Preparation, Characterization and D.C. Resistivity of $y(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4)+ (1-y) Ba_{0.9}Sr_{0.1}TiO_3$ Magnetoelectric Composites

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Abstract: *The magnetoelectric composites having chemical formula $y(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4)+(1-y)$ $Ba_{0.9}Sr_{0.1}TiO_3$ were prepared by standard double sintering ceramic method . Presence of constituent phases in the ME composite is confirmed by X-Ray Diffraction pattern. Purity of composition in ME composite was studied by EDAX spectrum. The Microstructural of the samples was studied by scanning electron micrographs. The effects of ferrite content on average grain size and DC resistivity were also studied which is important regarding the improvement of properties of the product materials.*

Keywords: *ME composite; XRD; EDAX; SEM; DC Resistivity*

I. Introduction

In Magnetoelectric effect (ME), magnetic field induces strain in the ferromagnetic phase which is transferred to ferroelectric through elastic coupling, due to which results in induce voltage in ferroelectric phase due to piezoelectric effect. Thus ME effect is result of coupling between ferromagnetic phase and ferroelectric phase. The ME effect was firstly observed in single phase ME material Cr_2O_3 in 1961[1, 2]. ME materials are classified into single phase ME material and Composite material. Single phase ME material possess both ferromagnetic and ferroelectric properties simultaneously while in composite material these two properties are present in two different phases. Due to weak magnetoelectric coupling of single phase materials in comparison with composite materials, these materials are not suitable for practical applications. Many researchers have worked on composite materials to improve ME voltage coefficient. Due to potential of convertibility of magnetic field into electric field and vice versa [3- 8], ME composite have large technological applications like transducer and sensor [9-14].

In our present study we have focused on preparation of pure ferrite, ferroelectric phases and their ME composites without any impurity. We have studied DC resistivity of individual phases and ME composites with chemical formula y $(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4)+(1-y) Ba_{0.9}Sr_{0.1}TiO_3$, where y=0.15,0.30 and 0.45.

II. Materials and Methods

Both ferromagnetic phase $(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4)$ and ferroelectric phase $(Ba_{0.9}Sr_{0.1}TiO_3)$ were prepared by double sintering ceramic method using carbonates and oxides. Nickel cobalt cadmium ferrite phase $(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4)$ was prepared by using carbonates of nickel, cobalt and cadmium and iron oxide. These powders were taken in appropriate molar proportion and mixed & ground in agate mortar for 2-3 hours. These compositions are presintered at 950^0C for 12 hours. The ferroelectric phase is also prepared by same procedure using carbonate of barium, strontium and titanium oxide in appropriate molar proportion and presintered at 1100^0C for 12 hours. The powders of ferrite and ferroelectric phases were mixed in the ratio 15:85, 30:70 and 45:55 respectively, so as to prepare ME composite. The mixer powders were presintered at 1150^0C for 12 hours. The pellets of ferrite , ferroelectric and ME composites having thickness 2-3 mm and diameter 10-15 mm were prepared by using hydraulic press. The remaining powder and pellets were finally sintered at 1200^0C for 12 hours.

To confirm the formation of ferrite phase, ferroelectric phase and ME composites without any impurity, XRD patterns were taken by using X-Ray diffractometer (Bruker D8 Advance) by using Cu-Kα radiation (1.5418Å). Purity of composites was also studied by taken using EDAX spectrum (JEOL JED 2300). The average grain size was calculated by recording micrographs using scanning electron microscope (SEM model –JEOL JSM 6360). DC resistivity of all the samples was calculated by two probe method up to 900^0C .

III. Result and Discussion

Fig.1 represents X-Ray diffraction pattern of $Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4$ ferrite phase and $Ba_{0.9}Sr_{0.1}TiO_3$ ferroelectric phase and their composites. XRD pattern shows that ferrite phase having intensity maxima (311) plane without any impurity peaks. The XRD of ferroelectric phase confirms the formation with maximum intensity peak (110) plane without any impurity peak. X-Ray diffraction patterns of ME composites show well

defined peaks of ferrite, ferroelectric phase without any intermediate phase. The most intense peak (311) and (110) confirms the formation of spinel ferrite phase and tetragonal ferroelectric phase respectively. It is observed that with increase in ferrite content the intensity peak of ferrite increases while that of ferroelectric phase decreases which obeys law of mixture [15]. The lattice parameters were calculated and reported in table. 1 , shows very slight deviation in lattice parameters of ferrite and ferroelectric phases may be due to stress exerted on each other by the two phases [16].

Fig.2 represents the EDAX spectrum of $0.15(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4)$ + $0.85(Ba_{0.9}Sr_{0.1}TiO_3)$ ME composite which confirmed the formation without any additional impurity. EDAX spectrum of all samples show same results.

Fig.3.represent scanning electron micrographs of $y(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4)+(1-y) Ba_{0.9}Sr_{0.1}TiO_3$. Average grain sizes of all the samples were calculated using Cottrell's method. From fig.2 a) and e), the average grain size of ferrite phase is of the order of 1.29 μm and 1.46μm that of ferroelectric phase. The variation of average grain size with ferrite content shown in fig.4 conforms that average grain size increases with increase in ferrite content may be due to average grain size of ferrite phase is large than that of ferroelectric phase

Fig.5 shows DC resistivity (ρ) of all the samples. Graph shows that ferroelectric phase has maximum resistivity than that of ferrite phase and DC resistivity of ME composite decreases with increase in ferrite content [17, 18]. For all samples the plots shows the decrease in DC resistivity with increase in temperature and increase in ferrite content, show two regions. The first region at low temperature is attributed to ferroelectric state while the second region at high temperature which is attributed to paraelectric state due to thermally activated polaron hopping mechanism [19, 20]. By using Arrhenius relation activation energies of all samples were calculated and listed in table 1. For all the samples activation energies are greater than 0.2eV which clearly indicates that the conduction is due to hopping of charge carriers. It is also observed that the activation energy increases with increase in ferrite content.

IV. Conclusions

Ferrite and Ferroelectric phases with their ME composites with chemical formula $y(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4)+(1-y) Ba_{0.9}Sr_{0.1}TiO_3$ were successfully prepared by standard double sintering ceramic method. The XRD pattern of all the samples confirms the presence of the cubic spinel structure of ferrite phase and tetragonal phase formation of ferroelectric phase. EDAX spectrums confirm the purity of individual phases and compositions of ME composites without any additional impurity peaks. The average grain size increases with increase in ferrite content may be due to average grain size of ferrite phase is large than that of ferroelectric phase The temperature dependence DC resistivity shows semiconducting nature of all the samples. The activation energies are greater than 0.2eV which clearly indicates that the conduction is due to hopping of charge carriers and the activation energy increases with increase in ferrite content.

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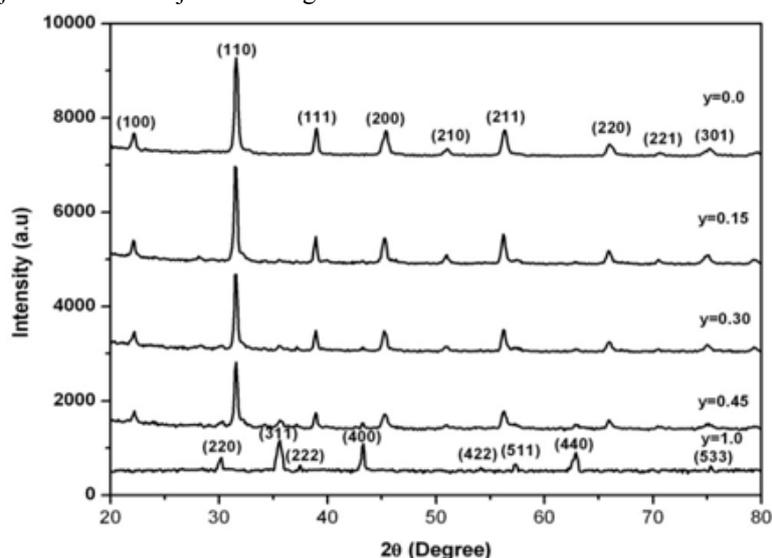
Fig.1. XRD patterns of $y(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4)+(1-y) Ba_{0.9}Sr_{0.1}TiO_3$ ME composites.
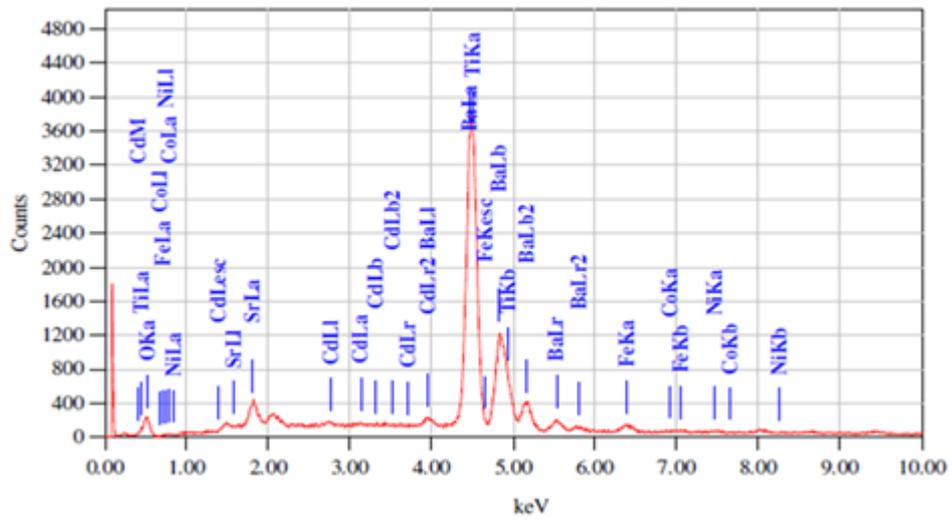
Fig.2. EDAX spectrum of $0.15(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4)+0.85(Ba_{0.9}Sr_{0.1}TiO_3)$ME Composites
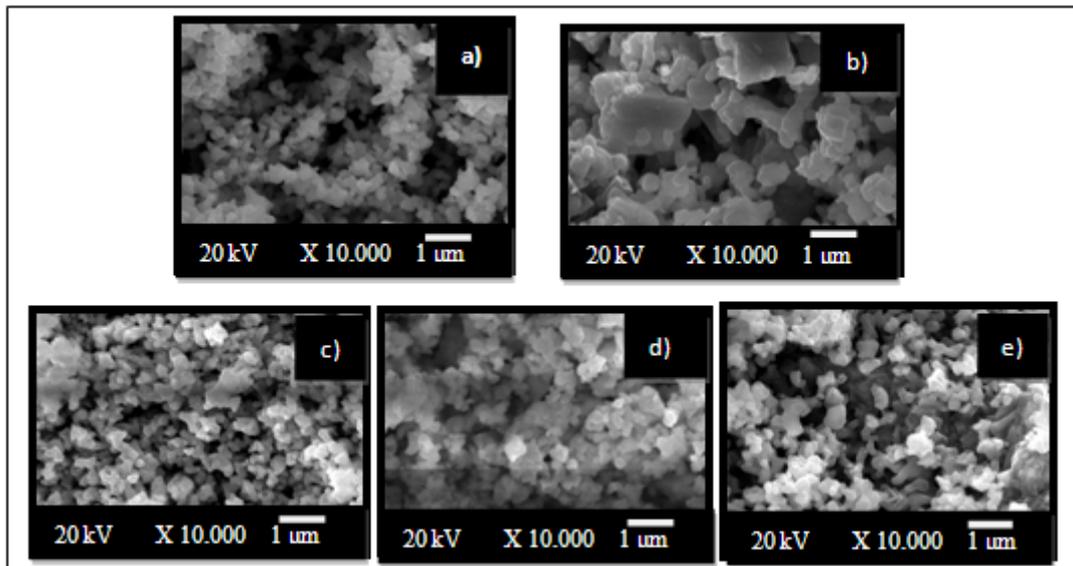
Fig.3. Scanning Electron Micrograph of $y(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4)+(1-y) Ba_{0.9}Sr_{0.1}TiO_3$ ME composites.
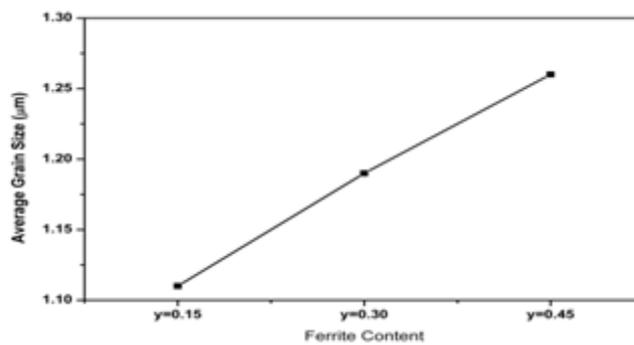
Fig.4. Variation of Average grain size with Ferrite content for $y(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4) + (1-y) Ba_{0.9}Sr_{0.1}TiO_3$ ME composites.
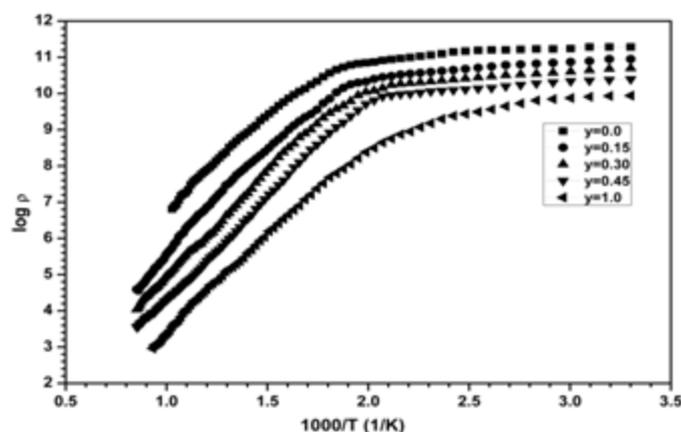
Fig.5. Variation of DC resistivity with temperature for $y(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4) + (1-y)$ $Ba_{0.9}Sr_{0.1}TiO_3$ ME composites.

Table.1. The lattice parameter, average grain size, resistivity and activation energy of $y(Ni_{0.7}Co_{0.2}Cd_{0.1}Fe_2O_4) +$ $(1-y)$ $Ba_{0.9}Sr_{0.1}TiO_3$ ME composites.

Composition	Lattice Parameters (A)			Average Grain size ($\square$m)	$\square_{RT} \times 10^{10}$ ($\square$ cm)	Activation Energy (eV)
	Ferrite	Ferroelectric	c/a			
y=0.0	-	a=3.99, c=4.03	1.010	1.29	19.2	0.649
y=0.15	8.367	a=4.00, c=4.02	1.004	1.11	8.9	0.784
y=0.30	8.366	a=4.01, c=4.04	1.009	1.19	5.03	0.798
y=0.45	8.372	a=4.01, c=4.02	1.003	1.26	2.5	0.809
y=1.0	8.370	-	-	1.46	0.9	0.763

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