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Effect of different templates and texture on structure evolution and strain behavior of <001>-textured lead-free piezoelectric BNT-based ceramics

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ABSTRACT

In this work, <001> oriented $0.90(Bi_{0.5}Na_{0.5})TiO_3$–$0.05KNbO_3$–$0.05SrTiO_3$ (BNT–KN–ST) ceramics with high degree of texturing were produced by templated grain growth using different perovskite plate-like $(Bi_{0.5}Na_{0.5})TiO_3$ and $SrTiO_3$ as template particles. The effects of different template particles on the grain orientation, structure evolution, phase stability and strain response was systematically investigated to develop the lead-free piezoelectric materials with an excellent actuating performance. The strain response increased monotonously with the increase in the degree of texturing and the optimized microstructure was found to provide excellent strain properties with a large strain of 0.38% and normalized strain $S_{max}/E_{max} = 540$ pm/V for the textured samples with BNT template particles at room temperature as compared to the random counterparts. The temperature-dependent strain response is also compared to randomly oriented ceramics and the results demonstrated that the textured piezoceramics with BNT template particles exhibited the enhancement in the temperature stability over the temperature range room temperature-120 °C.

1. Introduction

In recent years, there has been an tremendous effort in developing lead-free piezoceramics from the viewpoint of environmental protection [1–9]. Among them, due to the recent discovery that the BNT-based materials with a reversible phase transition between an ergodic relaxor state and a ferroelectric under high fields possess a giant electric field induced strain that surpass those reached by $Pb(Zr,Ti)O_3$ (PZT) [2,5], BNT-based perovskites with complex compositions have received strong interest and shed light on the potentiality of lead-free piezoceramics for actuator applications driven at off resonance frequencies, representatively multilayer actuators. Several years ago, a giant strain of 0.45% at 80 kV/cm was observed in $0.92(Bi_{1/2}Na_{1/2})TiO_3$–$0.06BaTiO_3$–$0.02(K,Na)NbO_3$ (BNT–6BT–2KNN), which is even higher than that of a commercial soft lead zirconate titanate PZT ceramic (e.g. PIC151, PI Ceramics) [10]. A series of *in situ* diffraction studies under an electric field environment have demonstrated that the structural origin of the giant strain is a reversible phase transformation between metrically cubic and non-cubic phases [11–14], where the addition of KNN decreased the ferroelectric-relaxor ferroelectric transition temperature (T_{F-R}) below the ambient temperature. Since the discovery of the giant strain in BNT–BT–KNN pseudoternary system, large strains of the same origin have been found in other BNT-based solid solutions with effective chemical modifier [15–25].

Considering the fact that the piezoelectric single crystal exhibits higher performance in comparison with their polycrystalline counterparts, there has been enormous interest in processing and understanding lead-free piezoelectric single crystals. In BNT-based piezoelectric single crystals, rhombohedral phase BNT–BT lead-free single crystals showed up to 0.25% strain along the <001> direction and the tetragonal phase crystals exhibited strain as high as 0.85% with large hysteresis along the same direction [26]. A giant electric-field-induced strain of 0.87% under a low electric field of 40 kV/cm reported by Teranishi et al.

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[27]was observed in tetragonal BNT–BKT–BT single crystal along <001> direction. Recently, Park et al. [28] reported the synthesis of 91BNT–6BT–3KNN single crystal via solid state single crystal growth and have demonstrated an improved strain response (0.57%) for the crystal with <001> orientation at 70 kV/cm. These results demonstrated that BNT-based single crystals possess giant strain along the <001> direction, and thus are suitable for use in actuator applications. However, due to high cost in the growth of single crystals and difficulty in controlling crystal stoichiometry, ceramics with textured microstructures have attracted significant scientific and commercial interest as it builds upon the known material systems with excellent performance that have demonstrated the potential to be practical [29–36]. Fancher et al. [37] reported the textured BNT–BT–KNN ceramics with a <001> pseudocubic (pc) orientation, and demonstrated that <001> textured BNT–7BT–2KNN (BNT–BT–KNN) reaches a maximum 0.47% strain response, an almost 50% increase compared to its randomly oriented counterpart. In our previous work, enhanced electric-field-induced strain was observed in <001> textured 79BNT–20BKT–1BKT(BNT–BKT–KNN) with the improved texture quality [34]. Subsequently, in research on similar compositions to the aforementioned BNT–BT–KNN and BNT–BKT–KNN piezoceramics, Zhang et al. [38] reported <001> oriented 91BNT–6BT–3AgNbO$_3$ (BNT–BT–AN) piezoceramics using plate-like BNT templates and showed that the electric-field-induced strain for the <001> textured BNT–BT–AN has a 78% increase and exhibits significantly reduced frequency dependence in comparison with its randomly oriented counterpart. Very recently, Hussain et al. [39,40] synthesized plate-like BNT particles from a bismuth layer-structured ferroelectric Na$_{0.5}$Bi$_{4.5}$Ti$_4$O$_{15}$ precursor by a topochemical microcrystal conversion method and the large as synthesized plate-like NBT particles were utilized to fabricate textured 99.4BNT–0.6BaZrO$_3$ (NBT–BZ) ceramics, and the results demonstrated that the highly <001> textured BNT–BZ ceramics exhibit ~100% improvement in the strain response as compared to its random ones. Based on the above mentioned, texture control of BNT-based ceramics offer a advantage in that the strain response can be improved because it enables the polycrystalline ceramics to resemble their single crystal counterparts so that favorable domain engineered states can be obtained for compositions close to MPB.

In general, anisotropic particles or templates play a significant role in the grain orientation process [33], and template particles must have a similar crystal structure and possess less than 15% lattice mismatch with the matrix particle [41]. However, the template used to the fabrication of textured BNT-based solid solutions focuses on the plate-like Bi$_4$Ti$_3$O$_{12}$ (BiT) particles in the templated grain growth (TGG) progress [34,42,43]. Generally, the use of BiT templates causes not only the fact that the ceramics are difficult to be fully densified due to the difference of the structures between the template and matrix but also the target compound deviation induced by the interdiffusion between template and the matrix, giving rise to a deterioration of the piezoelectric properties compared to the random counterparts. Therefore, it is necessary to obtain large anisotropic template with a perovskite structure and high symmetry to meet the fabrication of BNT-based ceramics. According to the double molten salt synthesis (DMSS) method, our group successfully synthesize plate-like BT, NaNbO$_3$(NN), SrTiO$_3$(ST), and BNT particles which have a perovskite structure with high symmetry [30].

A recent *in situ* electric-field-dependent investigation has demonstrated that a <001> crystallographic texture effects the poling-induced phase transformation in BNT–7BT [35]. However, recent works have highlighted the fact that the effect texturing has on the electric-field-induced strain response is not well understood. There are limited studies on the temperature-dependent strain response of textured BNT-based lead-free piezoceramics [30,37,38,44], and currently no systematic investigations of the effect different perovskite templates and texture has on the structure evolution and strain behavior of BNT-based ceramics. In our recent works, phase diagram for the (Bi$_{0.5}$Na$_{0.5}$)TiO$_3$–KNbO$_3$–SrTiO$_3$ ternary system has been constructed and analyzed based on macroscopic properties and structure analysis, and the results showed that the ternary solid solution 0.90BNT–0.05KN–0.05ST (BNT–KN–ST) prepared by the traditional solid state reaction exhibits a large strain response of 0.34% at room temperature [45,46]. In the present study, the TGG method was applied to fabricate <001> textured BNT–KN–ST piezoceramics in combination with tape casting using 5 mol% BNT and ST anisotropic particles as templates to further improve the piezoelectric performance. We investigate the effect different templates and texture has on the phase structure and electric-field-induced strain behavior, and textured BNT–KN–ST is compared to randomly oriented ceramics to explore the effect texturing has across the room temperature relaxor transition.

2. Experimental

BNT and ST template particles were processed from Bi$_4$Ti$_3$O$_{12}$ (BiT) platelets using DMSS method. Plate-like BiT precursors were first prepared by molten salt synthesis (MSS). The Bi$_2$O$_3$ (99.975%), TiO$_2$ (99.6%) and NaCl (98%) were selected as starting materials. First, Bi$_2$O$_3$ and TiO$_2$ power were mixed with a ball milling in a ratio according to a predetermined number. The mixture was dry-mixed with NaCl and heated at 800 °C for 1 h to melt NaCl and then at 1000 °C for 2 h to prepare plate-like Bi$_4$Ti$_3$O$_{12}$ (BiT) particles. The resulting product was washed several times with hot de-ionized water to remove trace of NaCl. Secondly, the synthesized BiT platelets were mixed with SrCO$_3$ (99%) and NaCl and heated to 800 °C for 1 h to melt the NaCl, and then the mixture was heated to 950 °C for 2 h to complete the SrTiO$_3$ formation topological reaction. The BiT template particles were weighted and mixed with Na$_2$CO$_3$, TiO$_2$ and NaCl flux in ethanol and heated to 800 °C for 1 h to melt the NaCl, and then the mixture was heated to 930 °C for 2 h. Finally, the mixture was also washed with hot de-ionized water repeatedly until no Cl$^-$ was detected by AgNO$_3$ solution and HNO$_3$ was utilized to eliminate the bismuth oxide (Bi$_2$O$_3$) by-product.

A conventional solid–state reaction method was used to prepare the BNT–KN–ST matrix. BNT–KN–ST ceramics were textured by TGG method in combination with tape casting using BNT and ST template particles, and BNT and ST template particles of 5 mol% were added to adjust the stoichiometric chemical formula of BNT–KN–ST. The TGG process has been described in detail elsewhere [29]. Random counterparts were prepared using an experimental procedure in our previous work [46]. The plate-like BNT and ST particles synthesized above and pre-fabricated matrix were mixed with the solvent (toluene and ethanol in the weight ratio of 2:1) for 12–15 h using tumbling mill, with different templates. Then the LS modelbond (produced by Lingguang Electric Chemical Materials Technology Corporation in Zhaoqing city, China) was added in the slurry and milled for anther 3 h. The obtained stable slurry was subjected to tape casting with the doctor blade height of 100 μm to align the BNT and ST platelets in the powder matrix. The tapes were dried, cut, and laminated into a multilayer sheet with a size of 12 × 12 × 2 mm under a pressure of 20 MPa at 50 °C for 30 min followed by a binder burnout process at 500 °C for 4 h to remove organic additives with a heating rate of 0.5 °C/min. To increase the green density, the samples after the binder burnout process were subjected to the cold isostatic press (CIP) under a pressure of 200 MPa for 10 min. Then the samples were sintered at