Synthesis and Characterization of BSLT-NZFO Multiferroic Composite

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(Received 13 February 2020; revised manuscript received 10 April 2020; published online 25 April 2020)

Magnetodielectric composites with composition $(1-y)Ba_{0.886}Sr_{0.1}La_{0.01}TiO_3 - yNi_{0.8}Zn_{0.2}Fe_2O_4(BSLT-NZFO)$ where $y=0.2,\ 0.3,\ 0.4$ have been prepared by Hydroxide Co-precipitation route followed by microwave sintering. The X-ray diffraction pattern of the composites shows the formation of cubic spinel structure for the ferromagnetic phase and tetragonal pervoskite structure for the ferroelectric phase. Scanning electron micrographs (SEM) were taken to understand the surface morphology, grain size and grain growth of the composite. Dielectric and magneto dielectric (MD) properties of the composites also investigated at room temperature with different frequencies.

Keywords: Hydroxide Co-precipitation, X-ray diffractogram, Dielectric Properties, Magnetodielectric effect.

DOI: 10.21272/jnep.12(2).02007 PACS numbers: 77.80. – e, 77.84. – s

1. INTRODUCTION

Recently Multiferroic materials have received interest, because they can simultaneously show ferroelectric and ferromagnetic order [1-6]. These Multiferroic materials are promising applications for many multifunctional devices such as transducers, sensors and memories [2-6]. For selection Multiferroic materials one phase is ferrite and other one is ferroelectric depends on the various parameters like high magnetostriction coefficient, piezoelectric coefficient, high dielectric permeability and dielectric loss[3-5]. Ferromagnetic materials with inverse spinel structure cobalt ferrite(CoFe₂O₄) is well known to have a moderate saturation magnetization, large magnetocrystalline anisotropy, high coercivity and chemically stability [1]. The pervoskite (BaSr)TiO₃ (ABO₃) is one of most well known and widely studied ferroelectric materials with excellent dielectric constant and low dielectric loss, due to its potential applications in capacitors, tunable filters and memories (DRAM)[1-3].

A small amount of La³⁺ ions replaces the [BaTiO₃] A-site ions of Ba to form a modified structure which enhances the dielectric and ferroelectric properties of the material. When lanthanum (La) doped in BaTiO₃ ceramics, the electrical and dielectric properties of ceramics can be modified. The doping of a small amount La³⁺ ions on (A-site)Ba²⁺ sites requires the formation of negatively charged defects and are possible charge compensating mechanism are occurred Barium Vacancies, Titanium vacancies and electrons [9-11].

Glass-ceramic materials have a pore-free and well grained microstructure which is highly attractive applications for ceramics used in capacitors [9]. The microwave sintering have number of advantages like low sintering time, more uniform structure, rapid heating and heating from inside to outside direction of the material [1, 8].

In present study, the La doping was selected to reduce the curie temperature of the $Ba_{0.885}Sr_{0.1}TiO_3$ (BST). The Dielectric, magnetic and Magnetodielectric (MD) performances, the $(1-y)Ba_{0.885}Sr_{0.1}La_{0.01}TiO_3$

yNi_{0.8}Zn_{0.2}Fe₂O₄(BSLT-NZFO) ferroelectric– ferromagnetic was studied.

2. EXPERIMENTAL

 $Ba_{0.885}Sr_{0.1}La_{0.01}TiO_3$ were synthesized via Hydroxide Co-precipitation route. The synthesis was based upon the co-precipitation of metal hydroxide $Ba(OH)_2$, $Sr(OH)_2$, $La(OH)_3$ and $Ti\text{-O-}(OH)_2$. The stoichiometric amounts of high purity $Ba(NO_3)_2$, $Sr(NO_3)_2$, $La(NO_3)_3.6H_2O$, $K_2TiO(C_2O_4)2.2H_2O$ were used as starting chemicals. KOH used as a precipitation agent. After drying the powder was calcinated at $850~^{\circ}C$ for 30~min in microwave furnace. And then sintering was carried out at $1150~^{\circ}C$ for 5~min., with an intermediate grinding.

Nickel acetate Ni(CH₃COO)₂.4H₂O, Zinc acetate, Iron nitrate Fe(NO₃)₃.9H₂O, were first dissolved separately into distilled water. And KOH and NH₄OH used as precipitation agent to maintain PH $_{\sim}$ 10. After drying the powder was calcinated at 900 °C for 30 min and then sintered at 1050 °C for 5 min. in programmable furnace.

2.1 Preparation of MD Composites

The composite were prepared by using formula (1-y) BSLT + y(NZFO) with the help of agate mortar and pistol sintered powder of BSLT and NZFO mixed together and grounded thoroughly. Small amount of diluted polyvinyl alcohol was added as binder. The pellets are formed by using hydraulic press having 1.2 cm diameter. Composite pellets were finally sintered at 1050 °C for 5 min in microwave furnace.

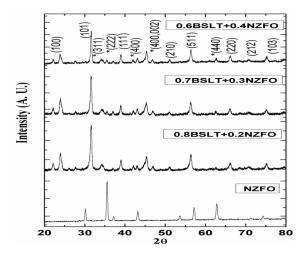
2.2 Characterization

To determine the X-ray diffractogram Bruker (D8 advance) XRD diffractometer was used. The measurements of dielectric constant as a function of frequency, temperature were carried out using (WK6500B) LCR-Q meter.

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3. RESULT AND DISCUSSION

The Fig. 1 shows XRD spectra of (1-y) Ba $_{0.885}$ Sr $_{0.1}$ La $_{0.01}$ TiO $_3-y$ Ni $_{0.8}$ Zn $_{0.2}$ Fe $_2$ O $_4$ where y=0.2, 0.3, 0.4. It is clearly seen that both the phases i.e. ferroe-lectric and ferrite phase. All peaks could be indexed for all samples and no any impurity peaks are seen in the x-ray diffractogram. '*' indicates ferromagnetic phase. The intensity of the major peaks, such as (1 0 1) for BSLT and (3 1 1) for NZFO depends in the composite. The observed value of lattice constant a=3.9912 Å, c=4.0132 Å and average crystallite size 40 nm for ferroelectric materials and for ferromagnetic phase lattice parameter a=b=c=8.4029 Å.



 $\label{eq:Fig.1} \begin{aligned} &\textbf{Fig. 1} - \textbf{X}\text{-raydiffractogramof} \ (1-y) \ Ba_{0.885}Sr_{0.1}La_{0.01}TiO_3 - \\ &yNi_{0.8}Zn_{0.2}Fe_2O_4 \ where \ y=0.2,\ 0.3,\ 0.4 \end{aligned}$

Figure 2 shows scanning Electron Micrographs of (1-y) Ba_{0.885}Sr_{0.1}La_{0.01}TiO₃ – yNi_{0.8}Zn_{0.2}Fe₂O₄ for y = 0.2, 0.3, 0.4. From this picture it is clearly seen that sample is dense with some void and grains are spherical in shape. As the ferromagnetic phase is increase, the surface morphology is changed and becomes more compact.

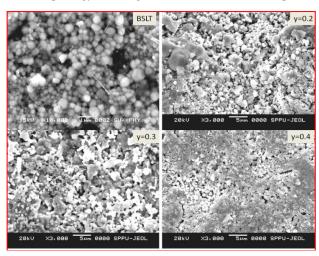


Fig. 2 – Scanning Electron Micrographs of (1-y) Ba_{0.885}Sr_{0.1}La_{0.01}TiO₃ – yNi_{0.8}Zn_{0.2}Fe₂O₄ where y = 0.20.3, 0.4

Fig. 3a shows variation of dielectric constant versus log f for (1-y) $Ba_{0.885}Sr_{0.1}La_{0.01}TiO_3 - yNi_{0.8}Zn_{0.2}Fe_2O_4$ where y = 0.2, 0.3, 0.4 at room temperature. From the Fig. 3a it is clear seen that the dielectric constant decrease rapidly with increasing frequency and then reaches a constant value at higher frequency. The high value of dielectric constant at lower frequency and low value of dielectric constant at high frequency due interfacial polarization and dispersion due to Maxwell-Wagner [1]. At lower frequencies, the values of dielectric constant are low and are explained on the basis of space charge polarization due to inhomogeneous dielectric structure. The inhomogeneities in the present system are impurities, porosity and grain structure. Fig. 3b shows the variation of loss tangent (tan δ) as a functions frequency (log f) at room temperature which has a similar dispersion as that of dielectric constant.

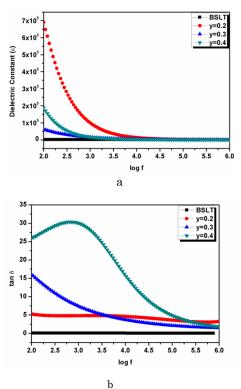


Fig. 3 – Variation of dielectric constant versus $\log f$ (a) Variation of loss tangent (tan δ) as a functions frequency ($\log f$) (b) and for (1-y) Ba0.885Sr0.1La0.01TiO3-yNi0.8Zn0.2Fe2O4 where $y=0.2,\ 0.3,\ 0.4$

4. CONCLUSIONS

Magnetodielectric composites with composition $(1-y)Ba_{0.885}Sr_{0.1}La_{0.01}TiO_3-yNi_{0.8}Zn_{0.2}Fe_2O_4(BSLT-NZFO)$ where y=0.2,0.3,0.4 have been prepared by Hydroxide Co-precipitation route. X-ray diffractogram patterns reveal the presence of the ferromagnetic and ferroelectric phases without any impurity phase in the composites. From the SEM studies, it is observed that average grain size is small and morphology is changed as ferromagnetic phase increased. Variation of dielectric constant with frequency dispersion due interfacial polarization.

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