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Structural and Magnetic Properties of $NiFe_2O_4$ + $BaZr_{0.2}T_{0.8}O_3$ ME Composites for Sensor Applications

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Abstract – Magnetoelectric (ME) composites with (x)$NiFe_2O_4$ + (1-x)$BaZr_{0.2}T_{0.8}O_3$ (x=0.1, 0.2 and 0.3) have been sunthesized by solid state reaction method to study the structural and magnetic properties of the composites. The presence of two phases in the particulate composites was confirmed by X-ray diffraction measurements; ferrite phase retains cubic spinel structure and ferroelectric phase retains tetragonal perovskite structure. The crystallite size of the composites were calculated using Debye-Scherer formula; crystallite size found to be increases. The lattice constant of the ferrites and ferroelctrics increases with increase in mole percentage of ferrite phase attributed to the difference in ionic radii of the elements present in the composites. The porosity of the composites were determined using Hendricks and Adam's method, which is increased with increase in mole% of the ferrite. The magneitc behavior of the composites were studied using saturation magnetization and magnetic moment and these are found to be increases with increase in applied field; the maximum saturation manetization 8.6839emu/gm is obtained.

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1. INTRODUCTION

Composite materials play a critical role in the modern technologies for unusual properties that they can achieve. Due to the exhaustive research that has gone into the development of these materials, it is likely that totally new composites will emerge. Many of our modern technologies require materials with a combination of properties that cannot be met by the conventional metal alloys, ceramics and polymeric materials [1]. ME composites are desirable for the synthesis of materials with unique or improved properties. The samples containing ferromagnetic and ferroelectric phases known as magnetoelectric (ME) composites show ME property, capable of conversion of energies stored in electric and magnetic fields. The ferrite-ferroelectric composites attracted the considerable attention of the researchers due to the observation of giant ME effects and facilitated by the sample response to electric and magnetic forces. ME composites are the smart structured materials that are attracted the attention of many researchers because of their better applications in potential sensors for magnetic field measurements and transducers for ME conversion [2]. Thus, the conversion mechanism of electric energy into magnetic energy and vice versa is responsible for the potential applications of ME composite materials such as memory devices, storage devices, waveguides, transducers, actuators and sensors and electrically controlled piezoelectric devices, etc. The selection of ferrite and ferroelectric phases mainly depends on the magnetostrictive coefficient and piezoelectric coupling constant. The successful incorporation of these two phases in particulate composite are expected to exhibit interesting magnetic properties. Thus, the soft ferrite $NiFe_2O_4$ with low anisotropy and high initial permeability is a promising phase for better magnetic properties. Furthermore, a large magnetic moment caused by the ion rearrangement favours the ME effect that can be obtained by putting an additional amount of magnesium oxide as one of the divalent component into nickel ferrite [3].

2. SYNTHESIS OF ME COMPOSITES

ME composites contains two individual phases, one ferrite and the other ferroelectric. The ferrite phase chosen was $NiFe_2O_4$. It was prepared through solid state reaction method by taking NiO and Fe_2O_3 in stoichiometric proportions. Similarly, the ferroelectric phase i.e. $BaZr_{0.2}Ti_{0.8}O_3$ were prepared through solid state reaction method starting from $BaCO_3$, ZrO_2 and TiO_2 which are mixed in stoichiometric proportion in liquid medium for homogeneous mixture. The constituent phase i.e. $NiFe_2O_4$ was presintered at 800°C and $BaZr_{0.2}Ti_{0.8}O_3$ was presintered at a temperature of 800°C for 8hours. After presintering, the individual phases were ground to fine powders. The ME composites were prepared by thoroughly mixing 10, 20 and 30% of ferrite with 90, 80 and 70% of ferroelectric respectively and were presintered at a temperature of 800°C for 8hours. 2% of polyvinyl alcohol (PVA) is added to the powder which acts as a binding agent and pressed into the form of pellets using hydraulic press; the pelletized samples were

final sintered at 1150°C for 12hours and furnace cooled to room temperature.

3. CHARACTERIZATION DETAILS

X-ray diffraction analysis carried out by Angle Dispersive X-ray Diffractometer (ADXRD) (model: Image plate MAR345, RRCAT Indore) using the energy of 15keV and the wavelength 0.80034Å using synchrotron radiation source in a range of Bragg angle 2θ ($10^0 \leq 2\theta \leq 60^0$). The magnetic properties of the composites studied using vibrating sample magnetometer (VSM) (model: VSM model 735, Lakeshore, NPL, New Delhi).

4. RESULTS AND DISCUSSIONS

4.1 XRD analysis

The structural formation of ME composites with general formula (x) NFO + (1-x) BZT (with x=0.1, 0.2 and 0.3) are confirmed by ADXRD. Thus, the ferrite phase $NiFe_2O_4$ retains a cubic spinel structure with (311) miller indices and ferroelectric phase $BaZr_{0.2}Ti_{0.8}O3$ retains a tetragonal perovskite structure with (101) miller indices. All peaks are indexed and identified using standard ASTM and JCPDS data.

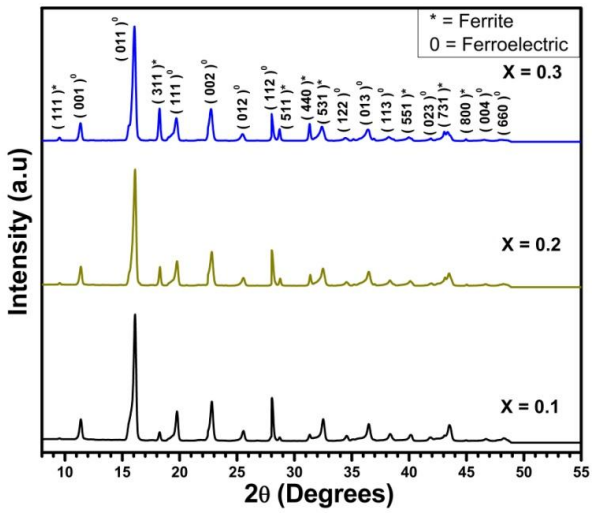

Fig 1. ADXRD plot of (x) NFO + (1-x) BZT composites.

The ADXRD pattern shows well defined peaks with no intermediate phase formation in the composites. (3 1 1) peak is more intense in case of pure ferrite sample and (101) peak is more intense in the case of ferroelectric sample. The ferrite and ferroelectric phases have cubic spinel and tetragonal perovskite structures [4].

These two sets of well defined peaks observed in composites are the characteristics of the constituent phases. There is no unidentified or impurity peaks are appeared in the spectrum which confirms the formation of the final product without any chemical reaction or structural changes occurred between ferrite and ferroelectric phases during the final sintering of the composites [5]. The crystallite size of the composites was calculated using Debye-Scherer formula is listed in table 1. We observed that the crystallite size found to be increases with increase in mole% of ferrites in composites. The maximum crystallite size of 2.48µm observed for (10%)$Ni_{0.9}Mg_{0.1}Fe_2O_3$ + (90%) $BaZr_{0.2}Ti_{0.8}O_3$.

Table.1 Lattice parameters of ferrite, ferroelectric and their composites, porosity and average grain size.

Composition (x)	Lattice parameters of the phases				% Porosity	Crystallite size (µm)	Average grain size (µm)
	Ferrite phase (Å)	Ferroelectric phase (Å)					
	A	A	c	c/a			
0.1	8.261	4.0296	4.0296	1.0000	10.66	2.48	1.95
0.2	8.370	4.0376	4.0378	1.0000	11.48	2.36	1.86
0.3	8.372	4.0373	4.0374	1.0000	12.41	2.29	1.78

We noticed that the lattice constant of ferrite and ferroelectric phases are increases with increase in mole% of ferrite in composites attributed to the difference in ionic radii of the elements present in the composites (table 1). Since the ferrite phase has the lattice constant of a=0.8372nm and ferroelectric phase have a=0.4037nm and c=0.4037nm. The ratio of c/a is found to be 1.0000. The non-variation of the tetragonal ratio (c/a=constant) indicates non-structural variations in composites [6]. By knowing the values of X-ray density and actual density of the samples, the porosity of the composites are estimated using the relation,

$$\%P = \left[\frac{d_x - d_a}{d_x}\right] \times 100 \quad (1)$$

where d_x is the X-ray density, d_a is the actual density of the samples. The estimated values of porosity of the composites are listed in table 1. It is observed that the porosity of the composites increases with increase in mole% of ferrite in composites [1]. The pores material provide insulating path to the electrons resulting in the increase of resistivity with porosity, which intern affects the ME response.

5. MAGNETIC PROPERTIES

The magnetic properties of ME composites are studied by DC hysteresis loop measurements that helps to understand the magnetic behavior of the samples under the influence of external magnetic field. The hysteresis loop of the composites are investigated at room temperature using VSM with an applied magnetic field of -6kOe ≤ H ≤ 6kOe is as shown in figure 6. The magnetic moment of the

composites in Bohr magneton calculated by using the relation,

$$\mu_B = \frac{M_{mole}}{5585} \times \sigma_s' \quad (2)$$

Where M_{mole} is the molecular weight of the sample, σ_s' is the saturation magnetization.

At room temperature, the samples show hysteresis loop type of magnetic behaviour, indicating the presence of an ordered magnetic structure can exist in the mixed cubic spinel– tetragonal perovskite structure system [7]. The saturation magnetization of ME composites increases with increase in mole % of ferrites attributed to the fact that individual ferrite grains acts as centre of magnetization and non-magnetic ferroelectric grains incorporate into the ferrite phase and break the magnetic circuit in samples. As the magnetic contact increases with the ferrite content, the net magnetization increases. Super exchange interactions in nickel ferrite via oxygen ion results in a high anisotropy layer in composites occurs when two magnetic ions separated by nonmagnetic oxygen ions.

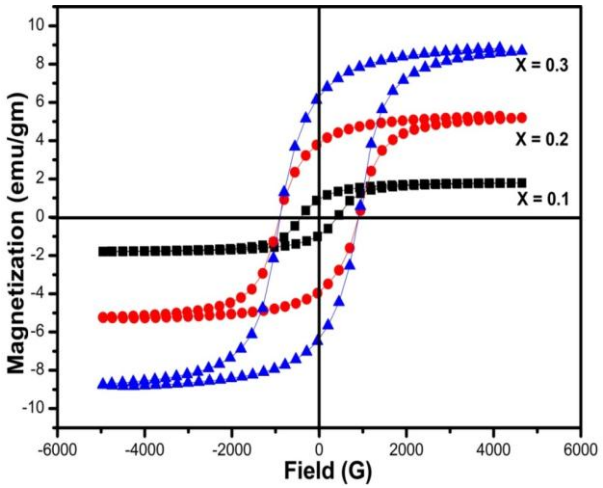

Fig. 6 M-H hysteresis loop of (x) $NiFe_2O_4$ + (1-x) $BaZr_{0.2}Ti_{0.8}O_3$ (x=0.1, 0.2 and 0.3) composites.

The magnetic ions have magnetic interaction mediated by the electrons in their common nonmagnetic neighbours, which is more important than their direct exchange interactions referred to as super exchange interaction [8]. The magnetization increases from 1.7876 emu/gm to 8.6839emu/gm with increase in mole% of ferrite as 10%, 20% and 30%. The maximum saturation magnetization found in the range of 8.684emu/gm for (30%) $NiFe_2O_4$ + (70%) $BaZr_{0.2}Ti_{0.8}O_3$ shows that the increase in magnetic parameters with decrease in ferroelectric concentration in the ME composites [9]. The magnetic moment of ME composites increases with in mole% of ferrite and is obtained in the range from 0.0898μ_B to 0.4205μ_B.

6. CONCLUSION

The ME composites having the formula (x) NFO + (1-x) BZT with different concentration have been synthesized successfully. The cubic structure of ferrite phase and tetragonal perovskite structure of ferroelectric phase are confirmed by XRD. The lattice constant of ferrites and ferroelectrics are determined by using interplanar distances and miller indices; hence, it increases with increase in mole percentage of ferrite. The porosity of the composites determined using Hendricks Adams method and it is found to be increased with increase in ferrite content. The magnetic properties of the composites have been studied succesfully; the saturation magnetization and magnetic moment found to be increases. The maximum saturation magnetization found in the range of 8.684emu/gm for (30%) $NiFe_2O_4$ + (70%) $BaZr_{0.2}Ti_{0.8}O_3$.

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