



Synthesis and enhancement of multiferroic properties of $(x)\text{Ba}_{0.95}\text{Sr}_{0.05}\text{TiO}_3-(1-x)\text{BiFe}_{0.90}\text{Dy}_{0.10}\text{O}_3$ ceramics



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ABSTRACT

Polycrystalline $(x)\text{Ba}_{0.95}\text{Sr}_{0.05}\text{TiO}_3-(1-x)\text{BiFe}_{0.90}\text{Dy}_{0.10}\text{O}_3$ [$(x)\text{BST}-(1-x)\text{BFDO}$] ceramics were synthesized by the standard solid-state reaction method. Samples prepared from these powders were sintered at various temperatures (900–1000 °C). The crystal structure, microstructure, magnetic, dielectric, complex impedance and magnetolectric properties of the $(x)\text{BST}-(1-x)\text{BFDO}$ ceramics were systematically investigated. The X-ray diffraction pattern indicated that $(x)\text{BST}-(1-x)\text{BFDO}$ ceramics showed a structural transition from rhombohedral to cubic phase. The average grain size increased to a maximum 2.86 μm for $x=0.20$ and then decreased. $M-H$ plot showed weak ferromagnetism for $x=0.00$ to 0.30. The magnetization value increased for increasing BST substitution up to $x=0.20$ due to the modification of crystal structure and then decreased for further increasing due to the nonmagnetic behavior of BST. A significant remnant magnetization (0.086 emu/g) and a coercive magnetic field (103.62 Oe) were also obtained for this solid solution. The complex initial permeability increased with the increase of both BST content (up to $x=0.20$) and sintering temperatures. The room-temperature dielectric constant increased whereas dielectric loss decreased with increasing BST content because of reducing oxygen vacancies. Impedance spectroscopy analysis confirmed the conducting grains and highly resistive grain boundaries affecting the conductivity but the grains had major contribution in the conduction mechanism. Magnetolectric voltage coefficient (α_{ME}) increased with the increase of BST content because of strong interaction between magnetic and ferroelectric domains. The highest value of $\alpha_{\text{ME}}=1.67 \text{ mV cm}^{-1} \text{ Oe}^{-1}$ was observed for $x=0.20$ solid solution sintered at $T_s=1000$ °C because of uniform grains in size and shape at this composition.

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1. Introduction

Multiferroic materials that exhibit more than one ferroic orders in the same phase have great attention in recent years because of their potential applications in multifunctional devices. There is a coupling between the ferromagnetic and ferroelectric orders which is known as magnetolectric (ME) coupling and this coupling is used in various practical devices such as multifunctional sensors, actuators, data storage, broadband magnetic field sensors, microwave phase shifters and magnetolectric memory cells [1]. Multiferroic materials are of two types: single-phase and composite multiferroic. The coupling effect between the ferroic orders can be easily realized in single phase multiferroics. But there are very few single-phase materials in nature which exhibit ferroelectric and ferromagnetic properties simultaneously. Among

them, BiFeO_3 (BFO) is one of the best candidate that exhibit multiferroic properties at and above room temperature [2]. The BFO demonstrates some promising and potential applications because of its large remnant polarization, the high ferroelectric Curie temperature (830 °C) and the high Neel temperature (370 °C). But the use of BFO have been restricted because of its some serious problems such as existence of secondary phase, large leakage current, low dielectric constant, weak magnetic behavior and poor phase (perovskite) stability. The BFO is a G-type antiferromagnetic material with cycloidal spin structure [3] and due to this structure its magnetic properties is poor comparing to ferromagnetic materials. Due to these problems, various attempts have been made by researchers to obtain BFO-based multiferroic materials with improved magnetic, electric and ME properties. For example, substitution of chemical compositions [4], applying high magnetic field [5], forming solid solution of BFO and other ABO_3 perovskite phases etc. Among these, BFO- ABO_3 solid solution has a great attention to increase structural stability, dielectric and magnetic properties [6]. For example, many reports have been published on

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the multiferroic systems of BFO–BaTiO₃ (BFO–BT) [6], BFO–PbTiO₃ (BFO–PT) [7], BFO–BiNaTiO₃ (BFO–BNT) [8] etc., however, the systems still did not meet the requirements. Although there is disappearance of secondary phases due to the formation of above mentioned solid solution, the solution show high dielectric loss, high leakage current, weak magnetic behavior etc. As a result the ME property is not improved significantly.

The BT is the most common ferroelectric oxide with ABO₃ perovskite structure. To improve dielectric constant and to reduce dielectric loss Sr atom is added into BT to form BaSrTiO₃ (BST) solid solution. The BST is promising because of its high dielectric constant, low dielectric loss and large tenability than BT. Tirupathi et al. [9] and Khelifi et al. [10] prepared the BST–BFO solid solution and studied the dielectric and magnetic properties. They found that BST helps to increase dielectric constant and magnetic properties but the dielectric loss is not reduced significantly. Also the systemic research on complex impedance and ME coupling of BST–BFO ceramics is not published yet. On the other hand, substitution of rare-earth elements in the B-site of BFO perovskite is rare but improved magnetic properties are expected by the substitution of magnetically active rare-earth atoms [11]. Although reports on multiferroic properties of Bi_{1-x}Dy_xFeO₃ is available but no results is found for the solid solution of Dy doped BFO and BST. On the other hand, enhanced magnetic properties are noticed for the case of Fe-deficient Gd doped BFO [12]. Due to the difference of magnetic moment of Fe³⁺ ($\mu_{\text{eff}}=5.92 \mu_B$) and Gd³⁺ ($\mu_{\text{eff}}=7.63 \mu_B$) and ionic radii between the cations, there is a significant lattice distortion and hence improve magnetic properties [11]. It is expected that the magnetically active rare earth atom Dy will play the same role. Thus the solid solution of BST–BiFe_{0.90}Dy_{0.10}O₃ (BFDO) may have enhanced multiferroic properties and probably observable ME coupling. In this study, we investigate the crystal structure, DC magnetization, complex initial permeability, dielectric properties, AC conductivity, impedance spectroscopy and ME effect of various (x)Ba_{0.95}Sr_{0.05}TiO₃–(1-x)BiFe_{0.90}Dy_{0.10}O₃ [xBST–(1-x)BFDO] ceramics with x=0.00–0.35.

2. Experimental details

2.1. Sample preparation

Polycrystalline (x)Ba_{0.95}Sr_{0.05}TiO₃–(1-x)BiFe_{0.90}Dy_{0.10}O₃ [(x)BST–(1-x)BFDO] ceramics with x=0.00, 0.10, 0.20, 0.25, 0.30, and 0.35 were prepared by the conventional solid state reaction technique. Raw materials of BaCO₃ (99.9%), SrCO₃ (99.9%), TiO₂ (99.9%), Bi₂O₃ (99.9%), Fe₂O₃ (99.9%) and Dy₂O₃ (99.95%) were mixed according to the stoichiometric formula and ball-milled in distilled water for 24 h to increase the degree of mixing. Then the mixed powders were calcined at 850 °C for 1 h. To obtain a homogeneous mixture the calcined powders were ground thoroughly and ball-milled again for 12 h. Later the powders were mixed with 10% polyvinyl alcohol (PVA) as a binder for granulation. Using these mixtures disk- and toroid-shape samples were prepared by applying a uniaxial pressure of 45MPa. Finally, the green pellets were sintered at various temperatures (900–1000 °C) in air for 1 h.

2.2. Characterizations

The crystal structure of the prepared samples were studied using a X-ray diffractometer (Philips PANalytical X'PERT-PRO) with CuK_α radiation ($\lambda=1.541 \text{ \AA}$) at room temperature and 1° per min scanning speed in a range of 2 θ from 20° to 60°. The lattice parameters were calculated from the x-ray diffraction (XRD) data. The microstructure of the sintered samples was examined by a

Field Emission Scanning Electron Microscopy (FESEM, model no. JEOL JSM 7600 F). The theoretical density (ρ_x) of the samples was determined by the formula, $\rho_x = \frac{n \times M_A}{N_A V}$, where, n is the number of atoms in a unit cell, M_A is the molar mass of the sample, N_A is the Avogadro's number and V is the volume of the unit cell. The bulk density (ρ_B) of each sample was calculated using the relation: $\rho_B = \frac{m}{\pi r^2 t}$, where, m is the mass, r is the radius and t is the thickness of the sample. The magnetic hysteresis (M–H) loops were measured using a vibrating sample magnetometer (VSM) (Micro Sense, EV9). The real part (μ_i') and imaginary part (μ_i'') of the complex initial permeability (μ_i^*) were measured as a function of frequency within the range of 10 kHz–100 MHz using a Wayne Kerr 6500B Impedance Analyzer. The values of μ_i' and μ_i'' were calculated using the relations: $\mu_i' = L_S/L_0$ and $\mu_i'' = \mu_i' \tan \delta$, where, L_S is the self inductance of the sample core and L₀ is the inductance of the winding of the coil without the sample and tan δ is the magnetic loss. L₀ is derived from the geometrical relations, $L_0 = \mu_0 N^2 S / \pi \bar{d}$, where, μ_0 is the permeability in vacuum, N is the number of turns of the coil (N=5), S is the cross-sectional area and $\bar{d} = (d_1 + d_2) / 2$ is the mean diameter of the toroid-shaped sample, where, d₁ and d₂ are the inner and outer diameter of the toroid-shaped sample, respectively. The dielectric measurements were carried out at room temperature within the frequency range of 1 kHz–1 MHz by using Impedance Analyzer. To measure dielectric properties the samples were painted on both sides by conducting silver paste to ensure good electrical contacts. The dielectric constant (ϵ') was calculated from the capacitance using the formula: $\epsilon' = Ct / \epsilon_0 A$, where, C is the capacitance of the pellet, A is the cross-sectional area of the electrode and ϵ_0 ($=8.85 \times 10^{-12} \text{ F/m}$) is the permittivity in free space. The AC conductivity (σ_{AC}) of the samples was calculated using the relation: $\sigma_{AC} = \omega \epsilon' \epsilon_0 \tan \delta$, where, ω is the angular frequency and tan δ is the dielectric loss. The ME effect was obtained by applying an ac magnetic field superimposed on a dc magnetic field on the sample, and then measuring the output signal with applied dc magnetic field. An electromagnet was used to provide a dc magnetic field of up to 0.77 T. A signal generator operating at a frequency of 50 Hz was used to drive the Helmholtz coil to generate an ac magnetic field. The output voltage generated from the sample under investigation was measured using a Keithley multimeter (Model 2000) as a function of dc magnetic field. ME voltage coefficient, α_{ME} , was calculated using relation, $\alpha_{ME} = (dE/dH)_{H_{ac}} = V_0 / h_0 t$, where V₀ is the ME voltage across the sample surface and h₀ is the amplitude of the ac magnetic field.

3. Results and discussion

3.1. Structural analysis

Fig. 1 shows the XRD patterns of BFO and various (x)BST–(1-x)BFDO ceramics sintered at 900 and 1000 °C for 1 h, respectively. The position of all peaks of XRD patterns of BFO matches well with the earlier reported result [13]. Moreover, two impurity peaks indicated by the symbols * and Δ are also observed because of slow phase formation kinetics of BFO. The impurity peaks might be formation of Bi₂Fe₄O₉ and Bi₂₄Fe₂O₃₉ and existence of such impurity peaks have been reported earlier by several authors [14,15]. The XRD patterns of BFO exhibit perovskite structure with rhombohedral unit cell and the lattice parameters are found to be a=b=5.58 Å and c=13.65 Å. On the other hand, the impurity peaks are found to be disappeared for the case of xBST–(1-x)BFDO ceramics as shown in Fig. 1(b) which indicate BST has been substituted in BFDO properly. From Fig. 1(b), it is also seen that XRD peaks shift towards the lower angle side with increasing BST up to x=0.20 and then the peaks shift towards the higher angle

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