

Structures and temperature-dependent electrical properties in $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3\text{--BiAlO}_3$ electrostrictive ceramics

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Abstract

Lead-free piezoceramics of $(1-x)\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3\text{--}x\text{BiAlO}_3$ [(1-x)BST-xBA ($0 \leq x \leq 0.12$)] have been synthesized and the structures and properties have been investigated systemically. X-ray diffraction patterns indicated that the solid solution limit is close to $x=0.04$, and a morphotropic phase boundary (MPB) separating tetragonal and pseudocubic phases exists near $x=0.02$. The ferroelectricity weakens monotonously with increasing x , accompanied by weakened butterfly shaped bipolar strain–electric field curves. The temperature dependent properties of the composition with $x=0.02$ have been typically investigated, showing that with increasing temperature, the ferroelectricity tends to be weakened. Pure electrostrictive effect has been found in the compositions with $x=0.02$ and 0.03 close to room temperature. Based on the results, the effects of BA on structures and electric properties were discussed.

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1. Introduction

At present most widely used piezoelectric materials are lead-based, such as $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$ (PZT). The lead-based materials have applications in actuators, sensors, and transducers due to their excellent electromechanical properties [1]. However, lead is toxic and its use is banned in many commercial applications [2]. To replace lead-based piezoelectric materials, huge efforts have been devoted to the search for lead-free counterparts with comparable properties. At present no lead-free piezoelectric materials can display comparable properties. Therefore, an alternative way is suggested to classify the required piezoelectric properties for various applications and develop lead-free piezoelectric ceramics targeted for each application accordingly [3].

For actuator applications, the generally requirements is high strain because $e_{\text{max}} \propto s_{\text{max}}^2$, where e_{max} is the strain energy, s_{max}

is the maximum field induced strain [1]. The common method to develop high strain materials is to form the morphotropic phase boundary (MPB) which generally shows improved strain [1–5]. However, the strain–electric field (S – E) curves of the reported MPB compositions still have hysteresis, which is detrimental for improving the fine position controlling, reducing energy consuming, etc., of electronic devices.

Actually, electrostrictive materials have hysteresis-free characteristics and, therefore, are of particular interest for actuator application [6]. But electrostrictive effect is very weak in most materials and the typical strain level is less than 0.10% [7]. However, in some relaxor ferroelectrics, the electrostrictive strain can reach 0.10–0.15%. Up to now, the reported lead-free electrostrictive materials are generally based on $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ [7,8]. One problem related with these BNT-based electrostrictive materials is that, extremely high field (larger than 8 kV/mm) is necessary to obtain $\sim 0.1\%$ strain level. The high field is one of the obstacles preventing its application. We note that recent theoretical and experimental works indicate modified BaTiO_3 (BT) based materials can also show electrostrictive effect with strain level of $\sim 0.1\%$ at low applied

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electric field [9,10], which means that it might be interesting to search for BT-based electrostrictive materials.

Barium strontium titanate, $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$, is a solid solution composed of barium titanate and strontium titanate. $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ has attracted much electronic interests mainly due to its high dielectric constant, alterable Curie temperature, low dielectric loss, high dielectric tunability, etc. Most of the reported works focused on the effects of Ba/Sr composition [11–16], etc., on dielectric properties, but few of them on its electrostrictive effect. It is noted that $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ (BST) has been reported to have tetragonal phase, and especially, show strong relaxations in terms of frequency dependence of the dielectric properties [12,14]. That means electrostrictive effects might be observed in BST-based materials.

On the other hand, an increasing attention has been paid to BiAlO_3 (BA) due to its large spontaneous polarization. However, it is known that the extremely structural instability has made BA difficultly available under normal conditions [17]. Moreover, its real crystal structure is still vague, it is being considered to own rhombohedral structure through theoretical calculations, but a tetragonal structure from experimentally prepared films [18]. Therefore, attempts have been made to stabilize BA by forming solid-solution with other stable perovskite materials, which results in a synergic effect that leads to a significant changes in structural and electrical properties [19,20].

Based on the above description, we have designed and prepared the solid solutions of $(1-x)\text{BST}-x\text{BA}$ with $x = 0, 0.01, 0.02, 0.03, 0.04, 0.06, 0.09$ and 0.12 . The solid solution limit, crystal structure, electrical properties and its temperature dependence have been investigated and discussed.

2. Experimental procedure

The $(1-x)\text{BST}-x\text{BA}$ ($x = 0, 0.01, 0.02, 0.03, 0.04, 0.06, 0.09$ and 0.12) ceramics were prepared using solid-state reaction method. Oxide and carbonate powders of BaCO_3 (99.8%), SrCO_3 (99.8%), TiO_2 (99.9%), Bi_2O_3 (99.8%) and Al_2O_3 (99.0%) were used as starting materials. Before being weighed, these powders were separately dried in an oven at 60°C for 24 h. For each composition, the oxides and carbonates were weighed according to the stoichiometric formula of $(1-x)\text{BST}-x\text{BA}$ with $x = 0, 0.01, 0.02, 0.03, 0.04, 0.06, 0.09$ and 0.12 . The mixtures were ball milled for 24 h in ethanol, the dried slurries were calcined at 900°C for 3 h in air and then ball milled again for 12 h. The dried powders were pressed into disks of 10 mm in diameter under a uniaxial pressure of 40 MPa using polyvinyl alcohol (PVA) as a binder. The pellets were sintered at $1250\text{--}1350^\circ\text{C}$ for 3 h in sealed Al_2O_3 crucibles. For electrical measurements, the sintered disks were grounded carefully to ensure the parallel surfaces. The circular surfaces of the disks were covered with a thin layer of silver paste and fired at 500°C for 30 min in air.

X-ray diffraction (XRD, Rigaku UltimaIII) and scanning electron microscope (SEM, XL30 Philips) were used for structure characterizations. The XRD measurements were carried on crushed, unpoled samples. The polarization–electric field (P – E) hysteresis loops and the strain–electric field (S – E)

curves were measured by TF Analyzer 1000 (AixACCT, Germany) in silicone oil with controllable temperature.

3. Results and discussions

Fig. 1(a) shows the XRD patterns of the $(1-x)\text{BST}-x\text{BA}$ ceramics. As can be seen, the ceramics have been crystallized to pure perovskite phase and no secondary phases can be identified when $x < 0.04$, indicating that the BA have completely diffused into the BST lattice to form solid solution. However, a second phases, indicated by the arrows and identified as $\text{Bi}_2\text{Al}_4\text{O}_9$ [10], tends to develop as $x \geq 0.04$. This means the solution limit is close to $x = 0.04$, which is significantly lower than that of $(1-x)\text{BaTiO}_3-x\text{BiAlO}_3$ system [20], one of the possible reasons is that BST and BaTiO_3 , though have same tetragonal crystal structure, have different lattice parameter and thus different accommodation for introduction of rhombohedral metastable BiAlO_3 . On the other hand, it is clear that with increasing x from 0 to 0.02, some diffraction peaks, e.g., that locating at $2\theta = 45.5^\circ$ in Fig. 1(a), become coalesced. Typically detailed composition-dependent XRD patterns near $2\theta = 45.5^\circ$ are plotted in Fig. 1(b). For the composition with $x = 0$ and 0.01, the diffraction peaks are split, which is the fingerprint of tetragonal structure. With x is increased to 0.02, these split peaks are coalesced into one peak with a weak shoulder at lower-angle side. With further increasing x , the shoulders tend to disappear, suggesting that the crystal structure of the $(1-x)\text{BST}-x\text{BA}$ ceramics evolves from the tetragonal to a possible pseudocubic symmetry, in other words, a tetragonal–pseudocubic MPB locates near $x = 0.02$.

Fig. 2 typically shows the SEM micrographs of the $(1-x)\text{BST}-x\text{BA}$ ceramics with $x = 0.01, 0.02, 0.03$, and 0.04 . The SEM observations confirm that all samples are dense with well-developed microstructure and grain morphologies. The average grain size is about $1\ \mu\text{m}$. No significant composition dependent grain size and microstructural morphologies can be observed, though the composition with $x = 0.04$ has second phases, which might indicate that the ratio of second phases is very low.

Electrical measurements have been carried out on all samples, however, it is found the composition with $x = 0$ is

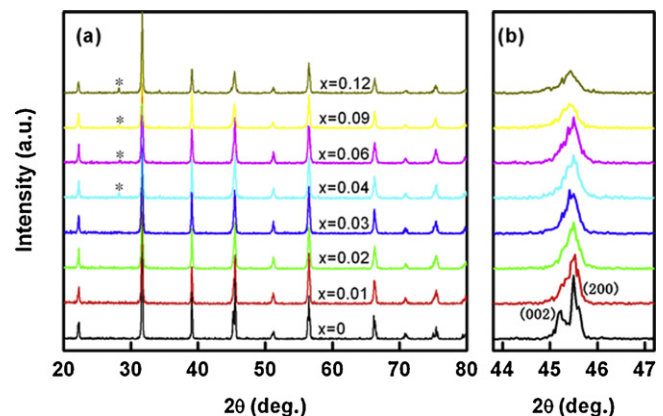


Fig. 1. (a) XRD patterns of all $(1-x)\text{BST}-x\text{BA}$ ceramics. (b) The local detailed XRD patterns, showing a phase transition.

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