Structural and Electrical Properties of $Ba_{0.7}Sr_{0.3}TiO_3/BaFe_{12}O_{19}$ nano composite

Noor S. Abed[1], Noor A. Ahmed [2] , Farouq I. Hussain[3], Sabah J. fathi[2]

[1] Director of Education of Kirkuk Kirkuk Iraq.

[2] Department of Physics College of Sciences Kirkuk University Kirkuk Iraq

[3] Department of Physics College of Education for Pure Science Ibn ALHaitham University of Baghdad Iraq

Abstract : The nanocomposite with the composition xBa07Sr03TiO3 + 1-x BaFe12O19 with ratio (x=90,70,50)% was synthesized by sol-gel auto composition method Both of electrical and structural properties of nanocomposite investigated and discussed and reported The nanosized composite was confirmed using XRD technique SEM and EDX The electrical properties such a dc and ac conductivity dielectric loss factor (ε_r) and dielectric constant (ε'_r) was investigated as a function of frequency from(50 Hz to 1 MHz) by using LCR meter Device The results of X-ray Diffract meter show that the samples have two phase tetragonal for $Ba_{0.7}Sr_{0.3}TiO_3$ and Hexagonal for $BaFe_{12}O_{19}$ and all prepared samples are in nanoscale size (21-54) nm and porosity decreases with increasing the overlay ratios to minimum value(28.1)% at ratio of composition (50%) Dielectric Strength was studied as a function of overlay ratio.
Keywords: Composites, ferroelectric, auto combustion, XRD, SEM, EDX, Structural Properties.

Introduction:

The Perovskite Barium Strontium Titanate ($Ba0.7Sr_{0.3}TiO_3$)(BST) and Hexaferrite Barium Ferrite ($BaFe_{12}O_{19}$)(BF) have been prepared and characterized in this present study using the Chemical method Sol-gel auto-combustion. Composite materials includes ferroelectric and ferromagnetic phases have recently entice a great deal of attentiveness because of their applications in electronic devices. At high Temperature phase transition [2]. The chemical formula Barium hexaferrite $BaFe_{12}O_{19}$ form the hexaferrite family $BaFe_{12}O_{19}$ is the best known representative Which is used widely in magnetic recording media microwave applications and permanent magnets because it classify as an one of the most important hard magnetic materials [3].

Multiferroic (MF) term mention to materials which have single phase which used in 1994 by H Schmid Referred to which have at same time two or more primary ferroic (ferroelastic ferromagnetic and ferroelectric) properties [4]. These types of materials have stimulated a researcher to increasing attention in present day.

Which is using for applications as multifunctional devices such as sensors transducers actuators and multiple state memory elements [5].

The Multiferroic (MF) composite materials properties depend on the details of phase connectivity grain form volume fraction and the component phase properties Different connective structures have been tried to enhance the magneto electric and electrical properties[6].

In this work we aimed to prepare and characterize the magneto electric nanocomposites of the formula ($xBa_{07}Sr_{03}TiO3$+ 1-x $BaFe_{12}O_{19}$) with (x=90, 70and 50%) to improve their structural and electrical properties where the present BST, BF ceramic are possible candidates for ME applications nanopowders obtained by auto-combustion technique.

Experimental part:

In this research Nano Barium Strontium Titanate $Ba_{0.7}Sr_{0.3}TiO_3$ Nano Barium Ferrite $BaFe_{12}O_{19}$ compounds was synthesized by sol-gel autocombustion using nitrate materials Stoichiometric quantity $Ba(NO_3)_2$, $Sr(NO_3)_2$, TiO_2, $Fe(NO_3)_3\cdot 9H_2O$, 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-x $BaFe_{12}O_{19}$) 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) The crystal structure of the attend nanocomposite and their constituent phases were determined by X-ray diffractometer SHIMADZV6000 with Cu-Kα radiation(λ =15406Å) in a extensive range of Bragg's angle 2θ ranging from (10˚ - 90˚) at RT Average particle size D was calculated using (101) peaks from X-ray line widening Williamson-Hall and Debye-Scherrer's equation Scanning electron microscope (SEM). The electrical properties such Dielectric Loss Factor, Dielectric Constant, Dielectric strength, AC and DC conductivity using LCR meter LRC8105G20 5MHz Taiwan as a function of Frequency from (50Hz-1MHz).

Results & discussion :

XRD manner of (x $Ba_{07}Sr_{03}TiO_3$+ 1-x $BaFe_{12}O_{19}$):

From the figure1 we show two phases: Hexagonal for $BaFe_{12}O_{19}$ ferrite and piezoelectric tetragonal Perovskite structure $Ba_{0.7}Sr_{0.3}TiO_3$,So there is no important chemical reaction occur during sintering of the mixed powders and the highest peak value of XRD intensity corresponding to perovskite phase which happen at 2θ = 31.57˚. The lattice constant ac value of Piezomagnetic phase are 58.86 Å and 23.182 Å while for tetragonal Piezoelectric phase the ac are 3.99 Å and 4.0093 Å severally and the average crystalline size D of the pellets are calculated by Debye Scherrer's formula[7] :

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

Where K : constant ,β : full width at half maximum (FWHM), θ : diffraction angle and λ is wavelength of the X-ray . The value of *K* is taken as 0.9, *λ* is 1.5406 ˚A and Williamson-Hall formula [8]:

$$B\cos\theta = (K\lambda/D_{W-H})+[4\,\mathcal{E}\,\sin\theta] \qquad (2)$$

Where Ɛ 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	*Porosity%*
%90	28.6
%70	28.3
%50	28.1

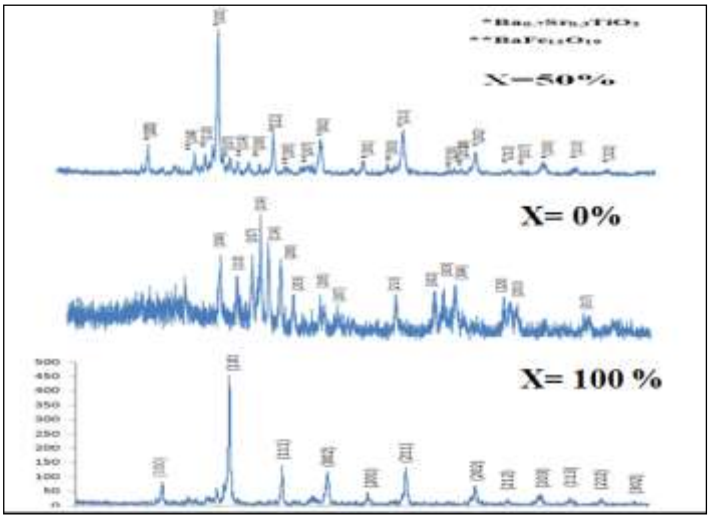

Figure (1): XRD pattern of (x $Ba_{07}Sr_{03}TiO_3$ + 1x $BaFe_{12}O_{19}$)

The Rietveld refinement data of nano composite (x $Ba_{07}Sr_{03}TiO_3$+ 1x $BaFe_{12}O_{19}$) with (x=0-100%) carried out using Full-Prof program The principle of the Rietveld method is to minimize the difference between the observed data and the calculated profile [9]The parameters Rp, Rwp, χ2, GOF, Phase and Space group obtained after refinement are show in table (2) and Figure (2):

Table (2): Rietveld refinement data of nano composite x $Ba_{07}Sr_{03}TiO_3$+ 1x $BaFe_{12}O_{19}$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%	35	30.7	1.29	1.14	P63mmc	Hexagonal

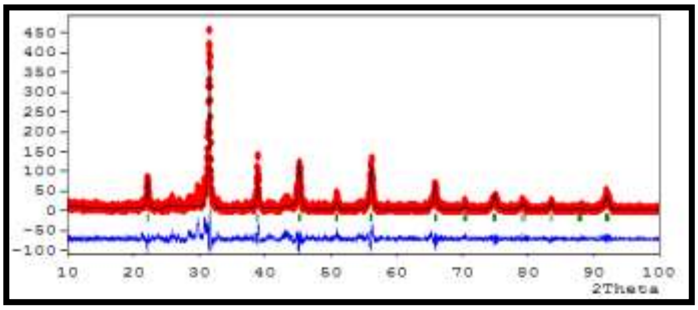

Figure (2a): Rietveld refinement of nano composite (x $Ba_{07}Sr_{03}TiO_3$+ 1x $BaFe_{12}O_{19}$) with x=100 %

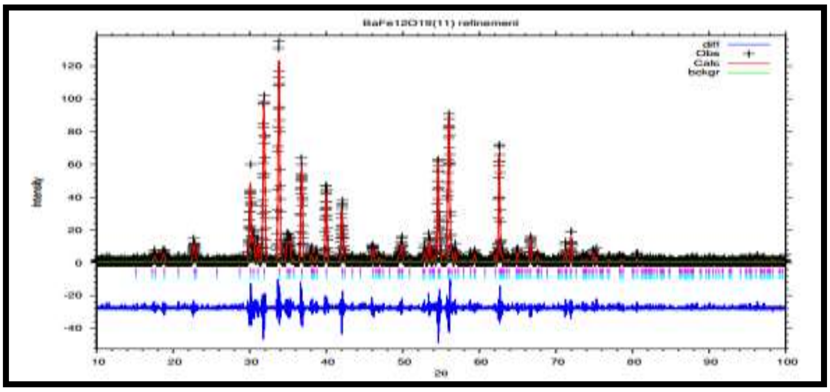

Figure (2b): Rietveld refinement of nano composite (x $Ba_{07}Sr_{03}TiO_3$+ 1x $BaFe_{12}O_{19}$) with x=0 %

Scanning Electron Microscope (SEM):

SEM is used to locate the surface morphology and the average crystallite size The SEM gives details about the grain size and the grain development and the details about the inter and intragranular pores and the distribution of grains in the bulk samples[10] Figure(3) shows scanning electron micrographs SEM of the nanocomposite sample 50%$Ba_{0.7}Sr_{0.3}TiO_3$+ 50%$BaFe_{12}O_{19}$.

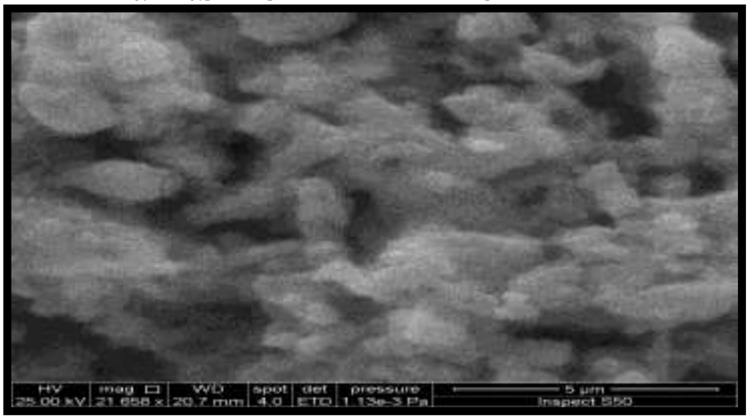

Figure (3): SEM for nanocomposite 50%$Ba_{07}Sr_{03}TiO_3$ + 50% $BaFe_{12}O_{19}$

Energy Dispersive X-Ray Spectroscopy:

EDX is an analytical technicality used for chemical characterization or the elemental analysis of samples As a type of spectroscopy It depend on the inquiry of a sample during interaction electromagnetic radiation with matter. The EDX attached to technique the SEM device[11]. To insure the presence of constituents of the nano particle composite (x$Ba_{07}Sr_{03}TiO_3$ + 1-x $BaFe_{12}O_{19}$) we used EDX which is show in the Figure below From Figure (4) we show present of component elements of the nanoparticles that form the composites.

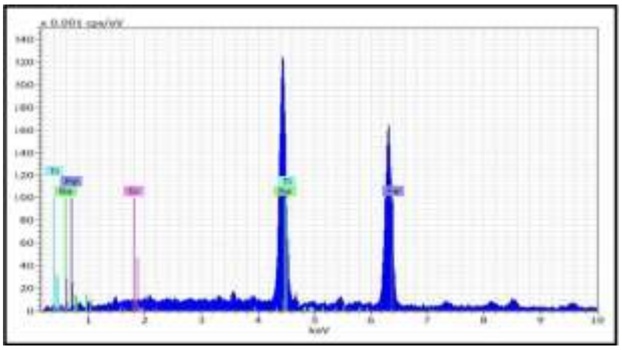

Figure (4) EDX image of nanocomposite 50%$Ba_{07}Sr_{03}TiO_3$ + 50% $BaFe_{12}O_{19}$.

Dielectric constant(ε'):

The ε' of all samples was calculate using this formula [12].

$\varepsilon'=ct/\varepsilon_o A$ (3)

C is pellet capacitance t is the thickness of pellet A is pellet cross sectional area of the flat surfaceε^0 is ($885*10^{12}$ F/m) the permittivity of free space Fig (5) shows the variance of dielectric constant ε' as function of frequency 50Hz1MHZ for nano composites(x $Ba_{07}Sr_{03}TiO_3$+ 1-x $BaFe_{12}O_{19}$) with variation ratio x=90,70,50% at RT. From Fig (5) we show that dielectric constant lessen with the increase in frequency and overlay ratio to reach minimum value at frequency (1MHz) and overlay ratio (50%) and the samples show usual dielectric dispersion behavior.

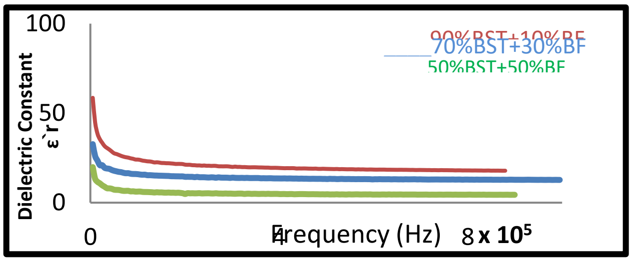

Figure (5): Effect of Frequency on (ε')for (x $Ba_{07}Sr_{03}TiO_3$+ 1x $BaFe_{12}O_{19}$)

Figure (6) shows the variation of dielectric lose factor ε" with frequency (50Hz-1MHz) at room temperature The dielectric loss factor lessen 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 lessen with the increase in frequency and overlay ratio to reach minimum value at frequency 1MHz and overlay ratio 50%.

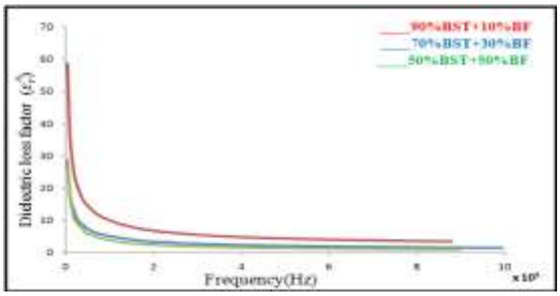

Figure (6): Effect of Frequency on Dielectric loss for $xBa_{07}Sr_{03}TiO_3$ + 1-x $BaFe_{12}O_{19}$

AC conductivity σ_{ac}: The ac part of the electrical conductivity was measured using the values of the dielectric loss factor and dielectric constant using the relation [13]

$\sigma_{ac} = \omega\varepsilon_{\circ}\varepsilon'_r \tan\delta$ (4)

ω is the angular frequency. Figure (6) shows the change of ac conductivity with frequency (50Hz-1MHz),The ac conductivity augmentation with increasing frequency and the overlay ratio reach to maximum value at frequency 1MHz and overlay ratio 50%.

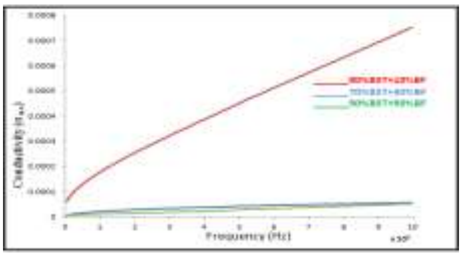

Figure (6): Effect of Frequency on ac conductivity for $xBa_{07}Sr_{03}TiO_3$ +1x $BaFe_{12}O_{19}$

DC conductivity: Dc conductivity was calculated using formula [14]:

$\sigma_{da} = t /RA$ (5).

From table (3) obtained that dc conductivity lessen with increasing the overlay ratio reach to minimum value at 50%.

Table (3): Effect of overlay ratio on the DC conductivity

Ratio composition	σ_{da} $(\Omega . cm)^{-1}$
%90	1.57E19
%70	1.51E19
%50	1.51E20

3.7. Dielectric strength:

Is the maximum electric field strength that it can hold out intrinsically without fiasco of and breaking down its insulating properties.

At fact the insulators cannot with standing undefined amounts of the voltage with sufficient voltage applied the insulating material finally will undergo to the electrical pressure and electron flux will happen [15]. Table (4) shows that dielectric strength increase with increasing Substituted Substance reach to maximum value at overlay ratio 50%.

Table (4): Dielectric strength for nanocomposite(x $Ba_{07}Sr_{03}TiO_3$+ 1x $BaFe_{12}O_{19}$).

Ratio composition	E_{br} *(Kv/mm)*
%90	3.025
%70	3.275
%50	4.4

Conclusions:

The ME nanocomposite ($x$$Ba_{07}Sr_{03}TiO_3$+1$x$ $BaFe_{12}O_{19}$) are successfully prepared by Sol-Gel auto combustion In addition to tetragonal ($Ba_{07}Sr_{03}TiO_3$)and Hexagonal phase is clearly observed in XRD patterns of ceramics. The Average crystallite size (D) was calculated as (16-29) nm using Williamson's Hall and Debye-Scherrer 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%. AC conductivity augment with increasing the frequency and overlay ratio, Porosity and DC conductivity decrease with augmenting overlay ratio amount to minimum value at overlay ratio 50% synthesized by this method are useful in many applications .

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