

Structural and Magnetic Study of Lead Free $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3/\text{NiZn}$ Ferrite Composite

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Abstract

In the present work, Lead Free $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3/\text{NiZn}$ Ferrite (NBT/NZF) composite has been synthesized by chemical route method. Structural analysis of composite has been done by X-ray diffraction (XRD) and Scanning Electron Microscope (SEM) study. The x-ray diffraction patterns has revealed rhombohedral perovskite structure for $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$, cubic spinel structure for NiZn Ferrite and both perovskite and spinel phases without any secondary phase for NBT/NZF composite. Magnetic Hysteresis curves for NiZn Ferrite and NBT/NZF composite show ferromagnetic behaviour at room temperature.

Keywords: Composite, XRD, SEM and M-H curve.

1. Introduction

The Magneto-electric (ME) composites mainly consist ferrite (piezomagnetic) and ferroelectric (piezoelectric) phases. Ferrites show piezomagnetic behavior due to their magnetostrictive properties in presence of ac magnetic field [1]. The cross mechanical coupling between ferrite and ferroelectric phases give rise to new materials with magnetoelectric (ME) property. The ME property of a composite is defined as appearance of electric polarization on the application of magnetic field or magnetic polarization on the application of electric field [2,3]. ME composites have considerable prospective for applications in multifunctional devices like sensors, transducers, magneto-electric memory devices etc. [4]. Many composites based on lead (Pb) based piezoelectrics and magnetic constituents like Ni, Zn based ferrites exist, but due to environmental concern lead free materials are preferred [5-7]. In this work, we have chosen lead free $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ (NBT) as the piezoelectric phase and $\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4$ (NZF) as piezomagnetic phase. Once the ferrite/ferroelectric composite is synthesized, most important is to achieve the intrinsic properties of both the magnetic and ferroelectric phases in it, without

any major change in interaction mechanism between the two phases. The present work only represents the structural and magnetic properties of NBT/NZF composite.

2. Experimental Details

2.1. Synthesis of NBT Phase:

For synthesis of NBT, stoichiometric amounts of sodium nitrate (NaNO_3), bismuth nitrate ($\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$) and Iron nitrate ($\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$) were dissolved in acetic acid with constant stirring. The 10 mol% excess of sodium and bismuth introduce to compensate Na and Bi loss during heat treatment process. Tetra-n-butyl titanate ($\text{Ti}(\text{OC}_4\text{H}_9)_4$) was mixed with acetyl acetone and 2-propanol in weight ratio of 1:3:5. This Ti solution was added drop by drop under constant stirring to produce Bi-Na-Fe-Ti complex solution. Then this complex solution was added to the aqueous solution of citric acid, which was taken 1:1.2 weight ratios to cationic mixture. The resulting solution was dried to obtain fluffy dry powder, which was further sintered at 650°C for 2 hours.

2.2. Synthesis of NZF Phase:

For the synthesis of NZF, stoichiometric amounts of Nickel-2-ethylhexonate, Zinc-2-ethylhexonate and Iron-3-ethylhexonate were dissolved in xylene. The mixture was heated at 80°C for 1 hr and 5-7 drops of polyethylene glycol (PEG) were added as surfactant. The final mixture solution was then dried at 300°C to get powder. The dried powder of NZF was presintered at 700°C for 3 hours under ambient condition.

2.2. Synthesis of NBT/NZF Phase:

NBT and NZF powders were mixed in 0.75:0.25 weight ratios to prepare $0.75(\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3)/0.25(\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4)$ Composite. After mixing, the NBT/NZF composite was sintered at 750°C for 1hr.

The crystallographic and microstructural properties of all specimens were studied by X-ray diffraction (PANalytical X'Pert PRO diffractometer) with $\text{CuK}\alpha$ radiation and scanning electron microscope (SEM)

Quanta 250, FEI Make - USA) respectively. The magnetic properties were measured by using vibrating sample magnetometer VSM (Microsense, USA) at room temperature.

3. Results and Discussions

X-ray diffraction patterns of NBT, NZF and NBT/NZF composite are shown in Figure 1.

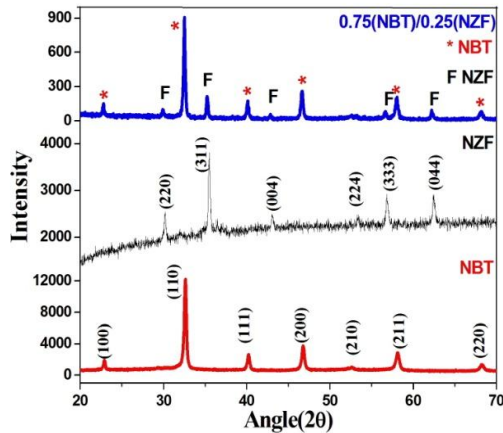


Figure 1. X-ray diffraction patterns for (a) NBT (b) NZF (c) NBT/NZF

XRD patterns reveal rhombohedral perovskite structure for NBT, cubic spinel for NZF. The XRD pattern of composite confirms the coexistence of both phases without any secondary phase formation, which emphasizes that the individual phases i.e. NBT and NZF have not chemically reacted in the composite formation.

The Scanning Electron Microscope (SEM) images of NZF powder, NBT/NZF composite samples sintered at 700°C, 750°C respectively are shown in Figure 2(a-b).

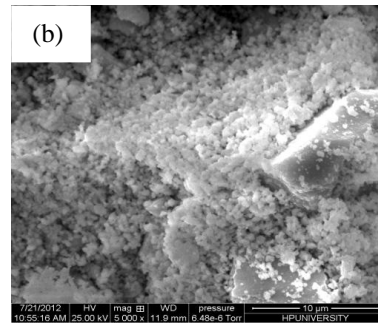
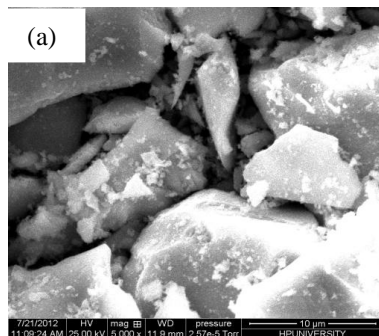


Figure 2. Scanning Electron Microscope Images for (a) NZF (b) NBT/NZF

The SEM micrograph of NBT/NZF composite shows the polycrystalline microstructures with different grain sizes and are non-uniformly distributed throughout the sample surface.

Figure 3 shows magnetic field dependence of magnetisation for NZF and NBT/NZF composite at room temperature. The values of M_s , M_r and H_c obtained from M-H curve for NZF are 39emu/g, 4.1emu/g and 58 Oe respectively. The values of M_s , M_r and H_c obtained from M-H curve for NBT/NZF composite are 11emu/g, 3.3emu/g and 109 Oe respectively.

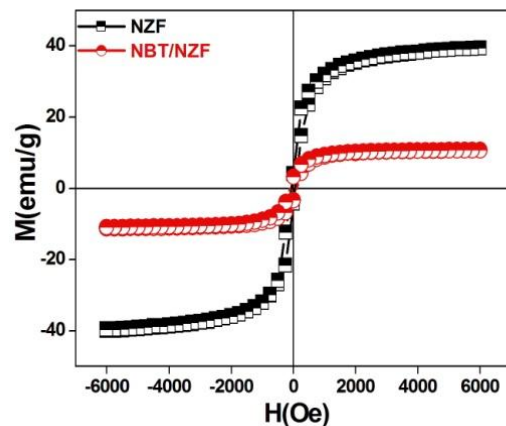


Figure 3. Magnetization vs magnetic field curves for (a) NBT (b) NZF (c) NBT/NZF

The coercive field H_c of the composite is much higher than that of the pure NZF. This variation in coercive field may arise due to the change in magneto-crystalline anisotropy energy of composite caused by lattice mismatch between NZF and NBT phases. The M-H curve for NBT/NZF demonstrates the ferromagnetic behaviour at room temperature. However, magnetic parameters (H_c , M_r and M_s) of composite have lower values than those for NZF. The

results may be explain on the basis that some of ferrite grains are connected to ferroelectric grains, which act as pores in the presence of applied magnetic field [5,8].

4. Conclusions

The NBT/NZF composite has been successfully synthesised by chemical route. The XRD pattern confirms the coexistence of both the phases (NBT and NZF) in NBT/NZF composite. The XRD results also reveal the formation of rhombohedral perovskite structure for NBT and cubic spinel structure for ferrite phase. The MH curve of Composite shows ferromagnetic behaviour at room temperature. The magnetic studies suggest NBT/NZF composite as an important multiferroic composite for further magneto-electric investigations. In order to study the ferroelectric and ferromagnetic domain interaction in the composite further investigations on ME coupling, ferroelectric properties in presence of magnetic phase and temperature dependent magnetic properties needed.

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