CHAPTER - V

Content

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Results and Discussion on magnetic and magnetoelectric properties of (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (X) $BaTiO_3$ Composite

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5.1 Introduction

The magnetoelectric effect is characterized by a variation of the electrical polarizations in response to an external magnetic field H or an induced magnetization by an external electrical field E. Hence ME effects originates from a product coupling between the magnetostrictive effect from the magnetostrictive phase and piezoelectric effect from the piezoelectric phase in the composite. It has been observed that the presence of ferroelectric phase in the composition affects the magnetic properties of ferrite to a large extent. Magnetic properties of ferrite can be discussed on the basis of magnetic parameters such as saturation magnetization, coercivity, susceptibility etc [1, 2]. As the ME composite consists of both the ferroelectric and ferrite phases as the ferrites are magnetic materials, it is necessary to study the magnetic properties of these ferrite materials, when present as a single phase and also when present as magnetostrictive component in the composites.

The ME effect is defined as an induced dielectric polarization of a material in an applied magnetic field or an induced magnetization in an

external electric field. The composite, which exhibit ME effect are known as the “Magneto-electric composites”. Composite material containing piezoelectric (ferroelectric) and piezomagnetic (ferrite) phases exhibit ME effect. It is due to the strain induced in the piezomangetic (ferrite) phase by the applied magnetic field, being mechanically coupled stress induced in the piezoelectric/ ferroelectric phase, the coupling resulting in an electric voltage [3,4].

The ME composite has been found a lot of technical applications in radio electronics, optoelectronics, microwave electronics and transducers in instrumentation. The selection of suitable combination of piezoelectric and piezomangetic material with a view to achieve ME effect itself is however a challenging task. In order to achieve better ME effect, the piezomangetic coefficient of ferrite phase and the piezoelectric coefficient of ferroelectric phase must be high [5].

Efforts are in progress to develop the magnetoelectric composite even that the theoretical understanding of the material is very limited. The theoretical treatment is very necessary in order to design new magnetoelectric composites for applications.

5.2 Experimental Procedure

a) Magnetization

Magnetic properties of ME composites were investigated using a pulse field hysteresis loop technique. The details of techniques are discussed in chapter III. The values of saturation magnetization (Ms), remenance magnetization (Mr) and coercive field (Hc) are obtained from hysteresis curve (M-H Plots). Curie temperature of ME composite materials was obtained using A.C. susceptibility and Loria technique.

b) Measurement of ME effect

The composite has to be poled electrically and magnetically before measuring the ME effect. The samples are heated at 151^0C which is approximately 30 ^{0}C above the ferroelectric Curie temperature (Tc) and their after it is cooled to room temperature in the presence of electric field of strength 3Kv/cm. The composite was magnetically poled by an external dc magnetic field of 5KOe at room temperature.

This poling was carried out in the step up in which magneto-electric conversion factor was measured by varying d.c. magnetic field.

5.3 Results and Discussion

5.3.1 Magnetic studies

a) Magnetic Hysteresis

The magnetic properties of (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$ composites materials were investigated using pulse field hysteresis loop technique. The hysteresis loop for all the samples of composite material under investigations are shown in Fig. 5.1-5.4. It is seen that the saturation magnetization and remanant magnetization of the composites increases and the saturation shifts towards high field with increase in volume fraction of ferrite phase [6]. At high magnetic fields, the magnetostriction gets saturated producing a nearly constant electric field in ferroelectric phase, thereby causing a decrease in polarization with increase in magnetic field.

The hysteresis loop for x= 0.00, 0.25, 0.50 and 0.75 at room temperature for x = 0.00 the saturation magnetization is 70.67 emu/gm and for x=0.25, 0.50 and 0.75 the values are 54.95 emu/gm, 35.59 emu/gm and 19.90emu/gm respectively. This indicates that the magnetic moment decreased with increasing ferroelectric content in the composite. It is clear from Fig 5.1-5.4 that all the sample exhibit typical magnetic hysteresis of the magnetic materials, indicating that the composites are magnetically ordered.

The hysteresis loops are used to obtain the values of saturation magnetization (Ms), remanent magnetization (Mr), and coercive field (Hc). The values of saturation magnetization (Ms), remanent magnetization (Mr) and coercive field (Hc) and remanance ratio (Mr/Ms) are given in Table 5.1. The saturation magnetization (Ms) decreases as ferroelectric content increases. The variation of the saturation magnetization (Ms) of the composites with compositions 'x' is plotted in Fig. 5.5. It is clear from Fig 5.5 that the saturation magnetization of the composites decreases linearly with increasing ferroelectric $BaTiO_3$ content. This is because the ferroelectric phase incorporated with the ferrite phase acts as pores in the presence of applied magnetic field which breaks the magnetic circuits resulting in the decrease of saturation magnetization with increasing ferroelectric content.

The decrease in saturation magnetization is due to non magnetic nature of $BaTiO_3$. Similar behaviour of the saturation magnetization (Ms) with composition 'x' is reported in the literature [7, 8]. The remanent magnetization (Mr) values obtained from M-H plots shows decreasing nature with increase in ferroelectric content. For x = 1.0 (pure ferroelectric), the M-H plot is not seen. The decrease in remanent magnetization (Mr) is due to increasing $BaTiO_3$ content. It is evident from Table 5.1 that the coerecivity of the composites decreases with

$BaTiO_3$ addition. The decrease in magnetic behaviour (as observed from the values of Ms, Mr and Hc) of composite under investigation is due to the reduction in the ferrite content of the composite.

b) Curie Temperature

The temperature dependence A.C. susceptibility plots for all the samples of the composite (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$ are shown in Fig 5.6-5.9. These susceptibility plots exhibits normal ferrimagnetic behavior which is only due to the ferrite content in the composite. The addition of ferroelectric $BaTiO_3$ compound leads to decrease in ferrimagnetic behavior of the composite samples, as it can be observed from the magnetization plots. As temperature increases the normalized susceptibility (χ_T/χ_{RT}) also increases slowly to a certain value and then start decreasing and finally reaches towards zero. The decrease in susceptibility behaviour is sharp [9, 10].

The temperature at which susceptibility reaches sharply towards zero is known as Curie temperature of the samples. At this temperature sample undergoes transition from ferrimagnetic to paramagnetic. The values of Curie temperature obtained from magnetization versus temperature plots are presented in Table 5.2**.** It is evident from the Table 5.2 that Curie temperature decreases with the addition of $BaTiO_3$ content 'x' (or due to decrease in ferrite content). The decrease in Curie temperature with

composition is slow. Curie temperature was also determined for all the samples by Loria technique and the values of Curie temperature are presented in Table 5.2. Curie temperature obtained by Loria technique is in good agreement with that obtained by susceptibility plots. The value of Curie temperature for pure cobalt ferrite phase (x = 0.00) is closely agrees with the literature value [11, 12].

5.3.2 Magnetoelectric (ME) effect

Fig. 5.10 shows the magnetoelectric effect voltage coefficient as a function of d. c. magnetic bias for all composites under investigation. The magnetoelectric (ME) effect is measured by changes in the resulting electric field in the magnetoelectric (ME) composites due to the applied external magnetic field [13, 14]. It is observed that magnetoelectric (ME) coefficient initially increases to a certain applied magnetic field (0.5KOe) and then start decreasing as applied field increases. The magnetoelectric (ME) effect is found to be maximum for x=0.75 (i.e. for 25% $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + 75% $BaTiO_3$). Similar behaviour of magnetoelectric (ME) coefficient was also observed in case of $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ + $BaTiO_3$ composite and other well known ferrite +ferroelectric composites [15, 16].

The magnetoelectric (ME) effect in composites is due to the strain induced in ferrite phase by the applied magnetic field which being

mechanically coupled to induce a stress in surrounding ferroelectric phase. This stress results the polarization of ferroelectric phase due to piezoelectric effect. Therefore, the magnetoelectric effect (ME) is not a property of either ferrite or ferroelectric but it is a property of ferrite + ferroelectric composite. This can be indirectly proved from the fact that no magnetoelectric (ME) effect was observed for x = 0.0 (ferrite phase) and x =1.0 (ferroelectric phase).The value of magnetoelectric (ME) effect are given in Table 5.3. It is evident from Table 5.3 that magnetoelectric (ME) effect increases as ferroelectric phase increases [17, 18]. The Table 5.3 also gives the resistivity values for the sake of comparison. The increase in magnetoelectric (ME) effect is related with the increase in resistivity of the composite. The number of ferrite grains increase with increase content of ferrite. The decrease in the resistivity leads to leakage of charges build up in ferroelectric phase through the surrounding ferrite grain [19, 20, 21].

Fig 5.11 shows the variation of electrical resistivity and ME coefficient with the ferroelectric content. As the ferroelectric content increases both the resistivity and ME coefficient of composites increase. This is due to the high resistivity of ferroelectric phase [22, 23].

5.4 Conclusions

I) The saturation magnetization, remanent magnetization, coercivity, Curie temperature of the composites decreases with decrease in ferrite content of the composites.

II) The Curie temperature obtained in the present system decreases with increase in ferroelectric contents of the composite material.

III) Magneto-electric conversion factor with varying magnetic field shows maxima in the curve at a lower magnetic field and then decreases continuously at higher magnetic field.

IV) As ferroelectric content increases both the resistivity and magnetoelectric conversion factor also increases.

V) The composite under investigation in the present study exhibits better values of magnetoelectric conversion factor.

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Table - 5.1

Saturation magnetization, Remnant magnetization, Coercive field and Remanence ratio for (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$ (x = 0.00 – 0.10) composite.

Comp. 'X'	**Saturation magnetization Ms (emu/gm)**	**Remnant magnetization Mr (emu/gm)**	**Coercive field Hc (Oe)**	**Remnance R= (Mr/Ms)**
0.00	70.67	31.42	508.77	0.44
0.25	54.94	16.70	232.61	0.30
0.50	35.59	14.70	506.00	0.41
0.75	19.90	08.27	393.99	0.41

Table - 5.2

Curie temperature obtained by Susceptibility and Loria method for (x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (1-x) $BaTiO_3$ (x = 0.25 – 1.00).

Comp. 'X'	**Curie Temperature (T_c) K**	
	Susceptibility	**Loria method**
0.00	670	655
0.25	650	634
0.50	628	611
0.75	605	587

Table- 5.3

Magnetoelectric conversion factor and Resistivity for (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$ (x = 0.25 - 0.75) composite.

Comp. 'x'	**Magnetoelectric conversion factor $[dE/dH]_H(\mu V/cm/O_e)$**	**Resistivity $\rho_{RT}(\Omega$ cm)**
75%$Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + 25%$BaTiO_3$ (x=0.25)	254	$3.30x10^8$
50%$Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + 50%$BaTiO_3$ (x=0.50)	351	$4.55x10^8$
25%$Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + 75%$BaTiO_3$ (x=0.75)	647	$6.25x10^8$

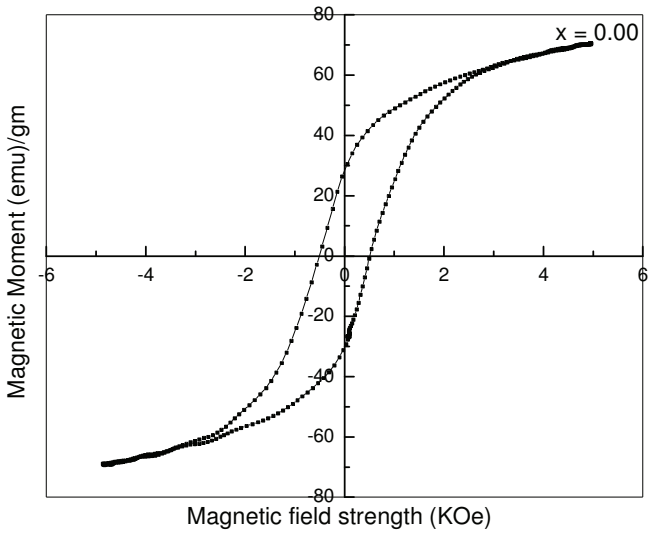

Fig. 5.1 Hysteresis loop for (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$. (x = 0.00)

Composite.

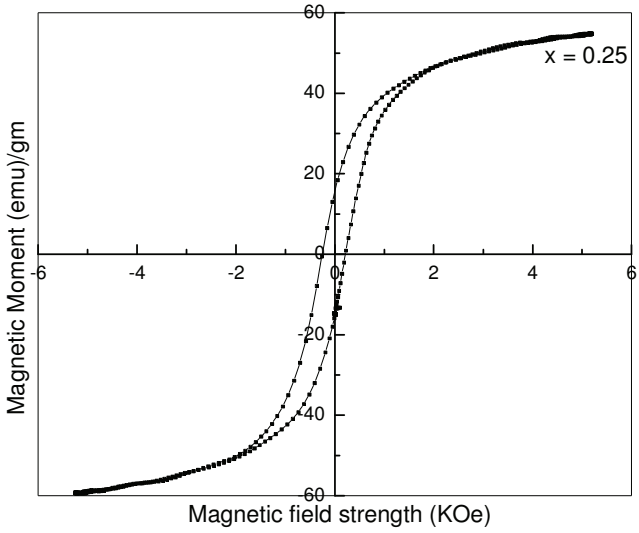

Fig. 5.2 Hysteresis loop for (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$. (x = 0.25)

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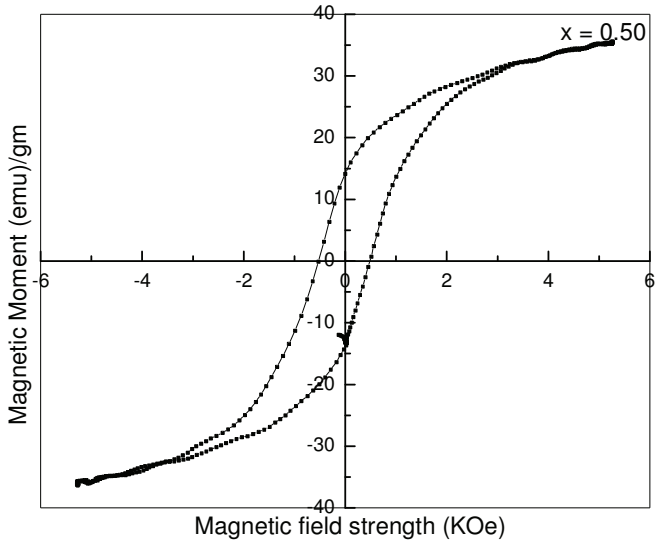

Fig. 5.3 Hysteresis loop for (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$. (x = 0.50)

Composite.

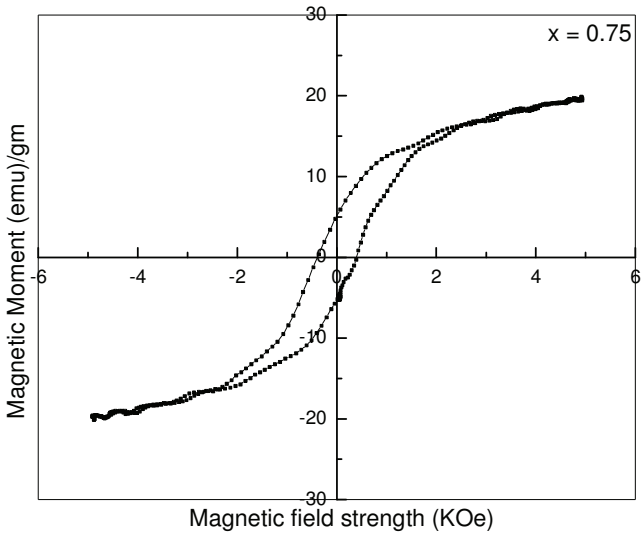

Fig. 5.4 Hysteresis loop for (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$ (x = 0.75) Composite.

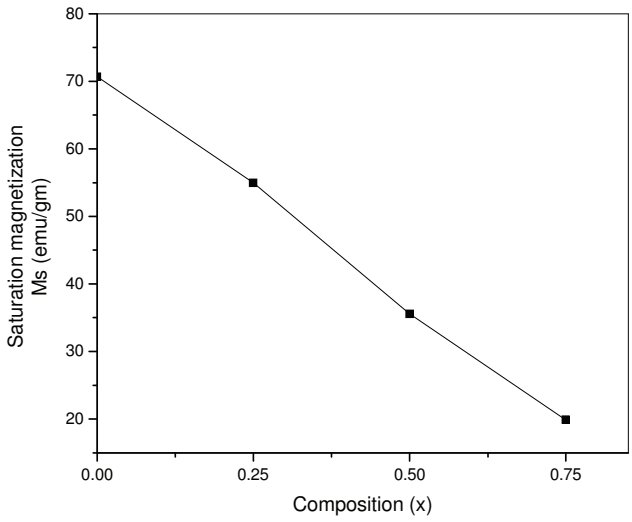

Fig 5.5 Variation of saturation magnetization with composition for (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$.

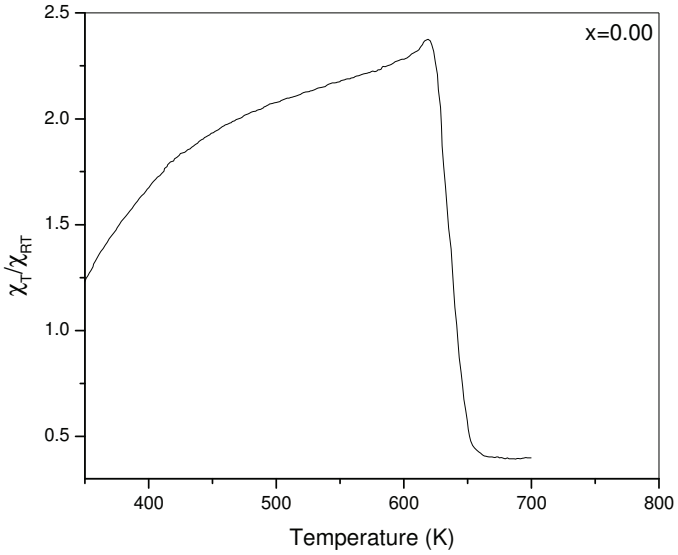

Fig 5.6 Variation of χ_T/χ_{RT} versus temperature of system (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$. (x = 0.0) composite.

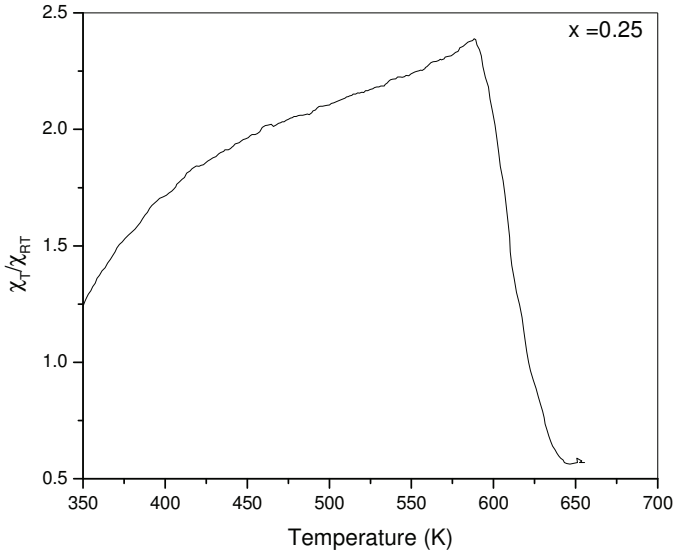

Fig 5.7 Variation of χ_T/χ_{RT} versus temperature of system (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$. (x = 0.25) composite.

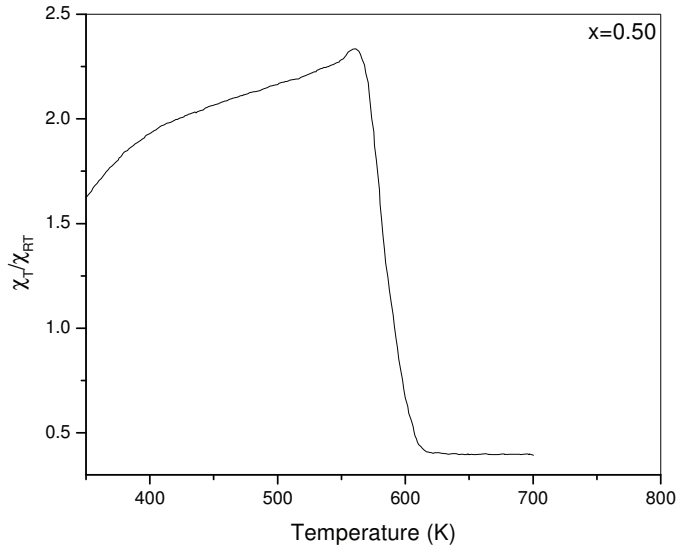

Fig 5.8 Variation of χ_T/χ_{RT} versus temperature of system (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$. (x = 0.50) composite.

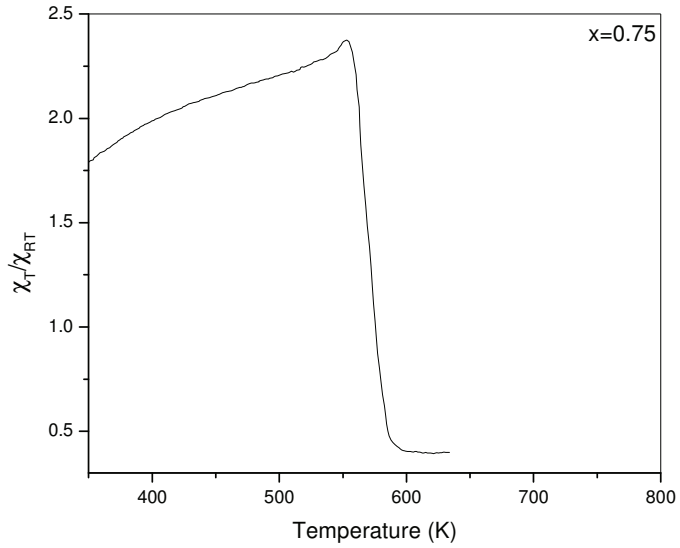

Fig 5.9 Variation of χ_T/χ_{RT} versus temperature of system (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$ (x = 0.75) composite.

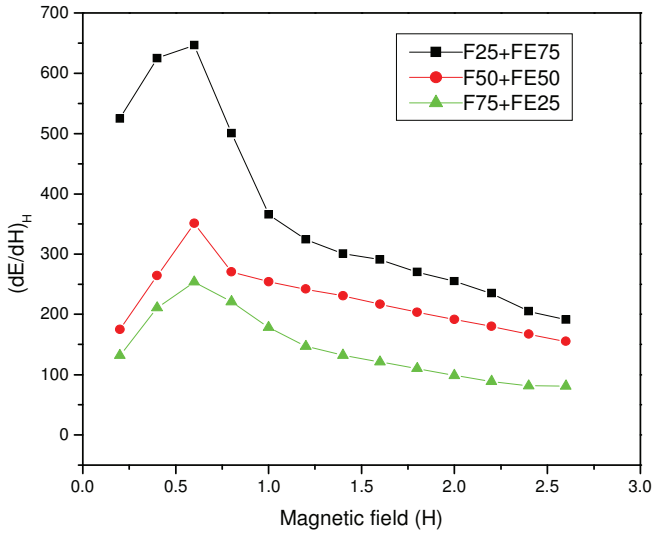

Fig 5.10 Variation of magneto-electric conversion factor with magnetic field for (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$.

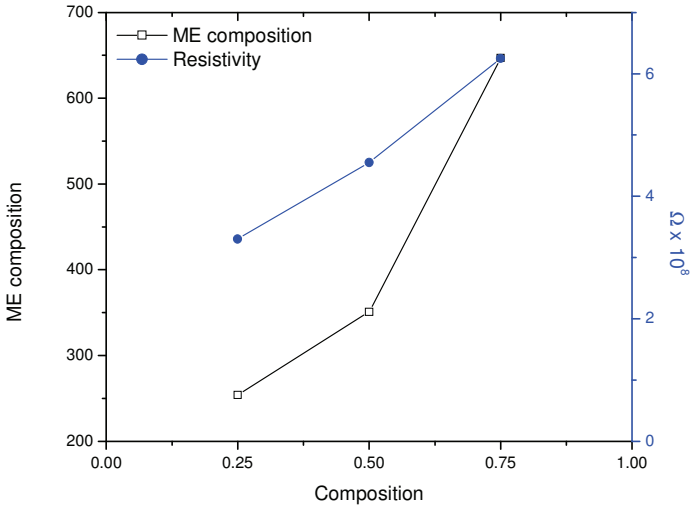

Fig 5.11 Variation of resistivity and Magneto electric effect with composition for (1-x) $Co_{1.2}Mn_{0.2}Fe_{1.6}O_4$ + (x) $BaTiO_3$.