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Dielectric behavior and A. C. Conductivity in Cu-Ti Ferrites

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ABSTRACT

Dielectric properties such as dielectric constant (∈') and dielectric loss tangent were studied as a function of composition, frequency and temperature for a series of $Cu_{1+x}Ti_xFe_{2-2x}O_4$ ferrite samples prepared by using conventional ceramic technique. Initial Ti^{4+} ion substitution results in decrease in dielectric constant while dc resistivity increases on substitution. The measurement of dielectric constant with frequency (100Hz-1MHz) shows decrease in dielectric constant with increasing frequency and tends to reach constant value. Variation of dielectric constant with frequency at different temperature show increase in dielectric constant in general and the tendency of dispersion in dielectric constant observed increase with temperature. The variation of dielectric constant with temperature at different frequencies was studied. Dielectric constant steadily increases reaches to maximum and thereafter decreases. The frequency dependence of dielectric loss tangent is found to be abnormal for some samples. The frequency dependence of dielectric loss tangent at various temperatures was studied, the abnormal behavior of dielectric relaxation processes was observed. A.C. Conductivity was derived from dielectric constant and loss tangent data. The conduction in this system is interpreted as due to small polaron hopping.

Key words: ferrite, loss tangent, dielectric constant, polaron hopping.

INTRODUCTION

The dielectric constant (∈') and dielectric loss (tanδ) for Cu-Cr ferrites were studied [1]. The results showed that there was an abnormal behavior of the dielectric constant giving dispersion peak at certain frequency. The dielectric constants for ferrites containing copper were studied [2]. The results showed that there is an abnormal behavior of dielectric constant with frequency. The a.c. conductivity and dielectric constant were studied for copper ferrite system [3]. The a.c. conductivity was found to be high for higher frequency and showed a trend to be expected for small polaron hopping. The dielectric constant and a.c. conductivity are hitherto not reported so far for Cu-Ti ferrites. Therefore, the present paper reports the study of the effect of composition, frequency and temperature on dielectric constant and a. c. conductivity of Cu- Ti ferrite prepared by the ceramic technique.

MATERIALS AND METHODS

Samples of the system $Cu_{1+x}Ti_xFe_{2-2x}O_4$ with x= 0.0,0.05, 0.1, 0.15, 0.2 and 0.3 were prepared by usual ceramic technique.[24] The samples were pressed in form of discs and rubbed with silver paste as contact material. Dielectric measurements as function of frequency in the range of 100 Hz – 1MHz at room temperature and also as a function of temperature in the range 300- 800 K for few selected frequencies, viz. 1 kHz, 10 KHz, 100 kHz, 500 kHz nd 1 MHz, were carried out using a LCR meter (HP-4284A model).The dielectric constant and a. c. conductivity ($\sigma_{a.c.}$) was calculated by using the formulae given in literature [3].

RESULTS AND DISCUSSION

The room temperature values of dielectric constant (∈') and dc electrical resistivity ρ_{dc} for the composition of $Cu_{1+x}Ti_xFe_{2-2x}O_4$ series are given in Table 1. It is seen that for Ti^{4+} substitution the dielectric constant decreases while the dc resistivity increases on substitution of Ti^{4+}. Murthy and Shobhandri [4] have investigated the dielectric properties of some Ni-Zn ferrites as a function of composition, frequency and temperature. Yu et. al [5] have studied the dielectric behavior of Ni-Zn ferrites as a function of composition and frequency. Ravi Kumar and Ravinder [6] have also investigated dielectric behavior of mixed Mn-Zn-Gd ferrites. Iwauchi [7] reported a strong correlation between the conduction mechanism and dielectric behavior of ferrites. Rezlescu and Rezlescu [8] studied composition, frequency and temperature dependence of dielectric constant of copper containing mixed ferrites such as Cu-Zn and Cu- Mn. They explained the compositional dependence of dielectric polarization on similar lines to that of conduction. They concluded that the electron exchange interaction $Fe^{2+} \Leftrightarrow Fe^{3+}$ results in the local displacement of electrons in the direction of an electric field, which determines the polarization of the ferrites. A similar explanation is proposed for the composition dependence of the dielectric constant of ferrites under investigation. Effects of Ge4+ substitution in CuFe2O4and Ti4+ in Ni-Zn ferrite have been reported earlier [9]. They have made similar observations. Thus the replacements of Fe^{3+} ions by tetravalent ions like Ti^{4+} reduce the total number of Fe^{3+}/Fe^{2+} ions available for electron exchange contributing the conduction enhancing the electrical resistivity. The decrease of dielectric constant (∈') with the tetravalent ion substitution in CuFe2O4 can thus be related to increase of ρ_{dc}.

The variation of dielectric constant (∈') with frequency at room temperature is shown in figure 1. The ε' decreases with increasing frequency and tends to reach a constant value. A comparative study of dispersion curves of these samples shows that the change in the values of ε' at lower frequencies is larger than that at higher frequencies. The dielectric constant ε' does not show much dispersion with frequency at high values. Similar results are observed in many ferrites and this is explained by Koop's phenomenological theory on the basis of space charge polarization due to inhomogeneous dielectric structure. It has been reported that the dielectric constant ε' shows similar dispersion behavior for other ferrites [10,11,12, 25]. The dielectric constant ε' decreases continuously with increasing frequency, which is but natural as any species contributing to polarizability, is bound to show polarization lagging progressively behind the field at higher and higher frequencies. The second feature of large ε' at low frequencies could be understood [13,14] to indicate the predominance of the species like Cu^{1+}, Fe^{2+}, O^{2-} vacancy, grain boundary defects, interfacial dislocation pile up etc. which show in ε, giving rise to higher values. In addition sensitivity of these species to 'x' would give rise to larger dispersion.

The variation of dielectric constant ε' with composition and frequency has been studied by Reddy and Rao and others [15-16]. They have explained the behavior of dielectric constant by assuming that the mechanism of dielectric polarization is similar to that of the conduction. They have concluded that the electron exchange interaction $Fe^{2+} \rightarrow Fe^{3+}$ results in local displacement of electrons in the direction of an electric field, determining the polarization of ferrites. In normal dielectric behavior, ε'decreases with increase in frequency and beyond a certain frequency of electric field, the electron exchange does not follow the alternating field. This might be the reason for ε'showing not much dispersion Ge series. Similar result has been reported by Patil [9].

Fig. 2 show variation of ε' with frequency at different temperatures. ε' decreases with increase in frequency in all samples. It is observed that as temperature increases, there is increase in ε' in general, and the tendency of dispersion in ε' also increases with temperature. This is to be attributed to the thermal activation of electron exchange wherein at higher temperature thermal activation would result in larger dielectric polarization 0and larger value of tan δ [4].

In fig. 3 variation of loss tangent (tan δ) as a function of frequency for all the samples are shown. It is seen that tan δ exhibits dispersion in the frequency range of the experiment. In general tan δ decreases with increase in frequency, showing a peak for some compositions at certain frequencies and then exhibiting decreasing trend.. In Ti series samples with x = 0.1 show small peak at 30 KHz and second peak is observed around 800 KHz.

This peaking behavior can be explained with the help of the relation ,

$\tan\delta = \sigma / \omega\varepsilon'\varepsilon_0 = 1/ \omega\varepsilon'\varepsilon_0\rho$

where σ Ω^{-1} cm^{-1} is the conductivity. ω is the angular frequency corresponding to maximum value of tan δ.

With the increase of frequency (ω), both ε'and ρ decrease, while ε_0 remains constant. Thus the decrease of tan δ is contributed more by increase of ω .The increase of tanδ before the peak appears, could be due to rapid decrease of ε'and ρ with the increase of frequency. The change in the relaxation frequency may most probably be related to change of activation energies of the conduction process, which in turn may stem from changing band gap in these heavily substituted ferrites.

Fig. 4 shows frequency dependence of tan δ at various temperatures. The abnormal behavior of dielectric relaxation processes is observed. The relaxation peaks were shifted to higher frequency on increasing temperature. This behavior is observed in all samples for the compositions of all the samples. The relaxation in dielectric loss tan δ has been observed previously for Mn-Mg ferrites [18], Li-Ni ferrites [19], Co-Zn ferrites [20] and Li-Zn ferrites [21] .The increase in relaxation frequency with increasing temperature is attributed to thermal activation of electron transport, responsible for dielectric behavior.

In fig. 5, temperature dependence of dielectric constant ε' at 100 KHz for Ti^{4+} series is shown. It is observed that the dielectric constant ε' increases with temperature, reaches a maximum value at about 450 K and then decreases. The ε'_{max} decreases with increase of Ti^{4+} content and accordingly T_{max} also decreases marginally.

The plots of a.c. conductivity with temperature for $Cu_{1.1}Ti_{0.1}Fe_{1.8}O_4$, is shown in fig.6 From these plots it can be seen that the σ_{ac} increase slowly up to 350 k, and then the frequency dependence increases rapidly. The conductivity is found to be high for higher frequency. This conduction can be attributed to localized charge carriers. According to localized model the electronics are strongly localized on cations. Theoretical work by several over the years has provided some understanding of conduction in oxides and transition metal compounds. For these materials, the interaction between electrons and optical phonons is strong and conduction is explained on the basis of polarons.The treatment of conduction by polarons is discussed by several workers [17, 22,26]. Polarons belonging to two categories, large and small polarons. In the large polaron model, the conductivity is by band mechanism at all temperatures and the a.c. conductivity decreases with frequency. The small polaron conduct in band like manner up to a certain temperature, and the conductivity increases with frequency. At higher temperature, the conduction is by thermally activated hopping mechanism. The localization may be attributed to electron-phonon interaction or strong magnetic interaction between carriers and magnetic sub – lattice [23]. An additional localization Fe^{2+} ions may arise from inhomogeneous distribution of ions over octahedral and tetrahedral sites in spinel lattice' The experimental results in present case also shows a trend expected for small polaron conduction.

Table 1: Compositional Variation of Dielectric Constant (ε') and DC Resistivity (ρ_{dc}) at room temperature for series $Cu_{1+x}Ti_xFe_{2-2x}O_4$.

Content x	ε' 100 Hz	ε' 1 KHz	ρ_{dc} KΩ
0.05	1457	762	9
0.1	982	583	16
0.15	843	560	20
0.2	791	551	22
0.3	508	308	24

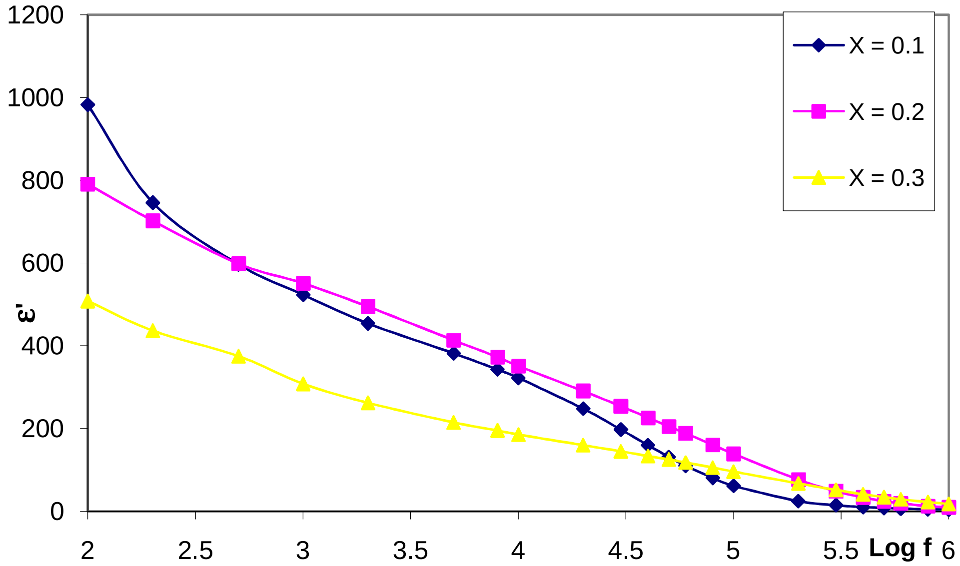

Fig: 1 Variation of dielectric constant(e')with frquency For Cu_{1+x} Ti_x Fe2-2_x O4 system.

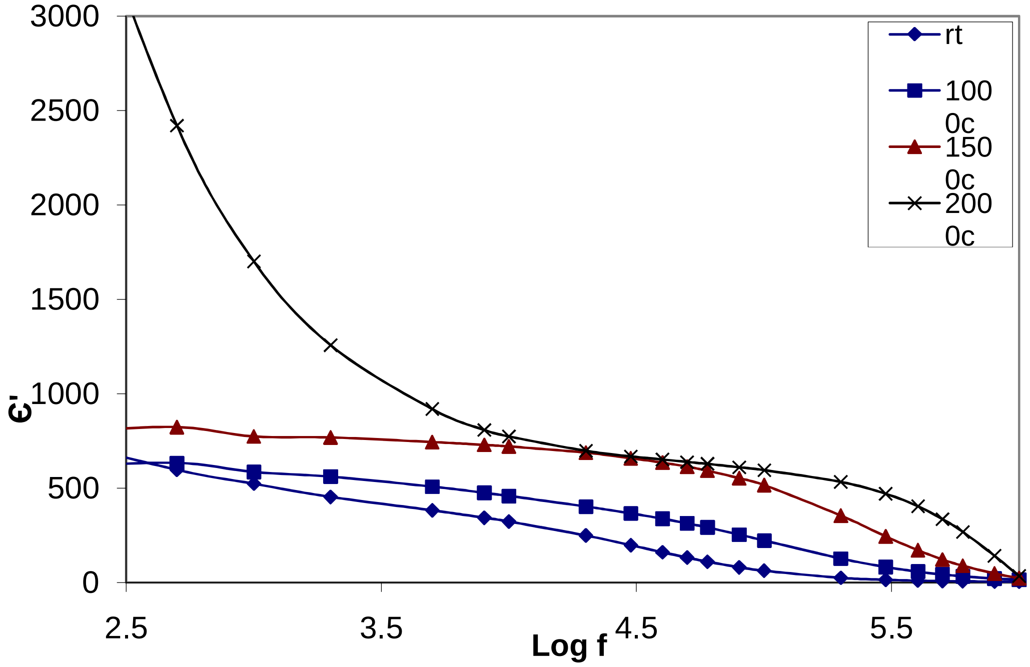

Fig: 2-Frequency dependance of Dielectric constant (e') at various temperatures for Ti.1 sample.

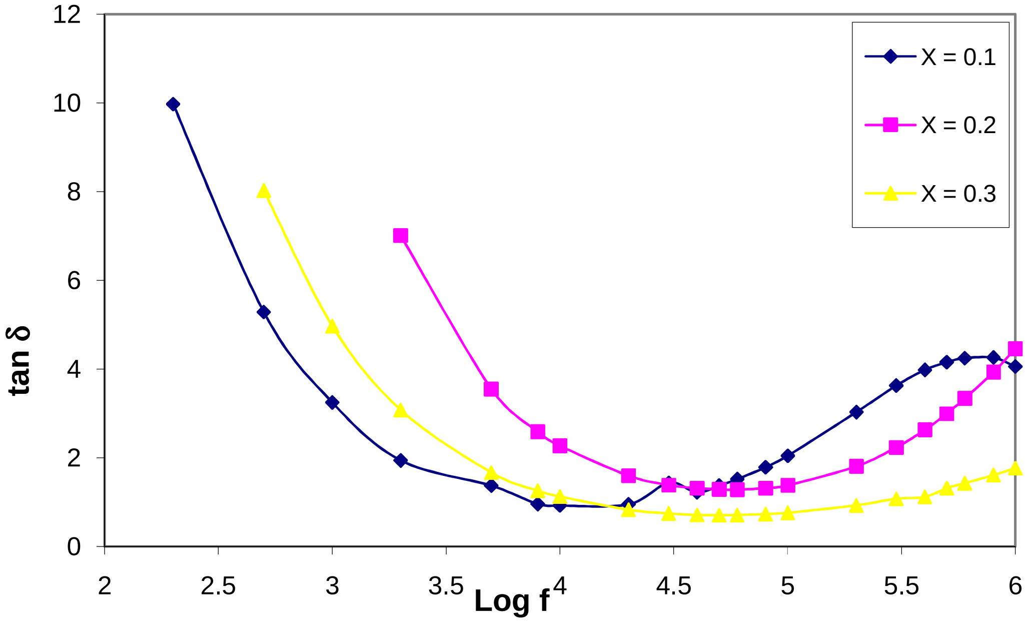

Fig:3-Variation of Loss tangent (tan d) with frequency for Cu_{1+x} Ti_x Fe_{2-2x} O4 system.

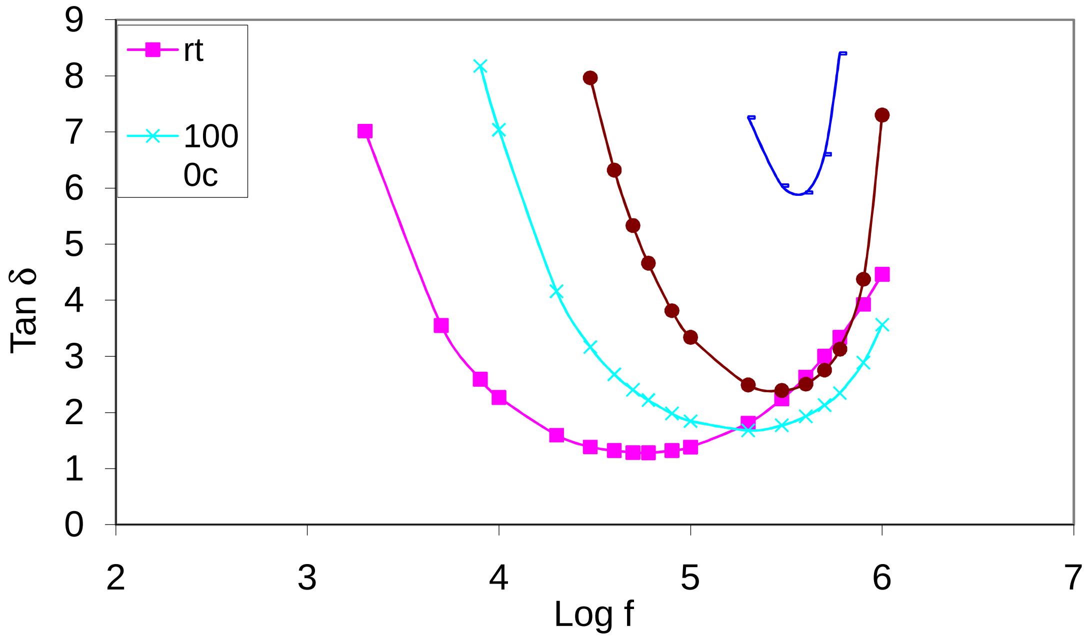

Figure 4- Frequency dependence of Dielectric loss tan δ at various Temperature for Ti 0.2 sample

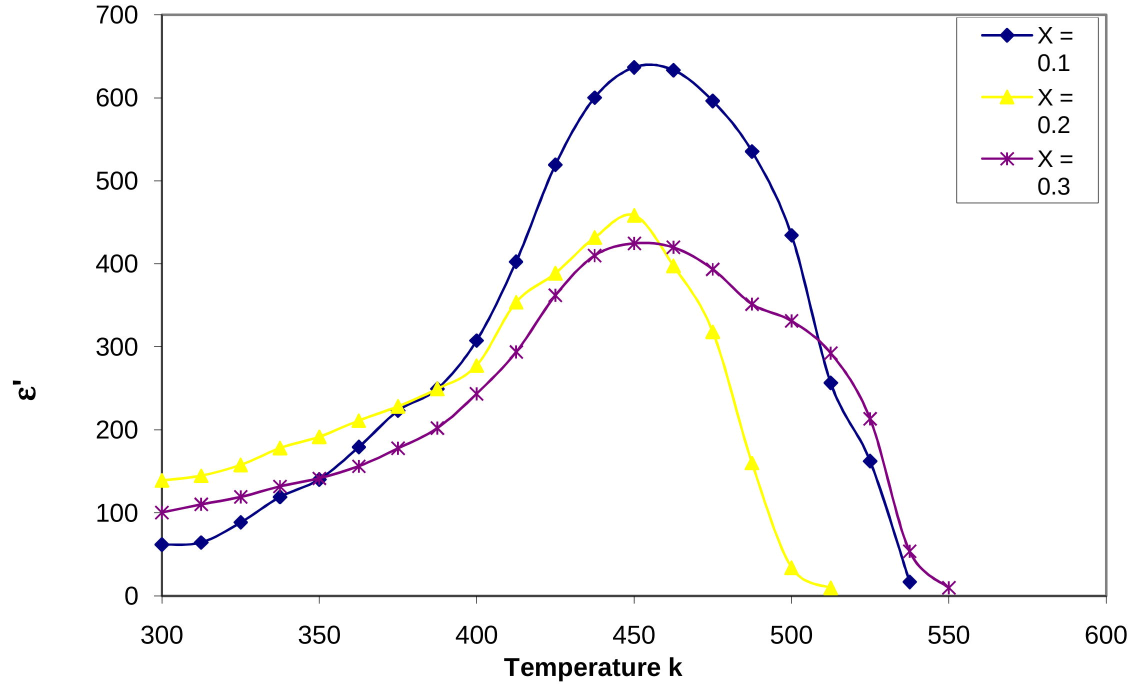

Fig5.-Temperature dependace of dielectric constant (e')with x for Ti series at 100 KHz.

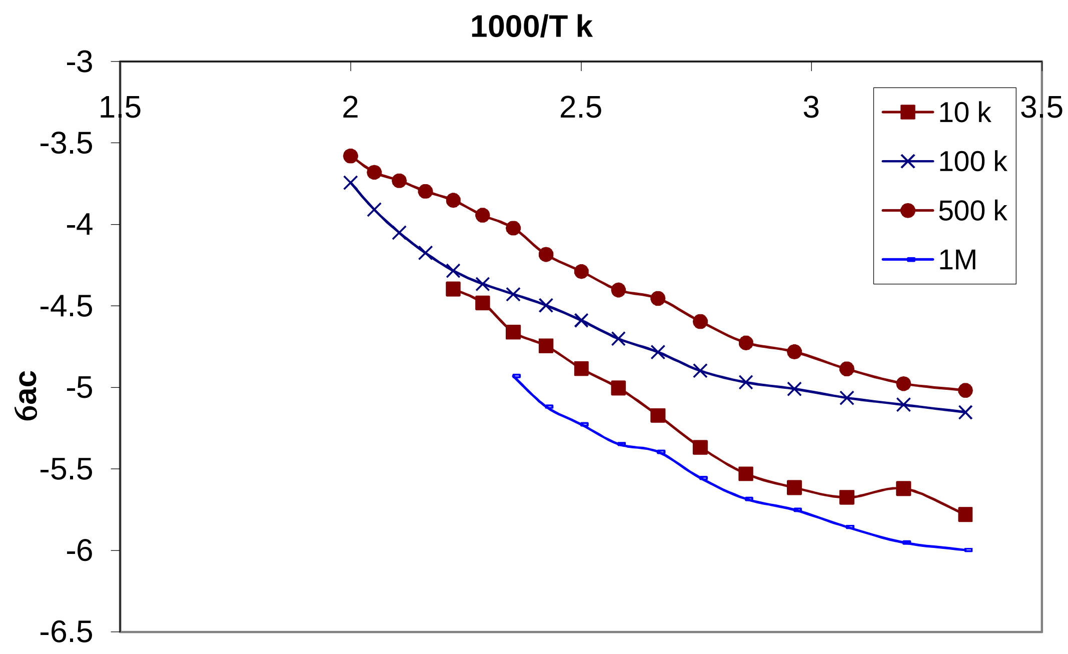

Fig:6-Thermal Variation of ac conductivity (6ac)at different frequencies for Ti.1sample.

CONCLUSION

- Real dielectric constant ε’ decreases and dielectric loss tangent tan δ decreases as the frequency increases which is normal dielectric behavior in ferrites could be explained on the basis of Koop’s theory.

- ε'and tan δ and dielectric relaxation frequency found to increase on increasing temperatures. This could be related thermal activation of the hoping frequency of localized electronic charge carriers.
- Abnormal dielectric behavior (peaks) was observed on tan δ curves at relatively high temperatures. These relaxation peaks take place when the jumping frequency of localized electrons between Fe^{2+} and Fe^{3+} polarization are similar. They could be explained on the basis of hoping conduction mechanism.
- The ac conductivity is found to be high for higher frequency. This conduction can be attributed to localized charge carriers. The experimental result in present case also shows a trend expected for small polaron conduction.

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