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Giant piezoelectricity and ultrahigh strain response in bismuth sodium titanate lead-free ceramics

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ABSTRACT

The obvious conflicts between large piezoelectricity and high strain could be solved by adding new components in BNT. Moreover, this urgent problem has been solved and induced a large strain as well as a high piezoelectricity in BNT. Large normalized strain (d_{33}^{*}=526–597 pm/V) and high strain (0.342%–0.388%) were achieved at the composition range of $0.0025 \le x \le 0.0050$, suggesting that such a system is a promising lead-free candidate for electromechanical actuator applications. Furthermore, high d_{33} values of 141–204 pC/N have also been attained at the above composition range due to adding new components.

1. Introduction

High-performance piezoceramics are some of the most paramount and widely used materials, and as such research into this topic is at the forefront of high-technology advanced materials [1]. $Pb(Zr_{1-x}Ti_x)O_3$(PZT)-based for many years have been the most widely used materials in piezoelectric and ferroelectric application owing to their superior piezoelectric properties [2]. However, increased cognizance for environmental safety has commenced considerable efforts to hamper the amount of hazardous substances in consumer products [3]. The research and development of lead-free piezoelectric materials have captured many countries' attention for protecting environment and human health.

Many attempts have been made to develop a new lead-free piezoceramics with desired properties [4,5]. However, there are few major breakthroughs in the piezoelectricity of BNT-based ceramics. In an effort to search for a high d_{33} in BNT, the MPB always plays a major and constructive role [3,6,7]. Extensive experiments have confirmed that a high d_{33} value can be attained in BNT-based ceramics by adding the second compositions at this boundary [8–11]. The electric field-induced strain of lead-free ceramics has recently become a new highlight because their strain values can exceed or are comparable to those of lead-based ceramics [12]. The coexistence of ferroelectric and relaxor ferroelectric order can induce large strain. However, as shown in Fig. 1, many researchers are confused by the problem of the unbalanced development of both d_{33} and strain [13–19]. An enhancement in d_{33} (141 pC/N) and d_{33}^{*} (597 pm/V) could also be observed in the Sr_2ZrMnO_6 modified BNT-based ceramics (see Fig. 1), showing the possibility of promoting both d_{33} and strain. Consequently, in order to achieve large scale applications of piezoelectric actuators, the target of this work is to promote both d_{33} and strain by constructing the coexistence state of ferroelectric and relaxor ferroelectric phases around MPB compositions.

In this work, a new lead free piezoceramics of $(1-x)(Bi_{0.5}Na_{0.5})_{0.935}Ba_{0.065}TiO_3$-$xSr_2ZrMnO_6$ (BNBT- x SZMO) was prepared by a conventional solid-state reaction method and the compositional dependence of the structure and electrical properties of all samples were investigated.

2. Materials and methods

The BNBT- x SZMO (x=0–0.010) piezoelectric ceramics were synthesized by conventional solid-state reaction method using analytical-grade metal oxide or carbonate powders as raw materials. These starting materials were weighed according to stoichiometric formula and ball milled for 12 h in ethanol. The dried slurries were calcined at 850 °C for 2 h and then ball milled again for 6 h. The powders were subsequently compacted into disk-shape and sintered at 1140 °C for 2 h in covered alumina crucibles. Structure and electrical characterizations are similar as that reported elsewhere [20].

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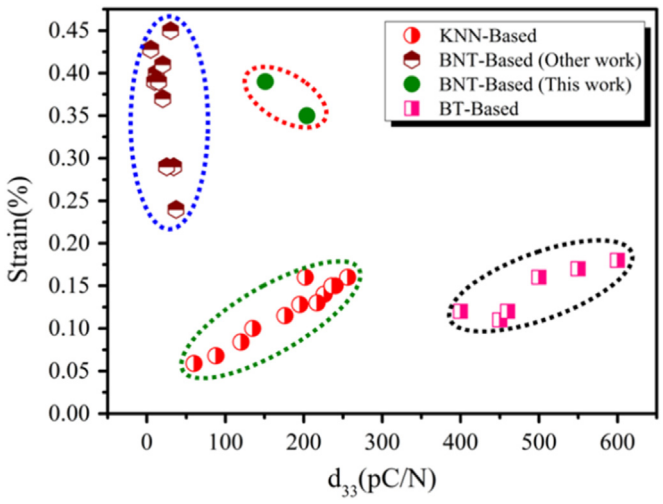

Fig. 1. Compared analysis of strain and piezoelectricity in lead-free piezoelectric materials.

3. Results and discussion

The XRD patterns with a 2θ ranging from 20° to 70° for BNBT- x SZMO samples are demonstrated in Fig. 2(a). All ceramics exhibited a pure perovskite structure without any traces of secondary phases. All peaks can be indexed based on the standard diffraction XRD of the tetragonal $(Bi_{0.5}Na_{0.5})TiO_3$ with P4bm space group (JCPDS#70-4760). Expanded XRD patterns in the 2θ range of 31°–34° is presented in Fig. 2(b). Obviously, the peaks position of BNBT- x SZMO solid solutions shifted to a lower angle with an increase of SZMO content. This could be attributed by the slightly expansion of the crystal lattices, which is caused the substitution of large Sr^{2+} ion (r=1.18 Å) for $(Bi_{0.5}Na_{0.5})^{2+}$ ($r_{(Bi_{0.5}Na_{0.5})^{2+}}$=1.025 Å) small ions on the A-site and Zr^{4+}, Mn^{4+} ($r_{(Zr\ Mn)}^{8+}$=0.625 Å) for Ti^{4+} (r=0.605 Å) on the B-site, respectively [21]. Similar results were also reported in BZ-modified [22] BNBT and BST-modified [23] BNT ceramics.

Room-temperature ferroelectric hysteresis loops (P–E) of BNBT-x SZMO (x=0–0.010) ceramics measured at 10 Hz are shown in Fig. 3(a). The P–E shape strongly rests with the composition of the ceramics. The characteristic values of E_c and P_r can be seen in the inset of Fig. 3(d). The P–E loop of pure BNBT displays a well-saturated typical ferroelectric behavior with a coercive field (E_c) of 40.6 kV/cm and a remnant polarization (P_r) of 29.0 μC/cm^2, respectively. At x=0.0050, E_c and P_r drop drastically down to 15.6 kV/cm and 22.6 μC/cm^2, whereas the maximum polarization (P_m) decreases only a little. It indicates that although the ferroelectric order of BNBT is disturbed by the SZMO doping, the free energy of the ferroelectric phase in BNBT appears so competitive with that of the relaxor ferroelectric phase at zero fields that it can be easily induced by an external electric field.

The field-induced strain level, piezoelectric constant d_{33} and normalized strain of BNBT- x SZMO ceramics are presented in Fig. 3(b). A maximum unipolar strain value of 0.388% is observed at x=0.005, and the corresponding normalized strain ($d_{33}^{*}=S_{max}/E_{max}$) attains a maximum value of 597 pm/V, giving a relatively large piezoelectric constant d_{33} of 141 pC/N. At x=0.0025, the materials exhibit a large d_{33} of 204 pC/N, and attains a relatively high strain of 0.342% along with normalized strain S_{max}/E_{max} of 526 pm/V (see Fig. 1). The origin of an enhancement in d_{33} and S_{max}/E_{max} could be attributed to the coexistence of ferroelectric and relaxor ferroelectric order.

The compositionally induced ferroelectric to relaxor-ferroelectric phase transformation can be well verified by the bipolar strain measurements (see Fig. 3(c)). Samples such as x=0 and x=0.0025 show butterfly shaped strain hysteresis loops with large negative strain which is typical for normal ferroelectrics. With increasing x, the measured strain increases remarkably, reaching its peak (S_{max}=0.39%) at x=0.0050, and then decreases slightly. While S_{neg} increases before x=0.0050 and then drops down steeply. The large strain in x=0.0050 suggests that the reduction in S_{neg} is interrelated with domain back-switching during bipolar cycling. Consequently, the significant enhancement in bipolar strain can be attributed to the ferroelectrics to relaxor ferroelectrics phase transition, which is supported by the P–E loops shown in Fig. 3(a).

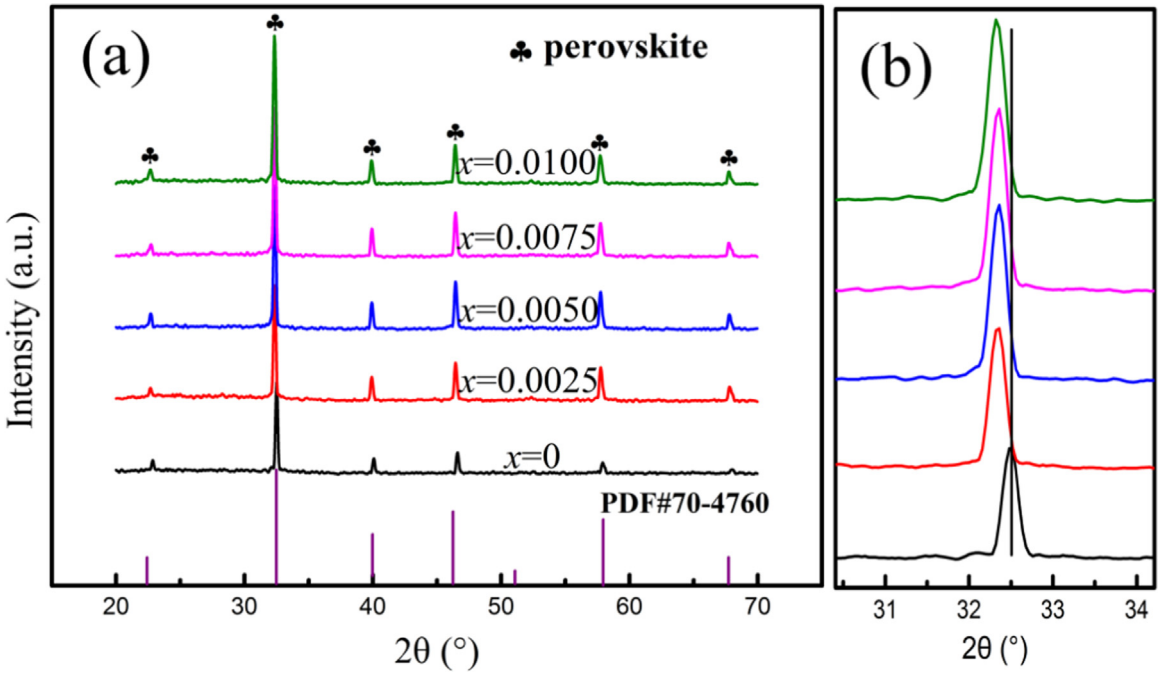

Fig. 2. XRD patterns of BNBT- x SZMO ceramics in the 2θ range: (a) 20°–70° and (b) 31°–34°.