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Pairing high piezoelectric properties and enhanced thermal stability in grain-oriented BNT-based lead-free piezoceramics

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ABSTRACT

$(Bi_{0.5}Na_{0.5})TiO_3$ (BNT)-based piezoceramics usually exhibit enhanced piezoelectric coefficient d_{33} together with the deterioration of depolarization temperature T_d, which is the common drawback limiting their use in practical application. Here, we demonstrate that harnessing the microstructure in BNT-based ceramics will be an efficient way to resolve this obstacle. < 00l > oriented piezoelectric ceramics $0.94(Bi_{0.5}Na_{0.5})TiO_3-0.06BaTiO_3$ was engineered by templated grain growth (TGG) using $NaNbO_3$ (NN) as templates. The manufactured textured ceramics with the optimized microstructure was characterized by not only approximately 200% enhancement in the magnitude of piezoelectric response (d_{33}~297pC/N) but also improved thermal stability (T_d~57 °C) in comparison to its randomly oriented counterparts (d_{33}~151pC/N and T_d~32 °C). Moreover, the enhanced piezoelectricity in grain oriented specimens primarily originated from a high degree of non-180° domain switching as compared to the randomly axed ones. The current study opens the door to pair high piezoelectric properties and enhanced thermal stability in BNT-related materials though texture technique.

1. Introduction

Piezoelectric materials have been demonstrated to possess myriad applications in daily life such as actuators and energy harvesting electromechanical devices [1]. Bismuth sodium titanate $(Bi_{0.5}Na_{0.5})TiO_3$ (BNT) has been deemed as a key lead-free alternative for currently dominated $Pb(Zr_{0.5}Ti_{0.5})O_3$ owing to excellent electromechanical and piezoelectric properties for their solid solution with $BaTiO_3$ (BT) or $(Bi_{0.5}K_{0.5})TiO_3$ (BKT) at morphotropic phase boundary [2–4]. However, it is challenging to completely replace Pb-based materials because of a common drawback that the enhancement of piezoelectric properties though solid solution or chemical doping is generally accompanied by obvious decrease in the depolarization temperature (T_d) for BNT-based materials [5–9]. Specially, this drawback limits working temperature and influences the property's temperature stability because thermal depolarization results in the disappearance of macroscopic piezoelectric performance [10]. Thus, the T_d reflects the upper-limit temperature range of operation and represents material figure of merit in practical application as actuators. In order to resolve the above mentioned common drawback, massive attempts using conventional methods on basis of forming solid solution and chemical doping have been exploited for BNT-based lead-free piezoelectric materials [11–21]. To exemplify, the composite of relaxor ferroelectric 0.94BNT-0.06BT and semiconducting ZnO nanoparticles was found to defer the thermal depolarization and enhance the temperature stability [11]. Hao et al. [22–24] investigated the temperature stability of piezoelectric properties and electrostrictive response in BNT-based ceramics by means of chemical doping. Recently, Dong et al. [12] reported that Mn doping BNT-BA ceramics exhibited enhanced ferroelectric properties and thermal stability. However, it should be pointed out that, most of the time, the increase on T_d is usually at the expense of decreased piezoelectric properties, which also restricts their use in practical application. Therefore, for transferral into real-world applications, there is no problem more important than searching for a method to increase T_d and improve piezoelectric properties simultaneously for BNT-based piezoelectric ceramics.

Grain orientation (texturing) is a very useful way of tailoring microstructure and electromechanical performance without doping not influencing phase transition temperatures (Curie temperature T_c and T_d) for electroceramics [5,25–32]. Moreover, prior works have demonstrated that grain orientation techniques can be utilized to pair targeted properties including high piezoelectric properties together with high phase transition temperature [5,33–37]. For example, Yan fabricated highly textured PMN-PT and PMN-PZT ceramics and the textured ceramics possessed extremely high piezoelectric coefficient d_{33} (> 1000pC/N) with good temperature stability [33–35]. The

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enhancement of d_{33} was attributed to domain engineering in < 00l > -oriented ceramics, which was analogous to that for single crystals [38]. In fact, the validity of this approach was proved by a recent work by Yan et al. on a grain-oriented Sm- and Mn-modified PT ceramics showing not only an extremely large piezoelectric voltage coefficient g_{33} (115×10^{-3} V mN^{-1}) but also a high T_C (364 °C) in comparison with random ceramics (T_c = 343 °C and g_{33} = 30×10^{-3} V mN^{-1}) [39]. In the case of BNT-based lead-free ceramics, appreciable improvements in the piezoelectric response and thermal stability have been observed in the textured BNT-BT near MPB with BNT as the seed template and textured BKT-BT-BNT on the tetragonal side with BT as templates [5,37]. Consequently, texturing is a viable method to tune large piezoelectric performance and high T_d in BNT-based electroceramics. Although < 00l > textured BNT-based ceramics had been investigated by some researchers and exhibited relatively high d_{33} compared to the non-textured counterparts, the temperature range of operation is restricted by lower value of T_d [36,40–42], as discussed earlier. Up to now, pairing large piezoelectric properties and high T_d is still challenging and has rarely been explored in BNT-based lead-free piezoceramics, mainly attributed to the fact that the researches mostly focused on doping strategies to optimize properties.

In this study, we provide fundamental aspects in the construction of large d_{33}- high T_d lead-free BNT-based materials by taking advantage of anisotropy and tailored microstructure. We select 0.94BNT-0.06BT (BNT-BT) as a matrix since it exhibits large piezoelectric properties together with relatively high T_d [43]. Previous works reported by Hiruma et al. and Fan et al. on BNT-NN and BNT-BKT-NN systems demonstrate that the introduction of NN into BNT-based ceramics has an obvious impact on electric properties and can effectively enhance the piezoelectric properties [3,44]. Therefore, it is expected that the BNT-BT composition was textured using $NaNbO_3$ (NN) template particles to improve the piezoelectric properties and thermal stability simultaneously. Here, we report < 00l > oriented BNT-BT-NN ceramics to demonstrate the feasibility of microstructure control in pairing superior piezoelectric performance and enhanced thermal stability.

2. Experimental procedure

For texturing, NN platelet templates were synthesized by topochemical microcrystal conversion method [26,40]. < 00l > textured BNT-BT ceramic samples were prepared by a cost-effective texturing approach named templated grain growth (TGG) using 1 mol% NN templates, abbreviated as BNT-BT-NN. The TGG texturing process for the fabrication of textured ceramics is presented in detail elsewhere [37,40,41]. For comparison, randomly oriented BNT-BT-NN ceramics were synthesized by a conventional solid state reaction method.

The crystal phase and microstructure of sintered ceramics were determined by X-ray diffraction (XRD, Bruker D8 Advanced, Germany) with Cu Ka radiation and scanning electron microscopy (SEM) (JSM, EMP-800; JEOL, Tokyo, Japan). The degree of < 00l > texture was achieved from the XRD pattern in 2θ range of 20–60° by Lotgering method [37]. To conduct the electrical experiment, both sides of the specimens with the thickness of ~0.5 mm were coated with silver paste on the flat faces and fired at 600 °C for 10 min. The ceramics were poled in a silicone oil bath under a dc field of 60 kV/cm at room temperature for 20 min. The dielectric properties of poled samples were conducted as a function of temperature and frequency by a high-precision LCR meter (HP 4284A; Agilent, Palo Alto, CA) with a heating rate of 1.5 °C/min. Thermally stimulated depolarization currents (TSDC) were utilized to determine T_d using a Keithley 6517A ampere meter (Keithley Instruments, Cleveland, OH) at the same heating rate. The polarization hysteresis loops and strain curves were tested at 10 Hz by using a FE test system (Precision Premier II; Radiant Technologies Inc, Albuquerque, NM) connected with a Miniature Plane-mirror Interferometer and the accessory Laser Interferometric Vibrometer (SP-S 120/500; SIOS Me b technik GmbH, llmenau, Germany). The *ex situ* thermal depoling experiments were conducted on the poled samples which are annealed at a selected temperature for 30 min to minimize measurement artifacts, and then the temperature were removed. The d_{33} was measured using a quasi-static d_{33} meter (Institute of acoustic, Chinese Academic Society, ZJ-6A, Beijing, China) when the specimens cooled to room temperature.

Table 1
Comparison of piezoelectric properties and phase transition temperature of BNT-BT-based lead-free materials.

Materials	d_{33} (pC/N)	T_d (°C)	Remarks	Refs.
BNT-6BT	110	–	CSSR	[43]
BNT-6BT	148	73	CSSR	[45,46]
BNT-BT-BNiT	160	39	CSSR	[47]
BNT-5.5BT	200	105	TGG	[48]
BNT-7.5BT	202	140	RTGG	[36]
BNT-8BT	98	116	RTGG	[49]
BNT-7BT	322	90	TGG	[37]
BNT-6BT	299	–	TGG	[42]
BNT-6BT	250	–	TGG	[50]
BNT-BT-BKT	192	165	TGG	[5]
BNT-5BT	280	125	Crystal	[51]
Mn:BNT-BT(Pt)	287	150	Crystal	[52]
BNT-5.6BT	380	130	Crystal	[53]
BNT-6.5BT	212	81	Crystal	[54]
BNT-BT-NN	297	57	TGG	this work

CSSR: conventional solid-state reaction; RTGG: reactive template grain growth.

3. Results and discussion

Some of the well-known BNT-BT-based piezoelectric materials (random ceramics, < 00l > grain oriented ones, and single crystals) and their corresponding piezoelectric response and phase transition temperature are summarized in Table 1. According to these reported data, we compared d_{33} at room temperature and depoling temperature T_d of the TGG BNT-BT-NN samples with those of other reported BNT-BT-based lead-free materials, as presented in Fig. 1. The results reveal that the construction of textured microstructure is a highly effective approach of tuning the piezoelectricity and thermal stability to the desired level. The d_{33} achieved in the < 00l > -oriented ceramics can reach as high as ~297 pC/N at room temperature, which is approximately two times larger than that of the randomly oriented counterparts (d_{33} = 151 pC/N). This giant value of d_{33} for 78% textured samples also exceeds that of many other lead-free BNT-based ceramics and is highly comparable to that of BNT-based single crystals (Table 1 and Fig. 1(a)). Fig. 1(b) shows room-temperature d_{33} as a function of T_d for various BNT-BT-based piezoelectric materials and also for the non-textured form of the present study. Evidently, the T_d magnitude is also observed to increase by a factor of approximately two as compared to its non-textured ceramics. More importantly, the BNT-BT-NN ceramics obtained by the TGG process in this study present giant piezoelectric response along with a reasonably high T_d. Thus, it is safe to say that pairing of giant d_{33} and enhanced thermal stability in BNT-based lead-free ceramics is realized. This performance reported here can promote the development of BNT-based piezoceramics, and has a great significance to the practical application as they couple a large piezoelectricity with improved temperature stability. Here, we prove the feasibility to maximize d_{33} keeping a relative high T_d in BNT-based materials though resorting to texturing method.

XRD patterns of random and textured BNT-BT-NN specimens with different degrees of texturing are presented in Fig. 2(a). Due to the low growth rate of oriented ceramics [5], we optimized dwell time of processing from 10 h to 60 h at 1175 °C to achieve texture development and higher grain orientation. All ceramics (random and textured) were found to possess a ABO_3 perovskite structure with no noticeable secondary phase. With increasing the sintering dwell time for textured ceramics, domination of intensities of < 00l > family peaks can be