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Short communication

Grain size dependent electrostrain in $Bi_{1/2}Na_{1/2}TiO_3$-$SrTiO_3$ incipient piezoceramics

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ABSTRACT

The electrical properties and microstructures of $(1-x)$ $Bi_{1/2}Na_{1/2}TiO_3$–$xSrTiO_3$ (BNT-ST100x, $0.20 \leq x \leq 0.30$) piezoceramics with different grain size distributions were investigated. The critical region separating the ferroelectric from ergodic relaxor is located around $x = 0.26$. Strains up to 0.2% could be achieved under a low driving fields ($E < 2$ kV/mm) in the BNT-ST26 composition resulting in excellent actuating performance of S_{max}/E_{max} >1000 pm/V. The electrostrain was largely depended on the grain size and an increment of strain up to ~38% can be realized by increasing the grain size from 5 μm to 13 μm in the BNT-ST26 samples. The results indicate that the field-induced strain performance of BNT-based incipient piezoelectric ceramics can be tailored via microstructure modifications, an alternative strategy to enhance the electromechanical properties.

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1. Introduction

Global awareness of environmental issues has stimulated exploration of lead-free piezoceramics as an alternative to traditional Pb-based ceramics in recent years [1,2]. In general, there are three kinds of most promising lead-free piezoceramics systems: $BaTiO_3$ [3], $Bi_{1/2}Na_{1/2}TiO_3$ (BNT) [4], and $(K,Na)NbO_3$ [5]. Unfortunately, at this moment, a complete substitution for PZT is still not feasible. However, since different applications demand various requirements, lead-free candidates may play a role in some specific circumstances. For example, multilayer piezoactuators are widely used as positioners with a high resolution of displacement up to 0.1 nm [6], which is based on the electric field-induced-strain behavior. Among the widely investigated lead-free piezoceramics, BNT-based compositions can generate large electric-field-induced strain due to the "non-polar" state to ferroelectric (FE) phase transition [7], demonstrating promising application potential in piezoactuators. However, in BNT-based ceramics, this strain often accompanies with a large driving field (e.g. 8 kV/mm for BNT-BT-KNN sample) [7]; while in multilayer stacked piezoactuator applications, the driving electric field does not exceed 3 kV/mm, but lies around 2 kV/mm in most cases [8]. Therefore, how to improve the actuating performance (S_{max}/E_{max}) under low electric field ($E \leq 3$ kV/mm) appears as an urgent problem.

Efforts have been carried out to reduce the driving field necessary for large strain in BNT-based system. For example, relaxor/ferroelectric composites were constructed in BNKT-BA system by Lee et al. [9], which resulted in a decrease of the driving field from 6 kV/mm to 4 kV/mm with a constant strain of ~0.24%. Another effective method is to utilize texturing technology. With a constant output strain ~0.17%, the corresponding driving field decreased from 6 kV/mm to 3.3 kV/mm when a polycrystalline BNT-BKT sample was transformed into <001>-textured ceramics [10].

Compared with the aforementioned approaches, the BNT-ST system outweighs due to its inherently low driving field ($E \sim 3$ kV/mm), as well as excellent field-induced-strain behavior ($d_{33}^* > 600$ pm/V). The large electrostrain of this system was originally reported by Hiruma et al. [11]; a phase boundary between rhombohedral ferroelectric and pseudocubic paraelectric was found in the vicinity of BNT-ST28, accompanying with a large strain (~0.19%) under $E = 3$ kV/mm. This phenomenon has been ascribed to the core-shell structure in a very recent publication, where the BNT-ST25 sample displayed a large piezocoefficient d_{33}^* ~600 pm/V at 3 kV/mm [12,13]. In the present study, enhanced

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piezoelectric property of d_{33}^* ~1100 pm/V ($E < 2$ kV/mm) was obtained in the BNT-ST26 composition, while the microstructure observation demonstrated homogeneous chemical distribution. Strong grain size effect on the electromechanical properties has been found in BNT-ST ceramics. Our results show that BNT-ST lead-free piezoelectrics with modified microstructures could deliver even larger strain than PZT under even lower driving fields, making the material system a promising candidate for application in piezoactuators.

2. Experimental

BNT-based lead-free piezoceramics with nominal compositions of $(1-x)Bi_{1/2}Na_{1/2}TiO_3$–$xSrTiO_3$ (abbreviated as BNT-ST100x) with $0.20 \le x \le 0.30$ were prepared by solid-state reaction method. For the powder synthesis, oxides and carbonates, Bi_2O_3 (99.9 wt%), TiO_2 (99.0 wt%), Na_2CO_3 (99.8 wt%), and $SrCO_3$ (99.0 wt%), were used as raw materials. The raw materials were mixed according to the desired stoichiometric composition by ball milling for 24 h in ethanol using ZrO_2 balls, and then the slurry was dried and calcined at 840 °C for 2 h. The synthesized powders were ball milled for 24 h in ethanol, then dried and pressed into small disks of 10 mm in diameter, followed by cold isostatic pressing at 200 MPa. Finally, the pellets were placed in a sealed alumina crucible and covered with a powder of same composition before sintered between 1220 °C and 1240 °C for 2 h; the samples with higher ST content favored slightly higher firing temperature [14]. Variations of the grain size of BNT-ST26 composition were achieved by alternating the sintering temperature between 1190 °C and 1270 °C. To obtain both a smaller grain size and high bulk density of the composition, a two-step sintering method was utilized [15], whereby the firing temperature was elevated to 1230 °C with a holding time of 10 min initially and then the temperature was reduced to 1180 °C for 1 h to finalize the densification process.

Secondary electron (SE) and backscattered electron (BSE) images of the samples were observed by a field emission scanning electron microscopy (FESEM, S-7001F; JEOL, Tokyo, Japan). Before performing the SEM, the samples were polished and thermally etched. Average grain size was obtained by measuring the sizes of 150 grains. The electromechanical properties were measured by the strain-field method, using the piezo-measurement system (aixACCT TF Analyzer 1000; aixACCT, Aachen, Germany) with a high voltage amplifier (TREK 610E, 10 kV, TREK Inc., Medina, NY). This measurement involved the application of a bipolar/unipolar triangular voltage waveform with a frequency of 0.1 Hz S_{max}/E_{max} was used to evaluate the electromechanical performance. Investigation of the temperature-dependent dielectric permittivity was carried out in a temperature-regulated chamber with a heating rate of 1 °C/min, which was connected to a precision LCR meter (TH2827, Tonghui, Changzhou, China) at 1 kHz, 10 kHz, and 100 kHz.

3. Results and discussion

Fig. 1 shows the electric-field-dependent strains and actuating performance S_{max}/E_{max} of the PZT-based (0.01PMS-0.99PNNZT) [16] and BNT-based piezoceramics, respectively. Among the investigated BNT-ST system, the BNT-ST26 sample demonstrates the best electric field-induced strain performance among the investigated compositions, indicating its potential capability for actuator applications. This result is consistent with the previous reports [11,12] that the compositions located around this region showed the coexistence of ferroelectrics and ergodic relaxors. Under a critical electric field, the transformation from the nonpolar state to the FE domain delivers the giant strain. As the representative of the investigated compositions, the S–E curve of the BNT-ST26 sample has been shown here, which was compared to that of PZT-based piezoceramics. The BNT-ST26 composition shows inferior field-induced-strain performance compared with the PZT-based sample at low electric field. However, when the field exceeds a critical value (E_{pol} ~ 1.5 kV/mm), the field-drived phase transition from ergodic relaxor to ferroelectric appears, which leads to a sharp increase in the strain and a significantly improvement of the piezoelectric property [20,21]. Under the driving fields between

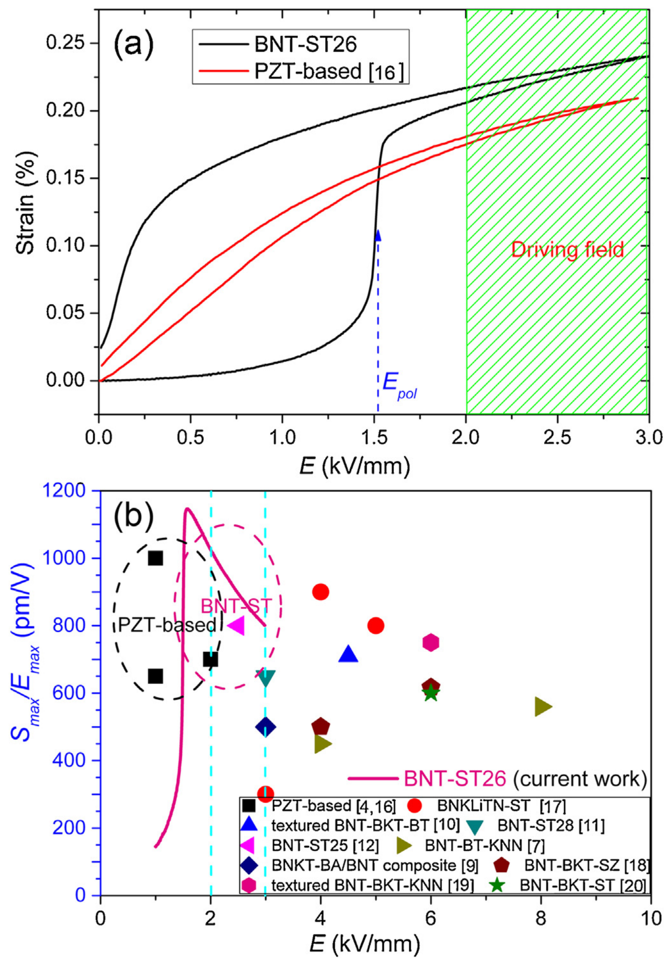

Fig. 1. E-dependent unipolar strains (a) for BNT-ST26 and 0.01PMS-0.99PNNZT, and the actuating performance S_{max}/E_{max} (b) for PZT-based and BNT-based piezoceramics [17–19].

Table 1
Physical properties of the BNT-ST26 samples.

Grain size (μm)	Sintering temperature (°C)	Sintering method	Holding time	Relative density (%)	$(S_{max}/E_{max})_{max}$ (pm/V)	E_{pol} (kV/mm)
5.0	1180	two-step	1 h	96.0	843	1.4
6.4	1190	normal	2 h	95.9	921	1.3
8.1	1210	normal	2 h	97.5	1042	1.4
10.5	1230	normal	2 h	98.0	1100	1.5
13.0	1270	normal	2 h	98.2	1163	1.5
2.2(ST25) Ref. [12]	1150	normal	2 h	~96.0	~810	2.5

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