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Studies on structural, dielectric, conductivity, magnetic and magnetoelectric properties of barium titanate doped with lithium ferrite



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ABSTRACT

In this paper, we report the synthesis of a multiferroic composite $BaTiO_3$ (BT) doped with $Li_{0.5}Fe_{2.5}O_4$ (LF) having chemical formulae (1-*x*) $BaTiO_3 + (x)$ $Li_{0.5}Fe_{2.5}O_4$ (x = 0, 0.05, 0.1, 0.15) using conventional solid state reaction method. We have focused on the structural and multiferroic properties of LF doped BT. We have characterized the grown composites by XRD, FESEM with EDAX, Piezoelectric coefficient, dielectric studies, A.C conductivity studies, magnetic properties and magneto-electric (ME) voltage coefficient. The XRD studies reveal the tetragonal nature of all composites. We have calculated the lattice parameters and the crystallite size from XRD pattern. The XRD patterns of all composites exhibit diphasic nature. The dielectric constant of all composites decreases steeply with an increase in the frequency reveals that at high-frequency region, the maximum conductivity decreases with an increase in the % of LF. The variation of conductivity with temperature reveals that the Curie temperature of the composite decreases as and when the % of LF is increased in BT. We have also studied the temperature dependence of the magnetization and the variation of the magnetic field. The transition temperature and Neel temperature of all composites increase LF content.

1. Introduction

Multiferroic composites exhibiting both magnetic and electric properties yield more than one ferroic order parameters like ferroelectricity, ferromagnetism or piezoelectric, piezomagnetic and ferroelasticity phases. Multiferroic composites possessing the above said ferroic parameters exhibit better and special properties [1]. The magnetoelectric properties of multiferroic composites exhibit strong coupling between ferrite and ferroelectric phases simultaneously, which enables the use of the composites in many potential applications like sensors, transducers, actuators, spintronics devices, actuators and magnetoelectric transducers [2–6]. Magneto-electric material facilitates the conversions of magnetic energy to electrical energy and vice versa. These materials require both permanent dipole moments in the form of piezoelectric phase and magnetic phase and in magneto-electric material, magneto-electric phase should be dominant [7]. The presence of

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magnetic order induces a magnetostriction effect in the composite which in turn induces the piezoelectric effect, causing a flow of an electric field in the system. Hence the strong magneto-electric effect (ME) could be realized in the composite [8].

The BaTiO₃ (BT) is a widely used material which possesses properties like ferroelectric, piezoelectric and dielectric [3]. The BaTiO₃ (BT) is widely used in applications like multilayer capacitors, Thermistors and electro-optic devices [3]. The BaTiO₃ (BT) material has high dielectric constant, low dielectric loss and large electro-optic coefficient [3]. A multiferroic material having a high Magneto-electric (ME) coefficient finds its use in applications like spintronics, sensors, actuators and micro electric devices [9–12].

In order to enhance the multiferroic properties of composite materials, these materials have to be synthesized by doping a suitable material. One such composite material is Lithium Ferrite [13–16] doped in BaTiO₃ (BT). LiFe₅O₈–BaTiO₃ system shows the different magneto-

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electric response, which can be varied with composition as well as microstructures. Therefore, a better control of composition and microstructure can give a better macroscopic property [15,16]. Li_{0.5}Fe_{2.5}O₄ (LF), an inverse spinel crystal, is used in microwave devices and memory core applications. The Li_{0.5}Fe_{2.5}O₄ (LF) is used in high-frequency devices due to its huge resistivity, the hardness of mechanical, square loop properties and high Curie temperature [17,18]. In Li_{0.5}Fe_{2.5}O₄, all the Li¹⁺ ions and 3/5 of the Fe³⁺ ions occupy the octahedral site and the remaining Fe³⁺ ions occupy tetrahedral sites, which consists of antiparallel magnetic moments. Octahedral sites influence in determining the net magnetization. In view of the above mentioned properties, we have doped LF in BT which is focused the influence of LF on the structural, morphological, dielectric, piezoelectric, magnetic and magneto-electric properties of BT.

2. Experimental method

The pure BT and LF doped BT were synthesized using conventional solid state reaction method. The analytical purity of compounds were BaCO₃ (98%, Merck), TiO₂ (99.5%), Li₂CO₃ (99%), Fe₂O₃ (98%, Loba Chemie). The BaCO₃, TiO₂, Li₂CO₃ and Fe₂O₃ powders were weighed in a stoichiometric ratio and mixed together using agate motor. These samples were grinded for 10 h to obtain a homogeneous mixing and distribution of the ingredients. Then samples were calcinated at 900 °C temperature for 3 h. The calcinated powders were again grinded for 3 h and a binder (prepared by PVA) was added to the calcinated powders followed by grinding process to obtain a finely powdered composite. The fine powder is shaped into pellets using a hydraulic press. These pellets were sintered at 1150 °C temperature for 3 h and the sintered pellets were characterized using XRD, FESEM with EDAX, piezoelectric coefficient, dielectric measurements, VSM (MT) and magneto-electric voltage coefficient. The flow chartdepicting the method of preparation of all samples is shown in Fig. 1.

The XRD Measurements were carried out using Bruker D8 Advance X-Ray Diffractometer, EDAX for quantitative elemental analysis of a sample is analyzed by Carlzeiss ultra-55. The strain varies with electric field for the piezoelectric coefficient is measured by TF analyzer 2000. The dielectric measurements have been obtained from LCR Meter, Wayne Kerr Electronics Pvt. Ltd., Model: 1J43100. The variation of magnetization with temperature (MT) of samples is measured by VSM Quantum Design PPMS, Model 6000 and the magneto-electric voltage coefficient (α_{ME}) was measured with respect to the DC magnetic field (H_{dc}) by superimposing 1 Oe AC magnetic field generated by Helmholtz coils at a frequency of 1 kHz. Using SR 830 DSP lock-in amplifier, the output voltage of all the samples was measured.

3. Results and discussions

3.1. X-ray diffraction

The X-ray diffraction (XRD) patterns of the BaTiO₃ (BT), (95) BT + (5) LF, (90) BT + (10) LF and (85) BT + (15) LF are shown in Fig. 2 (a). The diffraction peaks of pure BT and its composites are indexed using JCPDS no79-2263. The XRD pattern of pure BT and its composites reveal the formation of the tetragonal perovskite structure. The peaks at 34° and 63° strongly confirm the incorporation of LF in BT. The peak centered at 31.5° (shown in Fig. 2 (b)) is slightly shifted towards the higher diffraction angle when the LF is doped in BT material. But as and when the % of the LF content is increased, the peak centered about 31.7° does not shift which suggests the existence of ferroelectric nature of the samples with no chemical reactions between ferroelectric and ferromagnetic component [19]. We see from Fig. 2 (a) that, the peaks represents the ferroelectric phase and thus all the composites exhibit diphasic nature.

The lattice constants have been calculated from the predominant peaks of BT doped LF and are presented in Table 1. The lattice constant of BT shown in Table 1 agrees well with an earlier reported value [20]. From Table 1, we see that the lattice parameters in composites are nearly equal to its constituent phases representing indicating the absence of structural changes in the material and intensity of the ferrite peaks only increases with its ferrite in the composites [21]. Since no other extra peaks were identified, we conclude that highly crystalline composites are formed during the sintering process.

The amount of the constituent phases approximately present in the composite after sintering are calculated using the corresponding intensity peaks [22] and shown in Table 1. The percentage of phases are calculated using the relations



Fig. 1. Flow chart for the method of preparation.

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