C H A P T E R - V

SUMMARY AND CONCLUSIONS

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Since 1950, magnetic oxides, especially ferrites stired up tremendous interest because of their electrical, electronic, microwave and computer applications. As these materials are technologically important the same have been studied extensively from the point of view of their magnetic and electrical properties to check their suitaoility for certain applications. In many electronic applications ferrites with low coercive force and narrow hysteresis loop along with the higher sensitivity are required for minimising eddy currents and hysteresis losses. The other important factor in selecting ferrites for certain amount of magnetic effect in applications is permeability. Without any loss on the performance level, the miniaturisation can be done by using ferrites with high permeabilities. In magnetic switching devices the sequareness ratio is of prime importante.

Zinc ferrite shows the normal spinel structure with proved certainity and without much effect of conditions of preparations when heat treated below 1200^{o}C to 1400^{o}C. Nickel ferrite and copper ferrite are reported to be partially inverted ferrites. Copper ferrite exhibits electrical switching, lattice distortion, structural change with temperature, sensitivity to most of the conditions of preparations and quenching. The mixed nickel zinc ferrite system has been studied by many workers from the point of view of their electrical and magnetic properties.

However, much remains to be explored in this system as different workers have investigated various interesting properties with changing conditions of preparation. We have undertaken the study of mixed Ni-Zn ferrite and copper substituted mixed Ni-Zn ferrite to investigate the role of copper addition in changing structural, electrical and magnetic properties.

Following studies were, therefore, proposed.

(1) (A) Preparation of $Ni_{1-y-x}Zn_xCu_yFe_2O_4$

where $x = 0, 0.2, 0.4, 0.6, 0.8, 1.0$ for $y = 0$.

and $x = 0, 0.2, 0.4, 0.6, 0.8$ for $y = 0.2$

by ceramic method at an appropriate temperature due care being taken for non reduction of copper ion.

(B) X-ray diffraction studies to characterise the ferrite samples.

(2) D.C. resistivity studies to understand the mechanism of conduction in ferrites and also the determination of Curie temperatures of the samples which could be verified with the experimentaly determined values.

(3) Hystersis studies with a view to determining saturation magnetisation values.

Chapter I deals with the theory of crystal structure of ferrites, history and development of ferrites, Neel's theory of ferromagnetism and a brief review of electrical and magnetic

properties of ferrites. The factors that influence the properties of ferrites are also discussed. At the end of the chapter applications are discussed.

Chapter II is split up into three parts A, B and C.

In Part A preparation of the ferrite samples is described. We have prepared the ferrites by standard ceramic technique.

In Part B our data on crystal structure is presented. Observed and calculated 'd' values from the diffraction patterns indicate that the spinel structure is fully formed in our samples. All the samples exhibit cubic crystal structure which agrees with that reported by Patani et al.[1] The graphs of lattice parameter 'a' against content of zinc show that the lattice parameter 'a' increases with the addition of zinc for Ni-Zn ferrite and Ni-Zn ferrite, containing 0.2 Cu. It is observed that Vegard's law is obeyed by both the series.

In general the lattice parameter 'a' decreases with addition of copper in Ni-Zn ferrites. When the ferrites containing the same content of zinc are compared, the lattice parameter does not show either increasing or decreasing trend. The deviation in the lattice parameter was found to be approximately 0.01 A^{o} for all the copper containing Ni-Zn ferrites. The deviation in the lattice parameter was explained on the basis of ionic volume of the copper ion which in this case is replacing Zn ion.

In Part C the method for determining Curie temperature has been described which is a slight modification of the method reported by Loria and Sinha[2]. The values of Curie temperatures determined are reported. The graphs of Curie temperature (T_c) with the content of zinc are given. The nature of the plots is discussed in relation to resistivity and saturation magnetisation studies in chapters three and four.

In Chapter III, the studies on resistivity of the ferrite samples have been reported. The resistivity studies were carried out by two probe method. The graphs of log ρ versus $10^3/T$ are plotted. From these graphs, the values of activation energy, ΔE, and Curie temperatures T_c are obtained. The Curie temperatures so obtained are found to agree with the experimentally determined values.

The breaks and discontinuities occuring in log ρ versus $10^3/T$ plots are attributed to several sources[3,4,5].

The change in the activation energy is divided into four different regions. It is assumed that the activation energy is maximum at high temperatures and minimum in region of low temperatures, however the values of ΔE are comparable. The low values of ΔE in the region 1 (low temperature) are attributed to the impurity conduction. The slight increase in value of ΔE in region two is attributed to influence of phase transition[5] or some undetected phase formed at the time of sintering or at the time of resistivity measurement. The

breakes in the conductivity separating regions three and four or two and three in some samples give a temperature which nearly coincides with the Curie temperature, suggesting a magnetic ordering change taking place in the samples. The change in the activation energy is attributed to magnetic ordering which affects conductivity.

The electrical resistivity, the change in activation energy at Curie temperature and relationship of activation energy with the composition are explained and discussed on the basis of mechanism of hopping of polaron process which has been successfully employed to explain the electrical properties of ferrites[6].

The values of ΔE indicate that the hopping process due to polarons is favoured in case of our ferrites. The theory of conductivity has been explained on the basis of hopping of polarons due to thermal activation[7].

The compositional variation of resistivity and the compositional variation of M_s Fig.4 Chapter 3 bear one to one correspondance indicating the role of magnetic ordering in influencing the resistivity of ferrite samples. This statement is supported by the fact that there is a decrease in T_c with increasing zinc content. Guillauds et al.[8] have given compositional variation of M_s for series of ferrites[9,10]. The magnetisation behaviour of the zinc containing ferrites has

been explained on the basis of three sublattice model by Yafet and Kittel[9]. Recently Srivastava et al.[10] have used three sublattice model to determine exchange constants in spinel ferrites.

In Chapter IV studies on magnetisation have been reported. The values of saturation magnetisation M_s and theoretical and experimental values of Y-K angles have been found out.

The variation of Ms with the content of zinc for $Ni_xZn_{1-x}Fe_2O_4$ is explained on the basis of cation distribution and is not unexpected[11]. It is expected that B-B interaction is antiferromagnetic even in the mixed ferrimagnetic zinc ferrite. The effect of this interaction is usually masked by strong AB interaction which causes the spins on B site to be aligned parallel to each other. However, the substitution of zinc on A site weakens the A-B interaction and when 50 % of A site has been substituted by Zn^{+2} ions, the B-B interaction becomes comparable in strength with AB interaction and the magnetic moment decreases. The variation of Ms with zinc content for the mixed copper substituted Ni-Zn ferrite can be explained on the similar arguments.

The results on Y-K angles are correlated with the magnetisation variation in nickel-zinc and copper substituted nickel-zinc ferrites

In both the ferrite series as the content of zinc is increased the Y-K angles go on increasing. Thus the change in

magnetisation on zinc substitution occurs due to the presence of Y-K angles in the Spin system on B sites. The condition for Y-K angles to occur in Ni-Zn ferrite system has been investigated in the molecular field approximation by Satyamurthy et al.[12] using a non linear three sub-lattice model. The increase in Y-K angles indicate the increasing favouring of triangular spin arrangement on B sites leading to reduction in A-B interaction. B-B interactions are antiferromagnetic even in a mixed magnetic zinc ferrite. The effect of B-B interaction is usually masked by strong A-B interaction which causes the spin on B sites to be aligned parallel to each other. However, the substitution of zinc in excess of 20 % leads to canted type of arrangements on B site weakening the A-B interaction as suggested by Yafet and Kittel. When nickel in Ni-Zn ferrite is completely replaced by zinc, $\cos\alpha_{yk}$ becomes zero, suggesting that B-B interaction collapses leading to zero value of Ms for $ZnFe_2O_4$.

Our studies on D.C. resistivity show that copper substituted Ni-Zn ferrites system is of immence theoretical importance with a view to investigating its compositional behaviour, while our studies on magnetization reveal that there is a further scope for improvement in the Ms values with proper tailor making of the ferrites.

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