

**CHAPTER V**  
**SUMMARY AND CONCLUSIONS**

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The spinel ferrites are extensively used for miniaturization. The ferrites occupy an important place in the field of electrical, electronics, computer technology and microwave communication. The important properties of ferrites that make them suitable for these applications are their high electrical resistivity and low eddy current losses. They have advanced to a position of technological prominence in the last two decades. They are being extensively studied from the point of view of understanding their behaviour and applications.

There has been a growing interest in NiCuZn ferrites for the application in producing multilayer type chips. In the recent years much progress has been made in the control of ferrite material properties through chemical composition and preparation techniques. By the use of suitable substituents and control over the preparation conditions, the properties of Ni - Cu ferrites can be altered to make them suitable for wide range of applications. Hence much attention has been paid towards the study of spinel ferrites containing non-magnetic ion like Zn, Cd, Mg etc.

The mixed nickel zinc ferrite system has been studied by many workers from the point of view of their electrical and magnetic properties. The purpose of this study is to investigate the effect of nickel addition on the electrical and magnetic properties of NiCuZn ferrite.

The work involved the following studies -

1. Preparation of ferrites with the general formula  $\text{Ni}_x \text{Cu}_{0.2} \text{Zn}_{0.8-x} \text{Fe}_2\text{O}_4$  where  $x = 0.2, 0.3, 0.4, 0.5, 0.6, 0.7$  and  $0.8$ .
2. X-ray diffraction studies to confirm single phase formation of the material.
3. Studies on electrical properties viz. d.c. conductivity, a.c. conductivity to understand conduction mechanism, nature of charge carriers and dielectric dispersion.
4. Studies on magnetic properties like hysteresis and initial permeability to see the effect of Ni and Zn substitution on magnetic properties.

The subject matter is presented in five chapters.

Chapter-I is introductory. It gives information about the past, present and future of ferrites, classification and types of ferrites, spinel structure, electrical and magnetic properties along with historical developments. A brief survey of research work

carried out on NiCuZn ferrites is included. The orientation of work has been given at the end of the chapter after applications of ferrites.

The Chapter- II is divided into two parts. Part-A deals with the synthesis of ferrites, while Part- B deals with the XRD studies. In the present case, the ferrites  $\text{Ni}_x\text{Cu}_{0.2}\text{Zn}_{0.8-x}\text{Fe}_2\text{O}_4$  (where  $x = 0.2, 0.3, 0.4, 0.5, 0.6, 0.7$  and  $0.8$ ) were prepared using standard ceramic method. The samples were presintered at  $800^\circ\text{C}$  for 10 hours and finally sintered at  $1150^\circ\text{C}$  for 24 hours and then furnace cooled at a rate of  $60^\circ\text{C}/\text{hour}$ . Pellets of 1 cm diameter and torroids with inner diameter 1 cm and outer diameter 1.5 cm were prepared by applying a pressure of 5 tonnes per square inch for five (minutes) using hydraulic press.

X-ray diffraction patterns were obtained using Philips x-ray diffractometer (Model PW 1710) with  $\text{CuK}\alpha$  ( $\lambda = 1.518 \text{ \AA}$ ) radiation. X-ray diffraction patterns confirm the cubic spinel structure of all the samples. The absence of any extra lines indicates the formation of single phase in the sample. The observed and calculated 'd' value are in good agreement with each other. The lattice parameter decreases with increasing nickel concentration (decreasing Zn) from  $8.420 \text{ \AA}$  to  $8.364 \text{ \AA}$  which is attributed to ionic size differences of  $\text{Ni}^{2+}$ ,  $\text{Cu}^{2+}$  and  $\text{Zn}^{2+}$  ions ( $\text{Ni}^{2+} = 0.74 \text{ \AA}$ ,  $\text{Cu}^{2+} = 0.72$ ,  $\text{Zn}^{2+} = 0.83 \text{ \AA}$ ).

The XRD data has been used to calculate the bond length  $R_A$ ,  $R_B$  and  $r_A$  and  $r_B$  also found to decrease linearly with Ni content (decreasing Zn content). They lie in the range 1.939 Å to 1.925 Å, 2.039 Å to 2.024 Å, 0.589 Å to 0.575 Å and 0.687 Å to 0.672 Å respectively. The variation of these parameters with nickel is attributed to the variation in lattice parameter. In the present work the porosity lies between 10 to 16%.

Chapter-III is divided into two parts. Part-A deals with dc conductivity, while Part-B deals with the ac measurements. D.C. resistivity was measured by conventional two probe method from room temperature to 600°C. The plots of  $\log \rho_{dc}$  vs  $1/T \times 10^3$  show a break at a particular temperature which co-relates well with the Curie temperature of the samples. The activation energy in the paramagnetic region is more than that the ferrimagnetic region. They lie in the range 0.12 eV to 0.37 eV for ferri and 0.49 eV to 0.54 eV for para. It is attributed to the magnetic disordering on heating. The conduction is attributed to the hopping of electrons between  $Fe^{2+}$  and  $Fe^{3+}$  ions on B site. The increase in resistivity with Ni content is due to the decrease in  $Fe^{3+}$  ions and thereby reducing  $Fe^{2+} \leftarrow \rightarrow Fe^{3+}$  conduction pattern. The decrease in resistivity with further increase in Ni content is due to the transfer of hole from  $Ni^{3+}$  to  $Ni^{2+}$  ions. In NiCuZn ferrite, copper is a

John-Teller ion. The deviation can also be attributed to the effect of John-Teller ions on the ground states of  $\text{Fe}^{2+}$  and  $\text{Ni}^{2+}$  ions in spinel ferrites.

AC conductivity was measured at room temperature on LCR meter bridge HP 4284 in the frequency range of 100 Hz to 1 MHz. The variation of dielectric constant with Ni content reveals that  $\epsilon'$  goes on increasing with Ni content (decreasing Zn content), reaches maximum at  $x = 0.5$  and then decreases with further increase in nickel content. The dispersion in  $\epsilon'$  with frequency is of Maxwell-Wagner type.

Variation of  $\tan \delta$  with frequency shows very small values above 1 MHz. A maximum  $\tan \delta$  is observed when the jump frequency of electron between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  is equal to the frequency of applied external field. The compositional variation of  $\epsilon'$  and  $\tan \delta$  is attributed to change in resistivity with composition.

From the variation of ac resistivity with frequency for all compositions it is observed that the dispersion is large at low frequency side and tends to level off in the high frequency side and tends to level off in the high frequency region, which is in agreement with Koop's model.

Chapter-IV includes magnetic properties viz. hysteresis (Part A) and initial permeability (Part B). The magnetization

measurements were carried out on the ferrite system using high field hysteresis loop tracer. The models for magnetization are explained in the beginning of the chapter. It has been observed that the saturation magnetization ( $4\pi M_s$ ) and magnetic moment increase from 1497 to 2953 G and 1.18 to 2.20 Bohr magnetons and reach maximum at  $x = 0.4$  obeying Neel's model. They decrease from 2764 to 1788 G and 1.96 to 1.25 Bohr magnetons i.e. beyond  $x = 0.4$  with further increase in nickel content obeying three sublattice model suggested by Yafet and Kittel.

The decrease in saturation magnetization and magnetic moment beyond  $x = 0.4$  suggest the existence of Y-K angles in the present samples. It is observed that as the content of  $Ni^{2+}$  is increased the Y-K angles go on decreasing. For  $x = 0.2, 0.3, 0.4, 0.5$  and  $0.6$  the Y-K angles be  $61^\circ, 46^\circ, 35^\circ, 29^\circ$  and  $21^\circ$  respectively. No Y-K angles are found at  $x = 0.7$  and  $0.8$ .

The  $M_r/M_s$  ratio decreases from  $x = 0.55$  to  $0.40$  and becomes minimum at  $x = 0.4$  with the addition of  $Ni^{2+}$  content. It then increases from  $0.42$  to  $0.50$  with further addition of  $Ni^{2+}$ .

The initial permeability was calculated by measuring inductance values of torroidal cores using Aplab LCR-Q meter at a fixed frequency of 1 kHz. The sharp decrease near Curie temperature suggests the single phase formation of the ferrites.

For  $x = 0.3$  and  $0.4$  the  $\mu_i$  shows a slight decrease with increase in temperature unlike the case of  $x = 0.5$  to  $0.8$ . The peaking behaviour is more pronounced in composition  $x = 0.3$  and  $0.4$ . In case with  $x = 0.5$  a diffuse peak is observed before permeability drops to zero at Curie temperature. For the samples with  $x = 0.6$ ,  $0.7$  and  $0.8$  the  $\mu_i$  remains almost constant upto Curie temperature and suddenly drops to zero showing slight tailing effect.

For the present NiCuZn ferrite system, there is an increasing trend in initial permeability upto  $x = 0.4$  from 611 to 652. Afterwards the initial permeability decreases with increase of  $\text{Ni}^{2+}$  content from 199 to 44. It can be concluded that  $\text{Ni}^{2+}$  when present in lower proportion allows easy to free domain wall motion. Whereas it appears to cause impedance to domain wall motion and reduces the magnitudes of initial permeability on higher content.

Curie temperature exhibits increasing trend on addition of  $\text{Ni}^{2+}$  content.