



Piezoelectric properties and thermal stabilities of strontium bismuth titanate (SrBi₄Ti₄O₁₅)

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Abstract

Bismuth layer-structured ferroelectric (BLSF) compound strontium bismuth titanate (SrBi₄Ti₄O₁₅, SBT) with cerium modifications have been synthesized using conventional solid-state processing. X-ray powder diffraction (XRPD) analyses reveal that the cerium-modified SBT ceramics have a pure four-layer Aurivillius-type structure. The dielectric, ferroelectric, and piezoelectric properties of the cerium-modified SBT ceramics are investigated in detail. The results indicate that the cerium modifications into SBT increase the densities, decrease the sintering temperature, lower the dielectric loss $\tan\delta$, and reduce the coercive field E_c . The piezoelectric measurements show that cerium is very effective in promoting the piezoelectric properties of SBT ceramics. The SBT ceramics modified with 4 mol% CeO₂ (SBT-4Ce) exhibit the optimized piezoelectric properties, with a piezoelectric constant d_{33} of 27 pC/N, which is the highest value among the modified SBT-based piezoelectric ceramics ever reported. The temperature-dependent electrical impedance and electromechanical coupling coefficients (k_p and k_t) reveal that the electro-mechanical coupling characteristics have a significant deterioration at ~ 400 °C because of the high conductivity at high temperature. But the temperature-dependent frequency constants (N_p and N_t) and thermal annealing analyses indicate the cerium-modified SBT ceramics have good thermal stabilities of piezoelectric properties up to 450 °C. These results demonstrate that the cerium-modified SBT ceramics are promising materials for high temperature piezoelectric sensors applications.

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1. Introduction

Bismuth layer-structured ferroelectric (abbreviated as BLSF) oxides, first reported by Aurivillius [1–3], are one of the most important ferroelectric compounds, which are known to possess a structure expressed by the general formula $(\text{Bi}_2\text{O}_2)^{2+}(\text{A}_{m-1}\text{B}_m\text{O}_{3m+1})^{2-}$, where A is a monovalent, divalent, or trivalent metallic ion (or their combination) suited to dodecahedral coordination; B is a tetravalent, pentavalent, or hexavalent metallic ion suited to octahedral coordination; and m is an integer that represents the number of BO_6 octahedra in $(\text{A}_{m-1}\text{B}_m\text{O}_{3m+1})^{2-}$ between the $(\text{Bi}_2\text{O}_2)^{2+}$ layers [4]. BLSFs are attractive electronic materials because of their low dielectric

constant, high dielectric breakdown strength, high Curie temperature T_c , low temperature coefficient of resonance frequency, strong anisotropy, and low ageing rate [5]. These advantages enable the use of BLSFs as piezoelectric materials that can be operated at high temperatures and high frequencies [5–7].

Recently several BLSF ceramic oxides have attracted extensive attention for their potentials in high-temperature piezoelectric sensors applications. Such BLSF ceramic oxides include $\text{CaBi}_2\text{Nb}_2\text{O}_9$ (CBN, $m=2$) [8–10], $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ (BTO, $m=3$) [11–13], $\text{Na}_{0.5}\text{Bi}_{4.5}\text{Ti}_4\text{O}_{15}$ (NBT, $m=4$) [14–16], and $\text{CaBi}_4\text{Ti}_4\text{O}_{15}$ (CBT, $m=4$) [17–19]. However, these prototype BLSF oxides are greatly difficult to be poled, and they possess very low piezoelectric properties. These undesirable properties arise from the restricted two-dimensional rotation orientation of their spontaneous polarizations (P_s) and their high coercive fields (E_c). To overcome these shortcomings, many efforts have been

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taken to enhance the piezoelectric properties of these BLSF oxides by some additives modifications [8–20]. These investigations have indicated that *A*-site substitutions are more effective than *B*-site substitutions in enhancing the piezoelectric performance of BLSF oxides, which stem from the fact that the cations in *B*-site have the similar in size and hardly make a major contribution to the polarization process for BLSFs [21]. Consequently, most reports on enhancing the ferroelectric and piezoelectric properties of BLSF ceramics are concentrated on *A*-site rather than *B*-site modification [8–20].

Since it has been reported that the *A*-site cerium-modified BLSF $\text{Na}_{0.5}\text{Bi}_4\text{Ti}_4\text{O}_{15}$ ceramics possess remarkable enhanced piezoelectric performance, with a piezoelectric constant d_{33} of 25 pC/N [14], several BLSF oxides were investigated by *A*-site cerium modifications to improve the piezoelectric properties of BLSF ceramic oxides. Such cerium-modified BLSF oxides include $\text{CaBi}_2\text{Nb}_2\text{O}_9$ [9,10], $\text{BaBi}_4\text{Ti}_4\text{O}_{15}$ (BBT), and $\text{K}_{0.5}\text{Bi}_{4.5}\text{Ti}_4\text{O}_{15}$ (KBT) [22]. And all these BLSF oxides show enhanced piezoelectric performance by *A*-site cerium modifications, indicating that cerium is a very effective modifier in improving the piezoelectric performance of BLSF oxides. Among these BLSF oxides, the cerium-modified CBN piezoelectric ceramics possess enhanced piezoelectric constant d_{33} values of ~ 16 pC/N and high Curie temperature T_c of > 900 °C, while the BBT and KBT piezoelectric ceramics possess remarkable increased piezoelectric properties, with piezoelectric constant d_{33} values of ~ 24 pC/N and ~ 28 pC/N respectively, but their Curie temperature T_c is on the order of 500 °C.

The Aurivillius-type strontium bismuth titanate $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ (SBT), which has a high Curie temperature T_c of ~ 520 °C, is a member of the Aurivillius-type family, BLSF oxides. Structurally, SBT has orthorhombic symmetry with space group $A2_1am$ [23], containing four perovskite-like TiO_6 octahedron units stacked in between $(\text{Bi}_2\text{O}_2)^{2+}$ layers. Electrically, the prototype SBT ceramics present a low piezoelectric constant d_{33} of 10–13 pC/N, as shown in previous studies [24]. To improve the piezoelectric performance of SBT ceramics, the piezoelectric properties of metal oxides modified SBT ceramics have been investigated extensively. For example, the Dy-modified SBT (SBT-Dy) has a piezoelectric constant d_{33} value of 17 pC/N [25], the Er-modified SBT (SBT-Er) shows a d_{33} value of 16 pC/N [26], and the Ho-modified SBT presents a d_{33} value of 15 pC/N [27]. These results revealed that the rare-earth oxides can enhance the piezoelectric properties of SBT ceramics. However, compared with other modified BSLF oxides with similar Curie temperature T_c , > 500 °C, the piezoelectric performance of modified SBT ceramics are still not very high. To further improve the piezoelectric performance of SBT, in the present work, the dielectric, ferroelectric, and piezoelectric properties of *A*-site cerium-modified layer-structured SBT ceramics were investigated based on the above-mentioned available facts [24–27]. Furthermore, the temperature dependence of the frequency resonant characteristics, electromechanical coupling properties, and piezoelectric properties at elevated temperature were investigated in detail for high temperature piezoelectric applications. Additionally, the underlying physical mechanisms for enhanced piezoelectricity were also addressed.

2. Experimental procedure

Conventional mixed oxide processing route procedure was used to prepare strontium bismuth titanate $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ (SBT) and cerium-modified SBT piezoelectric ceramics. Analytical grades SrCO_3 (99.0%; Sinopharm Chemical Reagent Co. Ltd., SCRCL), Bi_2O_3 (99.8%, Chengdu Shudu Nano-Science Co. Ltd.), TiO_2 (99.8%, SCRCL), and CeO_2 (99.9%, SCRCL) were selected as starting materials. The compositions investigated in the present work were $\text{Sr}_{1-x}\text{Ce}_x\text{Bi}_4\text{Ti}_4\text{O}_{15}$ ($x=0.00, 0.02, 0.04, 0.06, 0.08$, abbreviated as SBT-100 x Ce, i.e. SBT, SBT-2Ce, SBT-4Ce, SBT-6Ce, and SBT-8Ce). The weighed chemicals were wet milled in polyethylene bottles with ZrO_2 balls for 12 h in ethanol. The milled powders were dried and calcined at 800 °C for 3 h. After calcinations, the mixture was milled again in the same conditions. The milled powders were dried, grinded, and granulated with polyvinyl alcohol (PVA) binder. The granulated powder was pressed into disks 12 mm in diameter by 1.0 mm in thickness. To prevent evaporation of Bi ions and keep stoichiometry, the green compacts were put into the sealed Al_2O_3 crucibles and fully surrounded with the powder of matching compositions. By the ordinary firing, the samples were sintered at 1080–1140 °C for 3 h, and then cooled to room temperature freely.

The densities of the sintered samples were determined by applying the Archimedes method. The X-ray powder diffraction (XPRD) patterns were obtained with an X-ray diffractometer (D8 Advance, Bruker AXS GMBH) using $\text{CuK}\alpha$ radiation. Samples for electrical measurements were polished parallel to 0.5 mm in thickness, and silver electrodes were screen-printed on both surfaces of the polished samples and fired at 600 °C for 30 min. Samples were poled in silicone oil under a DC electric field of 100–140 kV cm^{-1} at 180 °C, and then the piezoelectric constant d_{33} was measured using a quasi-static d_{33} meter (Sinoceramics, Inc. YE2730A). A modified Sawyer–Tower circuit and linear variable differential transducer driven by a lock-in amplifier (Stanford Research System, model SR830) were used to measure the polarization hysteresis loops. The dielectric spectra measurements were performed with 4294A impedance analyzer (Agilent Technologies, Santa Clara, CA) as a function of temperature. The resonance and antiresonance frequencies were obtained from 4294A impedance analyzer. The electromechanical coupling factors (k_p, k_t), and frequency constant (N_p, N_t) were calculated according to Onoe's equations [28]. Thermal annealing experiments were conducted by holding the poled samples at specific temperatures for 1 h. Finally, the thermal annealed sample was cooled to room temperature, and d_{33} was subsequently measured.

3. Results and discussion

Fig. 1 presents the XRPD patterns of the unmodified and cerium-modified SBT ceramic pulverized powder with different cerium contents. All the XRPD patterns conform with the diffraction data of SBT [29], which confirms the presence of a single phase $m=4$ BLSF structure. The diffraction peak with

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