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Study of dielectric, magnetic and ferroelectric properties in $Bi_{1-x}Gd_xFeO_3$

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Abstract

Gd-doped $BiFeO_3$ polycrystalline ceramics were synthesized by solid-state reaction method and their dielectric and magnetic properties were investigated. X-ray diffraction pattern showed that $Bi_{1-x}Gd_xFeO_3$ ($x=0$, 0.05 and 0.1) ceramics were rhombohedral. The Gd substitution has suppressed the usual impurity peaks present in the parent compound and we obtained single phase $Bi_{0.9}Gd_{0.1}FeO_3$ ceramic. Gd substitution reduced the antiferromagnetic Néel temperature (T_N) in $Bi_{1-x}Gd_x FeO_3$. An anomaly in the dielectric constant(ε) and dielectric loss(tan (δ)) in the vicinity of the antiferromagnetic Néel temperature (T_N) was observed. Ferroelectric and magnetic hysteresis loops measured at room temperature indicated the coexistence of ferroelectricity and magnetism. The room temperature magnetic hysteresis loops were not saturated, but the magnetic moment was found to increase with increase in Gd concentration.

Keywords: Ceramics; Dielectric constant; Néel temperature; Magnetization; X-ray diffraction

1. Introduction

Multiferroic materials exhibit simultaneous presence of (anti) ferroelectricity, (anti)ferromagnetism and ferroelasticity in the same phase and possess tremendous potential for applications in multiple state memory elements, electric field controlled ferromagnetic resonance devices and transducers with magnetically modulated piezoelectricity [1–5]. These novel devices exploit magnetoelectric property of multiferroics. By magnetoelectric effect one means coupling between electric and magnetic subsystems [6]. P–E loop and M–H loop at same temperature serve as confirmation of magnetoelectric effect.

$BiFeO_3$ is one such few materials [7] having an antiferromagnetic behavior with a relatively high Néel temperature ($T_N=370$ °C) and ferroelectric behavior with a high Curie temperature ($T_C=810$ °C) [8–10] and possesses a rhombohedrally distorted perovskite structure. $BiFeO_3$ is the most promising candidate among multiferroics for novel applications due to its high values of magnetic and ferroelectric ordering temperatures, still it has not found use in industrial applications due to following reasons. One problem is that pure phase of Bismuth ferrite is difficult to obtain [11,12]. Various impurity phases have been reported, mainly comprising of $Bi_2Fe_4O_9$, $Bi_{36}Fe_{24}O_{57}$ and $Bi_{25}FeO_{40}$ [13,15]. High leakage current in the samples, leads to poor ferroelectric behaviour. From previous work it is observed that partial substitution of Bi^{3+} by rare earth element ions (La^{3+}, Nd^{3+}, Tb^{3+}, Sm^{3+}) [11,14,16,17] result in room temperature multiferroic properties. Since rare earth doped bismuth ferrite is reported to undergo structural transformation from rhombohedral to triclinic or orthorhombic around $x=0.1$ [17,18], we have prepared $Bi_{1-x}Gd_xFeO_3$, $x=0$, 0.05 and 0.1 using solid-state-reaction method and report the ferroelectric and magnetic properties of the modified multiferroic.

2. Experimental

$Bi_{1-x}Gd_xFeO_3$ ceramics with $x=0$, 0.05 and 0.10 were prepared by conventional solid-state reaction method. Bi_2O_3, Fe_2O_3 and Gd_2O_3 of analytical grade were mixed in agate mortar for 2 h. This mixture was calcined at 800 °C for 1 h in a programmable furnace. The powders were then pressed into disks of thickness 1 mm and diameter 7 mm by using a uniaxial

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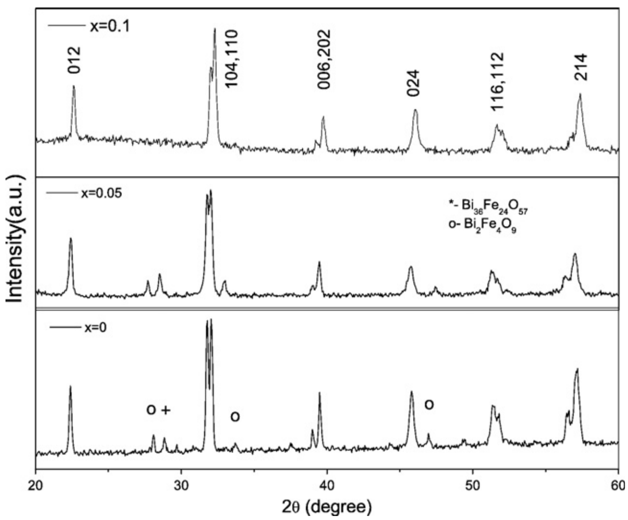

Fig. 1. XRD pattern of $Bi_{(1-x)}Gd_xFeO3$ samples.

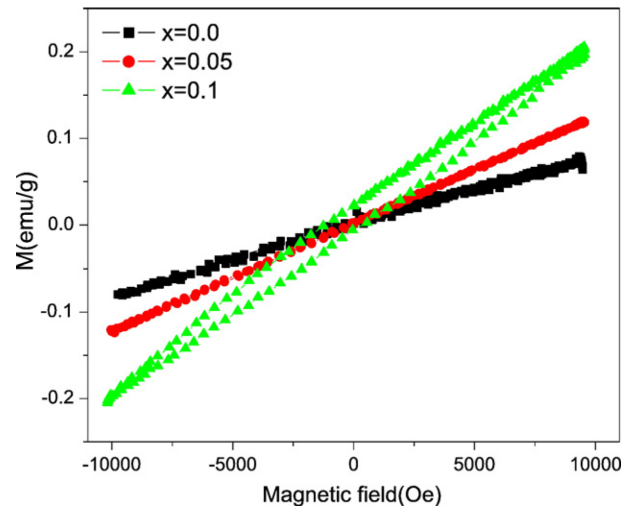

Fig. 3. Magnetization versus magnetic field curves for $Bi_{(1-x)}Gd_xFeO3$ samples at 300 K.

hydraulic press. The final sintering of the pellets was done at 820 °C for 2 h. X-ray diffraction (XRD) pattern of the samples were obtained using Cu K_α radiation at a slow scanning rate of 1°/min for phase identification. FE-SEM was used to study the microstructure of the ceramics. Dielectric characterization was done by using a LCR meter (HIOKI model 3532-50). The magnetic hysteresis (M–H) loops were measured using a vibrating sample magnetometer (VSM) supplied by Priston Applied Research (Model 155) with a maximum magnetic field of 10 kOe. Ferroelectric hysteresis loops were measured by using ferroelectric loop tracer at a frequency of 50 Hz.

3. Results and discussion

Fig. 1 shows the XRD pattern of $Bi_{1-x}Gd_xFeO_3$, (x=0, 0.05 and 0.1). It is evident from the figure that all the compositions have rhombohedral structure. $BiFeO_3$ is a meta stable compound and because of its chemical kinetics of formation it is always associated with some impurity phases as $Bi_{36}Fe_{24}O_{57}$ and $Bi_2Fe_4O_9$. These impurities are also evident in undoped sample.

These impurity peaks are still present in $Bi_{0.95}Gd_{0.05}FeO_3$. Increase in Gd content up to 0.1 has suppressed these impurities and we obtained pure phase $Bi_{0.9}Gd_{0.1}FeO_3$ with all the peaks corresponding to rhombohedral structure with R3c space group. Here Gd_2O_3 (a third component as a minor additive) in the solid solution accelerate the formation of $BiFeO_3$ [19].

The variation of dielectric constant (ε) and dissipation factor (tan (δ)) with temperature ranging from 40 °C to 350 °C at various frequencies (1 kHz, 10 kHz and 100 kHz) is shown in Fig. 2(a) for x=0.05 and in Fig. 2(b) for x=0.1 respectively.

The ε vs. T graph shows a broad peak around 220 °C. This dielectric anomaly is attributed to antiferromagnetic transition of the sample. This anomaly in magnetoelectrically ordered systems is predicted by Landau–

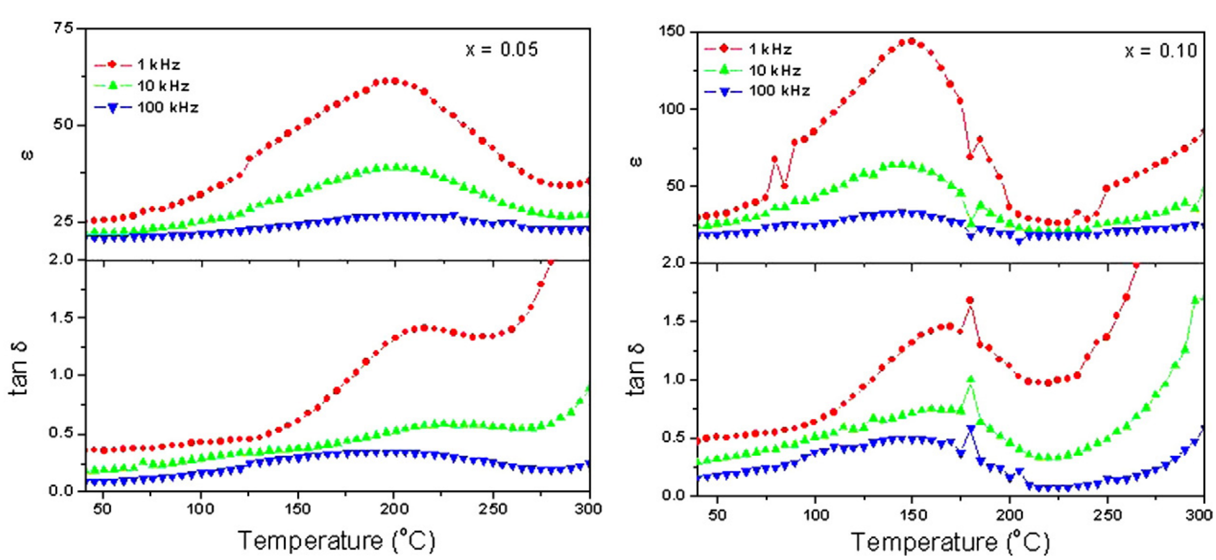

Fig. 2. Variation of ε and tan(δ) with temperature at frequencies 1, 10 and 100 kHz for (a) $Bi_{0.95}Gd_{0.05}FeO_3$ and (b) $Bi_{0.9}Gd_{0.1}FeO_3$.