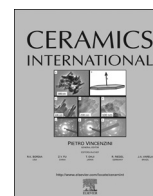




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# Phase structure, ferroelectric properties, and electric field-induced large strain in lead-free $0.99[(1-x)(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3-x(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3]-0.01\text{Ta}$ piezoelectric ceramics

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## ABSTRACT

Lead-free  $0.99[(1-x)(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3-x(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3]-0.01\text{Ta}$  piezoelectric ceramics were prepared by a conventional solid-state reaction process. The ferroelectric properties, and strain behaviors were characterized. Increase of the  $(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3$  content induces a phase transition from coexistence of ferroelectric tetragonal and rhombohedral to a relaxor pseudocubic phase. Accordingly, the ferroelectric order is disrupted significantly with the increase of  $(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3$  content and the destabilization of the ferroelectric order is accompanied by an enhancement of the unipolar strain, which peaks at a value of 0.35% (corresponding to a large signal  $d_{33}$  of 438 pm/V) in samples with 20 mol%  $(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3$  content. Temperature dependent measurements of both polarization and strain from room temperature to 120 °C suggested that the origin of the large strain is due to a reversible field-induced nonpolar pseudocubic-to-polar ferroelectric phase transformation.

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## 1. Introduction

Lead zirconate titanate (PZT) ceramics have been extensively used for piezoelectric transducers, sensors, and actuators due to their excellent piezoelectric properties around the morphotropic phase boundary (MPB) [1]. Nevertheless, due to the toxicity of lead oxide and its high vapor pressure during processing and the requirement of environmental protection, lead-free counterparts have been intensively studied [2–5]. Among various lead-free ceramic systems,  $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ -based lead-free ceramics have attracted large interest recently for their intriguing structural and piezoelectric properties [5]. In particular,  $(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3-(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3$  (BNT–BKT) solid solution exhibits optimum piezoelectric performance in the vicinity of MPB between rhombohedral ( $R3c$ ) and tetragonal ( $P4mm$ ) phases at compositions with the BKT concentration of 16–20 mol% [6]. The BNT–BKT system has many of the same characteristics present in PZT and is considered candidate to replace Pb-based materials.

Recently, the interest in the BNT-based system is closely related to its potentiality for actuator applications. Through slight chemical modification including incorporating a small amount of additional chemical entities such as ion dopants [7–10] or  $\text{ABO}_3$ -type compounds [11–25] in BNT-based MPB composition

and creation of moderate A-site vacancies in the MPB compositions [26,27], giant electric-field-induced strains even comparable with some PZT-based antiferroelectric ceramics were obtained in BNT-based materials. For example, Zhang et al. [11] published results of BNT– $\text{BaTiO}_3-(\text{K}_{0.5}\text{Na}_{0.5})\text{NbO}_3$  (BNT–BT–KNN) ternary solid solutions, which delivers a large strain response of 0.45% (the corresponding large signal  $d_{33}$  ( $S_{\text{max}}/E_{\text{max}}$ ) is 560 pm/V), when a small amount of BNT is replaced by certain amount of KNN. Later, Seifert et al. [12] introduced BNT in place of BT in the BNT–BKT–KNN system and reported a larger  $S_{\text{max}}/E_{\text{max}}$  of 600 pm/V. In these materials, although the emergence of this large strain is accompanied by a significant reduction in their piezoelectric activity characterized by the small signal  $d_{33}$  value, the material is considered to be highly promising for actuator applications where the large signal  $d_{33}$  is the figure of merit. Furthermore, the results showed that the sacrifice of piezoelectric activity is not only enhancing the actuating performance but also favors the temperature stability of the enhanced strain response [12,13]. To the origin of the observed large strain response in BNT-based ceramics, it was recently identified as a consequence of a drastic reduction in the remanent strain due to the presence of a “non-polar” phase (an ergodic relaxor state [14,28]) at zero field, which can transform to ferroelectric phase under electric-fields due to their comparable free energies and enables each bipolar cycle to fully utilize the inherently large poling strain of BNT-based materials [14].

Generally, the basic approach to achieve the optimized strain properties in BNT-based system is to place the composition of the

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material to the proximity of a composition-induced phase transition between ferroelectric and non-polar phases. Furthermore, an enhancement of the strain in BNT-based materials is accompanied by the disruption of ferroelectric order, and corresponded to the ferroelectric-to-relaxor transition point  $T_{F-R}$  at nearly room temperature (RT). Therefore, to achieve an expected large strain, one of evaluation criterion concerning the effect of the modifier is whether it can adjust the  $T_{F-R}$  to RT.

In our previous work [15], we disclosed that ternary BNT–BKT–KNN system exhibited a high strain response (0.32–0.45%) when the ferroelectric-to-relaxor transition point  $T_{F-R}$  was shifted to RT with the increase of BKT content. This gives information that change of BKT content in (BNT– $x$ BKT)-based materials can drive a greatly enhanced strain with the help of shift of  $T_{F-R}$  down to RT. Recent studies [29,30] showed that small amount of Ta doped into BNT-based ceramics plays the similar role as KNN dopant in BNT-based ceramics, which can also be helpful to shift the  $T_{F-R}$  down to RT and obtain the large strain. Based on the above,  $0.99[(1-x)(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3-x(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3]-0.01\text{Ta}$  (BNKT100 $x$ -1Ta) lead-free piezoelectric ceramics were designed and fabricated to achieve large strain response in BNT-based ceramics in the present study. The influences of composition on the ferroelectric stability and consequent changes in the electrical properties of the studied ceramics were investigated.

## 2. Experimental

$0.99[(1-x)(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3-x(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3]-0.01\text{Ta}$  (BNKT100 $x$ -1Ta;  $x=0.12-0.022$ ) lead-free ceramics were prepared by the conventional ceramic fabrication technique using reagent-grade metal oxides or carbonate powders of  $\text{Bi}_2\text{O}_3$  (99.98%),  $\text{TiO}_2$  (99.6%),  $\text{Ta}_2\text{O}_5$  (99.9%),  $\text{Na}_2\text{CO}_3$  (99.99%),  $\text{K}_2\text{CO}_3$  (99.0%). BNKT100 $x$  powders were first calcined at 850 °C for 4 h. After the calcinations,  $\text{Ta}_2\text{O}_5$  and BNKT100 $x$  powders were weighted according to the formulas and then ball-milled for 12 h. The resulting powders were mixed with a polyvinyl alcohol (PVA)

binder solution and then pressed into disk samples. After burning off the PVA, the disk samples were sintered at 1140–1160 °C for 2–4 h in air. To minimize the evaporation of Na and Bi elements, the samples were embedded in atmospheric powder of the same composition.

Phase structure of the ceramics was identified by using X-ray diffraction (XRD, Bruker D8 Advance, Germany) with  $\text{Cu K}_\alpha$  radiation. The surface morphology of the ceramics was studied by scanning electron microscope (SEM) (JSM-5900, Japan). For the electrical measurements, silver paste was coated on both sides of the sintered samples and fired at 650 °C for 10 min to form electrodes. The temperature dependences of the dielectric properties were measured using a HP 4294A precision LCR meter (Agilent, Palo Alto, CA). The electric-field-induced polarization ( $P$ - $E$ ) and strain ( $S$ - $E$ ) measurements were carried out using an aix-ACCTF2000FE-HV ferroelectric test unit (aix-ACCT Inc., Germany) connected with an accessory laser interferometer vibrometer (SP-S 120/500; SIOS Mebtechnik GmbH, Germany). For the temperature-dependent  $P$ - $E$  and  $S$ - $E$  measurements, the samples were submerged in a silicon oil bath with a temperature controller (aix-ACCT).

## 3. Results and discussion

Fig. 1 gives the microstructure of BNKT100 $x$ -1Ta ceramics observed on the surface of the compositions, showing dense microstructures. Change of BKT content has no significant influence on the microstructure of the ceramics, as the average grain size in these compositions is all about  $1.8 (\pm 0.5) \mu\text{m}$ .

Fig. 2 shows XRD patterns of BNKT100 $x$ -1Ta ceramics. A stable solid solution among BNKT100 $x$  and Ta with a pure perovskite structure is formed in all indicated compositions. In the compositions with lower BKT content ( $x < 0.16$ ), they reveals peak splittings at around  $40^\circ$  of (111)/( $\bar{1}\bar{1}\bar{1}$ ) and  $46^\circ$  of (002)/(200) reflections, which can be linked to a mixed rhombohedral-tetragonal crystal structure [6]. With increasing BKT content ( $x$ ), the split

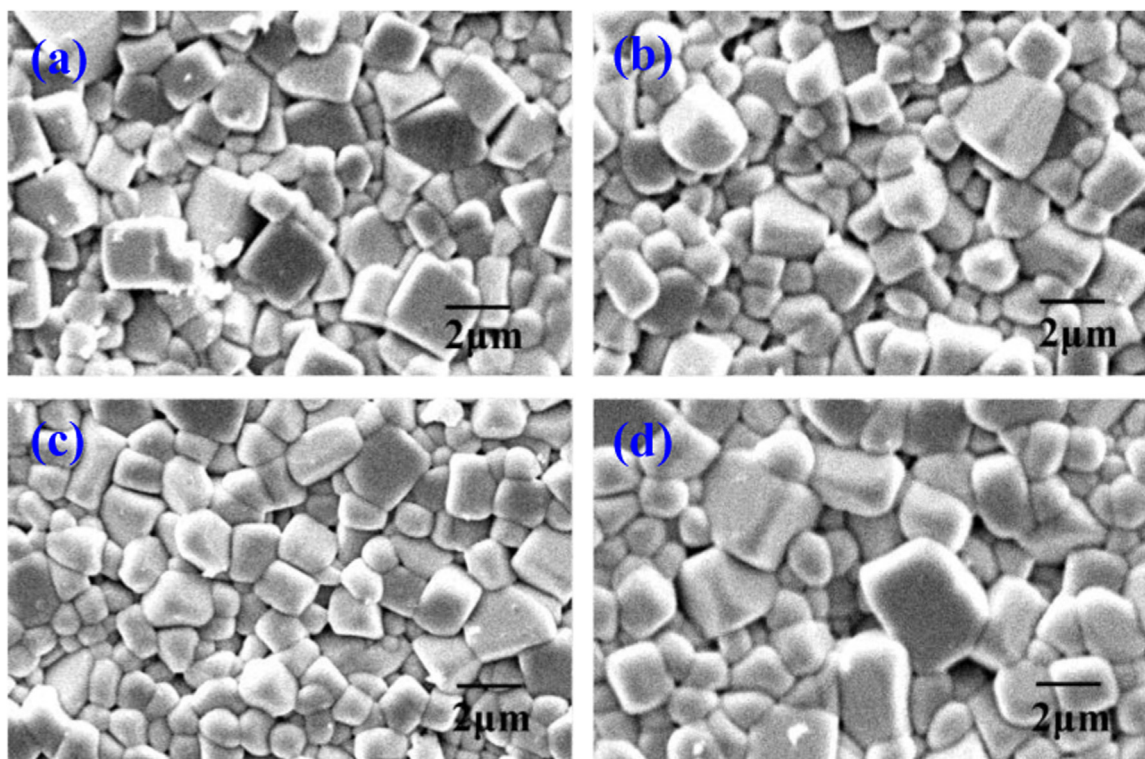


Fig. 1. SEM images of BNKT100 $x$ -1Ta ceramics with (a)  $x=0.12$ , (b)  $x=0.16$ , (c)  $x=0.20$  and (d)  $x=0.22$ .

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