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Study of structural and magnetoelectric properties of 1−x($Ba_{0.96}Ca_{0.04}TiO_3$)–x($ZnFe_2O_4$) ceramic composites

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Abstract Multiferroic ceramic composites of (1−x) $Ba_{0.96}Ca_{0.04}TiO_3$–(x)$ZnFe_2O_4$ (BCT-ZF) were prepared from ferroelectric (FE) barium calcium titanate (BCT) and ferromagnetic (FM) zinc ferrite (ZF) by using the solid state reaction method with different mol% fractions of x (x = 0.1 and 0.2). The preliminary structural studies carried out by X-ray diffraction at room temperature reveals that the samples have a tetragonal structure along with the cubic spinel ferrite phase. Raman spectra of the composites also confirm the existence of BCT phase and ZF phase. The room temperature ferroelectric polarization measurements as a function of magnetic field show the existence strong magnetoelectric coupling of 10.85 (mV/(cm.Oe).

1 Introduction

In the twenty-first century, researchers are aiming to discover such materials, whose magnetic and electronic functionality can be controlled by magnetic and electric stimuli in a single phase for novel magnetoelectric devices [1–11] Magnetoelectric (ME) multiferroic materials are the best materials for this novel multifunctional applications. $BiFeO_3$ (ferroelectric up to ~ 830 °C and antiferromagnetic up to ~ 370 °C) is one the rarest single phase room temperature multiferroic materials with many interesting physical phenomena. However, high leakage current of $BiFeO_3$ (BFO) and weak magnetoelectric coupling in limits its applications. A strong ME coupling is an important requirement for practical applications. In order to enhance the ME coupling, we synthesized an artificial multiferroics by combining a good ferroelectric and a ferromagnetic phases. The ferrite phase should be highly magnetostrictive possessing high resistivity state which is possible in ZF nanostructure [12], another phase should be highly piezoelectric which is BCT [13]. Such ME materials have a wide range of applications such as chip sensors, multistate memories, actuators and in biomedical field [14, 15]. The ME effect in composite materials is the result of the interaction between the two-phases (*i.e.* magnetoelectric and ferroelectric): magnetic field induces the structural distortion in the magnetic phase and the applied electric field produces the distortion in ferroelectric phase [16–18]. The ME coupling appeared in materials due to mechanical deformations in the magnetostrictive phase in applied magnetic field δH. Due to the piezoelectric effect, the strains transferred to the piezoelectric phase producing an induced electric field δE in the material. ME voltage coefficient $\alpha E = \delta E/\delta H$ characterizes the ME effect [19, 20].

Lead-free ceramics like $BaTiO_3$ (BT), $Bi_{0.5}K_{0.5}TiO_3$ (BKT), $K_{0.5}Na_{0.5}NbO_3$, $Bi_{0.5}Na_{0.5}TiO_3$ (BNT). have gained much attention due to their various properties like high dielectric constant [21], high piezoelectricity [22], good mechanical strength [23] and eco-friendly behavior [24–26]. At higher temperature, BCT is cubic (paraelectric), but on cooling it becomes tetragonal (ferroelectric) below Curie temperature of 410 K [27], orthorhombic below 290 K and rhombohedral below 190 K. In 2014 Verma et al. [13] has reported that polycrystalline BT with tetragonal

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