EFFECT OF Cu^{2+} IONS SUBSTITUTION ON DIELECTRIC BEHAVIOR OF NICKEL FERRITE

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ABSTRACT:

The samples of copper substituted nickel ferrite having the generic formula $Ni_{1-x}Cu_xFe_2O_4$ (x = 0.0, 0.4, 0.8) have been synthesized by standard double sintering ceramic technique using AR grade oxides. The formation of single phase cubic spinel structure of all the samples under investigation have been carried out using X-ray diffraction technique at room temperature. The dielectric constant (ε'), dielectric loss (ε''), dielectric loss tangent (tanδ) was measured as a function of frequency by using LCR–Q meter. The frequency dependence of dielectric measurements was carried out within the range 100 Hz to 1 MHz. The values of dielectric parameters (ε',$\varepsilon''$and tanδ)are much higher at lower frequencies but decreases with increase in frequency. At very high frequencies, its values become so small that it becomes independent of frequency. The decrease in dielectric parameters with increase of frequency may be due to the fact that beyond a certain frequency of the external electric field, the electronic exchange between ferrous and ferric ions cannot follow the alternating field.It is observed that dielectric constant (ε'), dielectric loss (ε'') and dielectric loss tangent (tanδ) appreciably increases with copper concentration x but decreases with increases in frequency.

KEYWORDS: Ni-Cu spinel ferrite, XRD, dielectric properties.

1. Introduction:

Ferrites are ferrimagnetic materials with good magnetic, dielectric properties and a large number of technological applications in satellite communication, memory device, computer, components, filter components, antenna rods, transformer core etc, because of their excellent electrical and magnetic properties [1].The high electrical resistivity, low eddy current and dielectric losses, moderate saturation magnetization, easy and low cost of preparation, high Curie temperature and high permeability are the remarkable characteristics of a ferrite material which makes them useful in variety of applications. The properties of ferrite depends on magnetic interaction, cation distribution in the two sub lattice, method of preparation, preparative parameters, type and amount of dopant [2-4]. The dielectric properties of ferrites are dependent upon several factors including the method of preparation, chemical composition and grain structure. Among the spinel ferrites, nickel ferrite is having special attraction because of their useful properties such as inverse spinel nature, high saturation magnetization and Curie temperature,

high electrical resistivity and chemically most stable. Copper is a Jahn Teller ion with magnetic moment 1μ_B.In the literature very few studies on copper substituted nickel ferrite are reported. We have prepared a series of copper substituted nickel ferrite with the formula $Ni_{1-x}Cu_xFe_2O_4$(x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) using ceramic technique. Here, we report our results on dielectric studies for x = 0.0, 0.4 and 0.8 samples.

2. Experimental:

The polycrystalline samples of $Ni_{1-x}Cu_xFe_2O_4$ (x = 0.0, 0.4, 0.8) were prepared using the standard ceramic technique [5]. A.R. grade oxides of corresponding ions (NiO, CuO and Fe_2O_3) were mixed in stoichiometric proportion. Grinding using agate mortar (4 h) was carried out for each sample. The samples were pre-sintered at 1293 K for 12 h. The sintered powder is again reground and sintered at 1353 K for 14 h. Then the powder of samples compressed into pellets of 10 mm diameter using a hydraulic press with pressure 6 ton/inch^2 and sintered at 1273K for 12 h. The samples were

furnace cooled to room temperature. The prepared samples were characterized by X-ray powder diffractometer in the 2θ range 20°-80°at room temperature to confirm single phase spinel structure. Dielectric constant (ε′) dielectric loss (ε″) and loss tangent (tanδ) as a function of frequency at room temperature were measured by LCR Meter. For dielectric measurements the pellets were coated with silver paste for good ohmic contact.

3. Results and Discussion:

3.1 XRD (X-ray diffraction): The XRD patterns of mixed spinel ferrites system $Ni_{1-x}Cu_xFe_2O_4$ (x = 0.0, 0.4, 0.8) under investigation shows that the samples have single phase cubic spinel structure. The figure 1 shows typical XRD pattern for x = 0.2.The Bragg's peaks are sharp and intense. Lattice constant calculated using XRD data increases very slowly with increase in copper content 'x'. The small variation in the lattice constant with copper substitution can be explained on the basis of very close ionic radii of nickel (0.69Å) and copper (0.72Å) [6]. The ionic radii of nickel and copper are approximately close to each other and hence there is no much variation in the lattice constant.

3.2 Dielectric constant (ε′): Variation of dielectric constant (ε′) of $Ni_{1-x}Cu_xFe_2O_4$ with frequency and copper content x is shown in Fig.2. It is observed from Fig.2 that dielectric constant increases with the increase in Cu^{2+} concentration. In Ni–Cu ferrites, copper ions occupy tetrahedral (A) sites and octahedral [B] sites where as Ni^{2+} ions prefer to go to octahedral [B] sites. Fe^{3+} ions which exist in 2+ as well as in 3+ states, occupy both A and B sites.
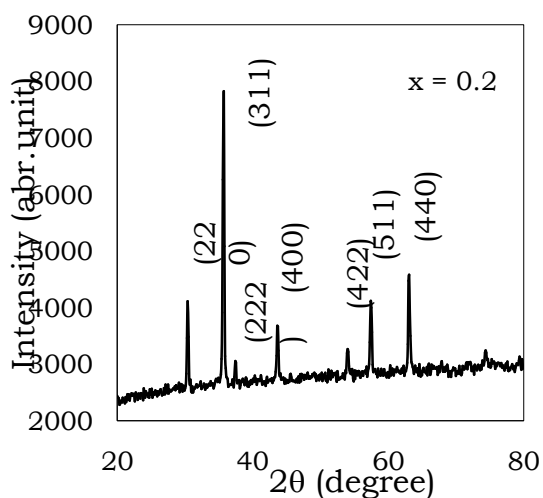
When Cu^{2+} is added in place of Ni^{2+} ions, some of the Fe^{3+} ions are converted to Fe^{2+} ions to maintain the charge neutrality. As a result, hopping between Fe^{3+} and Fe^{2+} ions increases hence the resistance of grains decreases. This increases the probability of electrons to reach the grain boundary. Consequently, polarization and dielectric constant increase [7]. The value of ε′ is much higher at lower frequencies. It decreases with the increase in frequency. At very high frequencies, its value becomes so small that it becomes independent of frequency. Other researchers have also observed similar kind of behavior [8, 9]. The variation in dielectric constant may be explained on the basis of space charge polarization. According to Maxwell and Wagner two-layer model [10, 11], space charge polarization is because of inhomogeneous dielectric structure of the material. It is formed by large number of well conducting grains separated by thin poorly conducting intermediate grain boundaries. The electronic exchange between Fe^{2+} and Fe^{3+} is due to the local movement of electrons in the direction of electric field which determines the polarization in ferrites. Polarization decreases with the rise in frequency and then attains a constant value. It is because of the fact that beyond a certain value of frequency of external field, the electron exchange $Fe^{2+} \leftrightarrow Fe^{3+}$ cannot follow the alternating field [12, 13].
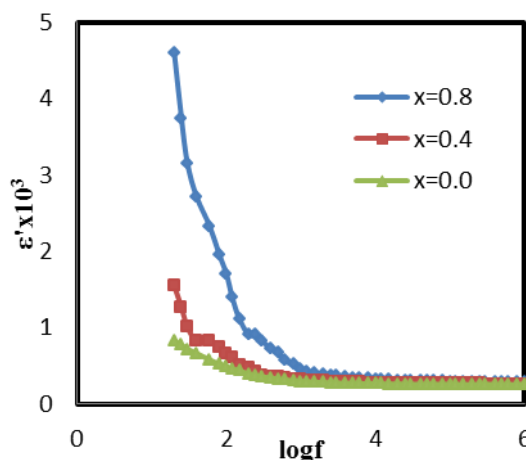
Fig.2: *Variation of dielectric constant (ε') with logarithm of frequency (LogF) of $Ni_{1-x}Cu_xFe_2O_4$.*

3.3 Dielectric loss (ε″): The variation of dielectric loss (ε″) is shown in Fig. 3. The variation of dielectric loss (ε″) with frequency can be explained on the basis of dispersion due to the Maxwell–Wagner interfacial polarization with Koop's phenomenological theory [14]. Dielectric loss decreases with the increase of frequency. It is because of decrease in polarization with the increase in frequency. Ultimately, it reaches a constant value. Other researchers [15] have quoted a similar kind of trend with the change in frequency. Rabinkin and Novikova [16] pointed out that polarization in

Fig. 1: *Typical XRD Pattern of $Ni_{1-x}Cu_xFe_2O_4$ (x=0.2)*

ferrites is similar to that of conduction. The electron exchange between $Fe^{2+} \leftrightarrow Fe^{3+}$ results the local displacement of electrons in the direction of the applied field that determines the polarization.
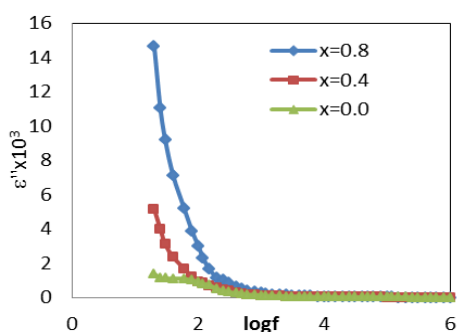

Fig.3: *Variation of dielectric loss (ε'') with logarithm of frequency (Log F) of $Ni_{1-x}Cu_xFe_2O_4$.*

Polarization decreases with the increase in value of frequency and then reaches a constant value. Dielectric loss (ε'') has large value at lower frequency. It is because of the predominance of species like Fe^{2+} ions, oxygen vacancies, grain boundary defects, interfacial dislocation pile-ups, voids, etc. [11- 17]. The decreasing trend in (ε'') with the increase in frequency is natural due to the fact that any species contributing to polarizability is found to show lagging behind the applied field at higher frequencies [7].

3.4 Dielectric loss tangent (tanδ): The values of tanδ depend on a number of factors such as stoichiometry, Fe^{2+} content and structural homogeneity, which in turn depend on the composition and sintering temperature of the samples [18, 19]. The variation of dielectric loss tangent (tanδ) is shown in Fig. 4. The initial decrease of tanδ with an increase in frequency may also be explained on the basis of Koop's phenomenological model [15].

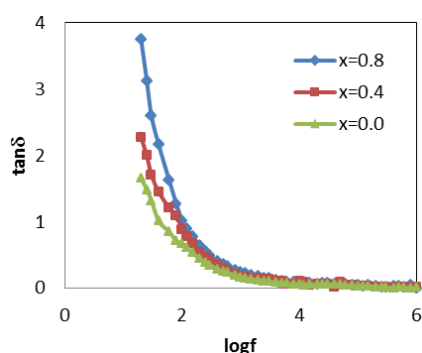**Fig.4:** *Variation of dielectric loss tangent (tanδ) with logarithm of (Log F) of $Ni_{1-x}Cu_xFe_2O_4$.*

Table 1 shows comparison of dielectric constant (ε′), dielectric loss (ε″) and dielectric loss tangent (tanδ) at room temperature at 100 Hz, and 1 MHz frequencies for $Ni_{1-x}Cu_xFe_2O_4$ system. It revealed

from Table 1 that dielectric constant, dielectric loss and dielectric loss tangent increases with Cu content and decreases with frequency.

Table 1: Room temperature dielectric constant (ε′), dielectric loss (ε″) and dielectric loss tangent (tanδ) at 100 Hz and 1 MHz of $Ni_{1-x}Cu_xFe_2O_4$

	Frequency					
	100 (Hz)			1(MHz)		
x	ε'	ε"	tanδ	ε'	ε"	tanδ
0.0	234	180	0.77	210	1.13	0.005
0.4	1350	2389	1.77	292	5.84	0.02
0.8	8266	52902	6.4	264	14.52	0.055

4. CONCLUSIONS:

1. Single phase cubic spinel structured samples of $Ni_{1-x}Cu_xFe_2O_4$ were obtained successfully using ceramic technique.
2. Substitution of copper (Cu^{2+}) ions increases the dielectric parameters of the system.
3. The values of dielectric parameters (ε′,ε″and tanδ) are high at low frequencies but decreases with increase in frequency.

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