ISO 9001 - 2015

ISSN 2349 - 4891

Monthly

IJRRAS

IF 4.665

Volume 4,Issue 6, June 2017

International Journal of Recent Research and Applied Studies

SURRAGH PUBLICATIONS

Original Article

ISSN: 2349 - 4891

International
Journal of Recent Research and Applied Studies
(Multidisciplinary Open Access Refereed e-Journal)

Structural and Electrical Properties of $Ba_{0.7}Sr_{0.3}TiO_3/Ni_{0.7}Zn_{0.3}Fe_2O_4$ Nano Composite

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Received 13th May 2017, Accepted 15th June 2017

Abstract

Composites materials containing both of ferroelectric and ferromagnetic phase with the composition **(x)**$Ba_{0.7}Sr_{0.3}TiO_3$+**(1-x)**$Ni_{0.7}Zn_{0.3}Fe_2O_4$ *(BST—barium Strontium Titanate), in which* **x** *varies as*(**x**=90, 70,50%)*.where the powder of compound perpetrated using Sol-Gel auto combustion technique in molar ratio[1:1] ,The Powder Calcination at 900ºC and Press at (7ton/cm^2). Then the samples were sintered at 1200°C for (2h)***.** *The structural and electrical characterizations of the investigated nanocomposite are discussed and reported. The formation of nanosized composite with two separate phases was confirmed by X-ray diffraction, scanning electron microscopy (SEM). XRD results show that the sample have two phase(tetragonal)for* $Ba_{0.7}Sr_{0.3}TiO_3$ *and (cubic)for* $Ni_{0.7}Zn_{0.3}Fe_2O_4$ *and all prepared samples are in nanoscale size (18-35nm) and porosity decreases with increasing the overlay ratios to minimum value(24%) at ratio of composition (50%) . The variation of dielectric constant (ε'), dielectric loss factor (ε") , the ac and dc conductivity of (x)*$Ba_{0.7}Sr_{0.3}TiO_3$**+** *(1-x)*$Ni_{0.7}Zn_{0.3}Fe_2O_4$ *was investigated as a function of frequency (50 Hz–1 MHz). This study enhances the use of this prepared system in memory applications.*

Keywords: Composites, ferroelectric, auto combustion, XRD, SEM, Structural Properties.

1. Introduction

Composite materials containing both ferroelectric and ferromagnetic phases have recently attracted a great deal of attention because of their potential applications in practical electronic devices[1]. The term multiferroic (MF) was first used by H. Schmid in 1994. His definition referred to multiferroics as a single phase materials which simultaneously possess two or more primary ferroic (ferroelectric, ferromagnetic and ferroelastic) properties. Today the term multiferroic has been expanded to include materials which exhibit any type of long range magnetic ordering, spontaneous electric polarization, and/or ferroelasticity. [2]. It is known that Perovskite such as Barium Strontium Titanate ($Ba_{0.7}Sr_{0.3}$TiO3) can stabilize in tetragonal phase at room temperature and it's an excellent ferroelectric material that has been used in capacitors for half a century [3] and Nickel-Zink ferrite is known as ferromagnetic material and has been used in the production of multilayer chip inductors [4]. There are many reported work on these materials. In this work we aimed to prepare and characterize the magnetoelectric nano-composites of the formula(x) $Ba_{0.7}Sr_{0.3}TiO_3$+ (1-x) $Ni_{0.7}Zn_{0.3}Fe_2O_4$ with x=90%, 70%and 50% to improve their structural and electrical properties, where the present BSTO-NZF ceramic are possible candidates for ME applications nanopowders obtained by auto-combustion technique . It was shown that auto-combustion synthesis is very convenient for obtaining the ferrite powder as a pure phase with good properties[5].To obtain barium Strontium Titanate by this method is not so simple due to the possible appearance of secondary phases which later may complicate the process of obtaining satisfactory properties of multifferoic composites[6]. A number of different methods were used to characterize obtained powders and ceramic composites in order to fabricate functional multiferroic material with both, ferroelectric and magnetic properties. The ferroelectric–ferromagnetic composites, as two-phase multiferroic materials, are desired not only for the fundamental research of magneto-electric effect, but also for the potential applications in many electronic devices [7].The most widely studied systems correspond to Co or Ni ferrites, with PZT, PNT, BT or BST [8]. Among them, the Ni-Zn ferrites/$BaSrTiO_3$ systems need to be further investigated because of high electrical resistivity, chemical stability and excellent electromagnetic properties of the Ni–Zn ferrites, and high permittivity, low dielectric loss.

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2. Experimental methods:

In this research, Nano Barium Strontium Titanate ($Ba_{0.7}Sr_{0.3}TiO_3$), Nano Nickel Zinc Ferrite

($Ni_{0.7}Zn_{0.3}Fe_2O_4$) compounds was synthesized by sol-gel auto-combustion using nitrate materials, Stoichiometric amounts(Ba(NO3)2 ,Sr(NO3)2, TiO_2, Fe(NO3)3·9H2O ,Zn(NO3)2), ammonia and citric acid ($C_6H_8O_7$) using molar ratios, [1: 1] [nitrate: fuel] where it was calcinations powders of nanoparticles at a temperature (900 C°) for (2h). Then the composites ($xBa_{0.7}Sr_{0.3}TiO_3$ + 1-$xNi_{0.7}Zn_{0.3}Fe_2O_4$) was prepared **by** mixing the powder with rate (x=90,70,50%) and the powder compress under pressure (7 ton / cm^2) and sintering temperature (1200 C°) for (2h) with a heating rate of 5˚C/min, The crystal structure of the prepared nanocomposite and their constituent phases were determined by *X*-ray diffractometer (SHIMADZV6000) with Cu-Kα radiation (λ = 0.15406nm) in a wide range of Bragg's angle (2θ) ranging from (10˚ - 90˚) at room temperature. The average particle size (D) was calculated from *X*-ray line broadening using (311)(101) peaks ,Williamson-Hall and Debye-Sherrer's equation . Scanning electron microscope (SEM) .The electrical properties such Dielectric Constant, Dielectric Loss Factor, Dielectric strength ,A.C and D.C conductivity using LCR meter (LRC-8105G(20), 5MHz, Taiwan) as a function of Frequency from (50Hz-1MHz).

3.1 Results and discussion :

XRD patterns of (x)$Ba_{0.7}Sr_{0.3}$TiO3 + (1-x) Ni0.7Zn0.3Fe2O4 ME nanocomposites is shown in Figure I. From the figure it is clear that, there are two phases: cubic structure $Ni_{0.7}Zn_{0.3}$Fe2O4 ferrite (piezomagnetic PM) and Perovskite tetragonal crystal structure $Ba_{0.7}Sr_{0.3}$TiO3 (piezoelectric PE). No third phase coexists in the prepared nanocomposites. This means that, no significant chemical reaction takes place during sintering of the mixed powders. The pattern revealed that, the highest peak value of XRD intensity corresponding to perovskite phase of PE which occurred at 2θ = 31.57˚. The value of lattice constant (*a*) for the PM phase is 8.359 Å, while those of (*ac*) for tetragonal PE phase are 3.99 Å and 4.0093 Å respectively. The average crystalline size (D) of the pellets are calculated using Debye Scherrer's formula[9] :

$$D = K\lambda/\beta\cos\theta \quad \ldots\ldots\ldots\ldots\ldots(1)$$

Where K is the constant, β is the full width at half maximum (FWHM), θ is the diffraction angle, and λ is wavelength of the X-ray, The K, λ values are taken as 0.9, 1.5406 ˚A for the calculations, respectively. and Williamson-Hall formula [10]:

$$\beta\cos\theta = k\,\lambda/D_{w-H} + [4\varepsilon\,\sin\theta] \quad \ldots\ldots\ldots\ldots\ldots(2)$$

Where ($\mathcal{E}$) is the micro strain.The crystalline size are (18-28 nm) and(31- 35 nm) respectively .Porosity decrease with increase of the ratio of composition reach to minimum value at overlay ratio (50%) which shown in table (1)

Table 1
Porosity of nanocomposite samples

Ratio of composition(x)	Porosity%
90%	30.2
70%	29.4
50%	24.47

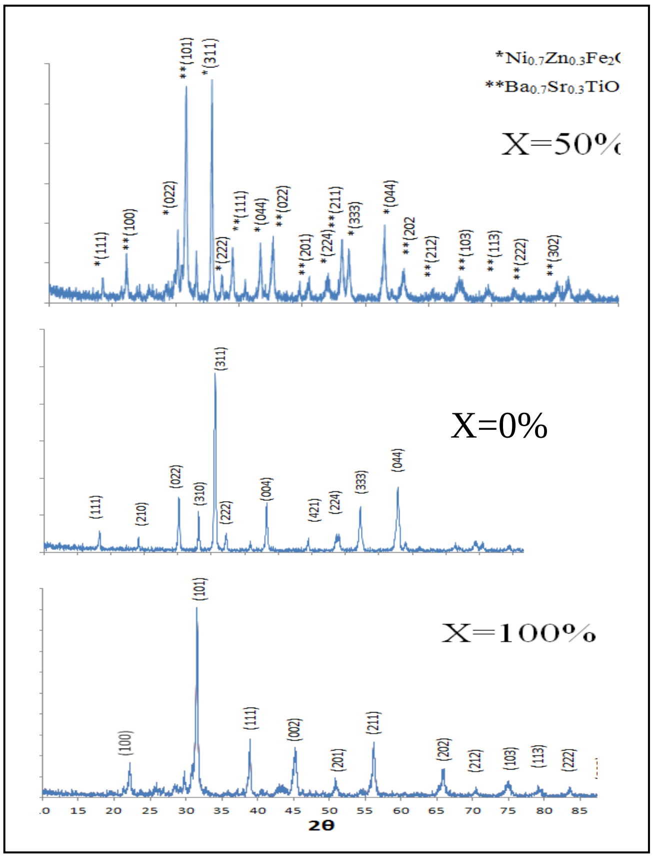


Figure I
XRD pattern of (x) $Ba_{0.7}Sr_{0.3}TiO_3$+ (1-x) $Ni_{0.7}Zn_{0.3}Fe_2O_4$.

The Rietveld refinement data of nano composite (x)$Ba_{0.7}Sr_{0.3}TiO_3$+ (1-x) $Ni_{0.7}Zn_{0.3}Fe_2O_4$ with (x=0,100%) carried out using FullProf program. The principle of the Rietveld method is to minimize the difference between the calculated profile and the observed data. The parameters Rp,Rwp,χ2,GOF,Phase and Space group, obtained after refinement are show in table (2) and Figure (II) .[11]

Table 2
Rietveld refinement data of nano composite $(x)Ba_{0.7}Sr_{0.3}TiO_3$+ (1-x) $Ni_{0.7}Zn_{0.3}Fe_2O_4$ with (x=0,100%)

Sample	R_{wp}	R_{exp}	χ^2	GOF	Space group	Phase
X=0%	39.0	27.1	2.07	1.43	P4mm	Tetragonal
X=100%	25	24.63	1.02	1.015	FD-3m	Cubic

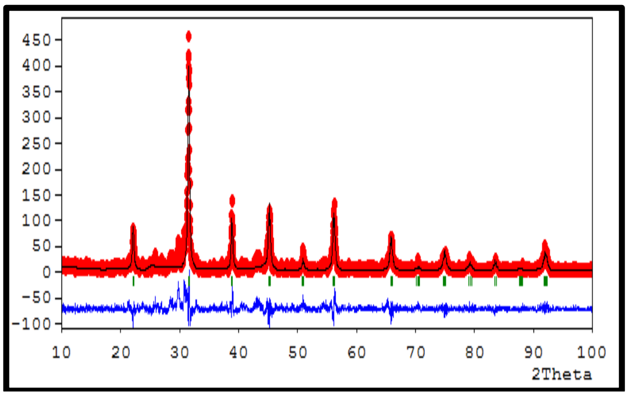


Figure IIa
Rietveld refinement of nano composite $(x)Ba_{0.7}Sr_{0.3}TiO_3$+ (1-x) $Ni_{0.7}Zn_{0.3}Fe_2O_4$ with (x=0 %)

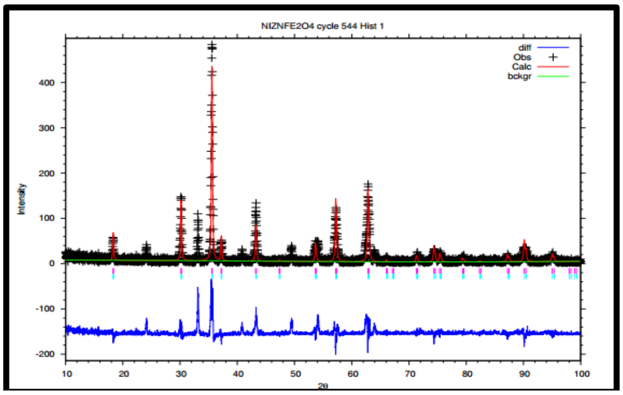


Figure II b
Rietveld refinement of nano composite $(x)Ba_{0.7}Sr_{0.3}TiO_3$+ (1-x) $Ni_{0.7}Zn_{0.3}Fe_2O_4$ with (x=100 %)

3.2 Scanning Electron Microscope (SEM).

The scanning electron microscope was used to determine the average crystallite size and the surface morphology. It gives information about the grain evolution and grain size. It also gives the information about the inter-granular and the intra-granular pores and the distribution of grains in the bulk samples. SEM measurements are based on the principle of irradiating the specimen with a finely focused electron beam. The secondary electrons, backscattered electrons, auger electrons, characteristic x-rays and several other radiations are released from the specimen. Generally, the secondary electrons are collected to form the image in the SEM mode[12]. Figure(III) shows scanning electron micrographs (SEM) of the nano-composite sample (50%$Ba_{0.7}Sr_{0.3}TiO_3$+ 50%$Ni_{0.7}Zn_{0.3}Fe_2O_4$)

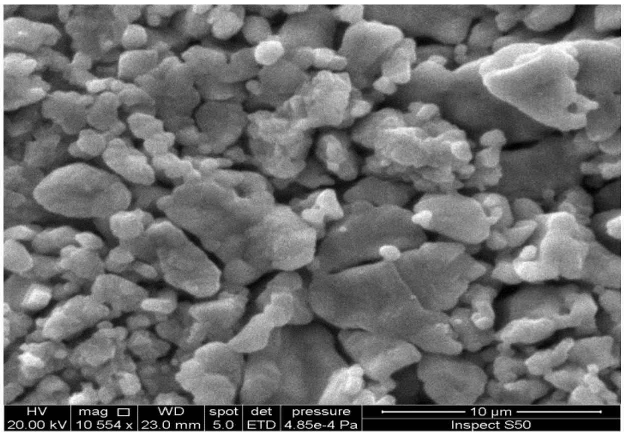


Figure III
SEM for nanocomposite (50%$Ba_{0.7}Sr_{0.3}TiO_3$ +50%$Ni_{0.7}Zn_{0.3}Fe_2O_4$)

3.4 Dielectric constant (ε'_r)

The Dielectric constant (ε') of the samples was calculate using this formula[12].

$$\varepsilon'_r = \frac{Cd}{\varepsilon_o A} \quad \cdots\cdots\cdots\cdots\cdots(3).$$

Where C is the capacitance of the pellet, d is the thickness of the pellet, A is the cross-sectional area of the flat surface of the pellet, and ε_0 is the permittivity of free space($8.85*10^{-12}$ F/m). Fig. 4 shows the variation of dielectric constant (ε'_r) as function of frequency(50Hz-1MHZ) for nano composites ($xBa_{0.7}Sr_{0.3}TiO_3$ + 1-$xNi_{0.7}Zn_{0.3}Fe_2O_4$) with variation ratio (x=90,70,50%) at room temperature. All the samples show usual dielectric dispersion behavior, where the dielectric constant decreases with the increase in frequency and overlay ratio to reach minimum value at frequency (1MHz) and overlay ratio (50%)

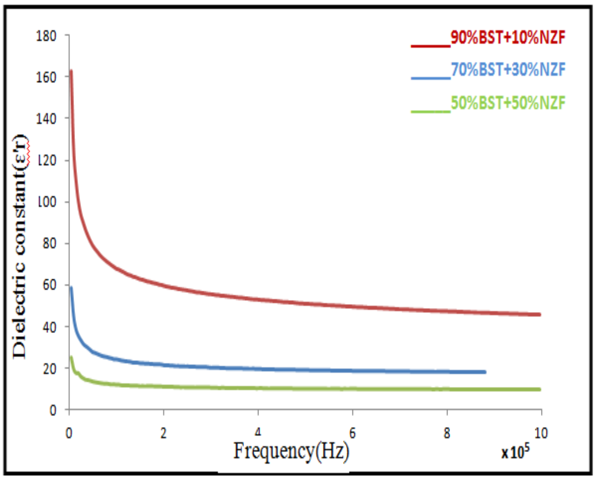


Figure IV
Effect of Frequency on Dielectric constant for ($xBa_{0.7}Sr_{0.3}TiO_3$ + 1-$xNi_{0.7}Zn_{0.3}Fe_2O_4$)

3.3 Dielectric Loss (ε''$_r$)

Figure (V) shows the variation of dielectric lose factor (ε''$_r$) with frequency (50Hz-1MHz)at room temperature .The dielectric loss factor decreases with increasing frequency ,the rate of decreasing is slow in higher frequency region show the potential applications of these materials in high-frequency microwave device .Dielectric loss decreases with the increase in frequency and overlay ratio to reach minimum value at frequency (1MHz) and overlay ratio (50%).

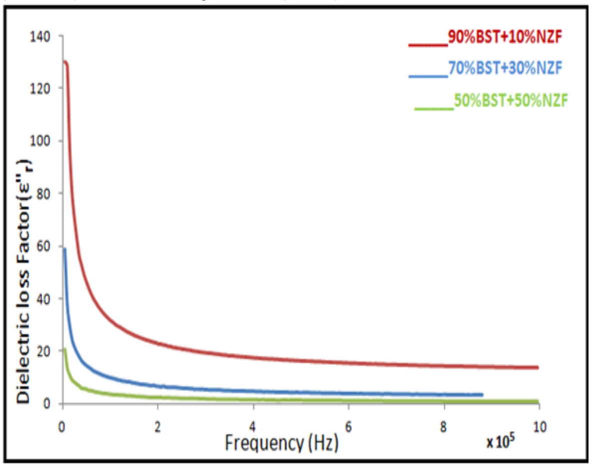


Figure V
Effect of Frequency on Dielectric loss for ($xBa_{0.7}Sr_{0.3}TiO_3 + 1\text{-}xNi_{0.7}Zn_{0.3}Fe_2O_4$)

3.4 A.C conductivity (σ_{ac})

The *ac* part of the electrical conductivity was calculated from values of dielectric constant and dielectric loss factor using the relation:[13]

$$\sigma_{a.c} = \omega\, \varepsilon_0 \varepsilon_r' \tan\delta \quad \text{.................(4)}$$

where ω is the angular frequency . Figure VI shows the variation of ac conductivity with frequency (50Hz-1MHz). The ac conductivity increases with increasing frequency and increasing the overlay ratio reach to maximum value at frequency (1MHz) and overlay ratio (50%) .

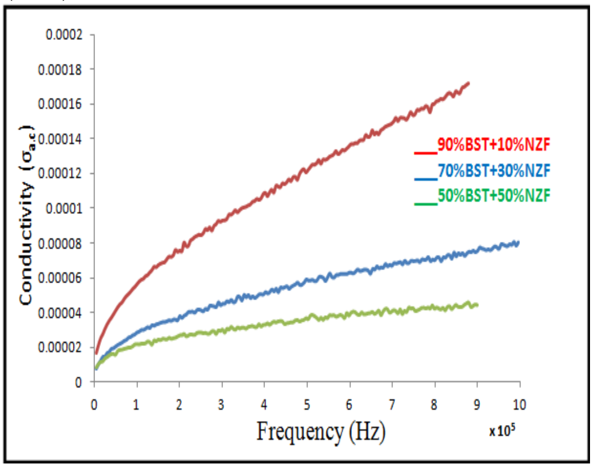


Figure VI
Effect of Frequency on ac conductivity for ($xBa_{0.7}Sr_{0.3}TiO_3 + 1\text{-}xNi_{0.7}Zn_{0.3}Fe_2O_4$)

3.5.D.C conductivity :

Dc conductivity was calculated using formula :[14]

$$\sigma_{d.c} = \frac{1}{R}\frac{t}{A} \quad \text{..............(5)}$$

From table (4) obtained that dc conductivity decrease with increasing the overlay ratio reach to minimum value at (50%)

Table 4
Effect of overlay ratio on the D.C conductivity

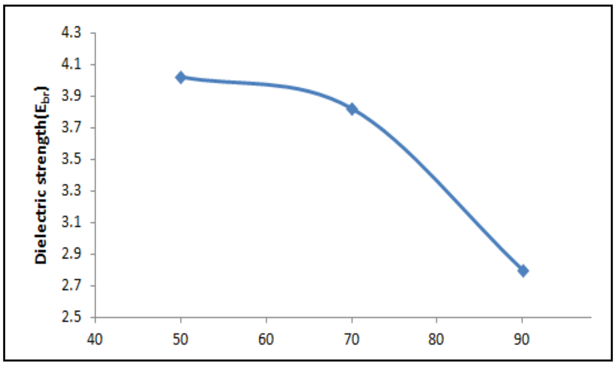
Ratio composition(x)	$\sigma_{D.c}$ $(\Omega.\text{cm})^{-1}$
90%	1.52E-19
70%	1.48E-19
50%	1.46E-19

3.6. Dielectric strength(E_{br})

Is the maximum electric field strength that it can withstand intrinsically without breaking down, *i.e.* , without experiencing failure of its insulating properties. The atoms in insulating materials have very tightly-bound electrons, resisting free electron flow very well. However, insulators cannot resist indefinite amounts of voltage. With enough voltage applied, any insulating material will eventually succumb to the electrical "pressure" and electron flow will occur.[15,16] Figure(VII)and Table(5) shows that dielectric strength increase with increasing Substituted Substance reach to maximum value at overlay ratio (50%).

Table 5
Dielectric strength

Ratio composition(x)	E_{br} (Kv/mm)
90%	2.8
70%	3.825
50%	4.025



Figure(VII) Effect of composition on Dielectric strength for nanocomposite(x) $Ba_{0.7}Sr_{0.3}TiO_3$+ (1-x) $Ni_{0.7}Zn_{0.3}Fe_2O_4$

Conclusions

The ME nanocomposites $xBa_{0.7}Sr_{0.3}TiO_3+(1-x)$ $Ni_{0.7}Zn_{0.3}Fe_2O_4$ .are successfully prepared by Sol-Gel auto combustion. In addition to tetragonal $Ba_{0.7}Sr_{0.3}TiO_3$, and cubic phase is clearly observed in XRD patterns of ceramics. The Average crystallite size (D) was calculated as (18-35nm) using Williamson's Hall and Debay-shere equation. Scanning electron microscope gives the morphological studies. Dielectric constant and Dielectric loss for all samples decreases with increases of frequency and overlay ratio to reach minimum value at frequency (1MHz)and overlay ratio (50%) . dielectric strength increase with increasing Substituted Substance reach to maximum value at overlay ratio (50%).A.C conductivity increases with increasing the frequency and overlay ratio. Porosity and D.C conductivity decrease with increasing overlay ratio to reach minimum value at overlay ratio (50%). synthesized by this method are useful in many applications.

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