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Preparation of Magneto Electric ($Bafe_{12}o_{19}$ / $Bifeo_3$) Composites using Sol-Gel Auto Combustion Method

G. Packiaraj, Rajshree B. Jotania, Jyothi Budida, Pankaj Kumar Modi

Abstract*: The coexistence of ferromagnetic and ferroelectric properties in single phase material leads to the existence of new type of material known as magnetoelectric. In present work, magneto electric composites $BaFe_{12}O_{19}$ / $BiFeO_3$ with different mass percentage of $BiFeO_3$ (0, 25%, 50%, 75% and 100 %) were prepared. $BaFe_{12}O_{19}$ (BHF) and $BiFeO_3$ (BiF) ferrites were prepared separately using Sol-gel auto combustion method and then physically mixed. Prepared composite samples were characterized using FTIR, XRD, SEM and VSM. XRD spectra reveal the mixed hexaferrite and bismuth ferrite phases. SEM micrograph of showed the formation of porous clusters of non uniform grains in the composites. Saturation magnetization of $BaFe_{12}O_{19}$ / $BiFeO_3$ composites decreased gradually with increasing in $BiFeO_3$ content and there is no systematic change in coercivity values.

Keywords* : Magneto-electric, hexaferrite, multiferroic, VSM and composite.

I. INTRODUCTION

Multiferroic material exibhit ferroelectric as well as ferromagnetic nature in a single phase [1]. In the presence of electric and magnetic field such material induces magnetization and polarization which is called as magnetoelectriceffect [2]. There is a lot of demand for multiferroic materials in manufacturing many devices like filters, oscillators, phase shifters, transformers, magnetoresistance and magnetic field sonsors, gradiometer, etc [3],[4]. However, a single phase multiferroic material may not accomplish the requirements of desirable propertries for specific application. These can be obtained by composite materials having magnetoestrictive phases as well as ferro-piezoelectric phases. Various factors determine the selection of composite materials like high dielectric permeability, piezoelectric coefficient and magnetostriction coefficient [5]. Bismuth ferrite ($BiFeO_3$) is one of the common multiferroic material exhibit both ferroelectric and antiferromagnetic nature simultaneously [6], [7]. At room temperature, Bismuth ferrite has rhombohedral lattice system with the space group R3c. It demonstrates strong ferroelectricity with high remnant polarization below 800 ˙C and transforms into the paraelectric state above this temperature. Also it has relatively high antiferromagnetic Neel transition temperature. However, implementing BiFeO3 for practical applications encounter difficulties in preparation of large quantity of nano structured BiFeO3 with pure phase.

The crystallization of M-type barium hexaferrite has 64 ions per unit cell and the 24 Fe^{3+} ions are placed at different crystallite sites. Finally, 4 Fe^{3+} ions with unpaired spin contribute the net magnetic moment of 20 μB per unit cell. In recent years, nano ferrites are important magnetic materials for making long lasting magnet, storage device and certain microwave devices [8]-[10]. In present work, magnetoelectric composites of two constituent phases $BaFe_{12}O_{19}$ (BHF) and $BiFeO_3$ (BiF) with different mass percentage (25%, 50% and 75%) of $BiFeO_3$ have been prepared and individual ferrites were synthesized by Sol-gel auto combustion method. Effects of magnetic coupling on structural and magnetic properties were investigated.

II. EXPERIMENTAL DETAIL

Barium nitrate $Ba(NO_3)_2$ (Merck, GR grade), Iron nitrate, $Fe(NO_3)_3.9H_2O$ (Sigma Aldrich, > 98 % purity), Citric acid, $C_6H_8O_7$ (Merck, GR grade) and 25 % (0.91 g/cm^3) aqueous solution of NH_4OH (Merck , GR grade) were used as raw materials to prepare $BaFe_{12}O_{19}$ hexaferrite. Stoichiometric amounts of iron nitrates and barium nitrates were dissolved in an optimum amount of double distilled water. The aqueous solution of citric acid was added drop by drop into the metal nitrate solution, where molar ratio of citric acid and metal nitrates was 1:1. Chelation of Ba^{2+} and Fe^{3+} ions in the solution was induced by constant strirring at 45 ^oC for some time. Then the solution with pH = 2 was neutralised by adding ammonia solution drop by drop. Then the final dark greenish solution was maintained at 80 ^oC for further evaporation. Once the water content was evaporated, the solution became thick gel and after few minutes, the gel got ignited itself and form ash structure. The obtained combusted powder was precalcined at 500 ^oC followed by final calcinations at 950 ^oC for 4 hours.

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***Correspondence Author(s)
G. Packiaraj*, Department of Physics, Aditya College of Engineering, Surampalem, Kakinada – 533 437, Andhra Pradesh, India. Email: packiaraj33@gmail.com, gpr331067@gmail.com
Rajshree B. Jotania, Department of Physics, University School of Sciences, Gujarat University, Ahmedabad – 380 009, Gujarat, India. Email: rbjotania@gmail.com
Jyothi Budida, Department of Physics, Aditya College of Engineering, Surampalem, Kakinada – 533 437, Andhra Pradesh, India. Email: budidajyothi2012@gmail.com
Pankaj Kumar Modi, Department of Physics, Patliputra University, Patna-800013, Bihar, India. Email: pm9651@gmail.com

For the preparation of $BiFeO_3$ ferrite, Barium nitrate was replaced by Bismuth nitrate, $Bi(NO_3)_3$ (Merck, GR grade) in the above procedure. Since bismuth nitrate is insoluble in water, initially it was dissolved in diluted nitric acid to get clear solution. Iron nitrate, citric acid and ammonia were added in the solution as per the procedure, and then the neutralized solution was kept in hot plate at 80 ^oC to get combusted powder. The combusted powder was annealed at 500 ^oC to get ordered bismuth ferrite. The $BiFeO_3$ ferrite thus obtained was added with prepared $BaFe_{12}O_{19}$ hexaferrite powder in different weight percentage (25%, 50% and 75%) of $BiFeO_3$. Then mixed powders were calcined at 500 ^oC for 4 hours in a muffle furnace. Hereafter, the prepared samples were coded as 100% BHF, 25% BiF, 50% BiF, 75% BiF and 100% BiF for pure $BaFe_{12}O_{19}$, composites with different weight ratio and pure $BiFeO_3$, respectively.

III. RESULTS AND DISCUSSION

A. Structural properties

Fig. 1 shows FTIR spectra of indivitual $BaFe_{12}O_{19}$ and $BiFeO_3$ ferrites and the composites with different mass percentage (25%, 50% and 75%) of $BiFeO_3$ and spectra were recorded to confirm the formation of final product and nature of the residual carbon is analysed in the samples. FTIR spectrum of pure $BaFe_{12}O_{19}$ shows two predominant bands around 580 and 450 cm^{-1}, which are characteristics peaks of ferrites on account of metal oxygen stretching [11]. In case of $BiFeO_3$ ferrite, the FTIR peaks at 410, 547, 649 and 750 cm^{-1} are corresponding to the vibrations bonds of Bi – O or Fe – O [12]. There is no indication of presence of any residual carbon in the samples as observed usually in the samples prepared by precursor method. The intensity and sharpness of these peaks were altered in composites, which reflect the strong interaction between $BaFe_{12}O_{19}$ and $BiFeO_3$ particles.

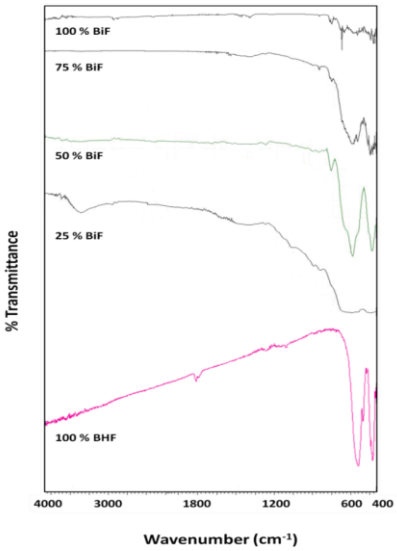

Fig. 1.FTIR spectra of $BaFe_{12}O_{19}$ / $BiFeO_3$ ferrite composites

Fig. 2 shows XRD patterns of $BaFe_{12}O_{19}$ and $BiFeO_3$ ferrites. The XRD pattern of $BaFe_{12}O_{19}$ (BHF) sample heated at 500 ^oC followed by 950 ^oC shows pure single phase M-type barium hexaferrite. In case of $BiFeO_3$ (BiF) heated at 500 ^oC, the prominent peaks in XRD plot reveal rhombohedral structure with the dimensions a = b = 5.577 Å and c = 13.861 Å, which are matched with the standard JCPDS file- PDF# 861518 [13]. Besides these prominent peaks, some other peaks are also observed, which assigned to Bi_2O_3. The loss of bismuth due to high temperature synthesis process causes the formation of impure phases like Bi_2O_3 [14]. Hence the calcination temperature was restricted to 500 ^oC for this sample.

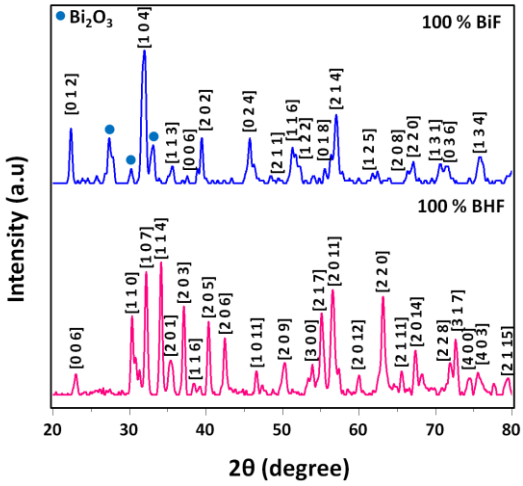

Fig. 2.XRD patterns of $BaFe_{12}O_{19}$ (BHF) and $BiFeO_3$ (BiF) ferrites

Table- I: Crystalline parameters of $BaFe_{12}O_{19}$ and $BiFeO_3$

Sample	Lattice constants		Unit cell volume ($Å^3$)
	a (Å)	c (Å)	
$BaFe_{12}O_{19}$ (BHF)	5.892	23.183	696.99
$BiFeO_3$ (BiF)	5.577	13.861	431.12

Fig. 3 shows the XRD spectra of $BaFe_{12}O_{19}$ / $BiFeO_3$ composites with various mass percentages (25%, 50% and 75%) of $BiFeO_3$. The XRD patterns of these three compositions reveal the mixed phases of hexaferrite and bismuth ferrite free from any noticeable third phase. So there was no any chemical reaction between two phases due to physical mixing and it was possible to make various composites with desirable properties. In the sample with 50% of BiF, the intensity of XRD peaks of both hexaferrite and bismuth ferrite is high compared to other two compositions. The intense peaks corresponding to BHF has been decreased with the increase of BiF content in the composition.

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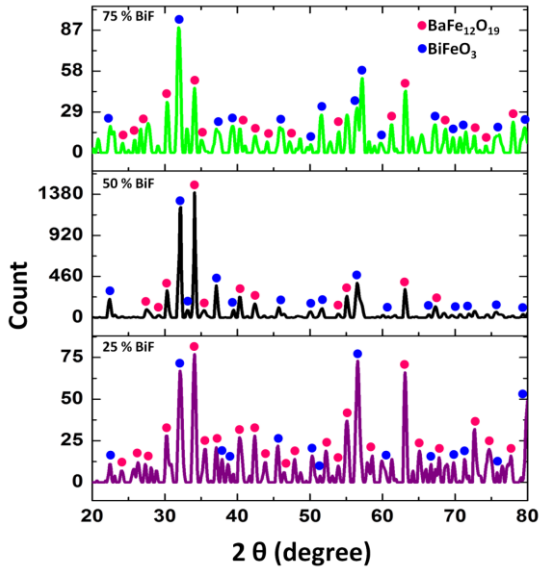

Fig. 3.XRD spectra of composites $BaFe_{12}O_{19}$/ $BiFeO_3$ with different mass percentage of $BiFeO_3$

B. Morphology

Fig. 4 shows the morphology of $BaFe_{12}O_{19}$, $BiFeO_3$ and the composite with 50% of $BiFeO_3$ powder calcined at 500 °C for 4 hours. The surface morphology of calcined $BaFe_{12}O_{19}$ powder sample revealed formation of dense grains. SEM micrograph of calcined $BiFeO_3$ ferrite shows the formation of porous clusters of non uniform grains. The SEM image of composite sample (50% of $BiFeO_3$ powder calcined at 500 °C for 4 hours) shows irregular, porous grains, which are randomly distributed.

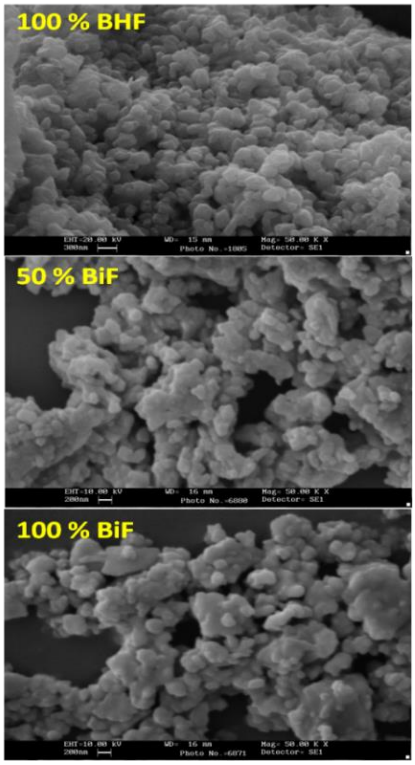

Fig. 4.SEM micrographs of BiFeO3 ferrites and the composite with 50% of BiFeO3

C. Magnetic properties

Hysteresis loops of $BaFe_{12}O_{19}$ and composites $BaFe_{12}O_{19}$ / $BiFeO_3$ are shown in Fig. 5 and magnetic values are given in Table 2. It is noticed that magnetic saturation (M_s) falls gradually with increasing in $BiFeO_3$ content. Similar results were also reported for $CrFe_2O_4$ - $BiFeO_3$ and $MnFe_2O_4$ - $BiFeO_3$ nanocomposites [15]. In ferrites, every grain is as source and responsible for magnetic moment and algebraic sum of all these individual moment result to net magnetism in the composites [16]. In present composites, the $BiFeO_3$ is being ferroelectric acts as tiny hole to passage of applied field and breaking magnetic circuitry. So, the incorporation of bismuth ferrite into hexaferrite system reduced the net magnetism.

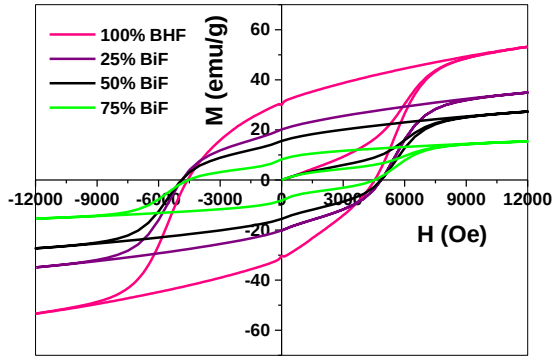

Fig. 5.Hysteresis loops of $BaFe_{12}O_{19}$ and composites $BaFe_{12}O_{19}$ / $BiFeO_3$ with different mass percentage

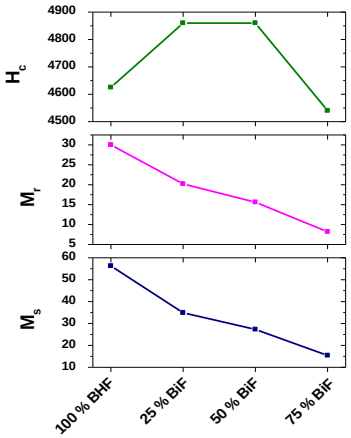

Fig. 6.Variation of Ms, Mr and Hc with increase in $BiFeO_3$ content

Table- II: Magnetic parameters of $BaFe_{12}O_{19}$ / $BiFeO_3$ composites

Sample	M_s (emu/g)	M_r (emu/g)	M_r / M_s	H_c (Oe)
100% BHF	56.24	30.00	0.533	4625
25% BiF	34.90	20.20	0.579	4860
50% BiF	27.30	15.60	0.571	4860
75% BiF	15.40	8.20	0.532	4540

Thus, total magnetic moment of the $BaFe_{12}O_{19}$/ $BiFeO_3$ composites decreases with addition of $BiFeO_3$ content and result in decrease of the net magnetization. However, there were no noticeable changes in coercivity values for composite samples with increase in $BiFeO_3$ content.

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Coercivity slightly increased for the 25% and 50% BiF composites and then decreased for 75% BiF composition compared to $BaFe_{12}O_{19}$ ferrite.

IV. CONCLUSIONS

The composites $BaFe_{12}O_{19}$ / $BiFeO_3$ with different mass percentage of $BiFeO_3$ (100% BHF, 25% BiF, 50% BiF, 75% BiF and 100% BiF) have been prepared by Sol-gel auto-combustion method. FTIR specta of $BaFe_{12}O_{19}$ / $BiFeO_3$ composites reveal the strong two absorption bands corresponding the metal – oxygen vibrations. XRD spectra of $BaFe_{12}O_{19}$ / $BiFeO_3$ composites reveal that mixed hexaferrite and bismuth ferrite phases have been formed. Micrographs of $BaFe_{12}O_{19}$ / $BiFeO_3$ composites show that irregular, porous and randomly distributed grains. Saturation magnetization (M_s) of $BaFe_{12}O_{19}$ / $BiFeO_3$ composites decreased gradually with increase of $BiFeO_3$ but coercivity values have not shown drastic changes.

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AUTHORS PROFILE

Dr. Packiaraj is currently working as Associate Professor in Aditya College of Engineerig, Surampalem, Kakinada since May, 2017. He has been awarded a Ph.D. degree in Physics from Gujarat University, Ahmedabad, Gujarat. He has participated and attended in various national and international conferences and workshops. His research areas include nano ferrite, Multiferroics and magnetic composites.

Prof. R.B. Jotania has joined Department of Physics, Elctronics and Space science, Gujarat University Ahmedabad, India on September 1997. She participated and attended in various national and international conferences and workshops. Her area of research is nanomagnetic materials: synthesis, cherecterization and applications. She has vast experiences on different types of hexagonal ferrites, hard-soft ferrite composites, ferrite -polymer composites, Microwave absorbing properties of ferrites.

Prof. Jyothi Budida has done Msc (Nuclear physics) from Andhra University and pursued M.tech(VLSI) from Bharath University and Pursuing PhD in physics from Dravidian University, Presently working as a Faculty of physics in Aditya College of Engineering, Surampalem. She has 10 national and International publications. Her area of interests are Antenna arrays , Radiation physics , Nano crystals.

Dr. Pankaj Kumar Modi, is working as Assistant Professor in Department of Physics, Patliputra University, Patna, Bihar, India**.** He has attended and presented paper in various national and international conferences. He has keen interest on Thin film and Solar cell.