Studies on Magnetoelectric Effect of $Ni_{0.5}Cu_{0.3}Zn_{0.2}Fe_2O_4$/$BaTiO_3$ ME Composite

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ABSTRACT

The piezomagnetic-ferrite and piezoelectric-ferroelectric Magnetoelectric ME composites with composition $(1-x)Ni_{0.5}Cu_{0.3}Zn_{0.2}Fe_2O_4$+ (x) $BaTiO_3$ in which x = 0, 0.2, 0.4, 0.6, 0.8 and 1 mol were prepared by conventional solid state reaction. The presence of constituent phases in the composites was confirmed by x-ray diffraction studies. In order to study the resistivity of composites it is measured in temperature range 300K to 800K. It is found that resistivity increases with increase in ferroelectric content in the composites. ME voltage coefficient was measured as function of applied dc magnetic field and variation in ME response has been explained in terms of the content of piezoelectric phase and the intensity of applied magnetic field. The magnetoelectric properties of these composites depend on the ferrite content, ferroelectric content and resistivity of constituent component of composites. The ME Voltage coefficient increases with increase in ferroelectric content in composites. The maximum ME voltage coefficient of 141mV/cm Oe was observed for composite with x=0.80.

Keywords : Magnetoelectric effect, Resistivity, Composites, X-ray diffraction, ferrite, ferroelectric.

1 INTRODUCTION

Composites consist of two or more phases mixed together. ME composites consists of piezomagnetic and ferroelectric phase mixed together. ME composites have two field coupled together i.e. electric and magnetic field, application of electric field induces magnetic field and application of magnetic field induces electric field[1,2]. It is due to strain induced in piezomagnetic / ferrite phase by applied magnetic field, being mechanically coupled to stress induced in the piezoelectric/ferroelectric phase, the coupling result in an electric voltage. The ME materials represents a kind of smart structures which have received a continuous attentions as potential sensors for magnetoelectric conversion[3].The ME was first observed in antiferromagnetic Cr_2O_3 compound, i.e. the single phase material[4] ME materials. Many ME material and structures have discussed and developed. ME composites have many applications. It is used as sensor, modulator, phase inverter, transducer, spintronics, waveguides etc [5]. Superior ME composite **is** that which give high ME voltage coefficient. In order to obtain high value of ME coefficient, the piezomagnetic coefficient of ferroelectric phase and piezoelectric coefficient of ferroelectric phase must be high. The resistivity of both the phases is comparable and mechanical coupling between two phases is perfect [6].The search for such systems was promoted by practical needs, due to the very low values of the magnetoelectric voltage coefficient found in single-phase materials of the order of 1-20mV/cm.Oe. [7]. The ME effect is realized in composites on the basis of the concept of product properties [8]. According to this principle, a suitable combination of two phases such as piezomagnetic and piezoelectric or magnetostrictive and piezoelectric phases, can yield a desirable ME property. The conceptual condition for the ME

effect in composites were pointed out by Boomgaard,[9]as the following: (i) The individual phases should be in chemical equilibrium, (ii)No mismatch between grains, (iii) High values for the magnetostriction coefficient of the piezomagnetic or magnetostrctive phase of the ferroelectric phase. (iv) There is no leakage of the accumulated charges through the phases, enabling a deterministic strategy for poling. Following this principal, various ME bulk composites have been prepared. The selection of suitable combination of pizomagnetic and piezoelectric material to achieve better ME effect is however challenging task. In present work, Nickel doped with copper and zinc having high magnetostriction coefficient was selected as a piezomagnetic material. A well known ferroelectric BaTiO3 which have better piezoelectric property is used as a ferroelectric phase. Structural measurements were done using x-ray diffraction and electrical measurements by D.C. resistivity. Electrical resistivities have been used to suggest the conduction phenomenon and identify the charge carriers responsible for conduction. ME voltage coefficient measured by electric and magnetic poling of composites.

EXPERIMENTAL:

The components of present composites are $BaTio_3$ as ferroelectric phase and $Ni_{0.5}Cu_{0.3}Zn_{0.2}Fe_2O_4$as a ferrite phase with general formula (1-x) $Ni_{0.5}Cu_{0.3}Zn_{0.2}Fe_2O_4$+ (x) $BaTiO_3$ in which x = 0, 0.2, 0.4, 0.6, 0.8,1 mol were prepared by conventional solid state reaction. The ferrite phase was prepared by NiO, CuO, ZnO, and Fe2O3 in required molar proportions. These oxides were mixed and grind in agate mortar for couple

of hours. The ferroelectric phase was prepared by using BaO and TiO_2 as starting materials. These oxides are also mixed and grind in agate mortar. The ME composites were prepared by mixing 0.8,0.6,0.4 and 0.2 mol of ferrite phase with 0.2,0.4,0.6,and 0.8 mol of ferroelectric phase respectively. The required molar proportions were mixed and grind for 3 hour. The grind powder mixture was pressed into pellets using hydraulic press. The pelletized sample was final sintered at 850° for 24 hour in programmable furnace and slow cooled to room temperature to yield the final product.

The structural characterization of the samples was carried out by using X-ray diffractometer using Cu Kα radiation. The D.C. resistivity measurement was carried out using two probe methods with varying temperature from 800 K to 300K. The resistivity calculated by using formula

$$\rho = RA/t$$

Where R= Resistance
     A= Area of cross section of pellet
     t=thickness of pellet

For ME voltage coefficient, the samples were poled both electrically and magnetically. For electric poling the samples were heated approximately 30° above the ferroelectric Curie temperature and then allowed to cool slowly in presence of electric field. The ME voltage coefficient was measured by varying applied D.C. magnetic field at room temperature.

RESULT AND DISCUSSION:

1. PHASE DETERMINATION

Fig.1 and Fig.2 shows the XRD pattern of composites with x=0.4 and x=0.2 respectively. The peaks are characteristics of both ferrite and ferroelectric phases. The intensity as well as number of ferroelectric peaks increases with increase in ferroelectric content in composites. It may be due to increase of molar percentage of ferroelectric. The $Ni_{0.5}Cu_{0.3}Zn_{0.2}Fe_2O_4$ ferrite phase has cubic spinel structure. The ferroelectric phase has tetragonal pervoskite structure. The lattice parameters are shown in the following table 1. It is found that lattice constant of ferrite phase is decreasing as the composition of ferrite phase decreases and lattice constant of ferroelectric phase increases as the ferroelectric composition increases within the composites.

Particle size:
Particle size calculated by using formula

$$t = 0.9\lambda / \beta \cos\theta$$

Where, t is Particle size
    β is full width half maximum
    λ is wavelength of source used for x-ray diffraction
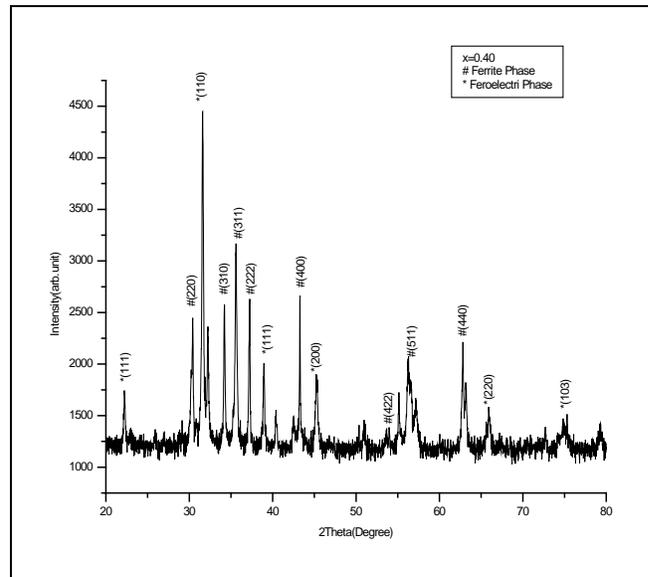   Ө is Bragg's angle.

Fig.1 XRD Patterns of of (1-x) $Ni_{0.5}Cu_{0.3}Zn_{0.2}Fe_2O_4$+ (x) BaTiO$_3$ (x = 0.4)
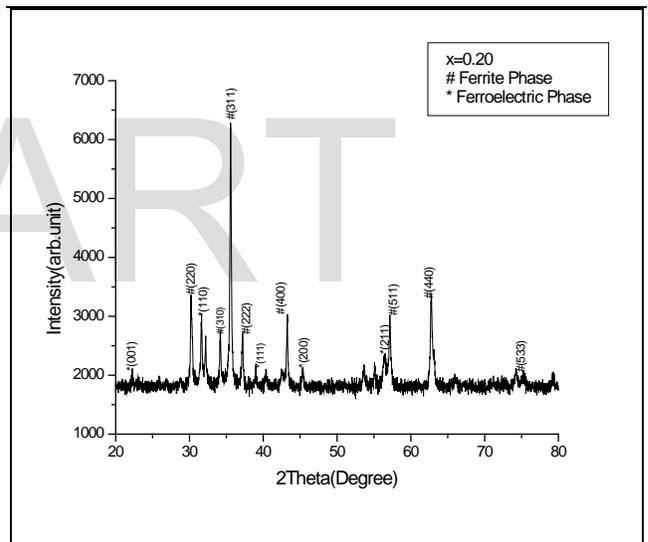

Fig.2 XRD Patterns of (1-x) $Ni_{0.5}Cu_{0.3}Zn_{0.2}Fe_2O_4$+ (x) BaTiO$_3$ (x = 0.2)

The particle size is calculated by using XRD and above formula as shown in table .1 the particle size decreases as the increase in ferroelectric composition. Ferroelectrics have smaller particle size than ferrite due to this particle size is decreases as ferroelectric content increases.

Table 1: Lattice parameters of Ferrite, Ferroelectric phases with their c/a ratios and particle size of (1-x) $Ni_{0.5}Cu_{0.3}Zn_{0.2}Fe_2O_4$+ (x) $BaTiO_3$ for (x = 0.0-1.0)

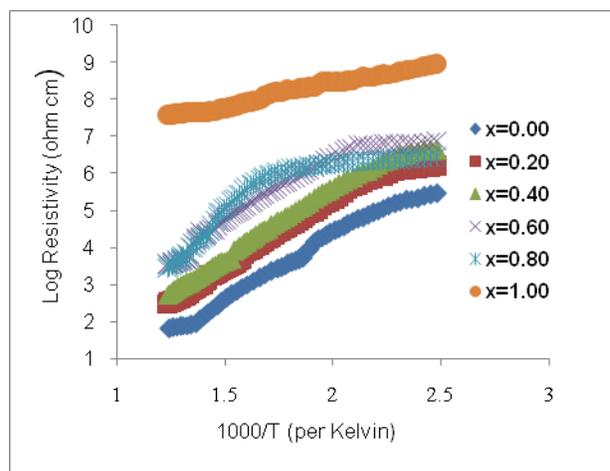| Composition	Lattice constant for ferrite a (A^0)	Lattice Constants for ferrolectrics		C/a ratio	Particle size
		a(A^0)	c(A^0)		
0.00	8.353	-	-	1.009	21.138
0.20	8349	3.990	4.025	1.011	15.274
0.40	8.343	4.000	4.042	1.010	14.344
0.60	8.319	4.005	4.049	1.011	13.629
0.80	8.237	4.006	4.049	1.010	13.206
1.00	-	4.020	4.060	1.009	8.311

Fig.3 Variation of dc resistivity (log ρ) with temperature (1000/T) for (1-x) $Ni_{0.5}Cu_{0.3}Zn_{0.2}Fe_2O_4$+ (x) $BaTiO_3$

2. ELECTRICAL RESISTIVITY:

Resistivity is important factor for ME effect. For ME effect the resistivity of composites must be high such that the accumulated charge carrier must not leak through the magnetostrictive phase. Thus electrical resistivity is an important property for ME composites. The temperature dependence of electrical resistivity is shown in Fig.3. The figure shows two trend of conductivity. The first trend observed at low temperature is due to impurities and it is attributed to order ferroelectric phase and the second trend occurs at high temperature this trend is due to the increase in the thermally activated drift mobility of charge carriers according to the hopping conduction mechanism [10]. In such a hopping process the carrier mobility is temperature dependant and is characterized by activation energy. In case of ferrites it is well known that electron hopping between Fe^{2+} and Fe^{3+} and hole hopping between Ni^{3+} to Ni^{2+} and Cu^{2+} to Cu^+ ions in B sites with an activation energy ≈0.2 eV are responsible for electrical conduction in bulk ferrite[11].With increase in ferroelectric content resistivity increases. This is due to fact that the ferroelectric have higher resistivity than as compare to ferrite phase. When the ferrite particles are dispersed throughout the composites it makes connected chains with ferroelectric particles and the resistivity is reduced to significantly due to low resistivity of ferrite Therefore good dispersion of ferrite particles in the matrix is required in order to sustain sufficient electrical resistivity of the composites.

3. ME EFFECT

The ME effect in composites having ferrite and ferroelectric phase depends on the applied d.c. magnetic field, electrical resistivity, mole percentage of constitute phases and mechanical coupling between the two phases [12]. Fig.4 shows the variation of ME voltage coefficient (i.e. dE/dH) and magnetic field with molar proportion of ferrite and ferroelectric content. Fig.4 shows the ME voltage coefficient as a function of D.C. magnetic field. From the plot it is observed that the ME voltage coefficient goes on increasing with increase in magnetic field and decreases afterwards. This ME coupling is achieved by electromechanical conversion in the piezoelectric phase and magneto-mechanical conversion in the magnetostrictive phase by stress transfer through the interface between the two phases. The ME voltage coefficient increases with increase in ferroelectric content in the composite. This increase is due to high resistivity of ferroelectric phase as compare to ferrite phase resulting in the leakage of charges developed in the piezoelectric grains through the low resistance path of surrounding ferrite grains [13-15].Thus ME effect is resistivity dependent property. At higher magnetic field, the ME output decreases due to saturation of magnetostriction at a certain applied field which produces a constant charge in the piezoelectric phase [16, 17]. In the present case maximum value of the ME voltage coefficient is 141 mv/cmOe observed for composite with x=0.8.
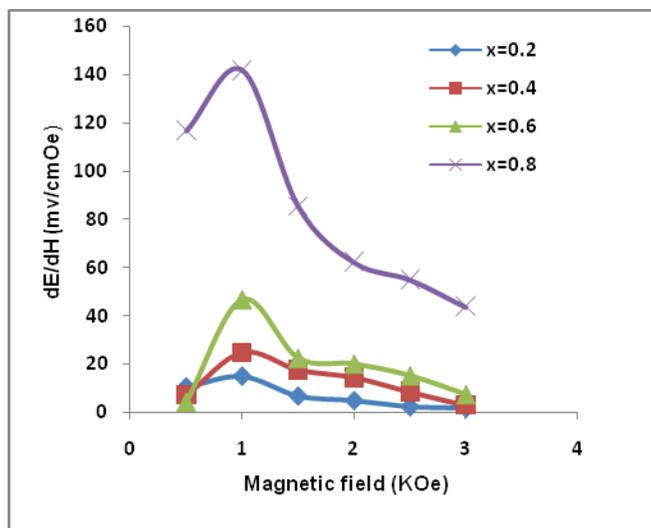
Fig.4 Variation of magneto electric coefficient with magnetic field for $(1-x)Ni_{0.5}Cu_{0.3}Zn_{0.2}Fe_2O_4 + (x) BaTiO_3$

CONCLUSION

The ferrite-ferroelectric ME composites were prepared successfully by standard ceramic method. The XRD pattern shows presence of both ferrite and ferroelectric phases. The intensity and number of ferroelectric peaks are observed to increases with increase in ferroelectric content in the composites. The D.C resistivity increases with increase in ferroelectric content in the composites because ferroelectrics have high electrical resistivity as compare to ferrite. The ME voltage coefficient also increases with increase in ferroelectric content.The maximum value of the ME voltage coefficient is 141 mv/cm. Oe. observed for this ME composite with composition x=0.8.

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