

## **CHAPTER VI**

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Summary and conclusions

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### SUMMARY AND CONCLUSIONS

The magnetoelectric effect is a coupled two field effect in which the application of an electric field induces magnetization and a magnetic field induces electric polarization [1]. However, recently the interest has been shifted to use the composites for the electrical applications [2] such as actuators, sensors, modulators, switches, rectifiers etc. The development of composite material has been subject of intensive interest since last 45 years.

Presently two types of composites, bulk and layered have been attracted the attention of number of scientist. Bulk composites have the advantage of superior mechanical strength over the layered sample [3]. One could also control physical, magnetic, electrical and ME parameters with proper choice for the two phases and their volume fractions.

The thesis is presented in six chapters.

Chapter I deals with three sections. Section A consists of the introduction of ME composites, involves the brief introduction of the ME composites along with the basic conditions of the Boomgaard for getting good ME effect. The brief survey of the work carried out on the ME composites is also listed in this section.

Section B involves the introduction to ferrites, their classification based on chemical composition and crystal structure, information about spinel ferrites and their classification and applications. The present work involves (y)  $\text{Co}_{1-x}\text{Cd}_x\text{Fe}_2\text{O}_4$  + (1-y) PZT ME composites where  $y=0.15, 0.30, 0.45$  and  $x = 0.1, 0.3$ . Therefore the literature survey of the Co-Cd ferrite is also included in this chapter. The ferroelectrics are described in the section c which also contains the orientation of the problem.

The discussion of the synthesis, structural analysis by x ray diffraction and microstructural characterization of the (y)  $\text{Co}_{1-x}\text{Cd}_x\text{Fe}_2\text{O}_4$  + (1-y) PZT is given in

Chapter II. It is divided into two parts. Section A contains the synthesis of ME composites for which the stepwise description of the ceramic method is given. Formation of the composites as well as the constituent phases takes place by solid state reaction. So that the solid state reaction and flow chart of synthesis of the ME composites are listed in this chapter. For preparation of ferrite phase AR grade  $\text{CoCO}_3$ ,  $\text{CdCO}_3$ ,  $\text{Fe}_2\text{O}_3$  were taken in molar proportions as starting materials. These were mixed and ground for 2-3 h. with the help of agate and mortar, presintered at  $900^\circ\text{C}$  for 10 h. To synthesize the ferroelectric phase (PZT), AR grade  $\text{PbO}$ ,  $\text{ZrO}_2$  and  $\text{TiO}_2$  were used as starting materials. The composites were prepared by thoroughly mixing 45, 70 and 55 mole% ferroelectric phase with 15, 30 and 55 mole% of ferrite phase. Pelletized samples of 2-3 mm thickness and 15mm diameter were sintered at  $1000^\circ\text{C}$  for 10h.

Section B is devoted to structural analysis by X-ray diffraction. The XRD patterns of the samples were taken on Philips X-ray diffractometer (Model PW 1710) using  $\text{CuK}\alpha$  radiation ( $\lambda = 1.5418\text{\AA}$ ). They reveal that the ferrite phase has cubic spinel and ferroelectric phase has tetragonal perovskite structure. The XRD patterns of composites show the presence of both the ferrite and ferroelectric phases in it. The calculated and observed 'd' values are in good agreement with each other. The intensity and number of the ferrite peaks were observed to increase with increase in ferrite content. It is reported [4] that the ionic radius of  $\text{Co}^{2+}$  ions is 0.82 which is smaller than that of  $\text{Cd}^{2+}$  ions so that the lattice parameter must be increase with increase in Cd content. The lattice parameters of  $\text{Co}_{0.9}\text{d}_{0.1}\text{FeO}_4$  and  $\text{Co}_{0.7}\text{Cd}_{0.3}\text{Fe}_2\text{O}_4$  are  $a = 8.37\text{\AA}$  and  $8.40\text{\AA}$  respectively are found to increased with Cd content.

The surface morphological studies by SEM were listed into Section C of this chapter. The study was carried out with the help of SEM (Model JEOL-JSM 6360). All samples were scanned at a magnification 10000. The average grain size was calculated by Cottrel's line intercept method. From the micrographs of ferrites and ferroelectric it is clearly seen that the grain size of the  $\text{Co}_{0.7}\text{Cd}_{0.3}\text{Fe}_2\text{O}_4$  is (1.81

$\mu\text{m}$ ) is greater than that of the ferroelectric phase ( $0.52\mu\text{m}$ ). From the micrographs of the composites it is seen that there is uniform distribution of the ferrite and ferroelectric phases through out it which may help to enhance the ME effect. The grain size is maximum for composite with  $y = 0.30$  while the grain size of the ferrite phase is maximum than that of the ferroelectric phase. The increase in the Cd content caused to increase the grain size in respective composites.

Chapter III deals with the sum properties such as dielectric constant, ac conductivity, dc resistivity of the composites as well as their constituent phases, ferrite and ferroelectric. This chapter is divided into two sections.

Section A consists of the study of dc resistivity. The plots of variation of the resistivity with temperature show two regions of conductivity. The first region observed at lower temperature region is attributed to the ordered state of ferroelectric phase and second region that occurs at higher temperatures is attributed to disordered paraelectric state. The resistivity of all the samples is decreased with the increase in temperature shows the semiconducting behavior of them, which is due to thermally activated mobility of charge carries according to the hopping conduction mechanism. The resistivity of the ferrite  $\text{Co}_{0.7}\text{Cd}_{0.3}\text{Fe}_2\text{O}_4$  is greater than that for  $\text{Co}_{0.9}\text{Cd}_{0.1}\text{Fe}_2\text{O}_4$ . By calculating the activation energy for both the ferro and para regions, it is observed that the activation energy of the para region is greater than that for the ferro region. In  $(y) \text{Co}_{0.9}\text{Cd}_{0.1}\text{Fe}_2\text{O}_4 + (\text{PZT})$  ME composites the resistivity fir the composite with  $y = 0.30$  is greater than that for  $y = 0.15$  and  $y = 0.45$ . Out of all the composites from the series  $(y) \text{Co}_{0.7}\text{Cd}_{0.3}\text{Fe}_2\text{O}_4 + (1-y) \text{PZT}$  ME composites the composite with  $y = 0.15$  has greater resistivity than that for the composites with  $y = 0.30$  and  $y = 0.45$ .

Section B is focused on the dielectric properties. The dielectric measurements of the pelletized samples with frequency at room temperature were carried out. It is also seen that the decrease is relatively sharp at lower frequencies than that at higher frequency regions all composites the dielectric constant shows the dispersion in certain lower frequency range. The dielectric constant is 242 for

(y)  $\text{Co}_{0.9}\text{Cd}_{0.1}\text{Fe}_2\text{O}_4$  and 520 for (y)  $\text{Co}_{0.7}\text{Cd}_{0.3}\text{Fe}_2\text{O}_4$ . This may be due to two possible reasons viz. space charge polarization and the migration of ferric ions. In (y)  $\text{Co}_{0.9}\text{Cd}_{0.1}\text{Fe}_2\text{O}_4$  + (PZT) ME composites the dielectric constant for the composite with  $y = 0.30$  is greater (2106) than that for  $y = 0.15$  (427) and  $y = 0.45$  (556). The variation of dielectric loss with frequency also shows the same behavior to that of the dielectric constant with frequency.

The ac conductivity at room temperature was calculated in the frequency range 20Hz-1MHz. It reveals the conduction mechanism. The conductivity increases linearly with increase in frequency and plots confirms that the conduction mechanism is due to the small polaron.

Chapter IV includes the study of magnetic properties of the ferrites and ME composites in which brief introduction of the magnetic materials and their properties such as magnetic moment; magnetization is given which is followed by the short information about domains, domain boundaries, magnetic anisotropy, magnetostriction and magnetic hysteresis.

For the measurement of magnetic properties, the high field hysteresis loop tracer designed by Tata Institute of Fundamental research, Bombay was used.

From the hysteresis plots for the ferrites with  $x = 0.1$ , and  $0.3$  resultant magnetization is found to increase with Cd content while due to the behavior of the ferroelectric material as a non magnetic material, the saturation magnetization of the composites is less than that for the ferrite phase and goes on increase with increase in ferrite content in the composites. The  $M_s$  values for (y)  $\text{Co}_{0.9}\text{Cd}_{0.1}\text{Fe}_2\text{O}_4$  + (1-y) PZT ME composites with  $y = 0.15, 0.30, 0.45$  are 6.52 emu/gm, 12.55 emu/gm and 18.18 emu/gm respectively. They show the variation of saturation magnetization with Cd content. The saturation magnetization for  $\text{Co}_{0.9}\text{Cd}_{0.1}\text{Fe}_2\text{O}_4$  is 52.04 mu/gm and that for  $\text{Co}_{0.7}\text{Cd}_{0.3}\text{Fe}_2\text{O}_4$  is 58.20 emu/gm. This is due to the fact that,  $\text{Cd}^{2+}$  ion is a non magnetic ion and has tendency to occupy tetrahedral site. The substitution of  $\text{Co}^{2+}$  by  $\text{Cd}^{2+}$  causes the migration of  $\text{Fe}^{3+}$  ions to octahedral sites (B sites). When the concentration of magnetic ions

such as  $\text{Fe}^{3+}$  with higher spin magnetic moment than  $\text{Co}^{2+}$  by  $\text{Cd}^{2+}$  which leads an increase in the resultant magnetization [5]. The value of the remanent magnetization for the present samples suggests that most of the magnetization vectors have turned in the magnetically preferred direction making a small angle with the direction of applied field. This helps in increasing the magnetization [6].

The study of magnetoelectric effect as a product property of the composites is described in Chapter V. The ME coefficient of the composite was measured as a function of dc magnetic field. The composites have to be poled electrically and magnetically before the ME measurement. The electric poling was carried out at room temperature in the dc electric field 2 kV/cm. The samples were magnetically poled at the dc magnetic field of  $\sim 5\text{kOe}$  at room temperature. The measurement of the static ME voltage coefficient  $(dE/dH)_H$  was measured using Keithley electrometer (Model 4514/E) in dc magnetic field. For  $(y) \text{Co}_{0.9}\text{Cd}_{0.1}\text{Fe}_2\text{O}_4 + (1-y) \text{PZT}$  ME composites the ME voltage initially increases, reaches a maximum and again decreases with applied magnetic field. It is attributed to the saturation of magnetic phase so that constant strain production gives the constant electric field. From the plots it is clearly observed that the ME effect is strongly influenced by the connectivity, resistivity, and microstructure of the composites [7, 8, 9]. The maximum ME voltage coefficient was observed for the composite with  $y = 0.30$  due to its high resistivity.

Summary and conclusion of the present work is presented in the sixth chapter.

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