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STRUCTURAL AND MAGNETIC PROPERTIES OF Cu SUBSTITUTED Ni-Cu-Zn FERRITES

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ABSTRACT

Fe- deficient Ni-Cu-Zn ferrites with composition $Ni_{0.28}Cu_{0.10+x}Zn_{0.62-x}Fe_{1.98}O_4$ (where x = 0.00, 0.03, 0.06, 0.09 & 0.12) were prepared by the standard double sintering ceramic technique using high purity Fe_2O_3, CuO, ZnO and NiO. The samples were sintered at 1150°C for 3 hrs. The analysis of XRD patterns indicates that the samples have the single phase cubic spinel structure. The lattice parameter 'a' is found to decrease linearly with the increase of Cu content following the Vegard's law. The bulk density is lower than the X-ray density. This may be due to existence of pores, which were formed and developed during the sample preparation or the sintering process. Density is observed to increase with increasing Cu content whereas the porosity increases. The compositional dependence of Curie temperature T_c was obtained from the permeability versus temperature plots of different compositions. The Curie temperature was observed to increase with increasing Cu content.

Keywords: *Ferrites, Lattice parameter, Porosity, Bulk density, Curie temperature.*

1. INTRODUCTION

Polycrystalline spinel ferrites present an important set of properties: high permeability in the radio frequency level, the high Curie point, more stable, high electrical resitivity, low eddy current, low dielectric losses, low cost and environmental stability (Ovidiu Caltun *et al.,* 2001; Goldman *et al*., 1999; Akther Hossain *et al*., 2007; Rajath *et al*., 2008; Lee *et al.,* 1998). Ferrites exhibit a substantial spontaneous magnetization at room temperature, like the normal ferromagnetic materials. They have two unequal sub-lattices called tetrahedral (A-site) and octahedral (B- site) and are ordered antiparallel to each other. In ferrites the cations occupy the tetrahedral (A) and octahedral (B) sites of the cubic spinel lattice and experience competing nearest neighbour exchange interaction (J_{AB}) and the next nearest neighbour (J_{AA} and J_{BB}) interactions with $|J_{AB}| >> |J_{BB}| > |J_{AA}|$.

The magnetic properties of ferrites are dependent on the type of magnetic ions residing on the A and B sites and the relative strengths of the inter (J_{AB}) and intrasublattice (J_{BB}, J_{AA}) interactions. Most of the magnetic spinels are inverse type like Ni Fe_2O_4, Cu Fe_2O_4, Mn Fe_2O_4, etc . Zn Fe_2O_4 is a normal spinel. Ni Cu Zn ferrite is a solid solution of inverse and normal ferrites. Due to the favorable fit of charge distribution Ni^{2+} and Cu^{2+} ions show strong preference of B sites. Zn^{2+} ions show a strong preference for A sites due to electron configuration. The net effect is the significant increase in magnetic moment in octahedral site as well as in the unit cell. The present work focuses on the effect of Cu substitution on the structural and magnetic properties of Ni-Cu-Zn ferrite.

2. EXPERIMENTAL DETAILS

Cu-substituted Ni-Cu-Zn ferrites $Ni_{0.28}Cu_{0.10+x}Zn_{0.62-x}Fe_{1.98}O_4$ where x = 0.00, 0.03, 0.06, 0.09 and 0.12 sintered at 1150°C for 3hrs are prepared by the standard double sintering ceramic methods at the materials Science Division, Atomic Energy Centre, Dhaka. Single-phase cubic spinel structure was confirmed by XRD using a Phillps (PW 3040) X'pert PRO X-ray diffractometer. From the XRD data, the lattice constant was determined by using Nelson-Riley function and then X-ray density is determined using the lattice constant. The bulk density was calculated by considering the cylindrical shape of the pellets. The Curie temperature (T_c) was measured on the wound toroid-shape samples as a function of frequency and temperature using an HP4192 impedance analyzer.

3. RESULTS AND DISCUSSIONS

A phase analysis using XRD technique was performed to confirm the formation of single-phase cubic spinel structure as shown in Fig-1. The XRD pattern of all the samples of $Ni_{0.28}Cu_{0.10+x}Zn_{0.62-x}Fe_{1.98}O_4$ with the [hkl] values corresponding to the diffraction peaks of different planes (111), (220), (311), (222), (400) (422), (511) and (440), which represent either odd or even indicating the sample are spinel phase. All the samples show good crystallization with well defined diffraction lines with no extra lines corresponding to confirm the formation of single phase spinel structure. All the samples with increasing trend of the lattice parameter as the

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Cu content is increased. Since no ambiguous reflection other then the spinel structures is evidenced, this demonstrates the homogeneity of the prepared samples.

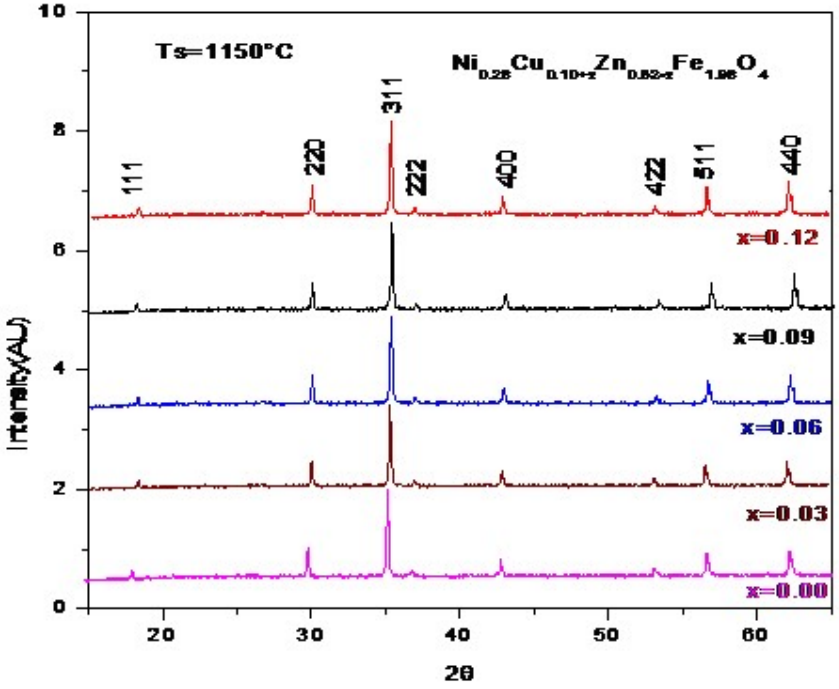

Figure 1: XRD patterns of $Ni_{0.28}Cu_{0.10+x}Zn_{0.62-x}Fe_{1.98}O_4$ ferrites sintered at 1150°C

The lattice parameter 'a' and the X-ray densities were calculated by using the X-ray data. The average values of 'a' were found by plotting 'a' against Nelson-Riley function.The lattice parameter of all the samples has been precisely determined considering the reflection with the Cu-K_α component using the extrapolated Nelson- Riley function F(θ) = 0 at θ=90° (J. B. Nelson *et. al.* 1945), $F(\theta)=\frac{1}{2}\left[\frac{\cos^2\theta}{\sin^2\theta}+\frac{\cos^2\theta}{\theta}\right]$, where θ in the Bragg's angle.

Variation of lattice parameter with Cu content in shown in Fig 2 and also shown in Table 1. From the Fig 2 it is observed that the lattice parameter decreases linearly with increase of Cu content as the ionic radii of Cu^{2+} and Zn^{2+} are 0.70 Å and 0.82Å respectively. Since the radius of the substituted ions in smaller than that of the displaced ions which decrease the lattice constant.

The dependence of bulk density d_B and X-ray density d_x upon Cu content (x) is represented in Fig 3. The bulk density d_B was measured by usual volume and mass using the equation.

$$d_B=\frac{m}{v}=\frac{m}{\pi r^2 h},$$

Where as X-ray density d_x was calculated from the molecular weight and the volume of unit cell for each sample by using the relation

$$d_x=\frac{8m}{Na^3}gm/cm^3.$$

Where N is Avogadro number ($6.023\times10^{23}mol^{-1}$) and m is molecular weight in gm. The bulk density slightly increases with increasing Cu content as shown in Fig 3. The densities of Cu^{2+} and Zn^{2+} are 8.96 gm/cc and 7.14 gm/cc respectively. It was also observed that bulk density was less than x-ray density.

The percentage of porosity was also calculated using the equation

$$P=\left(1-\frac{d_B}{d_x}\right)\times100\%.$$

Porosity changes slightly with changes of Cu content (x). It is understood from the data from Table 1 porosity values increases significantly with increasing Cu concentration.

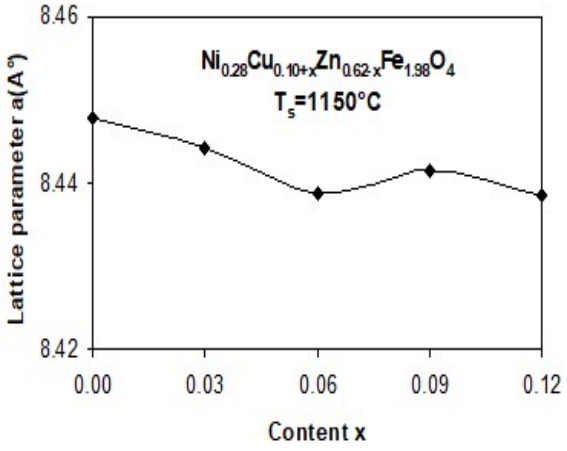

Figure 2: Variation of lattice constant 'a' as a function of Cu content (x) of $Ni_{0.28}Cu_{0.10+x}Zn_{0.62-x}Fe_{1.98}O_4$ ferrites sintered at 1150°C

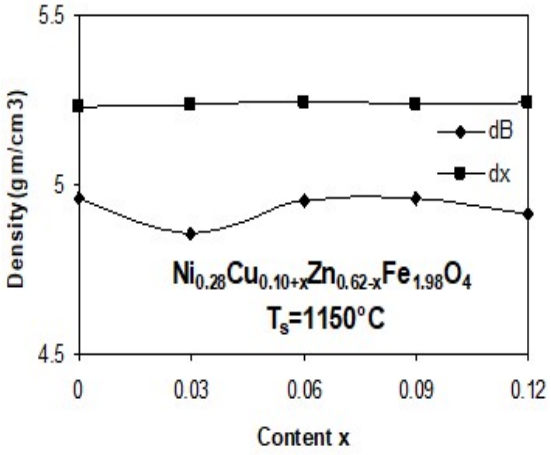

Figure 3: Variation of density with Cu content (x) of $Ni_{0.28}Cu_{0.10+x}Zn_{0.62-x}Fe_{1.98}O_4$ ferrites sintered at 1150°C

Fig. 5 shows the thermal variation of initial permeability (μ') for the toroid shaped samples found that the initial permeability increases with the increase in temperature, while it falls abruptly close to the Curie point. The Curie temperature T_c is a property which is relevant for inductor applications and very sensitive to composition.

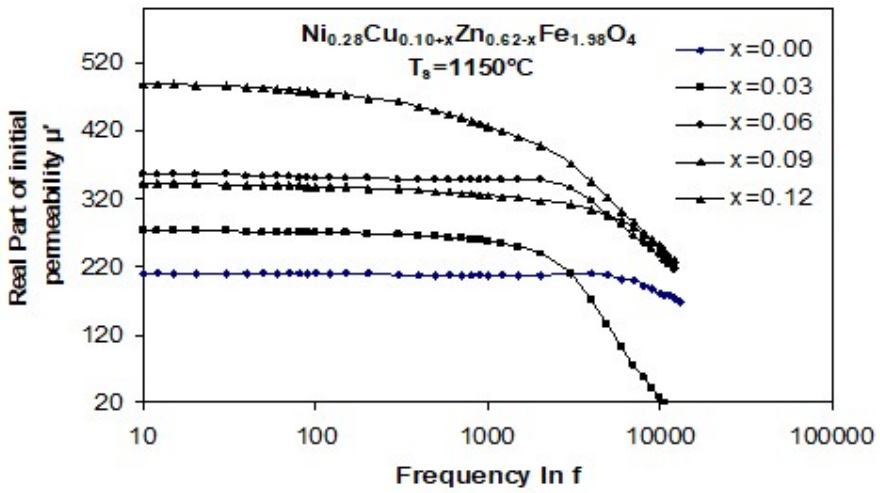

Figure 4: Variation of initial permeability as a function of frequency of $Ni_{0.28}Cu_{0.10+x}Zn_{0.62-x}Fe_{1.98}O_4$ ferrites sintered at 1150°C/3hrs

Table 1: Lattice parameter (a), x-ray density (d_x), bulk density (d_B), porosity (p%), Curie temperature (T_c) of $Ni_{0.28}Cu_{0.10+x}Zn_{0.62-x}Fe_{1.48}O_4$ samples.

Cu Content (x)	a(Å)	d_x (gm/cm^3)	d_B (gm/cm^3)	p%
0.00	8.4477	5.231	4.9598	5.1824
0.03	8.4441	5.236	4.8562	7.2622
0.06	8.4387	5.245	4.9559	5.5154
0.09	8.4416	5.238	4.9608	5.3034
0.12	8.4386	5.243	4.9176	6.2064

In Fig 5 and Fig 6 show the variation of T_c as a function of Cu content of $Ni_{0.28}Cu_{0.10+x}Zn_{0.62-x}Fe_{1.98}O_4$ system. It is observed that T_c increases continuously with the increase of Cu^{2+} content, It is known that in Ni-Cu-Zn ferrites Ni ions prefer octahedral B- sites, Zn ions prefer tetrahedral A- sites and Cu ions prefer octahedral B-

sites (Cullity 2nd ed. 2009). Substitution of Zn by Cu strengthening A-B interaction which increases the value of T_c.

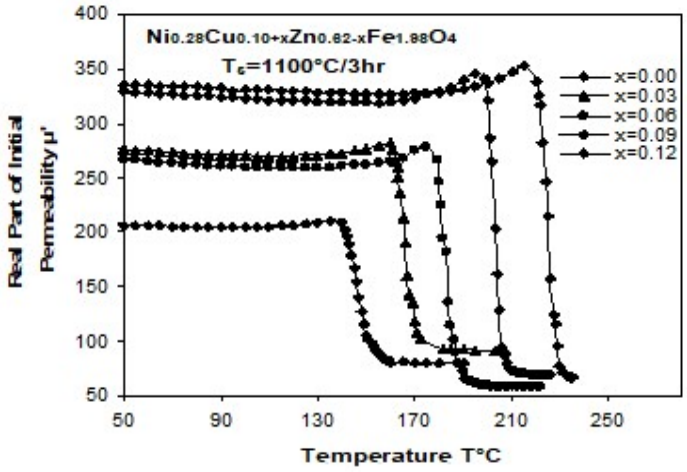

Figure 5: Temperature dependence of initial permeability (μ') for $Ni_{0.28}Cu_{0.10+x}Zn_{0.62-x}Fe_{1.98}O_4$ Sintered at 1150°C/3hr

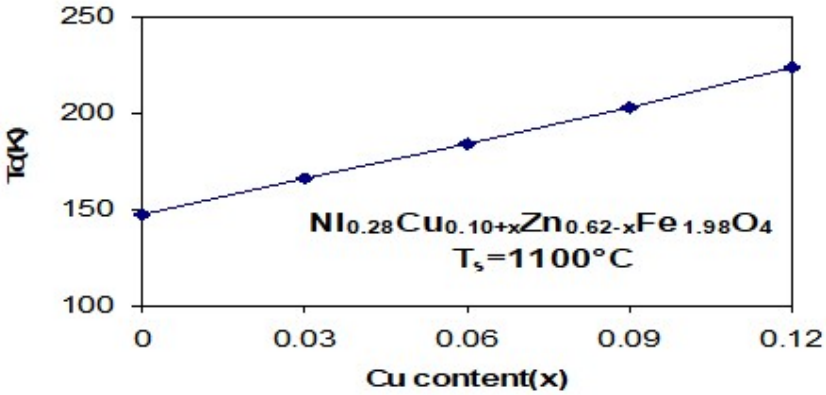

Figure 6: Variation of Currie temperature T_c with Cu content (x) of $Ni_{0.28}Cu_{0.10+x}Zn_{0.62-x}Fe_{1.98}O_4$ ferrites sintered at 1150°C/3hr

4. CONCLUSIONS

The XRD analysis reported in the study confirmed the single phase cubic spinel structure of the prepared samples. The lattice constant decreases with increasing Cu content. The bulk density was less than the X-ray density. Curie temperature and initial permeability increases as Cu^{2+} content increases. The initial permeability proportional changes with density.

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