



Photoluminescence, enhanced ferroelectric, and dielectric properties of Pr³⁺-doped SrBi₂Nb₂O₉ multifunctional ceramics



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ABSTRACT

Pr³⁺-doped SrBi₂Nb₂O₉ (SBN) multifunctional ceramics were synthesized by the conventional solid state method. The photoluminescence (PL) excitation and emission spectra, enhanced ferroelectric and dielectric properties were investigated. The X-ray diffraction (XRD) and FESEM analyses indicated that the samples were simple phase and uniform flake-structure. Under the 250–350 nm ultraviolet (UV) excitations, the red emission was obtained and the optimal emission intensity was investigated when Pr doping level was 0.005 mol. With the increasing of the Pr³⁺ ion contents, the ferroelectric properties were enhanced obviously. As a new multifunctional material, the Pr³⁺-doped SBN ceramics could be used for a wide range of application, such as integrated electro-optical devices.

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1. Introduction

Bismuth layer structured ferroelectrics (BLSFs) have attracted great interest in the recent years because of their potential application in technical devices [1–3]. For example, BLSFs ceramics are considered as superior candidates for nonvolatile random access memory (FRAM) [4] and high temperature piezoelectric application [1] due to their excellent fatigue-resistant properties and high Curie temperature. The BLSFs by the chemical formula Bi₂A_{n-1}B_nO_{3n+3} consist of perovskite layers (A_{n-1}B_nO_{3n+1})²⁻ sandwiched between bismuth oxygen sheets (Bi₂O₂)²⁺, where A and B represent the different cation of low and high valences and *n* indicated the number of perovskite layers [5–7]. The solid solutions, SrBi₂Nb₂O₉ (SBN), consist of a oxygen sheets (Bi₂O₂)²⁺ with two perovskite layers of oxygen octahedra, (SrNb₂O₇)²⁻ in the c-direction [8,9]. Due to the similarities both in structure and ferroelectric, and dielectric ferroelectric properties between SrBi₂Nb₂O₉ and a large family of fatigue-free layered perovskite oxide, exemplified by SrBi₂Ta₂O₉, SrBi₂NbTaO₉, and SrBi₄Ta₄O₁₅, SrBi₂Nb₂O₉ can act as a model materials for fatigue-free ferroelectric oxide [8,10,11].

Many studies have confirmed that the oxygen vacancies play a significant role in the severe leakage currents [12]. Doping with rare earth, such as La³⁺, Ce³⁺, Nd³⁺, Pr³⁺ etc., can effectively decrease the vacancies concentration, hence effectively modified the electrical properties [13,14]. On the other hand, some rare earth ions (such as Eu³⁺, Pr³⁺) have been extensively used as luminescence centers over the past few decades due to their potential applications [15,16]. Thus, by rare earth doping, some ferroelectric materials have multifunctionalities of significant enhanced electrical properties and strong luminescence [17–19].

In this paper, we report on the preparation of Pr³⁺ doped SBN ceramics by conventional solid-state reaction route, and associated effect of Pr³⁺ doping on their crystal structure, luminescence, ferroelectric and dielectric properties.

2. Experimental

In our experiments, Pr³⁺ doped SrBi₂Nb₂O₉ ceramics according to a stoichiometric composition of SrBi_{2-x}Pr_xNb₂O₉ (where *x* = 0, 0.002, 0.005, 0.008, 0.01, 0.02, or 0.03) were synthesized using conventional solid-state reaction techniques. The starting materials were Bi₂O₃ (99.999%, Sinopharm Chemical Reagent Co. Ltd.), Nb₂O₅ (99.9% Kojundo Chemical Lab), SrCO₃ (99.99% Sinopharm Chemical Reagent Co. Ltd.), Pr₆O₁₁ (99.99% Alfa Aesar). The raw materials were mixed with the addition of alcohol, dried, heated at 850 °C for 4 h (in which Bi₂O₃ excess of 2 mol% out of the stoichiometry was added to compensate for the volatilization

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of Bi^{3+} during sintering at high temperatures). After calcinations, the ground powders, added with 10 wt% polyvinyl alcohol (PVA) binder, were pressed into disk-shaped pellets of 10 mm in diameter and about 1.5 mm in thickness. Finally, they were sintered at 1200 °C for 2 h in air.

The sample crystallization behavior was examined by XRD (XRD, Bruker D8 Advanced, Germany) and scanning electron microscopy (SEM, S-4700, Hitachi, Japan). The photoluminescence (PL) and photoluminescence excitation (PLE) spectra were recorded using a fluorescence spectrofluorometer (F-7000, Hitachi, Japan). After the luminescent measurement, electrodes for electrical property measurements were fabricated with silver paste. The polarization–electric field (P–E) hysteresis loops were measured with a precision ferroelectric analyzer (Premier II, Radiant Technologies Inc. Albuquerque, NM). The temperature dependence of dielectric constant and loss was measured at 1 MHz using an Agilent HP4284A LCR meter connected to a lab-made furnace with a heating rate of 2 °C/min.

2.1. Results and discussion

Fig. 1 shows the X-ray diffraction patterns of Pr^{3+} doped SBN samples. All the recorded peaks of the ceramics agree well with orthorhombic phase of $\text{SrBi}_2\text{Nb}_2\text{O}_9$ (JCPDS 49-0607), indicating that the obtained samples are single phase and there is no impurity phase associated with the Pr doping. As shown in the insets of the Fig. 1, with the increase in Pr concentration in SBN ceramics, the strongest peak (115) obvious shifts to the lower-angle side, which is attributed to the lattice expansion because of the bigger Pr^{3+} ion substitution for Bi^{3+} in $[\text{Bi}_2\text{O}_2]^{2+}$ sheet ($r_{\text{Pr}^{3+}} = 0.114 \text{ nm}$, $r_{\text{Bi}^{3+}} = 0.103 \text{ nm}$, for a coordination number of 12) [17,20–22].

Fig. 2 shows the surface morphologies of the samples sintered at 1200 °C for 2 h. Flake-like grains are observed in the ceramics. Normally, the thickness direction of the plate is along c-axis and the grains grow layer by layer along c-axis. It can be seen that the thickness is $\sim 1.2 \mu\text{m}$. For the ceramics $x = 0.005$, the thickness is approximately $1 \mu\text{m}$, which is typical morphology of bismuth-layer structured ferroelectric [23].

The photoluminescence properties of Pr doped BTN ceramics were investigated. The PL and PLE spectra of Pr doped SBN ($x = 0.005$) are shown in Fig. 3. The excited spectra monitored at 610 nm exhibits two excitation band: a strong broad band at 260–380 nm with peak around 330 nm and a relatively weak band ranging from 430 to 500 nm with peaks around 450, 473, and 491 nm. The strong broad excitation band, locating at the emission

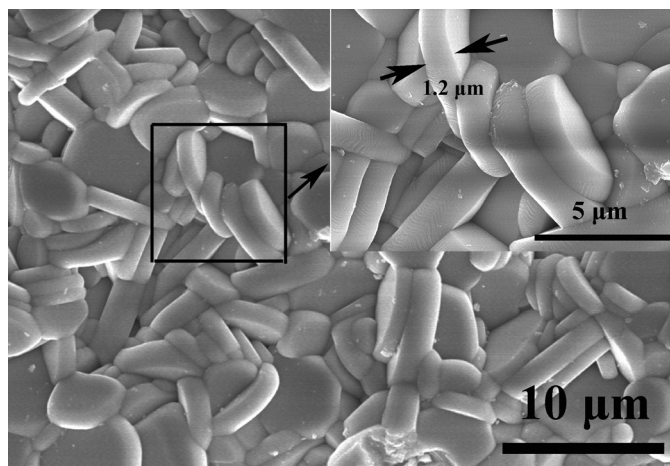


Fig. 2. Surface morphologies of the Pr doped SBN ceramics with $x = 0.005$.

of the Ultraviolet, is attributed to the absorption of the matrix SBN or Bi^{3+} ions [24]. The weak excitation band, which covers the emission of blue light, is attributed to the typical f – f transition from $^3\text{H}_4$ ground state to the $^3\text{P}_J$ ($J = 0-2$) [25,26]. With the excitation of 330 nm, the emission spectrum consists of a sharp band with peak at 610 nm, which was attributed to the typical transition from $\text{D}_2 \rightarrow ^3\text{H}_4$ of Pr^{3+} ions [27]. The strong red emission band of Pr^{3+} ions excited by UV light, suggesting the effective energy transfer, should be useful in various display devices [28,29].

The influence of Er^{3+} content on the emission spectra (with $\lambda_{\text{ex}} = 330 \text{ nm}$) of SBN ceramics for $x = 0.002-0.03$ is shown in the spectra of Fig. 4. There is an obvious influence of Pr^{3+} content on the intensity of the emission spectra. With the increasing Pr^{3+} content, the PL intensity of the Pr^{3+} was found to increase and then decrease because of concentration quenching [30], which indicated that the maximum emission intensity occurs at $x = 0.005$.

Fig. 5 shows the P–E hysteresis loops of Pr doped SBN samples measured with 100 Hz at room temperature. With the Pr doping, the polarization properties of the SBN ceramics improved obviously, for which the hysteresis loops of higher doping samples exhibit better shapes. The corresponding $2P_r$ (where P_r is the remanant polarization) values increased with increasing the Pr^{3+} content, which could be attributed to the substitution of large-radii cation (Pr^{3+}) for Bi^{3+} that assisted the elimination of oxygen vacancy and vacancy complex defects [31,32].

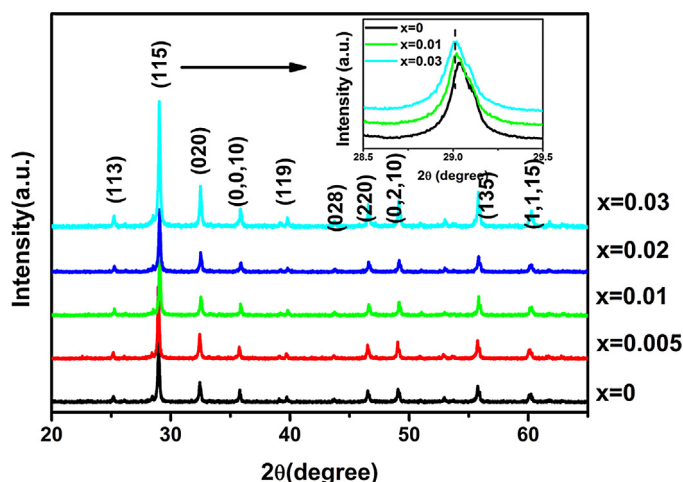


Fig. 1. XRD patterns of $\text{SrBi}_{2-x}\text{Pr}_x\text{Nb}_2\text{O}_9$ ceramics. The insets show the magnified (115) peaks.

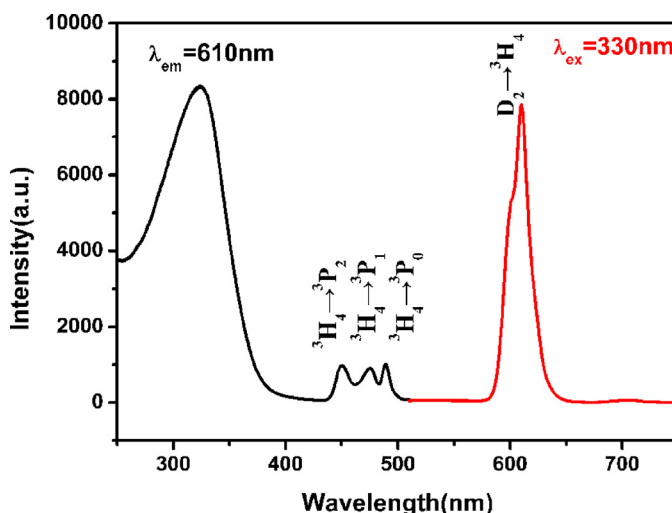


Fig. 3. PL and PLE spectra of Pr doped SBN ceramics ($x = 0.005$).

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