Structural Analysis and Dielectric Properties of (x) $BaTiO_3$ + (1-x) $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ Magnetoelectric Composites

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Abstract : Magnetoelectric phenomena have been observed in two phase layered, laminated, bulk composites. Composites with varying magnetic Phase ($Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ ferrite) and electric phase (BaTiO3 ferroelectric) were prepared by using Chemical and Solid State Method. From XRD pattern it is clear that, there are presence two phases; cubic structure of piezomagnetic phase and perovskite tetragonal crystal structure of piezoelectric phase. No third phase coexists in the prepared magnetoelectric composites The lattice paramenters and crystallite size can be calculated from XRD. From Scanning Electron Microscope, It is observed that there are two dissimilar particle shapes corresponding to the two different phases. The variation of the dielectric constant with frequency ranging from 10 Hz to 1 MHz at room temperature has been studied. It shows dispersion due to Maxwell-Wagner type interfacial polarization.

I. INTRODUCTION

In magnetoelectric composites, induced polarization takes place with respect to magnetic field that gives rise important parameter i.e. ME voltage. ME composites have large technological application such as storage data device, sensor, transducer etc. The effect was first found in antiferromagnetic material such as Cr_2O_3 single phase compound at room temperature (α = 20mV/cm.Oe). To enhance magnetoelectric (ME) response of several orders of magnitude higher than that in those single phase compounds even above room temperature, Magnetoelctric composites were prepared by combining piezoelectric and magnetic phases [1]. In such composites, the individual phases like ferrite and ferroelectric interact with one another mechanically, so that a strain-induced ME coupling in magnetoelctric composites can be produced. The resulting effective ME couplings in composites, which are not present in their individual phases which defined by product properties [2]. In case of a bulk composite the electro-mechanical coupling of the ferroelectric phase has to be poled in order to allow for significant strain-induced interactions.

In present paper the magnetoelctric bullk composite such as (1-x) $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ + (x) $BaTiO_3$ composites were prepared by Solid state Ceramic method. This ceramic method is simple and properties of composites depend upon constitute phases, particle size and processing parameters [3]. Due to the microscopic interactions between the individual phases, the arising ME coefficient strongly depends on the microstructure of the composite. Characterization of ME composites has been done by XRD and SEM. The polarization state of the ferroelectric phase strongly influences the ME coupling. The electric measurement such as dielectric constant with respect to frequency was carried out.

II. EXPERIMENT

A) Preparation

The piezomagnetc phase $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ were prepared by taking starting material carbonates of nickel, cobalt, manganese and iron oxides were mixed and grounded ion agate motar and presintered at 950ºC and 8hr. Similarly Piezoelectric phase BaTiO3 were prepared by barium carbonate and titanium oxide were mixed and presintered at 1000ºC and 10 hr. ME Composites (1-x) $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ + (x) $BaTiO_3$ were prepared by mixing 10%, 15%, 20%, 25% ferrite phase with 90%, 85%, 80% and 75% ferroelectric phase respectively. These composites were finally sintered at 1200ºC for 12hr.

B) Characterization

X-ray diffraction of all the samples was carried out for to confirm the phase formation and crystal structure of the individual phases. Microstructural analysis of the sample were performed by Scanning Electron Microscope (SEM). The variation of dielectric constant (έ) will be measured at room temperature in the frequency range from 1kHz to 1MHz.

III. Results, Discussion and Conclusion

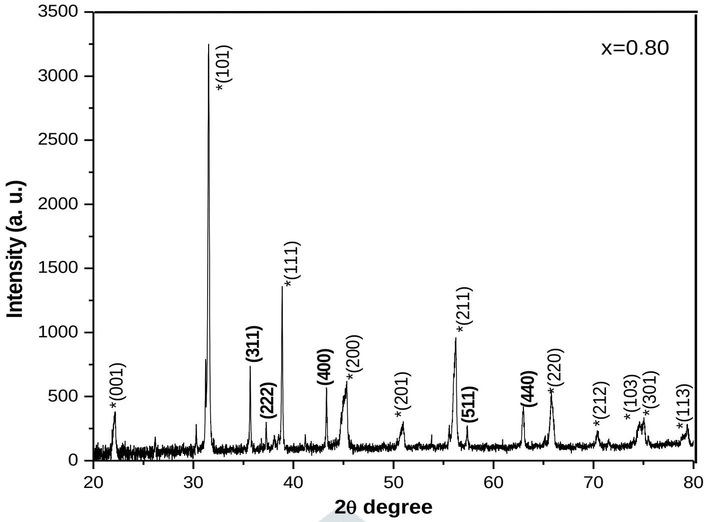

Fig 1 : XRD pattern of 20 % $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ + 80 % $BaTiO_3$ ME Composites

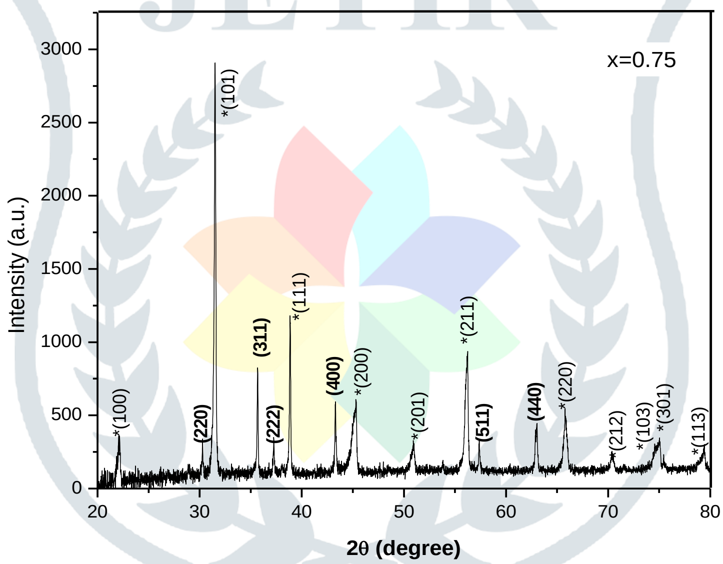

Fig 2 : XRD pattern of 25 % $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ + 75 % $BaTiO_3$ ME Composites

Figure 1 and 2 shows the X - ray diffraction pattern of 20% $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ + 80% $BaTiO_3$ and 25% $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ + 75% $BaTiO_3$ ME composites. It is observed that the peak (101) and (311) confirms that both ferroelectric and ferrite phase were present in composites. Intensity of ferrite phase peak increases and ferroelectric phase decreases as the content of ferrite increases in composite as per law of mixture. The peak of ferrite phase has cubic spinel structure and ferroelectric has pervoskite structure [4]. The lattice parameter of ferrite phase is 8.33Ǻ.

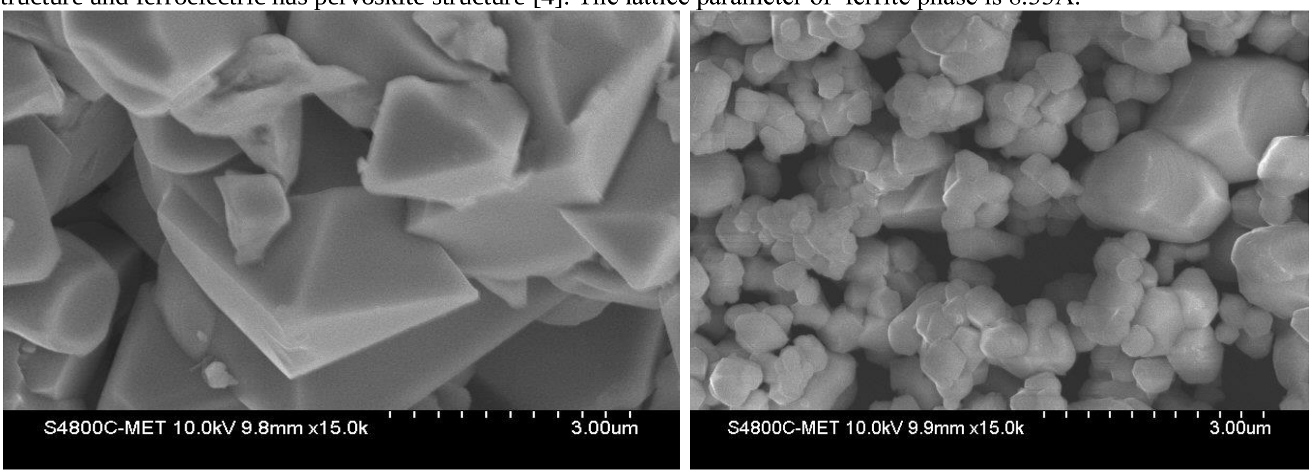

Fig 3 : XRD pattern of $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ ferrite and $BaTiO_3$ ferroelectric phase.

Figure 3 shows SEM micrographs of the ferrite and ferroelectric phase. The average grain size of all composites was calculated by using Cottrell's method. The grain size of ferrite is 3.37µm and grain size of ferroelectric is 0.71µm. The average grain size of ferrite phase is large as compare to ferroelectric phase. It is observed that the grain size of composite increases as the ferrite content increases [5] The results will be correlated with microstructural and phase properties because ferroelectric properties of composites are highly influenced by nonferroelectric grains surrounding the ferroelectric grains. The dielectric and ferroelectric properties can be influenced by domain wall motion. For larger grains, where size of the domain is smaller than the size of the grains, the movement of the domain walls is easier. On the other hand, in small grains, the movement of the domain walls is hindered by the grain boundary

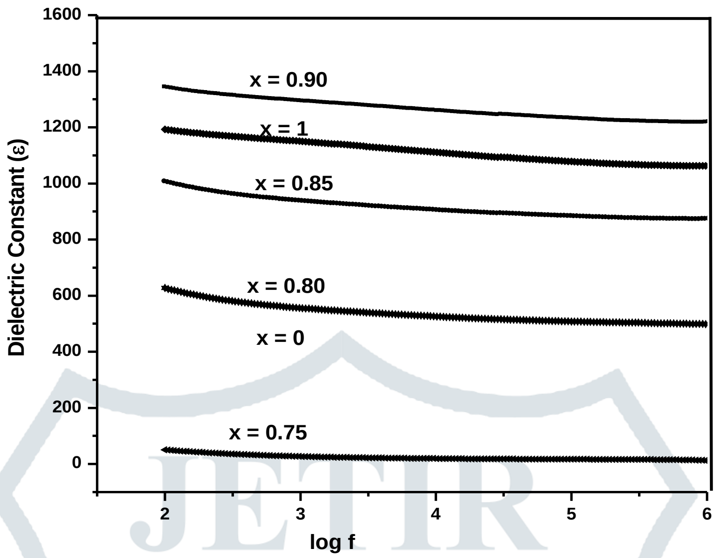

Fig 4 : Variation of dielectric constant with frequency of (1-x) $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ + x $BaTiO_3$ ME Composites

Figure 4 Shows the frequency dependence dielectric constant. In all cases the dielectric constant decreases with increase in frequency and at higher frequency it remains constant. Dielectric constant for ferroelectric phase is high as compare to ferrite phase. Similarly as ferrite content increases in composites the dielectric constant decreases due to increase the conductivity. At lower frequency there is electronic polarization and at higher frequency dispersión due to Maxwell Wanger type of interfacial polarization[6]. The values of dielctric constant with two different frequencies were as shown in Table.

TABLE

Sr. No	ME Composites	Dielectric Constant (at 100 Hz)	Dielectric Constant (at 10 KHz)
1	$BaTiO_3$	1190	1112.8
2	10 % $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ + 90% $BaTiO_3$	1346	1263.15
3	15%$Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ + 85% $BaTiO_3$	1007	906.82
4	20% $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ + 70% $BaTiO_3$	634	522.65
5	25% $Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$ + 75% $BaTiO_3$	54.96	15.99
6	$Ni_{0.94}Co_{0.01}Mn_{0.05}Fe_2O_4$	634	522.65

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