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A study of electrical and magnetic properties of Cu substituted Ni-Zn Ferrites

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Abstract: *Ferrites, which are a class of magnetic oxide compounds that contain Iron oxide as a major component, have found a large number of applications in various electronic and communication devices. The ferrite system $Ni_xZn_xFe_{2-y}O_4$; x=0.50, y=0.00, 0.05, 0.10, 0.15, 0.20.and 0.25 is prepared by citrate gel method. Influence of Cu substitution in the prepared samples (Ni-Zn ferrites) enhances the electrical and magnetic properties. The Curie temperature, Saturation Magnetization values are found to decrease with* Cu *concentration. The variation in Curie temperature can be explained on the basis of exchange interactions. The density of samples is low up to y=0.10 and then increases gradually with the increase of dopant content. The value of Dielectric constant slowly increases initially up to y=0.10 and then increases sharply at higher* Cu *concentration. The variation of dc resistivity as function of temperature for the concentration of Cu at y=0.00, 0.05, 0.10, 0.1, 0.20, 0.25 was studied. Structral properties studied by using XRD technique*

Keywords*: Ferrite, density, Saturation Magnetization, Curie temperature, dielectric constant, Resistivity*

I. Introduction

Ferrites which are class of magnetic oxide compounds that contain Iron oxide as a major component, have found number of applications in various electronic and communication devices. Ni-Zn ferrite is of greater commercial application in the electromagnetic interfaces known as EMI, which is used in hard disk drives in laptops. Generally ferrites have high resistivities (10^{11} ohm-cm)[1] as compared to metals. The electronic conduction in ferrites is mainly due to hopping electrons between the ions of the same element present in more than one valence state distribution randomly over crystallographic ally equivalent lattice sites [2]. The electrical resistivity of ferrites drops exponentially with rising temperature. It has been since the investigators of Blechschmidt [3]in 1938 ,that ferrites have high dielectric constants at low frequencies which exhibited dimensional resonance effects[4]. Higher values of dielectric constants are due to interfacial polarization [5].

In a ferrite we can observe transition from ferromagnetic to paramagnetic state as temperature increasing. Although there are few reports on different properties of Ni-Zn ferrite a systematic investigation about electrical properties on Ni-Zn ferrite to understand general properties. Hence in this communication we are reporting electrical and magnetic properties of Ni-Zn ferrites synthesized by Citrate gel method. It is well known that Ferrites possess cubic closed packed arrangement oxygen atoms.

Ni- Zn ferrite is mixed spinel in which the tetrahedral (A) sites are occupied by Zn^{+2} and Fe ions, and the octahedral sites (B) are occupied by Ni^{+2} and Fe^{+3} ions in the spinel formula AB_2O_4.The magnetic & dielectric proper-ties depend on this distribution of these ions on tetrahedral and octahedral sites. It is also believed that addition of Zn ions alter the saturation magnetization, lattice parameter and Curie temperature. Several researchers synthesized Ni-Zn ferrite by conventional methods like double sintering method, solid state reaction method, and chemical co precipitation method, hydro thermal process etc. In this context, the objective of this work is to synthesize zinc doped nickel ferrite produced by Citrate gel method.

II. Experimental

The following ferrite ceramic compositions are processed through the standard conventional ceramic route [6-7] with small changes. The starting materials are analytical reagent grade Nickel nitrate, Zinc nitrate, iron nitrate and Copper nitrate with the purity of 99.9%.

1. $Ni_{0.50}\ Zn_{0.50}\ Fe_2O_4$
2. $Ni_{0.50}\ Zn_{0.50}\ Fe_{1.95}\ Cu_{0.05}\ O_4$
3. $Ni_{0.50}\ Zn_{0.50}\ Fe_{1.90}\ Cu_{0.10}\ O_4$
4. $Ni_{0.50}\ Zn_{0.50}\ Fe_{1.85}\ Cu_{0.15}\ O_4$
5. $Ni_{0.50}\ Zn_{0.50}\ Fe_{1.80}\ Cu_{0.20}\ O_4$
6. $Ni_{0.50}\ Zn_{0.50}\ Fe_{1.75}\ Cu_{0.25}\ O_4$

Five different compositions of $Ni_xZn_xFe_{2-y}O_4$ with y = 0.0, 0.05, 0.10, 0.15,0.20 and 0.25are prepared by citrate gel method. The citrate process is simple, easy and doesn't require any elaborate and expensive experimental setup. The main advantages of this method is

1. Capacity to yield a homogenous mixture of the con-stituent ions.
2. As no ball milling is required in this process, there is a little scope of contamination of materials.
3. In case of conventional methods, there is a possibil-ity of introducing iron impurities during milling this leads inhomogenity in sample, which affects the mag-netic property.
4. This is a simple method which offers a significant saving in time and energy consumption.

The stoichiometric amounts of metal nitrates are dissolved in minimum amount of deionized water and stirred vigorously on a magnetic stirrer for 1 h at temperature 60◦C. Now, the citric acid is added to the solution slowly with gradual rise in temperature.. The resulting mixture was pre sintered in air at 800^0c for 6 hours. This pre sintered ferrite was again grounded for few hours added 5%PVA (Poly Vinyl Alcohol) as a binder and pressed into disk shaped pellets at a pressure of 5 tons per square inch using a hydraulic press. These disk shaped pellets placed on a platinum foil and sintered in air at 1100^0c for 10 hours. These samples were painted with air dried silver paste and heated at 600^0c for 1 hour to establish good electrical contact. The X-ray diffraction patterns of the ferrite powder was taken on powder X-ray diffractometer (X-RD) using Cu-Kα radiation

The density of sintered ceramic samples was determined by Archimedes principle. The D.C.Resistivity of processed compositions under investigation was carried out by two terminal method. It was calculated by using the Conductivity cell. The resistivity was calculated by using the relation ρ =RA/l, where A is area of cross section, R is Resistance. The Resistivity is generally changed by impurities substitution in basic ferrites [8]. The measurements of Curie temperature are carried out by using the technique [9] of attaching a small piece of ferrite material to the lower end of an iron rod attached to an electromagnet. The temperature was measured using Cr-Al thermocouple. The capacity and the dielectric loss of ferrite materials were measured at room temperature using LCR meter. The dielectric constant was calculated [10] using the following formula $\varepsilon = d.C/\varepsilon_0 A$, where ε_0 is the free space permeability, 'A' is the area'd' is the thickness of the sample. The saturation magnetization was measured using the pendulum method described by Rathonev and Snoek[11]. The magnetic moment per unit volume is equal to $M=K(\omega^2 - \omega_0^2)/V$, where 'M' is the saturation magnetization, 'K' represents a constant of the system and can be obtained by using standard sample of known magnetization, here 'ω' and ω_0 are the pendulum frequencies with and without ferrite sample respectively.

III. Results And Discussions

XRD analysis: The powder XRD pattern of the $Ni_xZn_xFe_{2-y}O_4$ with y = 0.0, 0.05, 0.10, 0.15,0.20 and 0.25 and sintered powders were recorded using a Pan-analytical X'Pert-PRO diffractometer using Cu-Kα radiation (λ = 1.5406 A). The formation of the spinel ferrite phase was confirmed by the JCPDS files [13]. The inter-planar spacing was obtained from the Intensity Vs. 2θ plot using the Braggs Law .The X-ray diffractogram of Ni-Zn ferrite is shown in **F**ig1, which reveals a single phase cubic spinel structure. The lattice parameter of

$a = d(h^2+k^2+l^2)^{1/2}$ where, a = lattice constant, d = inter planar distance and, (h, k, l) are the Miller indices

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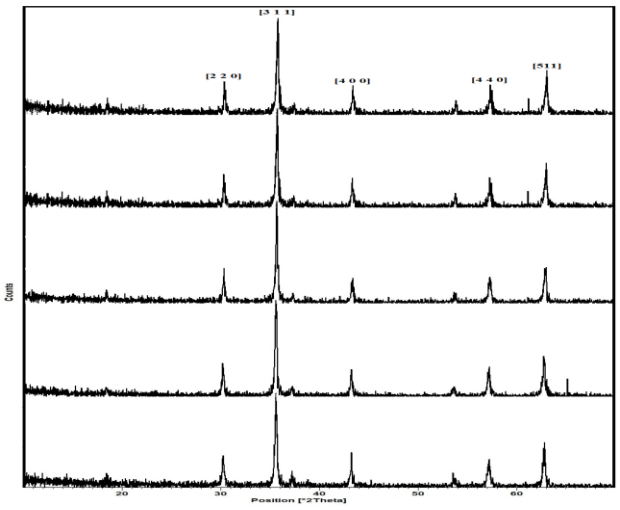

Fig1.XRD Pattern

Table1.variation of lattice constant, crystallite size ,strain with Cu content.

S.NO	Cu content	Lattice constant (A^0)	Crystallite size(nm)	strain
1	0.05	8.4691	32.2	$1.075x10^{-3}$
2	0.10	8.4673	32.7	$1.06x10^{-3}$
3	0.15	8.4359	37.8	$0.91x10^{-3}$

4	0.20	8.3894	39.9	$0.867x10^{-3}$
5	0.25	8.3759	34.28	$1.01x10^{-3}$

In table 1 it is clear that the variation of lattice constant decreases with increase of concentration of Cu content. It is also represent in Fig 2.This behavior of decrease lattice constant with Cu content is due to the difference in ionic radius.The average crystallite size of all samples is evaluated from the line broadening of most intense peak (311) using Debye Scherer equation.

$$D_{311} = 0.9\lambda/\beta\cos\theta$$

Where D average crystallite size,λ is the wavelength of incident X ray , β is FWHM at peak (311) The values of D increases with Cu concentration up to y=0.20 and decreases, because the micro strain increases at y=0.25. The strain was calculated by using $\beta\cos\theta/4$.

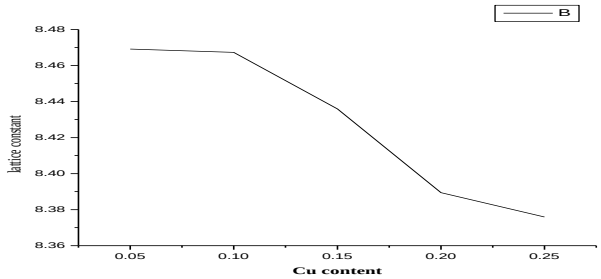

Fig 2 variation of lattice constant with Cu content

Density: Densities of the prepared compositions have been calculated. The variation of densities can be observed from Table2.

Table 2: Density variation with the Cu content

Cu Content	Density
0.00	4.73
0.05	4.64
0.10	4.55
0.15	4.82
0.20	4.91
0.25	4.95

Fig.3 shows the variation in density with the variation of Cu dopant concentration and it is clear from the figure that the density is low Upto 0.10 and then increased gradually with the increase of dopant content. The highest density is observed to be 4.95 gm/cm^3 which were found to be in good agreement with reported value of 4.88 gm/cm^3 for similar compositions as shown in table 2.

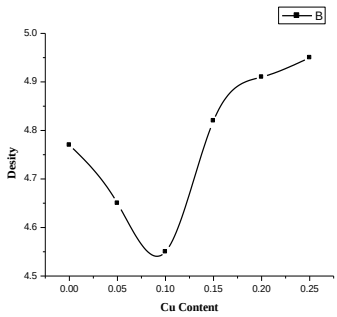

Fig.3 Density variation with Cu Concentration

Resistivity: The variation of Resistivity with the as a function of temperature for Cu^{+3} substituted different compositions have been calculated and show in the Table 3.

Table 3: Variation of resistivity of Cu concentrations as a function of Temperature

S.NO	1/T X10^{-3}	Log ρ values at various Cu concentrations					
		0.0	0.05	0.10	0.15	0.20	0.25
1	21	1.2	1.25	1.3	1.4	1.5	1.6
2	22	1.3	1.4	1.4	1.4	1.5	1.6
3	23	1.5	1.6	1.5	1.5	1.6	1.7
4	24	1.6	1.6	1.6	1.6	1.7	1.8
5	25	1.65	1.7	1.6	1.7	1.8	1.9
6	26	1.7	1.8	1.7	1.7	1.8	1.9
7	27	1.77	1.85	1.9	1.8	1.8	2
8	28	1.82	1.95	2	2	1.9	2.1
9	29	1.9	2	2.1	2.1	2.1	2.2
10	30	2	2.1	2.2	2.3	2.2	2.4

Fig.4 shows the variation of Resistivity as a function of temperature for the prepared samples. It has been observed the increase in resistivity for the prepared compositions from the table II. The increase in resistivity is because of formation of layers of Cu_2O_3 and the change in resistivity values due to hopping mechanism [12] in all the samples.

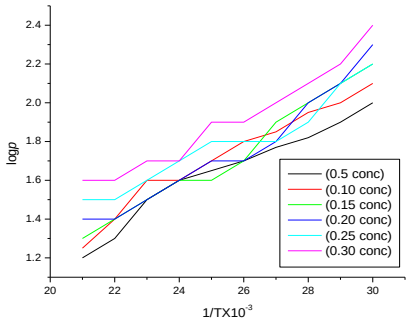

Fig.4 : log ρ Vs 1/T at various Cu^{+3} substituted concentrations

Curie Temperature: Variation of Curie temperature with the Cu^{3+} concentration have been calculated and indexed in the table4. The Curie temperature has been observed as decreasing with the increase of Cu concentration. The change in Curie temperature can be explained on the basis of exchange interactions. The non- magnetic Cu^{3+} dopant replaces the iron ions, which prefer to occupy B-sites [13]. The obtained value of Curie temperature for pure composition i.e., $Ni_{0.5}$ $Zn_{0.5}$ Fe_2O_4 is 370^0c which is almost in agreement with the reported value of 380^0c [14].

Table 4: variation of Curie Temperature for the Cu^{+3} substituted concentrations

Cu Content	Curie Temperature
0.00	375
0.05	365
0.10	353
0.15	339
0.20	305
0.25	270

The para magnetic Cu causes a decrease of magnetic interaction among the A and B sub lattices, which has been gradually seen in Curie temperature as shown in the fig.5

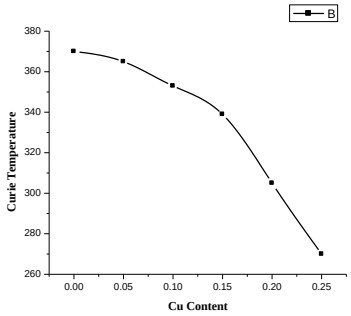

Fig 5: Variation of Curie temperature with Cu content

Dielectric constant: The variation of dielectric constant for the different concentrations of Cu content calculated and shown in the table 5. It is found that the value of dielectric constant slowly increased initially up to a Cu content of x=0.10 and then increases sharply thereafter at higher Cu concentrations

Table5: variation of dielectric constant at different Cu^{+3} concentrations

Cu Content	Dielectric Constant
0.00	18180
0.05	18681
0.10	19103
0.15	31488
0.20	40592
0.25	44654

It is very clean from the values of dielectric constant as shown in Fig.6 that the dielectric constant of the material increases by three orders in magnitude

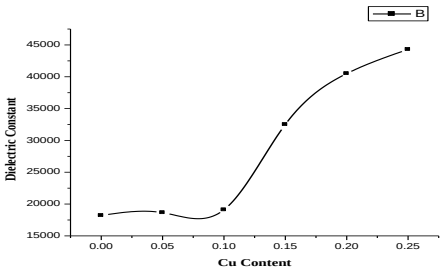

Fig.6 Variation of Dielectric constant with Cu content

Dielectric loss: Dielectric loss have been calculated and shown in the table6. Variation in Dielectric loss with increasing Cu concentrations is presented.

Table6: Dielectric loss for different concentrations of Cu^{+3}

Cu Content	Dielectric loss
0.00	0.039
0.05	0.025
0.10	0.031
0.15	0.034
0.20	0.030
0.25	0.031

It is evident from the Fig.7 that the dielectric loss decreased in the beginning and then increased with the increase of Cu concentration. The trend observed in the present study which is in agreement with the reported behavior.

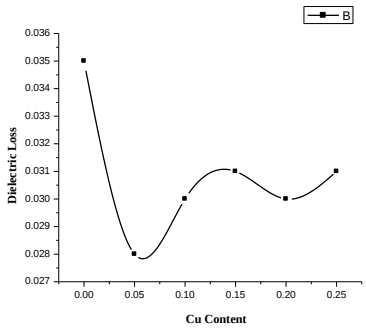

Fig.7 Variation of Dielectric loss with Cu content

Saturation Magnetization: The studies on Saturation Magnetization with the variation of dopant content are carried and calculated as shown in the Table 7 The change in Saturation Magnetization with dopant concentration is given in fig.8. The gradual decrease in saturation magnetization is observed with the Cu content. The variation of Saturation Magnetization can be explained on the basis of exchange interaction of cataions like Fe+3-O-2-Fe+3.The obtained value for basic composition is 71emu/gm which is almost reported

value [15].And further it is observed a systematic decrease in Saturation Magnetization in all samples due to addition of Cu Content

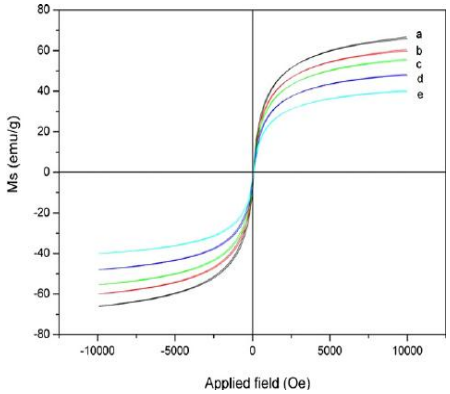

Table.7: variation of Saturation Magnetization of cu content.

Cu Content	Saturation Magnetization(emu/gm)
0.00	76
0.05	66
0.10	59
0.15	54
0.20	49
0.25	40

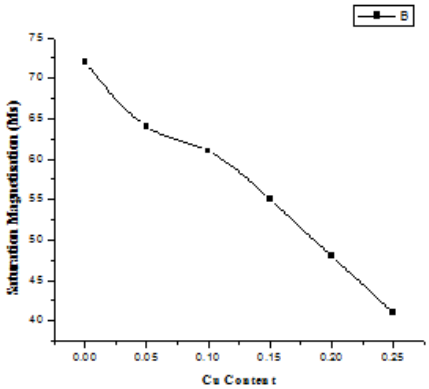

Fig.8 Variation of Saturation Magnetization with Cu content

IV. Conclusions

Samples of $Ni_{0.5}Zn_{0.5}$ Fe_2O_4 has been successfully synthesized using Citrate gel method. It is observed that the electrical properties like dielectric constant and dielectric loss changes with increasing Cu concentration. The d.c. resistivity changes with respect to temperature. Magnetic properties like curie temperature and Saturation magnetization also changes with increasing Cu content. The lattice constant also decreases with Cu Content.

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