CHAPTER VI

CHAPTER - VI

SUMMARY AND CONCLUSIONS

Ferrite-ferroelectric composites have recently attracted considerable attention owing to their wide range of practical applications. They are useful as sensors, phase inverters, optical modulators, ferroelectromagnetic wave generators, optical wave guides, etc. One of the most important scientific applications of these composites is that they give us precious information for determining the magnetic point groups and space groups and also the determination of magnetic and electric field induced phase transitions [1].

The above applications of magnetoelectric composites are due to the phenomenon of magnetoelectric effect (ME effect). The ME effect is magnetic field induced electrical polarization. This effect is not observed in the individual phases [2]. The recent interest and development of ferrite-ferroelectric composites results from the recognition of the potential for enhanced performance and the increasing need for a combination of desired material properties that often cannot be obtained in single phase materials. These composites having high values of magnetoelectric conversion factor are of practical interest because of their use in electronic devices[3].

The studies of ME composites in which (Ni, Co)Fe_2O_4 is ferrite phase and $BaTiO_3$ as ferroelectic phase have been carried out by many workers [4-9]. This composite is most widely used because of its high value of magnetoelectric conversion factor.

Although there has been an emphasis on the practical development of such composite materials, work related to electrical conduction is relatively scant. Hence it was decided to carry out studies on electrical conduction in $x(Ni_{0.75}Co_{0.25}Fe_2O_4)$ + $(1-x)(Ba_{0.8}Pb_{0.2}TiO_3)$ composites with x = 1, 0.85, 0.70, 0.55 and 0. The work involves preparation, characterization and measurement of electrical properties of these composites. The work is presented in six chapters.

Chapter - I is introductory. It includes definition and classification of composites, need and advantages and magnetoelectric composites. It also includes a review of literature, applications and orientation of the problem. The references are given at the end of the chapter.

Chapter - II deals with constituent phases namely ferrite and ferroelectric. It has been divided into two sections. Section A includes ferrite phase in which introduction, historical developments and structure of spinel ferrite is given. It also includes theory of ferrimagnetism and applications of ferrites.

Section B includes ferroelectric phase in which introduction, definition and classification etc. are given. The crystal structure of $BaTiO_3$ is explained. It includes dielectric properties, theory and application of ferroelectrics.

Chapter III deals with the preparation and characterization of ferrite - ferroelectric composites by XRD technique. It has been subdivided into two sections. Section A includes the methods of preparation and actual preparation of the samples. The ferrite phase was prepared by standard ceramic method using AR grade $NiCo_3$, $CoCo_3$ and Fe_2O_3 powders. The ferroelectric phase was prepared using AR grade BaO, PbO and TiO_2 powders. The ferrite was presintered at 700^0C and ferroelectric at 900^0C for 12 hrs. After presintering, the individual phases were grounded in an agate mortar to fine powder. The ME composites were prepared by thoroughly mixing 85, 70 and 55 mole % of ferroelectric with 15, 30 and 45 mole % of ferrite respectively. The composite mixture presintered at 800 ^{0}C again. The composite were ground to a fine powder and pressed into pellets (1cm diameter and 2-3 mm thickness). The compacts so prepared were finally sintered at 1100 ^{0}C for 24 hrs. The furnace was cooled at the rate of 80 ^{0}C per hour.

Section B deals with the XRD studies. The diffractogram were recorded on Phillips X-ray Diffractometer (Model PW 1710) using CuK_α

(λ = 1.5418 Å) radiation. Indexing of the patterns was done by comparing the present data with the standard ASTM data. The lattice parameters and interplanar distances were calculated using the standard relations. The lattice parameters of the ferroelectric phase in composites match fairly well with the reported values [10,11]. Also, the lattice parameter of ferrite phase in composites match fairly well with the reported values [9]. The calculated and observed d values are in good agreement with each other. As reported earlier, this composite result with cubic spinel structure for ferrite phase and tetragonal perovskite structure for ferroelectric phase [9,12]. Similar is the case with the present composites.

The lattice parameters and the c/a ratios are found to vary slightly with change in mole % of either phase. Similar observation have been made earlier [13]. The porosity of the samples lies in the range of 10 - 15 %.

Chapter IV deals with the sum properties of the composites. It is divided into three parts. The first part deals with the measurement of the dielectric properties and ac conductivity, second part gives the measurement of dc resistivity and third part the TEP measurements.

AC conductivity measurements were carried out in the frequency range 100 Hz to 1 MHz on LCR meter bridge HP 4284 A. The variation in the dielectric constant with frequency reveals that dispersion is due to Maxwell-Wagner type interfacial polarization [14,15]. Dispersion is large in compositions with large values of ε' in comparision to those with smaller values of ε'. The changes in dielectric constant are more at lower frequencies.

The plots of tan δ versus frequency show that for all the samples it decreases continuously with frequency. All the samples show dispersion in tan δ at lower frequencies. The variation of AC conductivity with frequency shows that it increases with increase in frequency. The results are similar to those reported by other workers [16].

The variation of dielectric constant with temperature for all the samples show that initially it increases with increase in temperature, reaches a maximum value at the critical temperature T_C and thereafter decreases. The transition temperature does not show any frequency dependence but the dielectric constant exhibits decrease in values for higher frequencies. The dielectric constant for ferrite is less as compared to the ferroelectric. The T_C for the present ferrite and ferroelectric is 540 ^{0}C and 190 ^{0}C respectively. In the three composites, the dielectric maxima are observed at temperatures 490 ^{0}C, 495 ^{0}C, and 515 ^{0}C respectively for x = 0.85, 0.70 and 0.55. The transition temperature are close to ferrite T_C. This observation is similar to that reported earlier for the other composites [13].

The dc resistivity was measured using two probe method, from room temperature to 600 ^{0}C. The plots of log resistivity vs reciprocal of temperature are drawn. For x = 0 and x = 1 compositions there is linear decrease in resistivity with increase in temperature, showing a break at the Curie temperature. The variation for x = 0.85, x = 0.70 and x = 0.55 compositions show that the resistivity initially increases with increase in temperature and thereafter decreases linearly with temperature again showing a break at their respective ferrimagnetic Curie temperatures. Activation energy below T_C is calculated and it is maximum for x = 1 and minimum for x = 0. The activation energy decreases as the ferrite content in the composition increases. The conduction in ferrite and ferroelectric phases is due to polaron hopping as reported earlier [9].

The thermoelectric power measurements of the samples were carried out from room temperature to 600 ^{0}C keeping the temperature of 20^{0}C across the cold and hot junctions. The sample x = 1, shows p - type conduction. The samples x = 0.85, 0.70, 0.55 and 0 show p -type conduction at lower temperatures but very soon show p to n type transition. The results are explained in terms of ions of variable valency.

Chapter V is concerned with the introduction to magnetoelectric effect as a 'product property' of composites. The measurement of ME conversion factor as the function of magnetic field is carried out. There is a gradual decrease in dE/dH with increase in magnetic field. Also dE/dH decreases with increase in volume fraction of ferrite at all the field strength. The variation is similar to that obtained by other workers [5, 8, 12]. The maximum value of dE/dH obtained for $x = 0.85$, $x = 0.70$ and $x = 0.55$ composites are 140, 120 and 80 μV/ cm/ Oe respectively.