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Preparation and electrical properties of $(1-x)SrBi_2Nb_2O_9-xBiFeO_3$ lead-free piezoelectric ceramics

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Abstract

Lead-free piezoelectric ceramics, $(1-x)SrBi_2Nb_2O_9-xBiFeO_3$ $[(1-x)SBN-xBFO]$ ($x=0.0$, 0.03, 0.05, 0.07, 0.10) were prepared by a conventional solid-state reaction method. The crystal structure, microstructure and electrical properties were systematically investigated. All compositions formed layered perovskite structure without any detectable secondary phases. Plate-like morphology of the grains which is characteristic for layer-structure Aurivillius compounds was clearly observed. The excellent electrical properties (e.g., $d_{33} \sim 19$ pC/N, $2P_r \sim 18.8$ μC/cm^2) and a high Curie temperature (e.g., $T_c \sim 449$ °C) were simultaneously obtained in the ceramics with $x=0.05$. Additionally, thermal annealing studies indicated that the BFO modified SBN ceramics system possessed stable piezoelectric properties, demonstrating that the modified SBN-based ceramics are the promising candidates for high-temperature applications.

Keywords: C. Electrical properties; Piezoelectric ceramics; Solid-state reaction; X-ray diffraction

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1. Introduction

Bismuth layer-structured ferroelectrics (BLSFs), known as Aurivillius family of oxides, have been investigated in high temperature piezoelectric device and ferroelectric non-volatile random access memory (FRAM) storage devices because of their attractive electrical properties such as high Curie temperature, low dielectric loss, excellent fatigue endurance and low aging rate [1–3]. Compared with traditional perovskite structure, the structure of these ferroelectrics, represented as $(Bi_2O_2)^{2+}(A_{m-1}B_mO_{3m+1})^{2-}$, consists of $[Bi_2O_2]^{2+}$ layers interleaved with perovskite block of $[A_{m-1}B_mO_{3m+1}]^{2-}$ units along the *c*-axes, where the *A*-site in the perovskite block can be occupied by 12-coordinated cations such as Bi^{3+}, La^{3+}, Ba^{2+}, Sr^{2+}, Ca^{2+}, etc, and the *B*-site can be occupied by 6-coordinated cations such as Ti^{4+}, Nb^{5+}, Ta^{5+}, W^{6+}, Mo^{6+}, Co^{3+}, etc. [4–6]. In addition, the *m*, in the range of 1–5, represents the number of perovskite blocks, and strongly influences the ferroelectric and dielectric properties of BLSFs [7–9].

Among numerous BLSFs, $SrBi_2Nb_2O_9$ (abbreviated as SBN), as the $m=2$ member of the Aurivillius family with Sr and Bi ions at the *A* sites and Nb ions at the *B* sites of the perovskite $(A_{m-1}B_mO_{3m+3})^{2-}$ block $((Bi_2O_2)^{2+}$ $(SrNb_2O_7)^{2-})$, has drawn a special attention because of its less distortion, low dielectric loss and low aging rates compared to other bismuth layer Aurivillius oxides [10,11]. In spite of several advantages of SBN ceramic, as other Aurivillius phase materials, it still suffers from several defects such as a relatively low remanent polarization and piezoelectric coefficient. To improve their piezoelectric and ferroelectric behavior, Sugandha et al. [12] and Wei et al. [11], who introduce Pr^{3+} to substitute for Bi^{3+}, and Er^{3+} to substitute for Sr^{2+}, respectively, observed the enhancement of dielectric and ferroelectric properties, together with a decline in sintering temperature.

Multiferroic, $BiFeO_3$ (BFO) is a promising lead-free piezoelectric material applied in multilayered electronic devices due

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to its high T_c (~830 °C) and ferroelectric–antiferromagnetic coupling effect at room temperature [13,14]. Gu et al. [15,16] reported a significant dielectric enhancement in BFO-modified $SrBi_2Nb_2O_9$ ceramics. Based on the above, we fabricated lead-free $(1-x)SrBi_2Nb_2O_9-xBiFeO_3$ ceramics [$(1-x)$SBN–xBFO] and expected to achieve improved ferroelectric piezoelectric properties in the present work. The $(1-x)$SBN–xBFO were synthesized by conventional solid-state reaction method over a wide range of compositions ($x=0.0$–0.10) and their structure, dielectric, piezoelectric and ferroelectric properties were investigated systematically. The structure–property relationships and possible mechanism are additional discussed.

2. Experimental

$(1-x)SrBi_2Nb_2O_9$–$xBiFeO_3$ [$(1-x)$SBN–xBFO] ($x=0.03$, 0.05, 0.07, 0.10) ceramics were prepared by conventional solid-state reaction method using reagent-grade metal oxides or carbonate powders of Bi_2O_3 (99.99%, Sinopharm Chemical Reagent Co., Ltd., China), $SrCO_3$ (99%, Sinopharm Chemical Reagent Co., Ltd., China), Nb_2O_5 (99.5%, Sinopharm Chemical Reagent Co., Ltd., China) and Fe_2O_3 (99.49%, Sinopharm Chemical Reagent Co., Ltd., China) as starting materials. All raw materials were weighed at stoichiometric proportion and then mixed homogenized by planetary ball milling in a polyethylene with stabilized zirconia balls for 15 h, using deionized water as liquid medium. After drying, the mixed powders were calcined at 800 °C for 2 h. After calcination, the mixture was milled again for 12 h. The powders were mixed with an appropriate amount of polyvinyl butyral (PVB) binder, and pressed into pallets with a diameter of 12 mm and a thickness of 0.7 mm under the pressure of about 10 MPa. After burning off PVB, the ceramics were sintered in an alumina crucible at 1100 °C for 3 h. For the electric measurements, disk samples with about 0.3 mm in thickness were used.

The density of the sintered ceramics was measured by means of the Archimedes method. The crystal structure of the ceramics was determined by X-ray diffraction (XRD) using a Cu Kα radiation ($\lambda=1.54178$ Å) (D8 Advance, Bruker Inc., Germany). The surface morphology of the ceramics was observed by scanning electron microscope (SEM) (JSM-6380, Japan). The ferroelectric hysteresis loops were measured through standardized ferroelectric test system (TF2000, Germany). The temperature dependence of dielectric properties and impedance spectroscopy for the samples was performed using a Broadband Dielectric Spectrometer (Novocontrol Germany). The samples were polarized in silicon oil in the range of 150–180 °C for 20 min, and piezoelectric measurements were carried out with a quasi-static d_{33}-meterYE2730 (SINOCERA, China).

3. Results and discussion

The XRD patterns of the $(1-x)$SBN–xBFO ceramics in the 2θ range of (a) 20–70° and (b) 28–30° were plotted in Fig. 1. As shown in Fig. 1(a), the ceramic possess a single crystalline-layered perovskite phase without any detectable secondary phases, which indicates that Fe^{3+} and Bi^{3+} has diffused into $SrBi_2Nb_2O_9$ lattices to form a new solid solution (1-x)SBN–xBFO. Meanwhile, it is obvious that the highest diffraction peak of $(1-x)$SBN–xBFO ceramics is (115) orientation, which is consistent with the highest diffraction peak of $(11\underline{2m}+\underline{1})$ in BLSFs phase [17] and belongs to orthorhombic structure (JCPDS 49-0607). Fig. 1(b) shows the enlarged XRD patterns of the ceramics in the 2θ range of (b) 28–30°. The (115) peaks gradually move towards the higher angles with increasing content of $BiFeO_3$, which is owing to the relatively smaller ionic radius of Fe^{3+} (0.55 Å) than that of Nb^{5+} (0.69 Å), and smaller ionic radius of Bi^{3+} (1.03) than that of Sr^{2+} (1.18 Å) [18], indicating the shrinkage of cell volumes.

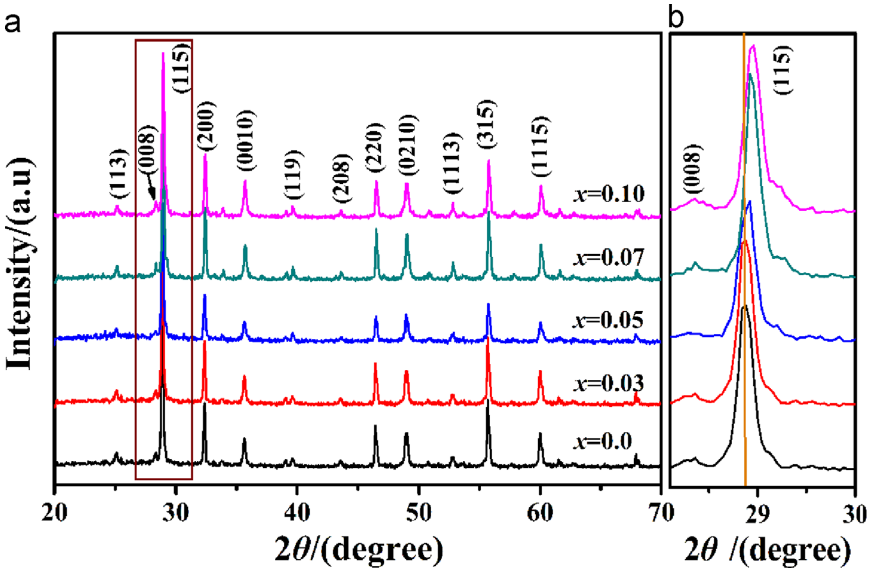

Fig. 1. XRD patterns of $(1-x)$SBN–xBFO ceramics sintered at 1100 °C.

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