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**ORIGINAL ARTICLE**

# STUDIES ON STRUCTURAL MAGNETIC AND MAGNETOELECTRIC PROPERTIES OF THE $COFE_2O_4$–$BA_{0.7}SR_{0.3}TIO_3$ COMPOSITES

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## Abstract:

*The magnetoelectric composites containing cobalt ferrite* ($CoFe_2O_4$) *and barium strontium titanate* ($Ba_{0.7}Sr_{0.3}$-$TiO_3$) *phases were investigated. The composites were prepared by standard double sintering ceramic method. The X-ray diffraction analysis was carried out to confirm the phases formed during sintering and also to calculate the lattice parameters. The hysteresis measurements were done to determine saturation magnetization (Ms), remenance magnetization (Mr) and coercivity (Hc) of the samples. The magnetoelectric voltage coefficient (dE/dH)H was studied as a function of intensity of the magnetic field. A large ME voltage coefficient of about 569 µVcm-1Oe-1 was observed for 20%* $CoFe_2O_4$ *+ 80%* $Ba_{0.7}Sr_{0.3}TiO_3$ *composite.*

## KEYWORDS:

Structural, magnetic properties;

## INTRODUCTION:

The development of composite materials has been a subject of intensive research today. The composite material is a mixture of at least two chemically distinct materials. The ME composites consisting of piezoelectric and piezomagnetic materials, the magnetoelectric effect is characterized by the appearance of an electric polarization on applying a magnetic field or a magnetic polarization on applying an electric field [1]. On the other hand the electrical polarization of piezoelectric material causes change in magnetization of piezomagnetic phase due to the mechanical coupling of the piezomagnetic and piezoelectric phases. This effect would make the conversion between electric energy and magnetic energy possible [2]. This provides opportunities for potential applications of these as ME memories, wave-guides, transducers, actuators and sensors. Research into magnetoelectric took off during the latter half of the twentieth century. In 1957, the linear magnetoelectric coupling coefficient was predicted to occur in $Cr_2O_3$ [3-6] and later some single phase crystal families were found to show the ME effect. However, these single-phase materials show such a weak ME effect that they have not yet found any technological applications [7]. An important strategy for engineering enhanced magnetoelectric effects is to introduce indirect coupling, via strain, between two materials such as a ferromagnet and a ferro-electric. Each phase may then be independently optimized for room temperature performance. Strain coupling requires intimate contact between a piezomagnetic (or magnetostric- tive) material and a piezoelectric material. This can be achieved in the form of composites, laminates or epitaxial multilayers. The necessary condition for the ME behaviour by the two-phase material is a strong mechanical coupling between the phases transferring elastic strains and ideally without loss [8-10]. Such coupling may occur between the phases when solid phases are grown by directional solidification from a eutectic melt. The major advantage of this type of in

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situ grown two phase composition is its well defined crystal orientation with respect its mutual growth direction. Magnetoelectric composites may be prepared by sintering powders of piezoelectric barium titanate (PE) and piezomagnetic spinel (PM) particularly if both phases can co-exist up to room temperature from the temperature at which they are sintered. Sintered composite materials are much easier as well as cheaper to prepare than unidirectionally solidified in situ composites [11-12]. As regard to the ME effect it was found that ME composites made by unidi-rectional solidification always gave a higher value than those prepared by solid state sintering of the presintered component phases for a given composition. The first method of preparation is undoubtedly good, nevertheless it is commercially unviable. In situ growth of the ME composites from the respective oxides is equally difficult due to the complicated reaction kinetics, thermodynamic constraints and all sorts of possible random diffusions but can be made commercially viable when suitably processed.

As the ME composites consist of both ferroelectric and ferrite phases and the ferrites are magnetic materials, it is also necessary and important to study the magnetic properties of these materials, when present as a single phase and also when present as magnetostrictive component in the composite. In the present study Cobalt ferrite ($CoFe_2O_4$) is chosen as a ferrite phase because of its low anisotropy and high permeability with high resistivity and low eddy current losses [13] and $BaSrTiO_3$ as a ferroelectric phase because it has been considered to be an important material for tunable microwave devices because of its high dielectric constant, large electric field tenability, relatively low dielectric loss, variable Curie temperature from 400 K to 30 K depending on composition of strontium [14, 15]. In the present paper the magnetic and magnetoelectric properties of (x) $CoFe_2O_4$ + (1 - x) $Ba_{0.7}Sr_{0.3}TiO_3$ composites with x = 0.20–0.60 are reported

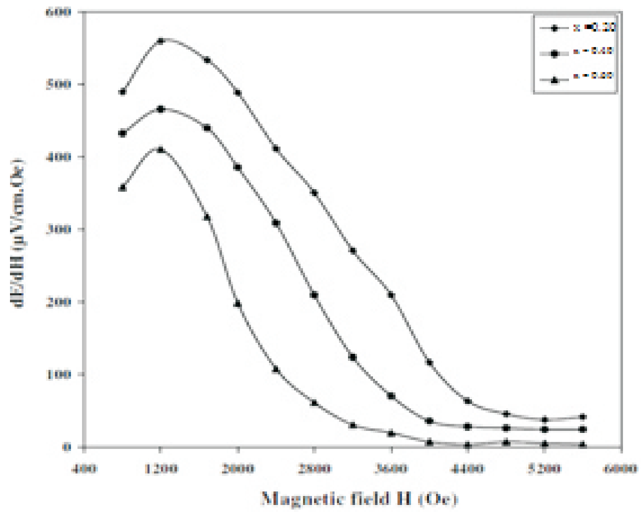
## 2. EXPERIMENTAL DETAILS

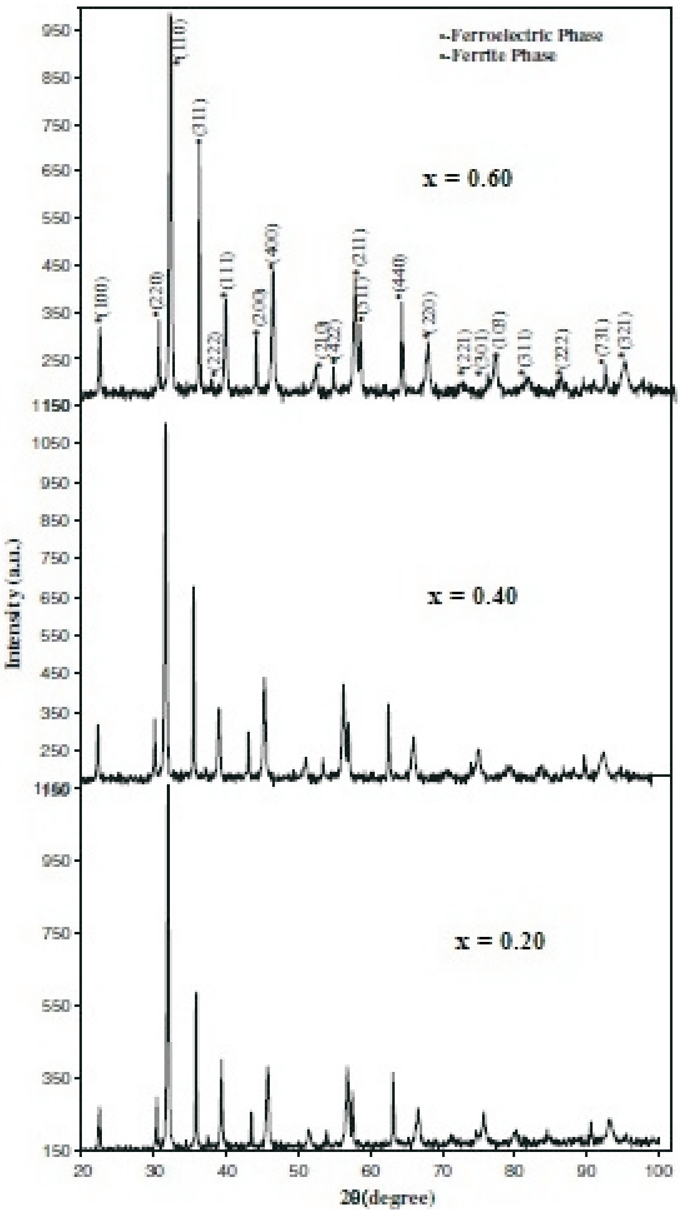
### 2.1 Preparation

The composites containing the two individual phases viz. ferrite ($CoFe_2O_4$) and ferroelectric ($Ba_{0.7}Sr_{0.3}TiO_3$) were prepared by standard double sintering ceramic method. The ferrite phase was prepared through solid-state reaction using AR grade CoO and $Fe_2O_3$ in appropriate molar pro-portions as starting materials. These basic oxides were mixed and ground in agate mortar for couple of hours and presintered at 925°C for 10 h. The ferroelectric phase was prepared following the same route by mixing AR grade $BaCO_3$, $SrCO_3$ and $TiO_2$ in their appropriate molar pro-portions and presintered at 1000°C for 12 h. The ME composites were prepared by mixing 20, 40 and 60 mole% of $CoFe_2O_4$ phase with 80, 60 and 40 mole% of $Ba_{0.7}Sr_{0.3}TiO_3$ phase respectively. The composite mixtures were presintered at 1100°C for 14 h. The pellets having 3–4 mm thickness and 10 mm diameter were prepared using the hydraulic press. The pellets were final sintered at 1000 °C for 10 h to yield the final product.

### 2.2 Characterization

The samples were characterized by using X-ray diffractometer (Rigaku Mini-Flux-II) using CuKa radiations (k = 1.5418 A°). The hysteresis parameters were measured by using high field hysteresis loop tracer interfaced with a computer at room temperature. The magnetoelectric coefficient of the composites was measured as a function of dc magnetic field. The composites had to be poled electrically and magnetically before measuring the ME effect. The electric poling was carried out in a dc field of 2 kV/cm during constant cooling of samples from 130 °C to room temperature. Magnetic poling was done at a constant DC magnetic field (5 kOe) for 15 min by mounting the samples centrally between the poles of a dc electromagnet using a laboratory made sample holder. The stray charges developed during poling were removed by grounding the plates of sample holder. The ME signal was registered by measuring the developed electric potential across the sample as a function of applied increasing DC magnetic field. The output ME voltage generated in the sample was measured using Keithley's electrometer (Model 2000). The static ME voltage coefficient (dE/dH)H is calculated by change in ME output voltage with respect to dc bias


x =0.20
x = 0.40
x = 0.60
dE/dH (µV/cm.Oe)
600
500
400
300
200
100
0
400
1200
2000
2800
3600
4400
5200
6000
Magnetic field H (Oe)


-Ferroelectric Phase
-Ferrite Phase
x = 0.60
x = 0.40
x = 0.20
(100)
(220)
(110)
(311)
(222)
(111)
(200)
(400)
(210)
(422)
(511)
(211)
(440)
(220)
(221)
(301)
(108)
(311)
(222)
(731)
(321)
Intensity (a.u.)
950
850
750
650
550
450
350
250
1150
1050
150
20
30
40
50
60
70
80
90
100
2θ(degree)

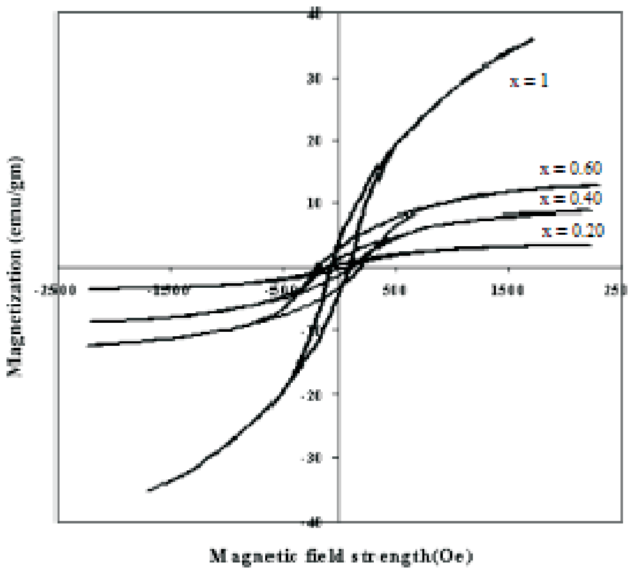
field.

## 3. RESULTS AND DISCUSSION

### 3.1 Structural Properites:

Figure 1 shows the X-ray diffraction patterns of the (x)$CoFe_2O_4$ + (1-x)$Ba_{0.7}Sr_{0.3}TiO_3$ ME composites. It is clearly seen that two phases viz. ferroelectric and ferrite phases can be clearly identified in the composites. No phases other than $Ba_{0.7}Sr_{0.3}TiO_3$ ferroelectric and $CoFe_2O_4$ ferrite are observed. This suggests that no significant chemical reaction has taken place during sintering of the mixed powders, retaining the presence of distinct ferroelectric and ferrite phases. This is very important for the preparation of ferroelectric–ferrite composite materials so that the dielectric and magnetic properties of ferroelectric and ferrite phases do not degrade on sintering.

Fig 2. Hysteresis loop for the (x) $CoFe_2O_4$ + (1-x)$Ba_{0.7}Sr_{0.3}TiO_3$ ME composites



A comparison between the XRD patterns of composites reveals that the intensity of ferrite peaks increase with increasing percentage of ferrite in the composites. The data on lattice constants for all the composites are given in Table 1. The lattice parameters match fairly well with the lattice parameters of the components when present as single phases.

### 2.2 Magnetic properties

Figure 2 shows magnetic hysteresis loops for (x) $CoFe_2O_4$ + (1 - x) $Ba_{0.7}Sr_{0.3}TiO_3$ samples. From the hysteresis loops the values of saturation magnetization (Ms) remenance magnetization (Mr) and coercivity (Hc) were obtained and is given in Table 1. The hysteresis behaviour of composites is due to the ferrite as a magnetic phase and ferroelectric as a nonmagnetic phase. It is seen from the Table 1 that in all composites the values of Ms,  and Hc increase as the content of the ferrite phase increases, as expected. Highest coercivity of 175 Oe (sample annealed at 750 °C) and highest saturation magnetization of 15.99 emu/gm were found in 60% $CoFe_2O_4$-40% $Ba_{0.7}Sr_{0.3}TiO_3$ composite. The values of MS and are found to obey the rule of mixtures [16 17]. However the slight variation is observed in the observed values and calculated values calculated by the rule of mixtures. This may be due to the fact the presence of the non-magneti ferroelectric phase and the interface effects was

Fig 3. ME conversion factor with magnetic field for (x)$CoFe_2O_4$ + (1-x)$Ba_{0.7}Sr_{0.3}TiO_3$ ME composites

expected to influence the magnetic properties in composites by changing the distribution of the magnetic ions and their spin orientation particularly at interfaces and consequently affect the magnetic interactions [18]. The presence of pores among the grains breaks the magnetic circuits and results in a reduction of magnetic properties with increasing pore concentration. In composites, ferroelectric material in the presence of magnetic field acts as a pore causing the reduction of magnetic parameters.

## 3.3 Magnetoelectric properties

Figure 3 shows the typical dependence of the ME conversion on dc magnetic field for the present composites.The magnetic bias dependence of dE/dH is similar for all three composites. As evidenced from the figure, the ME voltage coefficient increases with increasing magnetic bias, saturating at a bias level of *1.3 kOe. This saturation field corresponds to the field at which saturation magnetization is reached in cobalt ferrite. For all composites the highest ME value was obtained for 1.3 kOe DC field indicating that the magnetostrictive phase has reached a saturation value producing constant electric field in the piezoelectric phase, hence making dE/dH to decrease with increasing magnetic field.[19]. This indicates that magnetic saturation occurs at low stimulation and the samples are best suited in responding to relatively weak magnetic field. The behavior of the magnetic field dependence of ME voltage coefficient is similar to that for the magnetostrictive behavior. Although the ferroelectric and ferrite phases have no magnetoelectric effect; their composites have coupled magnetic-electric effect as a result of the elastic interaction between these two phases. The magnetoelectric effect, which can be observed as a product property of the composites has been strongly influenced by the connectivity between particles and the mole ratio of the two phases [21]. The maximum dE/dH also depends non-monotonically on the volume fraction (x) of ferrite, $CoFe_2O_4$, as expected, since the ME effect is a product property between piezoelectricity and magnetostriction. The maximum magnetoelectric voltage coefficient of the composites is observed for low mole ratio of the ferrite i.e., 20% ferrite phase. With increase in ferrite content dE/dH (a) decreases, though the magnetostrictively induced strain of the composites still increases with x. This drop in a with x is due to a higher concentration of the low resistance ferrite phase, which resulting in a low piezoelectric constant which on the other hand provides the conducting path for the charges developed in the ferroelectric phase [21].

**Table 1. Magnetoelectric data of (x) $CoFe_2O_4$ + (1-x) $Ba_{0.7}Sr_{0.3}TiO_3$ Composites.**

| Composition x | Lattice parameters (?) | | | Ms (emu/gm)(cal) | Ms (emu/gm)(The) | $H_C$ (Oe) | $(dE/dH)_H$ (µV/cm.Oe) |
|---|---|---|---|---|---|---|---|
| | Ferrite phase | Ferroelectric phase | c/a | | | | |
| 0.00 | -- | a = 3.97<br>c = 3.98 | 1.002 | -- | --- | -- | -- |
| 0.20 | a = 8.31 | a = 3.97<br>c = 3.99 | 1.005 | 3.51 | 5.21 | 80 | 569 |
| 0.40 | a = 8.31 | a = 3.95<br>c = 3.98 | 1.007 | 9.02 | 10.11 | 98 | 462 |
| 0.60 | a = 8.32 | a = 3.96<br>c = 4.01 | 1.012 | 12.31 | 15.99 | 175 | 419 |
| 1.00 | a = 8.33 | -- | -- | 35.52 | 35.41 | 33 | -- |

## 4 CONCLUSION

The ferrite ferroelectric composite materials are prepared by standard double sintering ceramic method. The phase identification was done by using XRD technique. No phases other than $Ba_{0.7}Sr_{0.3}TiO_3$ ferroelectric and $CoFe_2O_4$ ferrite were observed. The intensity of ferrite peaks increase with increasing percentage of ferrite in the composites. These composites are found to exhibit excellent magnetic and large ME voltage coefficient for x=0.20. The magnetic parameters such as saturation magnetization (Ms), remenance magnetization (Mr) and coercivity (Hc) were measured. The values of Ms and Mr are found to increase with increases in ferrite content in the composites. The presence of ferroelectric phase affects the values of Ms and Mr. The ME effect is found to decrease with increase in both magnetic field and ferrite content. In particular, the simple composite ceramics exhibit a very large ME response linearly varying with dc magnetic field, which makes these ceramics attractive for technological applications such as sensors, dynamic random access memories (DRAM), microwave phase shifters and active passive filters.

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