CHAPTER - III

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MAGNETIZATION STUDY.

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MAGNETIZATION STUDY

3.1 : Introduction :

The magnetic parameters like saturation magnetization (Ms), coercive field (Hc), remanence ratio. ($\frac{Mr}{Ms}$) and permeability (μ) are related with hysteresis loop. Therefore, hysteresis study for ferrites provide the information regarding all these magnetic parameters. These parameters are the suggestive of the nature of application of ferrites. The ferrites can be classified in to two groups on the basis of coercive force Hc. The ferrites, with low Hc are called as soft ferrites, where as with high Hc are called as hard ferrites. The soft ferrites are used in the manufacture of cores of transformers, motors and generators, high frequency inductance in which high permeability, low coercive force and small hysteresis losses are essential. The hard ferrites are applicable in loud speaker, telephones, TV and other apperatus for which high coercivity and high remanence are desirable. According to N'eel (1949) the coercive force(Hc) is related with the saturation magnetization, the internal stresses, the porosity and anisotropy.⁽¹⁾ Hysteress properties are highly sensitive to crystal structure, heat treatment, chemical constitution, porosity and grain size⁽²⁾ etc.

The saturation magnetization is an important basic magnetic property of ferrite which can be measured by ballestic method,⁽³⁾ the vibrating coil magnetometer^(4,5), vibrating sample

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magnetometer^(6,7) and microwave methods^(8,9). We have been obtained saturation magnetization for slow cooled ferrite system $Cu_x Ni_{1-x}Fe_2O_4$ ($0 \le x \le 1$) from hysteresis loop by using high field loop tracer HS 869 supplied by Electronics Corporation of India.

The theoretical aspects have been discussed in the next section in order to make the discussion of the results in terms of cation distribution and chemical composition, crystal structure etc.

3.2 : Theoretical Aspect :

The spontaneous magnetization was first explained by Weiss (1907)⁽¹⁰⁾ by postulating the existence of molecular field. However, the zero magnetization exhibited by ferromagnetic crystal led to the pridiction of randomly oriented domains. According to Barkhausen⁽¹¹⁾, the change in magnetization is discontinuous for the continuously changing applied field; which lead to argue that magnetization is due to rotations of magnetizations of whole domains. Landu and Lifshitr showed that, domain formation in any ferromagnetic material is a consequence of considerable reduction in magnetostatic energy from that of the saturation magnetization.

The attempt was made to over come limitations (as mentioned in 1.5 a) Weiss molecular field theory by Heisenberg (1928). He gave quantum mechanical treatment to explain the

alignment of moments in terms of exchange interaction between the un-compensated spins of electrons in the partially fielled According to Heisenberg, under certain conditions 3-d shells. the exchange energy produces effects similar to those of Weiss molecular fields. In this case electrons with parallel spins have lower energy than those with anti-parallel alignment. Spontaneous magnetization can also arise as a result of negative exchange interactions under favourable conditions of intervening ions. In this case the neighbouring magnetic moments are antiparallel (N'eel, 1948). This is the origin of spontaneous magnetization in ferrites; in which magnetic moment may be arranged in anti-parallel or some other complex fashion compensating partially. The magnetic moments associated to the magnetization are mainly spin magnetic moments due to the quenching of orbital angular momentum. However, the non-integral values of the magnetic numbers in case of Fe, Ni, Co at OoK could not be explained by the Heisenberg model. The explaination of this fact based on band theory of solids (Stoner, 1933). (14) The exchange energy is given by,

Wex =
$$2 JS^2 \ge Cos \phi_{ij}$$

 $i \neq j$

where S, total spin momentum per atom.

 \emptyset_{j} , the angles between spin momentum vectors of atoms i and j

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Here anisotropy is neglected and only nearest neighbour exchange interactions are considered.

The ferrimagnetic and antiferromagnetic materials had been investigated by extending the basic concepts associated with the explaination of ferromagnetic materials. N'eel developed theory of antiferromagnetism and ferrimagnetism by assuming the two sub-lattice model. Also he suggested the possibility of anti-parallel alignment of spins of nearest neighbours in contrast to the parallel alignment in the ferromagnetism. N'eel's theoretical contribution to the antiferromagnetism and ferrimagnetism was proved experimentally, (15) first by Gorter (1950) and also by Guillaud (1949, 1950, 1951). According to N'eel's two sub-lattice model, antiferromagnetism can be explained by quantum mechanical exchange interactions. (16) The antiferromagnetism may be regarded as a special case of ferrimagnetism in which magnetic moments of two sub-lattices are antiparallel and identical in magnitudes lead to net zero magnetization. In ferrimagnetism, magnetic moments of two sub-lattices are antiparallel but they differ in magnitude lead to net magnetization. The difference in magnitude attributed due to one, different spin magnetic moments are either magnetic ions of sub-lattice and (or), two, different crystallagraphic he sites that may occupied by the magnetic ions. The N'eel theory of ferrimagnetism has been explained in section (1.5 C). Also the structural explaination of magnetization in ferrites has

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been already explained in Section (1.5 b).

Neel theory had been extended by Yafet and Kittel⁽¹⁷⁾ by assuming triangular arrangement of spin, in which A-A and B-B interactions are comparable in magnitude to the A-B interaction. This theory also has been briefly mentioned in Section (1.5 d).

3.3 : Magnetic Anisotropy :

The term "anisotropy" is generally referred to describe "directional dependence: " of some physical properties like elasticity and strength; which are markedly dependent on crystallographic direction. On the similar line the magnetization exhibited by crystals of ferromagnetic metals and the ferrites has directional dependence of magnetization, the term 'magnetic anisotropy' is used. Experimentally⁽¹⁸⁾ it has been observed that, the magnetic materials can very easily magnetized along particular direction than others; known as easy direction of magnetization. Ultimately small value of applied field (consequently less energy) is required for the saturation magnetization along easy direction. The direction along which, it is most difficult to produce saturation is known as hard direction. Thus, the hard directions or axes of magnetization require the largest field or energy for achieving magnetic saturation.

The anisotropy energy (magnetocrystalline energy) is the difference between the energy required to magnetize the samples

to saturation along the hard direction and that required along an easy direction. The physical origin of this energy is spin orbit interaction. This energy is dependent upon orientation of magnetization with respect to crystallographic axis. For ferrites which are cubic crystals, the anisotropy energy is given by

 $\mathbb{E}_{\Lambda} = \mathbb{K}_{1} \left(\alpha_{1}^{2} \alpha_{2}^{2} + \alpha_{2}^{2} \alpha_{3}^{2} + \alpha_{1}^{2} \alpha_{3}^{2} \right) + \mathbb{K}_{2} \left(\alpha_{1}^{2} \alpha_{2}^{2} \alpha_{3}^{2} \right),$ where \checkmark denotes direction-cosines and K₁ and K₂ denote anisotrppy constant. The magnitude of K1 and K2 measures strength of anisotropy. The K1 and K2 changes from material to material and are temperature dependent, they also vary with structure and kind of magnetic ion. The electronic structure of magnetic ion contributes to anisotropy.⁽¹⁹⁾ Magnetic anisotropy may also arise due to stress and shape anisotropy. The stress anisotropy may be reduced by slow cooling of sample. This can be effectively reduced by incorporation of Fe²⁺ in small amount in the crystal such as it reduces the magnetostriction to zero. Shape anisotropy may be reduced by perparing ferrites in such a way as voids are spherical and few in number, with the help of prolonged sintering.⁽²⁰⁾ An anisotropy plays an important role in determining the properties like, hysteresis, permeability and magnetostriction. (21)

3.4 : <u>Magnetostriction</u> :

The dimensional change of the crystal as a function of magnetization is known as magnetostriction. The increase in

crystal energy due to interaction of the magnetization and the strain of the crystal; which is generally dependent upon the direction of magnetization and the crystalline axis is known as magneto crystalline (magneto elastic) energy. Due to the magnetostriction effective anisotropy energy changes. The ferrite exhibit magnetostriction.

3.5 Magnetic Domain :

Some magnetic materials exhibit zero magnetization in absence of applied magnetic field. However, the hypothesis of the internal molecular fields concluded that the whole specimen would be magnetized to saturation in one direction. The contrast between observed fact and hypothesis were put into end by Weiss after introducing the concept of domain. He suggested that the specimen is composed of smaller regions each of which is magnetized to saturation and these are randomly oriented with respect to each other, would led to zero magnetization. These tiny regions are known as magnetic domains. The concept of domain is one of the most important features of modern magnetic Theory.

In the polycrystalline samples domain patterns are irregular due to influence of crystalline properties. Where as in single crystal, the domains are more regularly arranged. The net magnetization exhibited in presence of small applied magnetic field is a consequence of orientation of direction of component of each magnetic domain in the direction of applied

field. In the magnetization process two mechanisms are involved, one, domain boundary displacement and second, domain rotation which is due to rotation of direction of magnetic dipoles simultaneously within an entire domain. The domain formation is a consequence of the process which minimizes the magnetic energy of the system which is in the four different forms, exchange energy, the anisotropic energy, magnetostriction energy and the magnetostatic energy.

The change in the direction of antiparallel spins in the neighbouring domains takes place via domain wall having thickness of few hundred of inter-atomic distances. Also the spins in the domain wall gradually changes their directions to aligne themselves in the direction parallel to direction of magnetization. There are two types of domain walls 180° and 90° walls. The spins rotate by 180° from one domain to the other in the former case and 90° in the latter case.

3.6 Magnetization curve and Hysteresis (Irreversibility) :

The magnetization curves both in terms of B (by full line from the origin) and M (by deshed line) in the first quadrant have represented in the Fig.3.1. As applied field H increases, both B and M continuously increases. For particular value of H, magnetization M becomes constant known as saturation magnetization Ms, where as B continues to increase with H, as H forms a part of magnetic induction B. However, beyond Bs, saturation induction, slope $\frac{dB}{dH}$ is unity as well as μ



Fig. 3.1

approaches to 1 as H approaches to infinity (since $\mu = B/H$). The curve from demagnetized state to saturation on B V_S H curve is called as "initial or normal" induction curve³⁹. The intgrinsic induction or ferric induction B_i = B - H may be plotted as a function of H. This curve differs from M - H curve by a factor of 4 π which is to be applied to the ordinate.

The hysteresis (or irreversibility) is a phenomenon observed in ferro or ferri magnetic materials in which after saturation a decrease in H to zero does not reduce M to zero. Thus ferro or ferri magnetic materials can be made permanent magnet. The variation of M and H determines susceptibility $(\chi = \frac{M}{H})$ and hence class of material, where as B-H curve provides permeability ($\mu = \frac{B}{H}$). The permeability is a very useful property as it immediately gives the induction B produced by applied field H.

The intercepts on B-axis $(+B_r \text{ and } -B_r)$ give value of retintivity or residual induction for applied field H=0. The intercept on negative H-axis (Hc) is reversed applied field necessary to 'coerce' the material back to zero induction, is known as "coercivity". Similarly reverse field required to reduce M to zero is called the interinsic coercivity H_{ci}. The saturation in the reverse direction has been represented by - B_s.

The interruption in the initial magnetization process at some intermidiate point such as 'a' and the corresponding field is reversed and the reapplied, the induction will travel along the minor hysteresis loop abcdea. Here 'b' is called 'remance' and C the 'coercive force'. There is possibility of number of symmetrical and non-symmetrical (as $\frac{1}{3}$ and $\frac{9}{2}$ k) minor loops inside the major loop.

The irreversibility and hysteresis in ferromagnetic material are attributed to the impedements to the motion of domain walls. The impedemento are the grain boundries, inclusions, dislocations and hetrogenetic due to other phases⁽²¹⁾ Magnetization increases slowely by reversible wall motion at lcw fields and above critical field by irreversible wall motion and at very high field by irreversible rotation. The rotation of domains have been observed by Barkhaµusen.⁽¹¹⁾ He has shown that discontinuous variation of nagnetization at low fields takes place where the field is continuously increasing. These are known as Barkhaµusen jumps. The wall energy is maximum at very high fields. This leads to irreversible increase in magneto-elastic and magneto-static energies of the material under the action of external magnetic field.

3.7 Losses :

The magnetic material under the influence of an alternating magnetic field absorbs magnetic energy and dissipated as heat. For alternating field H = Ho exp (iwt), the induction B can be written as,

$$B = B_{0} \exp i (wt + \delta) \qquad ----- \qquad (3.71)$$

The permeability, $\mu = \frac{B}{H}$ is,

$$\mu = \frac{B_0}{H_0} (\cos s + i \sin s) = \mu' + \mu'' ----- (3.72)$$

where, μ' is the component of flux which is in phase and μ'' component of flux which is 90° out of phase with the applied field. The energy loss per cycle can be proportional to μ'' . The power factor or loss factor is defined by, tan $\delta = \frac{\mu''}{\mu'}$, while the quality factor Q is defined as,

$$Q = \frac{\mu''}{\mu'} = \frac{1}{\tan \delta}$$
 (3.73)

The frequency dependence of μ ' and μ ", known as permeability or magnetic spectrum, is an important criterion as per applications are concerned.

The important mechanisms involved in the losses are discussed in the following sub-sections.

3.7 (a) : <u>Hysteresis Loss</u> : The energy dE required to change magnetization M to M + dM at a field H is given by dE = H d M. Thus the total energy absorbed for a complete hysteresis cycle is,

 $W = \oint H dM$ ----- (3.74)

which is equal to the area under the hysteresis loop. Low coercivity or high permeability results in a small area under the loop and hence a small loss. 3.7 (b) : <u>Eddy - Current Loss</u> : An induced electric current in the magnetic core material due to alternating magnetic field causes heating and power loss. The power loss per second has been calculated to be proportional to $\frac{f^2}{2}$, where f is the frequency and ? the electrical resistivity of the core material. The proportionality constant depends upon the geometry of the core.

3.7(c) : <u>Spin - Resonance Loss</u> : The precessional frequency of the electron spin vector in a magnetic material is

$$\mathbf{W} = \sqrt{\mathbf{H}_{\mathbf{k}}}$$

where, γ^{\prime} - gyromagnetic ratio and

Under the influence of external λ - f magnetic field H_i in a direction perpendiculat to that of H_k , the resonance sets in, when the λ - f frequency matches the precessional frequency and energy is absorbed from the applied field. In case of a material with negative crystal anisotropy constant the rotational processes are important, then the resonant frequency is inversely proportional to (μ -1). Therefore, higher the permeability of the material the lower the resonant frequency.

H_k - Internal anisotropy field.

3.7 (d) : <u>Relaxation Loss</u> : The loss exhibited at frequencies lower than resonance frequency is attributed to several relaxation processes. The major relaxation loss in ferrites is attributed to the electron exchange between Fe^{2+} and Fe^{3+} ions. As the magnetization changes its direction, then Fe^{2+} and Fe^{3+} ions tend to change their positions to attain the configuration that has the lower energy under the changed direction of magnetization. The readjustment of the Fe^{3+} , Fe^{2+} positions does nct require movement of ions, but merely that of electrons. This relaxation loss is frequency dependent, and is maximum when the applied frequency is close to the relaxation frequency for electron jump for a given material at a given temperature.

3.7 (e) : <u>Wall - Resonance Losses</u> : In certain samples, the low frequency loss has been identified as due to domain wall resonance. If the domain wall is disturbed from its equilibrium position, a restoring force sets in, which tries to bring it back. The wall, like stretched membrance, thus has a natural frequency of ascillation. If the frequency of the applied magnetic field matches this natural frequency, resonance absorption sets in.

3.8 : A) : <u>Curie Temperature Measurement</u> :

The spontaneous magnetization exhibited by ferrites was found to disappear above certain critical temperature known as Curie temperature. These temperatures have been measured by using simple experimental set up shown in the Fig. 3.2. The set up is similar to that of Sinha <u>et al.</u>,⁽²²⁾ The copper pan is suspended with the help of steel spring in a vertical muffle furnace. A mild steel rod of appropriate



diameter is introduced into the furnace from lower end which forms the core of an electromagnet. The space between lower end of the pan and upper end of the rod is adjustable. D.C. power unit is used to magnetize mild steel rod. The spring and lower end of rod has been connected by copper wire via d.c. source and bulb to complete electric circuit, when sample touches magnetic pole.

To measure Curie temperature of each sample of the series $Cu_x \operatorname{Ni}_{1-x} \operatorname{Fe}_20_4$, with x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0. Each sample was kept in the copper pan. At room temperature, copper pan with ferrite sample touches the upper end of delectromagnet which causes to complete electric circuit. The temperature of furnace was slowely increased (measured with the help of a calibrated Al-Cr thermocouple placed very close to sample). At certain temperature, the continuity of the electric circuit was broken due to demagnetization of the ferrite sample (i.e. sample becomes paramagnetic). Thus showing a magnetic transition and the temperature at which this phenomenon is observed is also called as "Curie temperature". The observed Curie temperature (Tc) for the present mixed ferrite series $Cu_x \operatorname{Ni}_{1-x} \operatorname{Fe}_2 O_4$ have been presented in the table No.3.1.

Table No. 3.1	: Curie lemperatures of the sptem CuxNipre Felle		
	 X 	Tc in °C	-
	0.0	580	
	0.2	565	
	0.4	524	
	0.6	498	
	0.8	473	
	1.0	441	

The variation in Tc with copper content (x) is shown in the Fig.3.3. It can be observed that as percentage of copper increases the Curie temperature decreases. Also the difference in Tc, for samples with x = 0.0 and 0.2 are perceptibily less. However, as x increases difference in Tc is higher.

3.8 - B) : <u>Saturation Magnetization Study</u> :

The measurement of saturation magnetization (Ms) provide saturation magnetic moment n_B by expression

$$n_{\rm B} = \frac{\rm Ms}{5585}$$
 (3.9.1)

where, W is the molecular weight of the sample. The knowledge of n_B values thus obtained can be made use of to evaluate the distribution of cations, which is explained in the following





Fig. 3.4



few lines; after explaining the set-up and actual method used.

3.8 B (a) : <u>Experimental Method</u> : For the measurement of saturation magnetization, HIGH FIELD LOOP TRACER, HS 869, supplied by Electronic Corporation of India Limited, Hyderabad was used. The instrument is compact and highly sensitive, consists of an electromagnet working on 50 H_Z mains supply. It produces a sinusoidal magnetic field of the order of 3500 Orsteds, the air gap of about 9 mm. A special balancing coil is used to detect the magnetization of the sample placed in the air gap. The experimental set-up is shown in the Fig. 3.4.

The signal from the balancing coil after integration (which is proportional to the magnetic moment of the specimen) has been fed to vertical plates of an oscilloscope after suitable amplification. Where as, the signal proportional to the magnetic field has been fed to the horizontal plates of the oscilloscope. The vertical deflection has been calibrated in terms of magnetic moment in e.m.u., where as the horizontal deflection has been calibrated in terms of Oe/div. The calibration was done with the standard nickel strip having magnetization of 55.25 e.m.u./div.

3.8-B (b) : <u>Display of Hysteresis Loop</u> : To display the hysteresis loop, the steps involved in procedure are as follows

- i. High voltage cable is connected from control unit to the C. core unit.
- ii. The balancing coil was slowely introduced in to the air gap after connecting it to 12-pin connector, associated with the C-core unit.
- iii. The out puts (vertical and horizontal) were connected to the inputs (vertical and horizontal) of the control unit.
- iv. The vertical out put of the control unit was connected to the vertical channel of the oscilloscope. And the horizontal out put was connected to the EXT. horizontal in put of the oscilloscope.
- v. The current range was kept at 1 ampere position, with current control at minimum position.

vi. The VER gain control was kept low positive.

With the help of current control knob, the current was increased upto 300 mA so as to obtain an ellipse on the screen. For an appropriate size of an ellipse horizontal gain potentiometer were adjusted. Then the ellipse was converted into a straight line by adjusting the vertical phase control. Now by varying "coarse" amplitude potentiometer phase difference was confermed (The straight line only will rotate) and which could be made zero by adjusting horizontal phase potentiometer. With this setting, balancing coil was slowely taken out of the C-core gap. The samples $\operatorname{Cu_XNi_{1-x}Fe_2O_4}$ (with x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) were kept in the coil in the form of pellet. The coil was then slowly inserted into the air gap. The corresponding hysteresis loop was displayed on the screen. The current was slowly increased to obtain saturation magnetization.

3.8-B (c) : <u>Measurement And Specimen Calculation</u> :

The alternating currents required for saturation magnetization of each sample of the series $Cu_xNi_{1-x}Fe_2O_4$, with x = 0.0, 0.2, 0.4, 0.6, 0.3 and 1.0, were noted. And the corresponding field excited in C-core were obtained from calibration plot of current (r.m.s.) in mA against excited peak field.

With the help of calibrated Y-plate of C.R.O. (1 mA = 0.1 volt), from corresponding effective lengths of hysteresis loop and excited fields in the C-core, saturation magnetization (Ms) in e.m.u./gm were obtained at room temperature. The values of Ms, thus obtained are comparable with the reported values. The saturation magnetic moments have been obtained by equation 3.9.1. The magnetic data are presented in the Table No.3.2.

The variation of Ms in e.m.u./gm with copper content for $Cu_xNi_{1-x}Fe_2O_4$ series is shown in the Fig. 3.5.

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X	:Saturation : Magnetization (Ms) at room temp. in e.m.u./gm	Saturation Magnetic moment n _B room temp. Bohr.magna en.	: Saturation Magnetization at (Ms) reported in at room tem. t- (Ref.23)	Saturation Magnetic moment(n _B *) reported at 0°K.(Ref.23)
0.0	53.0	2.22	47.60	2.182
0.2	48.2	2.03	44•97	2.066
0.4	41.3	1.75	42.52	2.033
0.6	37.0	1.57	41.33	1.978
0.8	28.7	1.23	37.58	1.660
1.0	23.2	0.99	29.15	1.393



The specimen calculation for Cu_{.2}Ni_{.8}Fe₂O₄ is given as follows :

The standard nickel strip of mass 0.85 gm, displayed the hysteresis loop of effective length 12 division. Therefore, for one gm. of standard nickel, corresponding divisions on -axis of C.R.O. become 14.1176. The e.m.u. per gram provided for standard nickel strip is 55.25, is used to obtain e.m.u. per division (say a = 3.9135 e.m.u./div.) for stand_nickel strip of one gram.

Now for the sample $Cu_{.2}Ni_{.8}Fe_{2}O_{4}$, of mass 0.7476 gm, observed effective length on Y-axis of C.R.O. is, 9.2 division. Therefore, division per gm. (Z) for the sample is Z_12.3060; lead to obtain saturation magnetization Ms as,

$$M_{S} = (Z) (a) e.m.u./gm$$

= 12.3060 x 3.91355
. $M_{S} = 48.1602 e.m.u./gm$.

Thus for each sample Ms has been calculated. As mentioned in B above (eq. 3.9.1) the values of saturation magnetic moment n_B in Bohr magneten can be estimated. The values obtained for present series are given in Table No.3.2.

3.9 : <u>Results and Discussion</u> :

As discussed in previous section the saturation magnetization (Ms) values have been calculated and presented in table number 3.2, for the system $\operatorname{Cu_XNi_{1-x}Fe_2O_4}$. In Fig.3.5 the compositional dependance of saturation magnetization Ms in emu χ gm is shown. It can be seen that the Ms value goes on decreasing as the composition becomes $\operatorname{Cu^{2+}}$ rich and is minimum for Cu Fe₂O₄ while maximum for NiFe₂O₄. Further it can be seen that a linear relation of variation of Ms with composition is exhibited. Our values of saturation magnetization are comparable with those reported earlier⁽²³⁾ of the same system. Whatever deviation in the values of Ms are seen from table No.3.2, can be attributed to the porosity, thermal treatment and other attendent factors like grain size, defects developed during the process, in short the method of preparation of ferrites, because the final electrical and magnetic properties depend upon these physical parameters.

Since the saturation magnetization goes on decreasing with increasing copper content, it suggests that the introduction of copper weakens the A-B interaction in the copper. Nickel system which lowers the Ms values with increasing Cu²⁺.

In Fig.3.3, the compositional variation of Curie temperatures, experimentally measured as discussed previously, is presented for the $Cu_xNi_{1-x}Fe_2O_4$ system. It can be noted that the Tc values also depend in the same fashion as the saturation magnetization i.e. the Tc values go on decreasing with increasing copper content. Further, this dependence also exhibits a nearly linear relationship. These our results on Tc measurement deviated a little from those previously reported ones; however, not much. It can be seen that the Tc value for NiFe₂O₄ is maximum (853°K) and minimum for CuFe₂O₄ (714°K).

The cation distribution in the slow cooled mixed ferrite system $Cu_xNi_{1-x}Fe_2O_4$ can be expressed as

$$\operatorname{Fe}_{(1-xy)}^{3+} \operatorname{Cu}_{xy}^{2+} \left\{ \operatorname{Ni}_{(1-x)}^{2+} \operatorname{Cu}_{x(1-y)}^{2+} \operatorname{Fe}_{(1+xy)}^{3+} \right\}$$

as it is well known that Ni²⁺ occupies essentially octahedral B-site, where as Cu^{2+} ion can occupy both A and B sites,²⁴ and therefore, y, the fraction of Cu^{2+} ion on A-site has been considered in the above distribution. Then y is given by

$$\mathbf{y} = \frac{\mathbf{n}_{B} - (1 - x) \mathbf{m}_{Ni} 2 + - x \mathbf{m}_{Cu} 2 + \frac{1}{2 \times \mathbf{m}_{Fe} 3 + - \frac{1}{2 \times$$

WHERE, m_{Ni}^{2+} , m_{Cu}^{2+} and m_{B}^{-3+} are the magnetic moments of respective elements and n_{B}^{*} the saturation magnetic moment at 0°K. Our values of saturation magnetization (Ms) for the system at room temperature are in good agreement with the earlier reported ones and therefore, the Ms values extrapolated by these authors to 0°K and hence n_{B}^{*} are used for calculation estimated thus has been presented in Table No.3.3.

From the table it can be seen that giving preference to B-site for Ni²⁺ ions, with increasing copper content up to

Table No.3.3 : Cation difficultion in the system

$$C_{4\times}N_{1+x}f_{e_{2}}o_{4}$$
, from magnetisation data.

Cation Distribution ± 0.002
0.0 $(Fe^{3+})^{A}$ $[Fe^{3+} Ni^{2+}]^{B} 0_{4}^{2-}$
0.2 $(Cu_{0.015}^{2+} Fe_{0.985}^{3+})^{A} [Ni_{0.8}^{2+} Cu_{0.185}^{2+} Fe_{1.015}^{3+}]^{B} 0_{4}^{2-}$
0.4 $(Cu_{0.040}^{2+} Fe_{0.960}^{3+})^{A} [Ni_{0.6}^{2+} Cu_{0.360}^{2+} Fe_{1.040}^{3+}] 0_{4}^{2-}$
0.6 $(Cu_{0.063}^{2+} Fe_{0.937}^{3+})^{A} [Ni_{0.4}^{2+} Cu_{0.537}^{2+} Fe_{1.063}^{3+}]^{B} 0_{4}^{2-}$
0.8 $(Cu_{0.053}^{2+} Fe_{0.947}^{3+})^{A} [Ni_{0.2}^{2+} Cu_{0.747}^{2+} Fe_{1.053}^{3+}]^{B} 0_{4}^{2-}$
1.0 $(Cu_{0.049}^{2+} Fe_{0.951}^{3+})^{A} [Cu_{0.951}^{2+} Fe_{1.049}^{3+}]^{B} 0_{4}^{2-}$

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x = 0.6, the tendency of copper ions exhibits an increasing preference for A-sites, while after x > 0.6 once again the Cu²⁺ ions increasingly migrate to B-site showing an increasing inversion tendency.

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