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"Dielectric Behavior and Magneto-Electric Effect in 20% CuFe₂O₄+80% KNbO₃ Composites"

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Abstract - ME composite of 20% CuFe₂O₄ as a ferrite phase and 80% KNbO₃ as a ferroelectric phase were prepared using a conventional ceramic double sintering process. The presence of phases was confirmed by X-ray diffraction (XRD) pattern. The dielectric parameter such as dielectric constant (ε'), dielectric loss (ε") and dielectric loss gent (tan δ) were studied as a function of frequency in the range of 1 KHz to 1MHz and also with temperature at constant frequency 1 KHz and 10KHz. The ME composites effect is measured by changes in the resulting electric field in the ME composite due to the applied external magnetic field.

Key Words: ME composites, X-ray diffraction, Magnetoelectric effect, Dielectric constant, Dielectric loss tangent.

1.INTRODUCTION

Composites are class of materials in which different phases mixed together and used for various applications. Most of the composite materials has been produced to upgrade mechanical properties of materials. Now a days the attentiveness is moved towards the use of composites for the electrical application [1]. The magnetoelectric effect is a phenomenon in which the application of magnetic field induces electric polarization. ME effect is a property of the posites which is not present in their constituent's phases [2]. Ferrite ferroelectric composites consists of two phases i.e. Piezomagnetic and Piezoelectric. The magneto electric effect is coupled, two field effects in which the application of electric field induces magnetization and magnetic field induces electric polarization [2]. It is because of strain induced in the Piezomagnetic phase by the applied magnetic field, being mechanically coupled to stress induced in the piezoelectric /ferroelectric phase, the coupling induces an electric voltage [3,4].

The ME composites have been used as sensors, waveguides, switches, phase invertors, modulators, etc. [5]. The selection of appropriate combination of piezoelectric and Piezomagnetic material as in to obtain ME effect is a challenging task. In the recent work we demonstrated the structural, dielectric and ME effect in 20%CuFe₂O₄ + 80%KNbO₃ composites. Which describe about the behavior of localized electric charge carriers. This also leads to a better comprehension of the mechanism of electrical conduction and dielectric polarization.

In the present scenario, copper ferrite is selected as a ferrite phase in order to achieve large Jahn-Teller distortion in ferrite lattice. Which will develop more mechanical coupling among the ferroelectric and ferrite phases that may result maximum ME signal. Low anisotropy favors magnetization mechanism in the crystal form such as domain wall moment and domain rotation which are the essential factors for magneto mechanical coupling that arises the ME effect and KNbO₃ is chosen as ferroelectric material for its high piezoelectric constant, high dielectric permittivity and high electromechanical coupling constant, as well as low dielectric and mechanical losses [6].

2. EXPERMENTAL

The ME composites contains two phases viz. ferrite and ferroelectric. In the present work, for the preparation of CuFe₂O₄ ferrite AR grade of copper oxide (CuO) and (Fe₂O₃₎ were taken in their required molar proportions. The mixture was ground thoroughly for 3 hour and pre sintered at 900°C for 12 hours. The powder is then reground again and finally sintered at 1050°C to obtain the single-phase copper ferrite (CuFe₂O₄). The potassium niobate (KNbO₃) phase was prepared through the solid-state reaction method using AR grade Potassium carbonate (K2CO3) and niobium pen-oxide (Nb₂O₅) in required molar proportions. The mixture was ground to fine powder. It was Pre sintered at 900°Cfor 12 hours. In the final sintering the material was held at 1050 $^{
m o}$ C for 12 hours. After sintering both the phases were grounded to fine powders. The ME Compositions were produced by mixing 20 Mol % of ferrite phases with 80Mol % of ferroelectric phase, respectively. All samples were sintered at 1050°C for 24 hour and naturally cooled to room temperature.

2.1 Characterization

The samples of X-ray diffraction patterns were taken on Philips X-ray diffractometer (Modal PW 1710) using Cu Kα radiation (λ = 1.5418Å). The parameter such as capacitance (c) and dissipation factor (D) were measured in the frequency range 1 KHz to 1MHz using LCR meter (HP 4284A). Measuring of ME output carried out by varying the applied dc magnetic field at room temperature with the help of a Keithley's electrometer (Model 2000).

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3. RESULTS & DISCUSSION

3.1. X-ray diffraction (XRD)

The composite was characterized by using X-ray diffractometer (Phillips Model PW 1710) using Cu $K\alpha$ radiation.

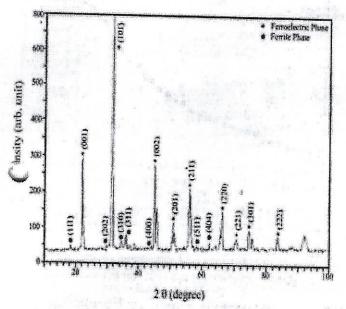


Fig. 1 XRD Patterns of 20% CuFe₂O₄ +80% KNbO₃

The X-ray diffraction (XRD) pattern of 20% CuFe₂O₄+ 80% (KNbO₃) composites are shown in fig. 1. It is observed that both the ferrites as well as ferroelectric phases are present. No irregular peaks are present in the pattern which affirm single phase formation of composites. Calculation for both the phases in lattice parameters being carried out. The ferrites CuFe₂O₄ is unique spinal ferrite it shows tetragonal nmetry in pure form and have cure point 457°C. The presence of John -Taller ion Cu2+ and is interesting to note that both CuFe₂O₄ and KnbO₃ possesses tetragonal structure. The lattice parameter of ferrite phase in the 20% ferrite composite are a=8.159 Å and c= 8.649 Å and c/a ratio equal to 1.060. The lattice parameter of ferroelectric phase in 60% ferroelectric composite are a=3.991 Å and c= 4.057 Å and c/a ratio equal to 1. 017. There are no structural changes noticed for either phase in the composites, as reported [7, 8].

3.2. Dielectric properties

a) Frequency dependence of dielectric properties

Fig.2 shows the variation of dielectric constant (E') with frequency for the composite 40% CuFe₂O₄ + 60% KNbO₃. From the fig.2 it is observed that the dielectric constant (E') rapidly decreases at low frequencies and fairly constant at higher frequencies. The large value of dielectric constant at low frequencies is because of dislocations and other defect [9, 10] In case of 40% CuFe₂O₄ + 60% KNbO₃ composite this large value of dielectric constant (E') has been ascribed to

the effect of heterogeneity samples like pores and layered structure. However, in case of composites it is attributed to the fact that, ferroelectric regions are enclosed by non-ferroelectric region similar to the relaxor ferroelectric material [11]. This arises to interfacial polarization which is reflected by sharp fall in dielectric constant (E').

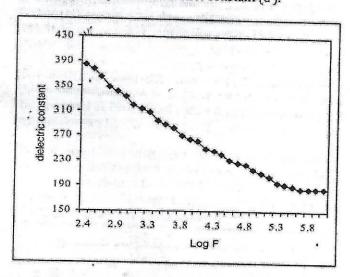


Fig.2 Variation of dielectric constant (ε') with frequency(F) for 20 % CuFe₂O₄ + 80 % KNbO₃

Figure 3. represents the variation of dielectric loss as a function of frequency. It is noticed from Fig. 3 has the dielectric loss also decreases with increase in frequency. At low frequency the decrease in dielectric constant is faster than at high frequency.

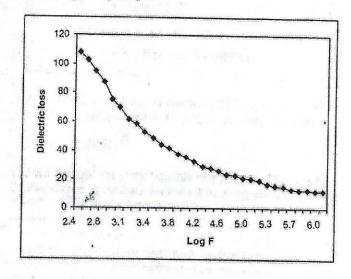


Fig.3 Variation of dielectric loss (ε") with frequency (F) for 20 % CuFe₂O₄ + 80 % KNbO₃

The variation of dielectric loss tangent (tan δ) with frequency for the composite shows the similar behavior as that of the dielectric constants with frequency. The dielectric loss tangent decreases exponentially with increase in

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frequency. Similar results have been reviewed by various workers in case of other composites [12, 13].

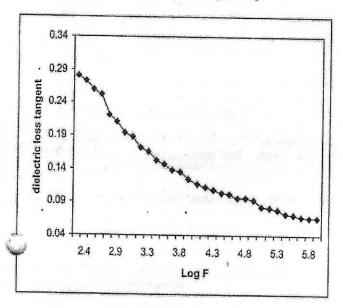


Fig.4 Variation of dielectric loss tangent (tan δ) with frequency (F) for 20% CuFe₂O₄ + 80% KNbO₃

b) Temperature dependence of dielectric properties

The variation of dielectric constant (ϵ ') with temperature is shown in Fig.5.The dielectric constant (e') increases with increasing temperature for 1Hz and 10 KHz frequency. It is noticed that for lower frequency the dispersion is more compared to the higher frequency.

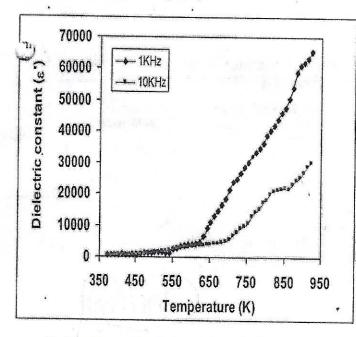


Fig.5Variation of dielectric constant (ϵ') versus temperature (K) for 20% CuFe₂O₄ +80% KNbO₃

4. MAGNETO-ELECTRIC (ME) EFFECT

Fig. 6 shows the magneto-electric effect voltage coefficient as a function of d. c. magnetic bias for all composites under investigation. The magneto-electric (ME) effect is measured by changes in the resulting electric field in the magnetoelectric (ME)-composites due to the external magnetic field [14]. It is noticed that magneto-electric (ME) coefficient initially increases to a certain applied magnetic field (0.5KOe) and then start decreasing as applied field increases. The ME effect is obtained maximum for 20% CuFe₂O₄ +80% KNbO3. Similar behavior of magneto-electric coefficient was also observed in case of CuFe₂O₄ + BaTiO₃ composite [15] and well-known ferrite +ferroelectric composites.

The ME effect in composites is because of the strain developed in ferrite phase by the impact of magnetic field which being mechanically coupled, results in stress around ferroelectric phase. This stress results in the polarization of ferroelectric phase due to piezoelectric effect.

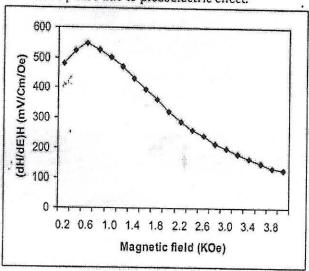


Fig. 6 Variation of magnetoelectric (ME) conversion factor with magnetic field for 20 % CuFe₂O₄ +80 % KNbO₃

5. CONCLUSION

The ME composites were successfully prepared by standard double sintering ceramic method. The X-ray analysis reveals presence of both the ferrite and ferroelectric phases in the composite. The dielectric behavior suggests the dielectric dispersion at lower frequency. This is because of the interfacial polarization and due to the heterogeneity of samples. The dielectric constant with temperature gives the dispersion for lower frequency is more compared to higher frequency. Our results on dielectric properties of the composite are analogous to literature. Magneto-electric conversion factor with varying magnetic field shows maxima in the curve at a lower magnetic field and then decreases continuously at higher magnetic field content increases magneto-electric conversion fa increases.

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