% AS-modified samples.

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## 1. Introduction

In the search for lead-free piezoelectric ceramics, bismuth sodium titanate (BNT) has been considered as a superior candidate material because of its large remnant polarization ( $P_r$ ) and Curie temperature ( $T_c$ ). However, its high coercive field  $E_c$  leads to difficulty in poling process, which limits its practical application [1]. To enhance the properties of BNT, it has been modified with other perovskite materials [2–7]. In particular, the solid solution between BNT and  $\text{BaTiO}_3$  (BT) [4,5] has many of the same characteristics present in  $\text{PbZr}_x\text{Ti}_{1-x}\text{O}_3$  (PZT) [8]. In  $(1-x)\text{BNT}-x\text{BT}$  system, a morphotropic phase boundary (MPB) between rhombohedral and tetragonal perovskite phases exists at compositions with  $x=0.06-0.07$ , at which samples show promising piezoelectric properties.

Besides the excellent piezoelectric properties, (BNT-BT)-based materials have been received more attention recently in the search for high-strain materials [9–13], since Zhang et al. [10] made a breakthrough in ternary  $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3-\text{BaTiO}_3-\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$  (BNT-BT-KNN) system. BNT-BT system is attractive as a base composition, since it is known that it has good piezoelectric performances along with inherently large poling strain at its MPB. To achieve high strain of BNT-BT system, a small amount of KNN [10],  $\text{SrTiO}_3$  [14],  $\text{BiXNbO}_3$  ( $X=\text{Ag}, \text{Li}$ ) [15] and etc have been doped into BNT-BT.

B-site complex ions substitution is also a good approach to achieve high-strain (BNT-BT)-based ceramics as reported in our recent works [16,17]. We disclosed that B-site complex ions  $(\text{X}_{0.5}\text{Y}_{0.5})^{4+}$  ( $X=\text{Fe}, \text{Mn}; \text{Y}=\text{Nb}, \text{Sb}, \text{Ta}$ ) are very effect modifiers. In this sense, if the X element is fixed as Al, it may play a similar role in the enhancement of the electrostrain since  $\text{Al}^{3+}$  has a close ion radius to that of  $\text{Fe}^{3+}$  or  $\text{Mn}^{3+}$ . Based on the above, the B-site complex ions  $(\text{Al}_{0.5}\text{Sb}_{0.5})^{4+}$  was designed and  $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.935}\text{Ba}_{0.065}\text{Ti}_{1-x}(\text{Al}_{0.5}\text{Sb}_{0.5})_x\text{O}_3$  was fabricated to achieve high-strain in the present study. Owing to the close ionic radius of  $(\text{Al}_{0.5}\text{Sb}_{0.5})^{4+}$  to that of  $\text{Ti}^{4+}$ , the complex ions  $(\text{Al}_{0.5}\text{Sb}_{0.5})^{4+}$  will enter the B-site of perovskite and replace  $\text{Ti}^{4+}$ . Accordingly, addition of  $(\text{Al}_{0.5}\text{Sb}_{0.5})^{4+}$  will induce the crystal lattice distortion and thus distort the local structure of BNT-BT. Therefore, destabilization of the ferroelectric order and an enhancement of the field-induced strain will be expected in this system.

## 2. Experimental procedure

Lead-free piezoelectric ceramics  $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.935}\text{Ba}_{0.065}\text{Ti}_{1-x}(\text{Al}_{0.5}\text{Sb}_{0.5})_x\text{O}_3$  (BNBT6.5- $x$ AS,  $x=0.005, 0.010, 0.015, 0.020$ ) were prepared by a conventional solid sintering method [17]. The crystal structures of the sintered ceramics were determined by X-ray diffraction analysis (XRD) using a  $\text{Cu K}\alpha$  radiation ( $\lambda=1.54178 \text{ \AA}$ ) (D8 Advance, Bruker Inc., Karlsruhe, Germany). The microstructure of the ceramics was observed by a scanning electron microscope (SEM, JSM-6380, Japan). The electric field-induced polarization ( $P$ -

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$E$ ) and strain ( $S$ - $E$ ) were measured by using an aix-ACCT TF2000FE-HV ferroelectric test unit (aix ACCT Systems GmbH, Aachen, Germany). The temperature dependence of dielectric properties was measured at temperatures ranging from room temperature to 500 °C (Novocontrol, Germany).

### 3. Results and discussion

Fig. 1(a) shows the XRD patterns of BNBT6.5- $x$ AS ceramics. All compositions exhibit a single perovskite structure indicating that AS has diffused into the B site of BNBT6.5 lattice to form a homogenous solid solution. The diffraction peaks of BNBT6.5- $x$ AS ceramics shift slightly to a higher angle with the increase of AS content. This suggests that AS substitution into the BNBT6.5 leads to the shrinkage of the unit cell due to the smaller radius of  $(Al_{0.5}Sb_{0.5})^{4+}$  (0.568 Å) than that of  $Ti^{4+}$  (0.605 Å). Fig. 1(b), (c) show the SEM micrographs of BNBT6.5- $x$ AS ( $x=0.005$  and 0.02) ceramics. The partial substitution of AS for Ti appears to have little influence on the morphology of BNBT6.5 ceramics. The average grain size in these compositions is about 2–3  $\mu$ m.

Fig. 2 shows the temperature dependence of dielectric permittivity and dielectric loss for poled BNBT6.5- $x$ AS samples. It is clear that poled BNBT6.5 exhibits two dielectric anomalies known as the ferroelectric-to-relaxor phase transition temperature ( $T_{F-R}$ ) and the  $T_m$  at which relative dielectric constant  $\epsilon_r$  reaches the maximum value [18]. With increasing AS content, the dielectric maximum decreases rapidly and  $T_m$  peaks become extremely broad. Moreover,  $T_{F-R}$  peak is shifted down to room temperature with the addition of AS, indicating a compositionally induced ferroelectric-to-relaxor phase transition. Due to the formation of ferroelectric-relaxor ( $F-R$ ) phase boundary, optimized strain response can be expected in a critical composition of the BNBT6.5-

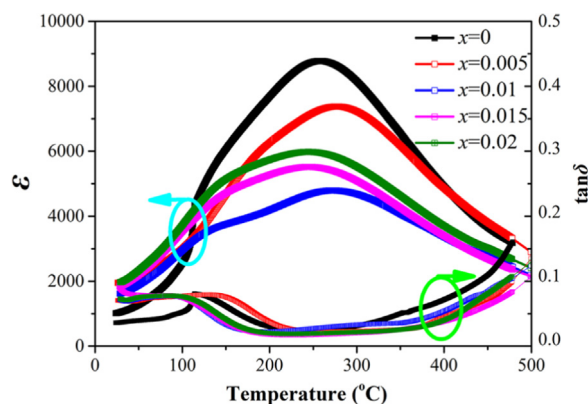


Fig. 2. Temperature dependence of the dielectric constant and loss of BNBT- $x$ AS ceramics.

$x$ AS system.

Fig. 3 shows (a) ferroelectric hysteresis ( $P$ - $E$ ) loops, (b) bipolar strain curves, (c) unipolar strain curves and (d) the large signal  $d_{33}^*$  of BNBT6.5- $x$ AS ceramics, respectively. The polarization hysteresis loop of BNBT6.5 displays a well-saturated typical ferroelectric behavior. With the increase of AS content, the shape of  $P$ - $E$  loops changes from “well saturated” to “constricted” which indicates that the BNBT6.5 ceramics are subjected to a phase transition from ferroelectric to relaxor phase [19]. The coexistence of two phases contributes to a large field-induced strain, as shown in Fig. 3(b) and (c). BNBT6.5 ceramic exhibits a typical ferroelectric behavior with butterfly shaped strain hysteresis loop and visible negative strain  $S_{neg}$  related to the domain back switching during bipolar cycles [20]. With the increase of AS,  $S_{neg}$  gradually disappears accompanied by a concurrent increase in the positive strain  $S_{pos}$ . Meanwhile, the butterfly like loop drastically changes

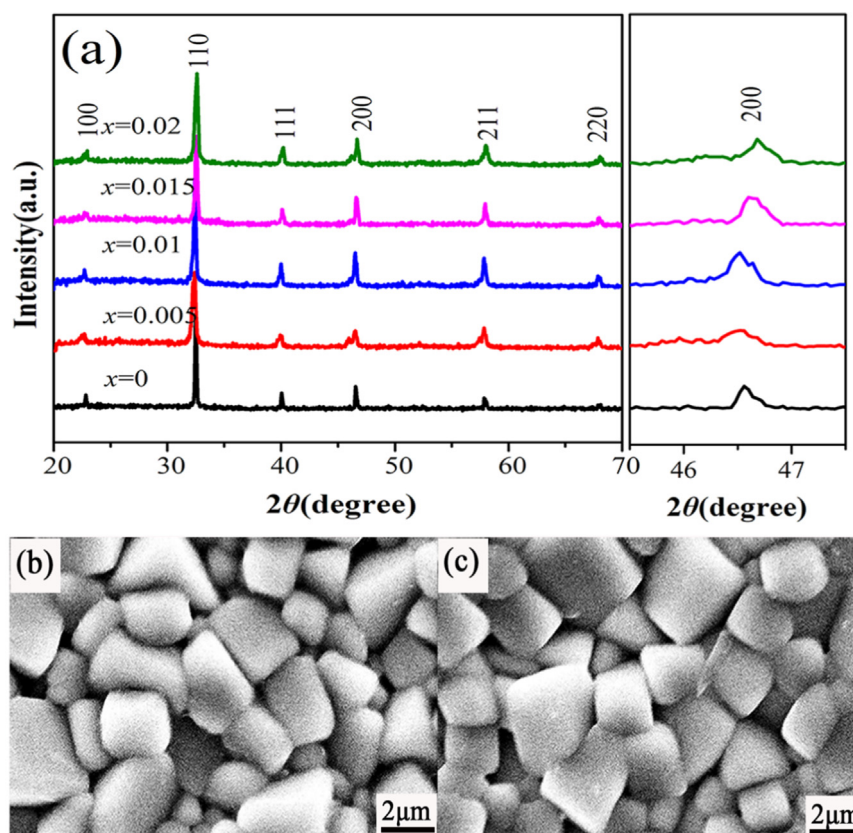


Fig. 1. (a) X-ray diffraction patterns of BNBT6.5- $x$ AS ceramics, (b),(c) SEM photographs of BNBT6.5- $x$ AS ( $x=0.005$  and 0.02) ceramics.

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