

# *Chapter-III*

---

---

## **Electrical Properties**

---

---

## CHAPTER III

# ELECTRICAL PROPERTIES

### PART A

## D.C. CONDUCTIVITY

#### 3.A.1 Introduction:

The mechanism of charge transport can be understood from the measurement of electrical conductivity, hall coefficient, thermoelectric power and magnetoresistance. The electrical conductivity of ferrites at room temperature varies from  $10^{-5}$  to  $10^{11}$  ohm<sup>-1</sup>-cm<sup>-1</sup>. The conduction mechanism in ferrite is quite different from that in semiconductors. In ferrites the temperature dependence of mobility affects the conductivity and the carrier concentration is almost unaffected by temperature variation.

Unlike semiconductors wherein the charge carriers occupy energy band, the charge carriers in ferrites are localized at the magnetic ions. In ferrites, the cations are surrounded by close-packed oxygen anions and as a first approximation can well be treated as isolated from each other. There will be an overlap of the anion charge cloud orbitals. Alternatively the electrons associated with particular ion will largely remain isolated

and hence a localized electron model is more appropriate in the case of ferrites rather than the collective electron model. This accounts for the insulating nature of ferrites.

Wang et al. [1] have measured the electrical resistivity of polycrystalline as well as single crystal lithium ferrite in the temperature range from 77 K to 300 K and found that  $\log \rho$  is a linear function of  $1/T$ . They found that

- a. The most effective process in raising the samples resistivity is the oxidation process.
- b. The ordered sample has a higher resistivity than the sample in the disordered state.
- c. Oxidation of the sample decreases the thermoelectric power.
- d. Lithium ferrite has low mobility.

They have attributed conduction mechanism to the residual  $\text{Fe}^{2+}$  ions. Austin and Mott [2] have attributed the conductivity of ferrite to polaron hopping.

The small activation energy values for the electronic conduction and the linear relation between  $\log \rho$  vs.  $1/T$  indicates the electron hopping motion between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$ . The resistivities, as low as 1/100 ohm-cm are found in iron- rich ferrites [3]. Suitable preparation techniques and chemical composition control the conductivity in the ferrite [4]. The frequency dependent dielectric behavior has created

considerable interest in the understanding of physical and chemical properties of ferrites [5-8].

In ferrites, the electron concentration is of the order of  $10^{22} / \text{cm}^3$ . The low mobility of electrons ( $10^{-4} \text{ cm}^2 / \text{V. sec}$ ) and holes ( $10^{-8} \text{ cm}^2 / \text{V. sec}$ ) limits their conductivity. The electrons in the crystal are wholly localized on specific cations. Therefore Bloch type wave function approach does not suit to explain the motion of electrons. The conduction in the ferrite is due to the exchange of 3d electrons, localized at the metal ions.

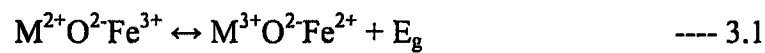
Jonker [9] has derived an expression for mobility from hopping conduction model, based on electron transition between localized cells. Small polaron model has been introduced by Haubenreisser [10].

Distribution of cations in the lattice, nonmagnetic and magnetic substitutions, amount of  $\text{Fe}^{2+}$  ions, sintering conditions, grain size and porosity etc. affects the electrical properties of ferrites. The electrical properties of nickel ferrites have been studied by many workers [11-14]. The spin alignment and electrical conduction relationship can be inferred from comparison of conductivity versus temperature studies observed below Curie or Neel temperature with high temperature studies for many oxides. The spontaneous magnetization does not affect the electrical properties. Ferrite exhibits a remarkable change in the slope of  $\log \rho$  versus  $1000/T$  relationship. Murthy and Sobhanadri [15] have

investigated the D.C. conductivity of some Ni-Zn ferrites as a function of temperature from room temperature to 300° C. The ferrites that contain iron in excess show n- type conduction and those with iron deficiency show p-type conduction.

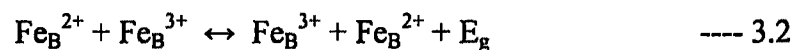
### 3.A.2 Conduction in ferrites

Conductivity of ferrites based on the mechanism of transport phenomenon and may be presented as

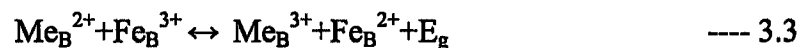


where,  $E_g$  is activation energy.

The high resistivity in ferrites is associated with the occupation of B sites by divalent metal ions and trivalent  $Fe^{3+}$  ions. The extra electron on  $Fe^{2+}$  ion requires very small amount of energy to move towards adjacent  $Fe^{3+}$  ion on the same site. The valence states of two ions get interchanged. Under the influence of an electric field, these extra-electrons constitute the conduction current. Verwey [16] mechanism can be written as



For divalent metal ions



where  $E_g$  is the activation energy, the energy required to transfer the electron from  $Me^{2+}$  to  $Fe^{3+}$  and vice versa. 'Me' denote divalent metal ions like Cu, Co, Mg, Mn, Ni and Cd etc.

The conductivity of ferrites can be explained in terms of electronic charge carriers like electrons and holes by the relation

$$\sigma = e (n_e \mu_e + n_h \mu_h) \quad \text{--- 3.4}$$

where  $n$  and  $\mu$  stand for concentration and mobility of electrons (e) and holes (h) respectively. As electrons moves in one direction, the holes move in opposite direction. Though the velocities of electrons and holes are the same, the mobility of electrons is much higher than the mobility of holes.

The hopping mechanism of conductivity in ferrite is associated with activation energy which can be calculated from  $\log \rho$  Vs  $1/T$  graph. As temperature increases, the conductivity increases. The magnetic state of ferrite changes from ordered ferrimagnetic state to a disordered paramagnetic state. This remarkable state change occurs at Curie temperature.

The lattice scattering increases with increasing amplitude of vibration at higher temperatures. The mobility is proportional to the mean free path between scattering events. Hence conductivity becomes temperature dependent and it is written as,

$$\sigma = \sigma_0 \exp(-\Delta E/kT) \quad \text{--- 3.5}$$

where  $\sigma_0$  = temperature dependent constant

$k$  = Boltzmann constant

Further, in the ferrite there is an interaction between orbital of neighbouring ions and electrons resulting into the polarization of lattice. This effect is more pronounced due to strong Coulomb interaction in ionic crystals such as ferrites. The combination of an electron and its polarization field is called as polaron. Due to such an effect the effective mass of an electron increases as it partially drags the ions along with it. Therefore mobility of electrons decreases and becomes highly temperature dependent.

The mobility is always less than  $1 \text{ cm}^2/\text{V}\cdot\text{sec}$  and is very low. With the help of small polarons the conductivity is related to  $E_g$ ,  $E_p$  and  $T$  as

$$\sigma = n\mu \exp(-E_g/kT) \exp(-E_p/kT) \quad \text{--- 3.6}$$

Due to polarization, the electrons are trapped at the potential well. When such a potential well is deep enough, the electron may be trapped at lattice site. Therefore its transition to neighbouring site may be governed by thermal activation. Hence due to thermal activation lattice is set into periodic vibrations which are quantized into photon.

At higher temperatures electrons jump from one site to another by thermal activation. The hopping mechanism contributes to conductivity at high temperature.

The AC conductivity ( $\sigma_{ac}$ ) is related to the dielectric relaxation caused by localized electric charge. The a. c. conductivity data was

calculated from dielectric constant and loss tangent measured using the relation

$$\sigma_{ac} = \epsilon' \epsilon_0 \omega \tan \delta \quad \text{---- 3.7}$$

where  $\epsilon'$ - the real dielectric constant

$\epsilon_0$ - the permittivity of free space

$\tan \delta$ - the loss tangent

$\omega$  - the angular frequency. (i.e.  $\omega = 2 \pi f$ )

The frequency response of these parameters ( $\epsilon'$ ,  $\epsilon_0$  and  $\sigma_{ac}$ ) are generally known as relaxation spectra or dispersion curves which characterize the materials.

### 3.A.3 Experimental techniques

The D C resistivity of sample was measured by two-probe method with the variation of temperature from room temperature to 550 °C. Silver paste was applied on either side of the pellets to give good ohmic contacts.

The resistivity was determined by the relation

$$\rho = \pi r^2 v / t I \quad \text{---- 3.8}$$

where t - thickness of pellet

r - radius of pellet

I - current through the sample



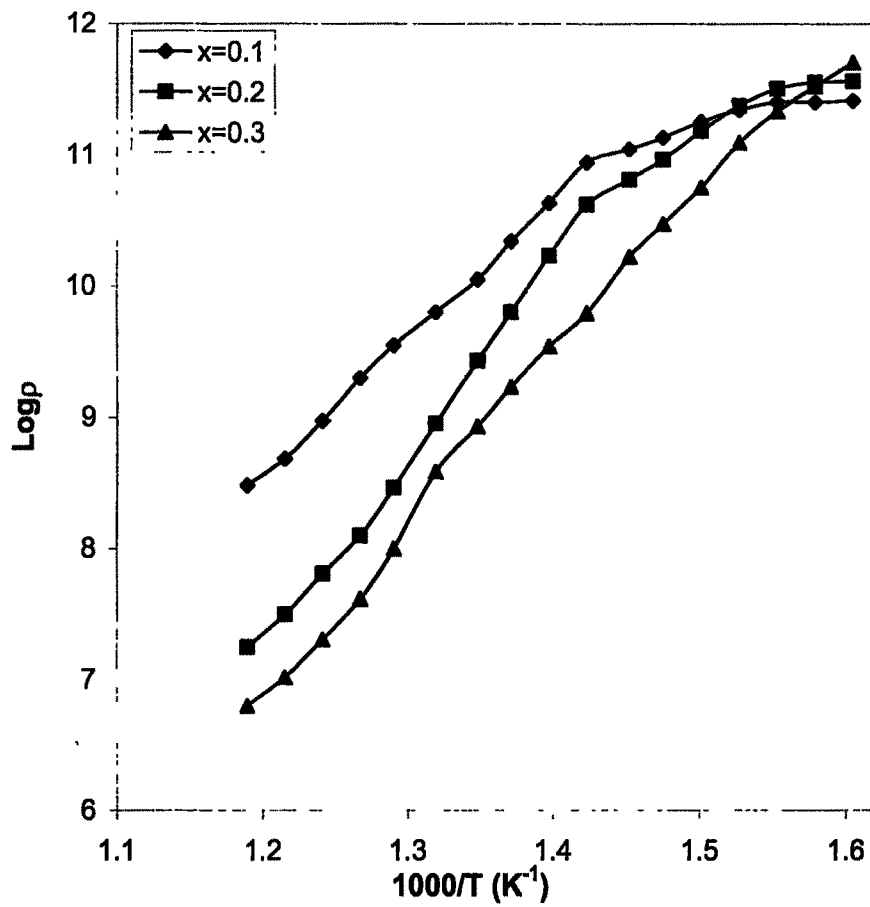
### 3.A.4. Results and discussion

The plots of  $\log \rho$  versus  $1000/T \text{ K}^{-1}$  for ferrite sample  $\text{Li}_{0.5}\text{Ni}_{1.5x}\text{Fe}_{2.5-x}\text{O}_4$  (where  $x = 0.1, 0.2, 0.3, 0.4$  and  $0.5$ ) are shown in Fig.3.A.1 and Fig. 3.A.2. From the plots it is observed that as the temperature increases the resistivity decreases linearly and shows a transition near Curie temperature. At this temperature the material changes from ferrimagnetic state to paramagnetic state. The change in slope is observed in all the ferrites. Similar results were reported by Bandyopadhyay and Fulrath [17].

The activation energies corresponding to paramagnetic and ferromagnetic regions were calculated from the plots of  $\log \rho$  versus  $1000/T$ . The data on DC resistivity and activation energy at paramagnetic and ferrimagnetic region is given in Table 3.A.1. This result is in agreement with the theory of Irkhin and Turov [18]. It is observed that the activation energy in paramagnetic region is greater than ferrimagnetic region.

The observed discontinuities in the plot of  $\log \rho$  versus  $1000/T$  have been attributed to several reasons. Irkhin [18] has stated that the change in slope of straight line occurs while passing through the Curie point. The separation of these two regions i.e. paramagnetic and ferrimagnetic regions occurs as the spin-spin interaction vanishes due to

thermal vibrations. Similar result is reported by Komar and Klivshin [19] in case of other ferrites.



**Fig. 3.A.1 - Variation of dc resistivity with temperature for  $\text{Li}_{0.5}\text{Ni}_{1.5x}\text{Fe}_{2.5-x}\text{O}_4$  ferrite**

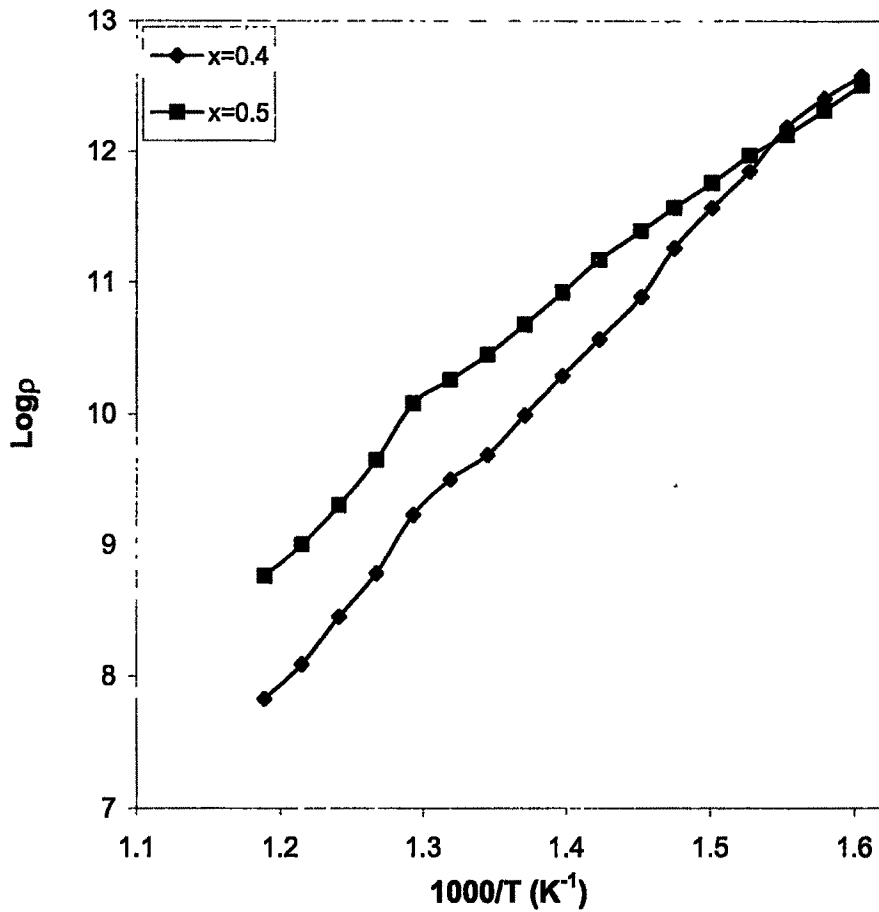


Fig. 3.A.2 - Variation of dc resistivity with temperature for  $\text{Li}_{0.5}\text{Ni}_{1.5x}\text{Fe}_{2.5-x}\text{O}_4$  ferrite

The electrical conductivity in Li-Ni Ferrites has been studied by Reddy et al. [20] and observed a transition at Curie temperature. Many researchers have reported the discontinuities in mixed nickel ferrites. These discontinuities shift with respect to Curie temperature of the material [21-23]. The electrical conductivity in ferrite is due to hopping of electrons from  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  ions on crystallographically equivalent site i.e. B-site. The lowering of activation energy is attributed to the effect of spin ordering. In other words, change in slope at Curie temperature is mainly due to spin ordering of electrons.

In the present case, as nickel content increases the room temperature DC resistivity increases. The Curie temperature at the transition of the ferrimagnetic and paramagnetic state is reduced as compared to the results of Reddy et al [20]. However the result is similar to the results reported by Chinnasamy [24]. Ponpandian et al.[25] have reported that  $\text{NiFe}_2\text{O}_4$  in the nanocrystalline form exhibits a mixed spinel structure with  $\text{Ni}^{2+}$  ions occupying both A and B sites, but the bulk  $\text{NiFe}_2\text{O}_4$  is a well known inverse spinel, with  $\text{Ni}^{2+}$  ions occupying only the B-sites.

Song et al. [26] have observed two regions of different slopes and is attributed the change to the change in conduction mechanism in case of substituted Li ferrites. Similar results were observed by Rane et al. [27]. The increase in electrical conductivity with increasing dopant

concentration has been attributed to decrease in the values of charge carrier mobility rather than to the carrier concentration. Satter [28] and Ravindra et al. [29] have observed similar variation in resistivity with change in slope at Curie temperature.

Many workers have reported three regions of conductivity [30-32] of which the first region has been attributed to the presence of impurities, the second region to the phase transition from tetragonal to the cubic and the third to magnetic ordering change.

The activation energy in the present case varies from 0.35 eV to 0.86 eV in the ferromagnetic region. For electron hopping the activation energy is less than 0.2 eV and for polaron hopping it is greater than 0.2 eV. In the present case the activation energies are greater than 0.2 eV indicating that the conduction is due to polaron hopping [33]. Similar value of activation energy has been reported by Mallpur [34]. Reduction of particle size may cause the change in resistivity of the samples and hence the activation energy is lower in both the regions.

It is seen that the Curie temperatures ( $T_c$ ) decreases with  $Ni^{2+}$  content. The Curie temperature mainly depends upon the strength of A-B interaction. This increase of Curie temperature attributes to strong A-B interaction. Similar results are observed in variation of saturation magnetization.

Table 3.A.1- Activation Energy and Curie temperature data for  $\text{Li}_{0.5}\text{Ni}_{1.5-x}\text{Fe}_{2.5-x}\text{O}_4$  ferrites

x	Curie Temperature. (T <sub>c</sub> ) °C	$\Delta E_{\text{ferr}}$ (eV)	$\Delta E_{\text{para}}$ (eV)
0.1	435	0.35	0.72
0.2	435	0.51	0.81
0.3	487	0.78	0.94
0.4	500	0.86	0.94
0.5	526	0.68	0.81

Table 3.A.2 - Room temperature D.C. Resistivity data

x	Resistivity $\rho_{\text{dc}} \times 10^5 \Omega \text{ cm}$
0.1	1.95
0.2	4.94
0.3	7.76
0.4	16.45
0.5	14.37

## **PART B**

# **Dielectric Properties**

### **3.B.1 Introduction**

The polycrystalline ferrites are very good dielectric materials. The dielectric properties of ferrites basically depend on several factors such as chemical composition, method of preparation, grain structure, additives, sintering temperature, sintering atmosphere etc. [35-37]. It was proposed that [38, 39] air sintered ferrites are characterized by relatively high conductive grains separated by high resistive grain boundaries. It has been pointed out that the resistivity and dielectric constant of polycrystalline ferrites exhibit dispersion with respect to frequency. As a consequence the divalent metal ions produced in the bulk. Since high conductivity grains separated by low conductivity layers so that the ferrites behave as an inhomogeneous dielectric structure.

Most of the high frequency applications related to electrical materials are concerned with dielectrics. The advantages of ferrites which are ceramic in nature over other available dielectric materials are elastic properties, temperature performance and greater resistance to environmental changes particularly at high temperature and their ability to form gas tight seals with metals [40].

### 3.B.2 Polarization in dielectrics

Jonscher [41] has interpreted the mechanism of polarization in ceramic materials that includes ferrites.

When the electric field is applied to a dielectric material the positive and negative charges within the material are displaced relative to one another. Hence the material has an electric dipole moment. The dipole moment per unit volume is called as polarization (P). The total polarization of the dielectric can be represented as the sum of the contribution of these mechanisms.

$$P = P_e + P_i + P_o + P_s \quad \text{---- 3.9}$$

where,

$P_e$  - Electronic polarization

$P_i$  - Ionic polarization

$P_o$  - Orientation polarization

$P_s$  - Space charge polarization.

In electronic polarization the center of gravity of electron is shifted relative to nucleus in an electric field. In ionic polarization there is displacement of positive and negative ions relative to one another. In orientational polarization electric dipoles orient by the application of an external field. The space charge also tends to turn its own polarization in the case of ferrites.

If the alternating field represented by,



$$E = E_0 e^{i\omega t} \quad \text{--- 3.10}$$

is applied to the ferrites, the polarization is not instantaneous. The time required for polarization shows up a phase retardation of charging current. Therefore instead of  $90^\circ$ , it advances only by  $(90-\delta)$ . Therefore, the dielectric displacement is given by,

$$D = D_0 e^{i\omega(t)} \quad \text{--- 3.11}$$

$$D = \epsilon^* \epsilon \quad \text{--- 3.12}$$

$$\text{where } \epsilon^* = \epsilon' - j \epsilon'' \quad \text{--- 3.13}$$

$\epsilon'$  - Real part of dielectric constant

$\epsilon''$  - Complex part of dielectric constant

For small values of phase angle, the loss is represented as,

$$\tan \delta = \epsilon'' / \epsilon' \quad \text{--- 3.14}$$

The phase shift arises due to the time lag between applied voltages and induced current. The energy is dissipated in the ferrites which require charge carrier migration.

If the period of the alternating field is more than relaxation time, then the dipole will not be able to follow the field. Debye stated the real and complex dielectric constant as to study the dielectric relaxation and is given as,

$$\epsilon' = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 + \omega^2 \tau^2} \quad \text{--- 3.15}$$

$$\varepsilon'' = \frac{(\varepsilon_s - \varepsilon_\infty)\omega\tau}{1 + \omega^2\tau^2} \quad \text{---- 3.16}$$

where  $\varepsilon_s$  - static (low frequency) dielectric constant

$\varepsilon_\infty$  - high frequency dielectric constant

Hence the loss angle  $\tan \delta$  can be written as,

$$\tan \delta = \varepsilon'' / \varepsilon'$$

$$\tan \delta = \frac{(\varepsilon_s - \varepsilon_\infty)\omega\tau}{\varepsilon_s + \varepsilon_\infty + \omega^2\tau^2} \quad \text{---- 3.17}$$

The loss tangent ( $\tan \delta$ ) is defined as the ratio of the loss or resistive current to the charging current in the sample. Also it is known that, there is a strong correlation between the conduction mechanism and dielectric constant (the polarization mechanism) in ferrites. From these considerations it is expected that  $\tan \delta$  decreases with increasing frequency.

### 3.B.3 Experimental techniques

The dielectric measurements were carried out using two probe method with the help of LCR meter bridge (HP 4284 A) in the frequency range 20 Hz to 1 MHz. The ferrite pellets were pelletized to about 15 mm in diameter and 2 - 3 mm in thickness. The Pellets were silvered pasted for good ohmic contact and were kept in the sample holder firmly. The values of capacitance ( $C_p$ ) and loss tangent ( $\tan \delta$ ) were noted directly for different frequencies at room temperature.

The dielectric constant was calculated by using the relation,

$$\epsilon' = (Ct/\epsilon_0 A) \quad \text{---- 3.18}$$

where, C = the capacitance

t = the thickness of the pellet

$\epsilon_0$  = Permittivity of free space

A = Area of cross section of Pellet

### 3.B.4. Results and discussion

The variation of dielectric constant ( $\epsilon'$ ) and loss angle ( $\tan \delta$ ) as a function of frequency for the ferrite samples in the system  $\text{Li}_{0.5}\text{Ni}_{1.5x}\text{Fe}_{2.5-x}\text{O}_4$  with  $x = 0.1, 0.2, 0.3, 0.4$  and  $0.5$  is shown in Fig. 3.B.1. A comparative study of the dispersion curves of these samples shows that the value of ( $\epsilon'$ ) at lower frequency of the applied field is larger than that at higher frequency. The dielectric constant decreases with increase of frequency in a certain range and thereafter remains constant beyond a certain frequency for all the compositions. Similar results were observed in many ferrites and this is explained by Koop's phenomenological theory.

The dielectric constant ( $\epsilon'$ ) at room temperature also depends upon the divalent content. In the present work, the decrease of ( $\epsilon'$ ) shows a marked preference of  $\text{Ni}^{2+}$  ions to the octahedral sites. Therefore with the increase of  $\text{Ni}^{2+}$  content the ferric ions migrate to the tetrahedral sites causing of the hopping charge carriers in octahedral sites, which lead to

decrease in dielectric constant values. The relative high values of the dielectric constant at low frequencies are due to the interfacial polarization ensuring that the samples can be fairly considered as formed of well conducting grains and poorly conducting grain boundaries in agreement with Koops model. Similar results are reported by El Kony et al. [42].

Reizlescu et al. [43, 44] have explained the dispersion of  $\epsilon$  by considering the multivalence ions such as  $\text{Fe}^{2+} \leftrightarrow \text{Fe}^{3+}$ ,  $\text{Cu}^{1+} \leftrightarrow \text{Cu}^{2+}$  (i.e. the contribution to polarization is due to n and p carriers). According to their model, local displacement of carriers takes part in the polarization in an opposite direction to that of external field. The contribution of p-type carries is lower than that by electron exchange  $\text{Fe}^3 \leftrightarrow \text{Fe}^{2+}$  and it has an opposite sign. In present case  $\text{Ni}^{2+} \leftrightarrow \text{Ni}^{3+}$ ,  $\text{Fe}^{3+} \leftrightarrow \text{Fe}^{2+}$  dipoles may be visualized as exchange of electrons. Formation of  $\text{Ni}^{3+}$  acts as carries and  $\text{Li}^{1+} + \text{Fe}^{2+}$  forms a stable bond which again helps in decreasing the dielectric constant. The variation of  $\tan \delta$  with frequency for the system under investigation is shown in Fig. 3.B.2. All the compositions show dispersion in  $\tan \delta$  with frequency. These observations are similar to those of  $\epsilon$  with frequency. Therefore the behavior of  $\tan \delta$  with frequency can be explained in the similar manner of  $\epsilon$  variation with frequency.

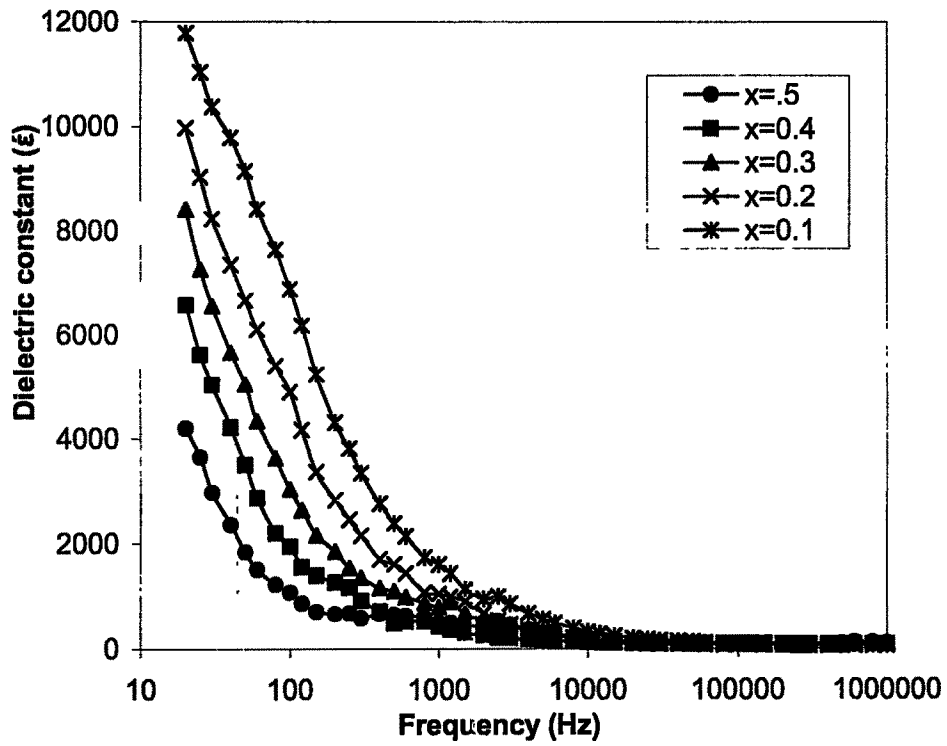


Fig. 3.B.1 - Variation of dielectric constant with frequency for  $\text{Li}_{0.5}\text{Ni}_{1.5x}\text{Fe}_{2.5-x}\text{O}_4$  ferrite

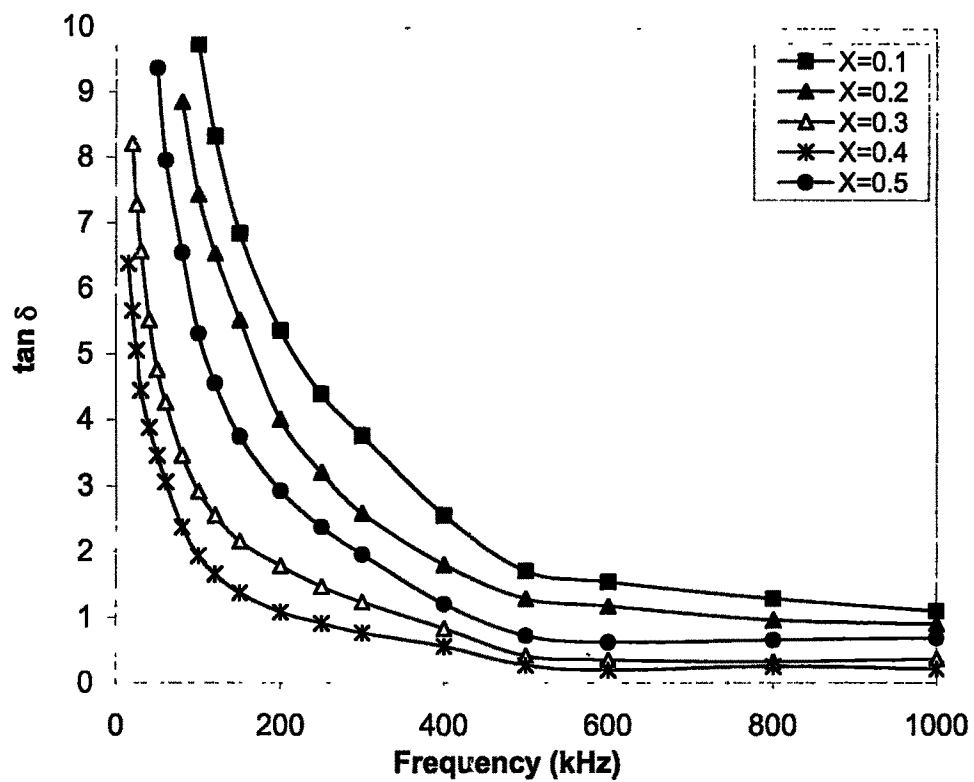
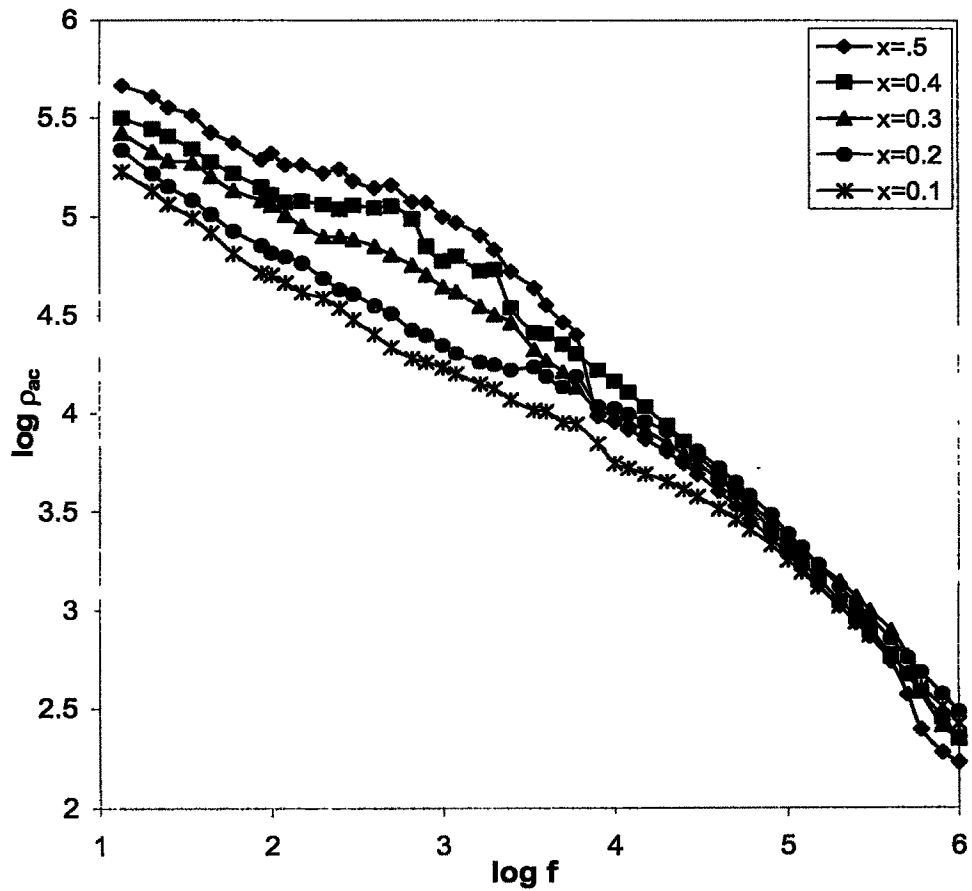


Fig. 3.B.2 - Variation of dielectric loss with frequency for  $\text{Li}_{0.5}\text{Ni}_{1.5x}\text{Fe}_{2.5-x}\text{O}_4$  ferrite



**Fig.3.B.3** Variation of ac resistivity with frequency for  $\text{Li}_{0.5}\text{Ni}_{1.5x}\text{Fe}_{2.5-x}\text{O}_4$  ferrites

The number of  $\text{Ni}^{2+}$  ions displaced from the B-site to the A-site increases when the grain size is reduced. This leads to a decrease in the number of  $\text{Ni}^{3+} \leftrightarrow \text{Ni}^{2+}$  pairs and hence number of available p-type carriers on the B-site. The same mechanism also explains the decrease of  $\epsilon$  with grain size reduction. The same result is reported by Ponpandian [25] for  $\text{Ni}_2\text{Fe}_2\text{O}_4$  ferrites.

The frequency dependent dielectric behavior with variation of temperature has been reported by Manglaraja [32].

The variation of AC resistivity ( $\rho_{ac}$ ) with frequency is shown in Fig. 3.B.3. All the samples show decrease in  $\rho_{ac}$  with increase in frequency which is normal behaviour of ferrites. The measurement of AC resistivity is carried out by Guyot et al. [46]. The dispersion of AC resistivity is discussed by Koops theory. Similar behavior has been reported in case of other ferrites [47]. Mallpur [34] has reported similar results for Ni-Mg-Zn nanocrystalline ferrites. Decrease in  $\rho_{ac}$  with increase in frequency is due to hopping of charge carriers.



**References**

- 1 F. F. Y. Wang, R. L. Gravel and M. Kestigian  
IEEE Trans. 103(1981) 665
- 2 I. G. Austin and N.F. Mott  
Adv. Phys. 18 (1969) 41
- 3 N. Miyata  
Jpn. J. Phys. Soc. 16 (1961) 206
- 4 L. Donald  
Proc. I.R.E. 44 (1956) 1303
- 5 L. G. Van Uitert  
Proc. I.R.E. 44 (1956) 1294
- 6 K. Iwauchi  
Jpn. J. Appl. Phys. 10 (1971) 1520
- 7 A. R. Van Hippel  
"Dielectric Materials and Applications"  
Chapman and Hall Ltd. London (1954)
- 8 J. Peter and K. J. Standley  
Proc. Phys. Soc. 71 (1958) 131
- 9 G. H. Jonker  
J. Phys. Chem. Solids 9 (1959) 165
- 10 W. Haubenreisser  
Phys. Status Solidi 21 (1961) 3905
- 11 C. Guillaud and R. Bertrand  
J. Des. Res. Cont. Mat. Res. Scientific 3 (1950) 73
- 12 L. G. Van Uitert  
J. Chem. Phys. a 61 (1950) 256
- 13 L. G. Van Uitert  
J. Chem. Phys. 23(1956) 1883

- 14 K. N. Subrahmanyam and S. A. Sweden  
Phys. Status Solidi A 61 (1980) 59
- 15 V. R. K. Murthy and J. Sobhanadri  
J. Mater. Sci. 36 (1976) K 133
- 16 E. J. W. Verway, F. deBoer and Vasanten  
J. Chem. Phys. 161 (1948) 1091
- 17 G. Bandyopadhyay and R. M. Fulrath  
Am. Chem. Soc. Bull. 54 (1975) 506
- 18 P. Irkhin and E. A. Turov  
Fiz. Met. Metalloved 4 (1957) 582
- 19 A. P. Komer and V. V. Klivshin  
Izv. Akad. Nauk. SSSR Ser. Fiz. 18 (1954) 56
- 20 P. V. Reddy, R. Satyanarayana and T. S. Rao  
Phys. Stat. Solidi (a) 78 (1983) 109
- 21 T. E. Whall, K. K. Young and Y. G. Prokova  
Phil. Mag. B (GB) 54 (1986) 505
- 22 M. A. Ahmed, M. E. El-Nimer, A. Tawfik and A. M. Hasab  
Phys. Stat. Solidi (a) 123 (1991) 501
- 23 J. M. Song and J. G. Koh.  
J. Magn. and Magn. Mater. 152 (1996) 383
- 24 C. N. Chinasamy, A. Narayanasamy, N. Ponpandian, K.  
Chattopadhyay, K. Shinosa, B. Jeyedevan, K. Tohji, K. Nkatsuka, T.  
Furubyashi and Nakatani  
Phys. Rev. B 63 (2001) 184108
- 25 N. Ponpandian, P. Balaya and Narayansamy  
J. Phys. Condens. Matter 14 (2002) 3221-3237
- 26 I. M. Song and J. G. Koh  
J. Magn. and Magn. Mater. 152 (1996) 383

- 27 K. S. Rane, M. S. Vernenkar and P. Y. Sawant  
Bull. Mater. Sci. 5(2001) 323
- 28 A. Sattar  
Egypt. J. Sol. 26 (2003) 113
- 29 D. Ravindra and R. T. Sheshagiri  
Cryst. Res. Technol. 25 (8) (1990) 963
- 30 S. A. Patil, B. L. Patil, S. R. Sawant, A. S. Jambhale and R. N. Patil  
Ibid 31(1992) 904
- 31 M. A. El-Hiti  
J. MJ. Magn. and Magn. Mater. 136(1994) 138
- 32 A. N. Patil, R. P. Mahajan, K. K. Patankar, A. K. Ghatage, V. L. Mathe and S. A. Patil  
Ind. J. Pure Appl. Phys. 38 (2000) 651
- 33 A. G. Rustamov, A. A. Samokhvalov and I. C. Fakidov  
Sovt. Phys. Solid State 5 (1963) 751
- 34 M. M. Mallpur  
M. Phil. Dissertation, Shivaji Univ., Kolhapur (2003)
- 35 E. Rezlescu, N. Rezlescu, P. P. Popa and L. Rezlescu  
Mater. Res. Bull. 33 (6) (1998) 915
- 36 M. Guyot  
J. Magn. and Magn. Mater. 18 (1980) 925
- 37 K. K. Patankar, V. L. Mathe, R. P. Mahajan, S. A. Patil, R. M. Reddy and K.V. Sivakumaar  
Mater. Chem. Phys. 72 (2001) 23
- 38 C. G. Koops  
Phys. Rev. 83 (1951) 121
- 39 R. Dias and A. L. Moreive  
J. Mater. Res. 12 (1998) 2190

- 40 G. P. Kothiyal  
Proc. DAE. BRNS National Symposium (1999) 25
- 41 A. J. Jonscher.  
"Dielectric Relaxation in Solid"  
Chelsea Dielectric Press, London (1963)
- 42 D. El Kony, S.A. Saafan, A. M. Abo and El Ata  
Egypt. J. Sol. 23(1) (2000) 137-146
- 43 N. Rezlescu, S. Istrate, E. Rezlăscu and E. Luca  
J. Phys. Stat. Solid (a) 23 (1974) 575
- 44 N. Rezlescu and E. Rezlescu.  
J. Phys. Chem. Solid 35 (1974)
- 45 R. V. Mangalaraja, S. Anantakumar, P. Manohar, F. D. Gnanam and  
M. Awano  
Mater. Lett. 58 (2004) 1593-96
- 46 M. J. Guyot  
J. Magn. and Magn. Mater. 15-18 (1980) 925
- 47 S. S. Bellad, S. S. Watve and B. K. Chougule  
Mat. Res. Bull. 34 (1999) 1089