

Structural and Magnetic Properties of Mn_{0.50-x}Zn_{0.50}Cu_xFe₂O₄

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ABSTRACT

 $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ (where x = 0.0 - 0.3) ferrites have been synthesized by auto combustion method. X-ray diffraction patterns reveal that all compositions are of single phase cubic spinel structure. The lattice parameter decreases with the increase in Cu^{2+} content obeying the Vegard's law. The bulk density, average grain size, initial permeability, Néel temperature and saturation magnetic induction of $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ increased with increasing Cu^{2+} content. It is observed that both density and initial permeability increase with increasing sintering temperature. The maximum initial permeability is found to be 1061 which is almost four times greater than that of the parent composition. The resonance frequency of all the samples shifts towards the lower frequency as the permeability increases with Cu^{2+} content. It is observed from B-H loops of $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ that coercivity decreases and retentivity increases with Cu^{2+} content. It observes are discussed.

Keywords: Combustion Method; Mn-Cu-Zn Ferrites; Initial Permeability; B-H Loop

1. Introduction

Polycrystalline spinel ferrites are technologically very important materials having potential applications and interesting physical properties. Mn-Zn and substituted Mn-Zn ferrites are pertinent magnetic materials due to their high permeability, high magnetization, relatively high Néel temperature, low losses, low cost and environmental stability. These ferrites have been widely used in electrical and magnetic devices for high frequency applications [1-4]. The physical and magnetic properties can be controlled by the preparation condition, chemical composition, sintering temperature and the amount of substitutions. Several investigations on the properties of Ni-Mn-Zn [5], Ni-Cu-Zn [6], Mg-Cu-Zn [7], Co-Mn-Zn [8] ferrites have been reported. It was found that the poor densification and slow grain growth rate can be remarkably improved and consequently initial permeability can be enhanced by the substitution of Cu [9]. No report has been found in the literature regarding the magnetic properties of Cu substituted Mn-Zn ferrites for Mn prepared by combustion method. In the present work, the influence of Cu^{2+} in place of Mn^{2+} on the properties of Mn-Zn ferrites has been investigated by studying the structure and some magnetic properties.

2. Experimental

The chemical compositions of $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ (with x = 0.0 - 0.3 at a step of 0.1) were prepared by combustion method. The stoichiometric amounts of commercially available analytical grade powders of $MnCl_2 \cdot 4H_2O$, $Cu(NO_3)_2 \cdot 3H_2O$, $Zn(NO_3)_2 \cdot 6H_2O$ and Fe (NO₃)₃ · 9H₂O were dissolved in ethanol to obtain a mixed homogenous solution. Ammonia solution was slowly added to adjust the pH at level 7. The solution was placed at constant temperature bath (70°C) followed by an ignition and formed a fluffy loose powders of the desired composition. The resultant powders were calcined at 700°C for five hours in air. The grounded fine powders were then pressed into disc- and toroid-shaped samples. The samples prepared from each composition were sin-

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tered at 1200°C, 1250°C and 1300°C for five hours in air. During sintering, temperature ramps were 10°C/min for heating and 5°C/min for cooling. The structural characterization was carried out with an X-ray diffractometer using CuK_{α} radiation ($\lambda = 1.54178$ Å). The lattice parameter was determined by using the Nelson-Riley extrapolation method [10]. The lattice parameters obtained from each reflected plane were plotted against Nelson-Riley function,

$$F(\theta) = 1/2 |\cos^2 \theta / \sin \theta + \cos^2 \theta / \theta|,$$

where θ is the Bragg's angle. The exact values of lattice constant, a_0 were estimated from the extrapolation of the best fitted line to $F(\theta) = 0$ or $\theta = 90^\circ$. The bulk density, ρ_B was determined using the expression

 $\rho_B = (W \times \rho)/W - W'$, where W and W' are the weight of the sample in air and water, respectively and ρ is the density of water at room temperature. The theoretical density, ρ_{th} was calculated using the relation:

 $\rho_{th} = (ZM/N_A a^3)$, where N_A is Avogadro's number, M is the molecular weight of the corresponding composition and Z is the number of molecules per unit cell, which is 8 for the spinel cubic structure. The porosity, P was calculated from the relation $P(\%) = \{(\rho_{th} - \rho_B) / \rho_{th}\} \times 100$. The micrographs of Mn_{0.50-x}Zn_{0.50}Cu_xFe₂O₄ were taken by a Scanning Electron Microscope (SEM). From these micrographs, the average grain size (D) was estimated by the linear intercept method. The frequency and temperature dependent complex initial permeability were measured using Wayne Kerr Impedance Analyzer (Model No.6500B) in the frequency range 100 Hz - 120 MHz. The real part (μ_i') and the imaginary part (μ_i'') of the complex initial permeability were calculated using the following relations $\mu_i' = L_s / L_o$, and $\mu_i'' = \mu_i' \tan \delta$, where L_s is the self inductance of the sample core and $L_o = (\mu_o N^2 h/2\pi) \ln(r_o/r_i)$, is derived geometrically. L_{a} is the inductance of the winding coil without the sample core, N is the number of turns of the coil (N = 4), h is the thickness, r_0 is the outer radius and r_i is the inner radius of the toroid-shaped sample. The relative quality factor (RQF) was calculated from the relation: $RQF = \mu_i^{\prime}/\tan \delta$, where $\tan \delta$ is the loss factor. B-H loops were measured at room temperature using an Automatic Magnetic Hysteresis Graph Tracer (Model no. AMH-300, Laboratorio Electrofisico).

3. Results and Discussion

3.1. Structural Analysis of Mn_{0.50-x}Zn_{0.50}Cu_xFe₂O₄

Figure 1 shows the X-ray diffraction (XRD) patterns for various $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ (with x = 0.0 - 0.3 at a step of 0.1) sintered at 1250°C. The positions of the peaks for the various compositions indicate a single phase cubic spinel crystal structure. It is seen that a_0 de-

creases linearly with increasing Cu²⁺ content and obeys Vegard's law [11].

The variation of a_0 as a function of Cu^{2+} content is shown in **Figure 2**. The values of a_0 are shown in **Table 1**. The decreasing a_0 with Cu^{2+} content may be explained in terms of ionic radii. As the ionic radius of Mn^{2+} (0.80 Å) ion is larger than that of Cu^{2+} (0.72 Å), a_0 decreases. [8,9,12]. As the smaller Cu^{2+} ions replace the bigger Mn^{2+} ions in $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$, the unit cell compresses without disturbing the symmetry of lattice [13].

A similar linear variation has been reported by Ravinder *et al.* [14]. The r-variant for $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ was calculated by the expression: *r-variant* = $(0.50-x)r_{Mn}$ + xr_{Cu} , where r_{Mn} and r_{Cu} are the radii of the Mn^{2+} and Cu^{2+} respectively. It is found that the r-variant decreases with increase in Cu^{2+} content which is also shown in **Figure 2**.

Figure 3 shows the effect of Cu²⁺ substitutions on ρ_{th} and ρ_B in Mn_{0.50-x}Zn_{0.50}Cu_xFe₂O₄ sintered at various sintering temperatures, T_s. It is found that ρ_{th} is larger in magnitude than corresponding ρ_B . This is because



Figure 1. XRD patterns of $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ sintered at 1250°C.



Figure 2. Variation of lattice parameter and r-variant of $Mn_{0.50\text{-}x}Zn_{0.50}Cu_xFe_2O_4$ with Cu^{2+} content.

x	Ts (°C)	$\overset{a_0}{(\mathrm{\AA})}$	$ ho_{th}$ (g/cm ³)	$ ho_{\scriptscriptstyle B}$ (g/cm ³)	P (%)	D (µm)	μ_i'	f_r (Hz)	B _r (T)	B _s (T)	H _c (A/m)	T _N (°C)
	1200			4.72	8.9	2.0	220	5.3×10^5	0.039	0.131	44	63
0.0	1250	8.4556	5.18	4.77	8.7		240	4.5×10^5				
	1300			4.85	7.7		285	4.2×10^5				
	1200			4.79	7.6	2.3	440	4.7×10^{5}	0.08	0.184	29	69
0.1	1250	8.4471	5.22	4.86	7.0		560	2.6×10^5				
	1300			4.95	6.0		600	2.3×10^{5}				
	1200			4.83	6.8	2.8	530	1.8×10^5	0.092	0.217	28	72
0.2	1250	8.4351	5.26	4.93	5.6		620	$1.7 imes 10^5$				
	1300			4.99	4.8		800	1.4×10^5				
	1200			4.87	6.1	3.5	675	1.0×10^5	0.098	0.228	27	79
0.3	1250	8.4212	5.30	4.98	4.5		800	9.0×10^4				
	1300			5.04	4.2		900	7.5×10^4				

Table 1. Lattice parameter, density, porosity, average grain size, initial permeability, resonance frequency, residual and saturation inductions, coercive field and Néel temperature of various $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ sintered at different sintering temperatures.

some pores may be developed in the samples during sintering process. It is also found that both ρ_{th} and ρ_B increase with increase in Cu²⁺ content while the *P* follows the opposite trend. The increase in ρ_B can be attributed to the difference in atomic weight and specific gravity of the Cu and Mn. The atomic weight and specific gravity of Cu (63.55 amu and 8.94 g·cm⁻³) is greater than that of Mn (55.94 amu and 7.21 g·cm⁻³) [15]. Also Cu²⁺ promotes densification of Mn_{0.50-x}Zn_{0.50}Cu_xFe₂O₄ during low sintering temperature. These combined factors may contribute to the enhancement of ρ_B for various

 $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ [16,17]. Similar results have been reported by Haque *et al.* [7]. On the other hand, *P* of $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ reduces due to increasing ρ_B . It is revealed from **Figures 3(a)** and **(b)** that ρ_B increases and *P* decreases with increasing T_s for various

 $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$. This is because during the sintering process, the thermal energy generates a force that drives the grain boundaries to grow over pores, thereby decreasing the pore volume and increases the density of the compositions. These results agree well with the reported value [7,18]. The increase in ρ_B and decrease in *P* for various $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ with the addition of Cu^{2+} for various T_s are tabulated in **Table 1**.

3.2. Microstructure of Mn_{0.50-x}Zn_{0.50}Cu_xFe₂O₄

The scanning electron micrographs as shown in **Figure 4** reveal that the grain size is influenced by the Cu²⁺ substitution, where the grain size increases with increase in Cu²⁺ content in $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ sintered at 1200°C. The D of $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ varies from 2 µm to 4 µm at constant T_s.



Figure 3. Variation of (a) density and (b) porosity of $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ as a function of Cu^{2+} content.

The values of D are presented in **Table 1**. Figure 5 shows the micrographs of $Mn_{0.40}Zn_{0.50}Cu_{0.10}Fe_2O_4$ at different T_s . It is seen that the D increases with increasing T_s . The grain size increases with increase in Cu^{2+} due to the melting point of copper (1084°C) is less than that of manganese (1246°C). During sintering Cu^{2+} influences

the microstructure by the formation of liquid phase. It facilitates the grain growth and grain growth reflects the competition between the driving force for grain boundary movement and the retarding force exerted by pores [19, 20]. D increases with T_s because of homogeneous grain growth.

3.3. Magnetic Properties of Mn_{0.50-x}Zn_{0.50}Cu_xFe₂O₄

3.3.1. Frequency Dependent Complex Initial Permeability

Figure 6 shows the complex initial permeability spectra



x = 0.0

x = 0.1



Figure 4. Scanning electron micrographs of Mn_{0.50-x}Zn_{0.50}Cu_xFe₂O₄ sintered at 1200°C.



1200°C



1250°C



Figure 5. Scanning electron micrographs of Mn_{0.40}Zn_{0.50}Cu_{0.10}Fe₂O₄ for various sintering temperatures.

for all $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ (where x = 0.0 to 0.3 at a step of 0.1) sintered at 1200°C, 1250°C and 1300°C, respectively as a function of frequency.

The complex permeability is given by $\mu = \mu_i^{\prime} - \mu_i^{\prime\prime}$. Here μ_i^{\prime} describes the stored energy expressing the component of magnetic induction B in phase and $\mu_i^{\prime\prime}$ describes the dissipation of energy expressing the component 90° out of phase with the alternating magnetic field H. It is found that μ'_i increases with increasing Cu^{2^+} content for various $Mn_{0.50\text{-}x}Zn_{0.50}Cu_xFe_2O_4$ at constant T_s. It is also observed that μ_i^{\prime} increases with increasing T_s. Highest μ_i^{\prime} was obtained 1061 for

Mn_{0.20}Zn_{0.50}Cu_{0.30}Fe₂O₄ at frequency 103 Hz sintered at 1300°C. μ_i^{\prime} remains almost constant in the frequency range up to a certain frequency, which is called the resonance frequency, f_r . There is a decrease in μ_i^{\prime} and increase in $\mu_i^{\prime\prime}$ above f_r observed for various

 $Mn_{0.50-x}Zn_{0.50}Cu_{x}Fe_{2}O_{4}$. The f_{r} is the range of frequency of the compositions up to which these can be used efficiently. The values of μ_i^{\prime} and f_r for all samples sintered at various T_s are presented in **Table 1**.

It is observed from **Figure 7** that the μ_i^{\prime} increases and f_r shifted towards the lower frequency with Cu²⁺ in Mn_{0.50-x}Zn_{0.50}Cu_xFe₂O₄ at different T_s. The increasing μ_i and shifting f_r to lower frequency at constant T_s



Figure 6. Frequency dependent Complex initial permeability of Mn_{0.50-x}Zn_{0.50}Cu_xFe₂O₄ at various T_s.

800

600

`∃⊺

400

200

follows the Snoek's relation [21]. The μ_i^{\prime} of ferrites depends on many factors like reversible domain wall displacement, the amount and the type of dopant ions, D and intragranular porosity, etc [22-24]. It is well known that μ'_i of polycrystalline ferrite is related to two magnetizing mechanisms: domain wall motion and spin rotation [25-27]. Globus et al. [22] studied several Ni-Zn ferrites and found a linear relationship between μ_i^{\prime} and D. Kakaktar *et al.* [28] studied the effect of D on μ_i and found that $\mu'_i \propto D$. The μ'_i can be expressed by Globus-Duplex relation $\mu_i' \propto \left(M_s^2 D / \sqrt{K_1}\right)$, where M_s is the saturation magnetization; K_1 is the anisotropy constant [22]. In our present study of microstructure, it is seen that the D increases significantly with Cu²⁺ content. Therefore, the increase of μ_i' with increasing Cu²⁺ content is justified. The increasing μ'_i with T_s is due to increase in both ρ_B and D.

Figure 8 shows the variation of μ'_i with Cu²⁺ content at different frequencies for various

 $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ sintered at 1200°C, 1250°C and 1300°C. It is found that μ'_i decreases at higher frequencies. This is due to the fact that at higher frequencies impurities between grains and intragranular pores act as pinning points and increasingly hinder the motion of spin and domain walls thereby decreasing their contribution to permeability and also increasing the loss [22].

For practical application the quality factor is often used as a measure of performance. The RQF increases with an increase of frequency, showing a peak and then decreases with further increase in frequency as shown in **Figure 9**. The variation of RQF with frequency showed a similar trend for all the samples. It is observed that the sample sintered at 1250°C has the highest RQF (5420) for $Mn_{0.40}Zn_{0.50}Cu_{0.10}Fe_2O_4$. The highest RQF for $Mn_{0.20}Zn_{0.50}Cu_{0.30}Fe_2O_4$ is 5420 sintered at 1250°C. This

is probably due to the growth of less imperfection and defects compared to those of other samples [9].



Figure 7. The variation of initial permeability and resonance frequency of $Mn_{0.50}$ - $_xZn_{0.50}Cu_xFe_2O_4$ at different T_s .



Figure 8. The initial permeability of $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ with Cu^{2+} content at different frequencies sintered at (a) 1200°C, (b) 1250°C and (c) 1300°C.

(c)

0.1 0.2 Cu content, x

03

0.0



Figure 9. The RQF of $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ with Cu^{2+} content sintered at 1200°C, 1250°C and 1300°C.

3.3.2. Temperature Dependent Permeability

The μ_i^{\prime} as a function of temperature for various $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ sintered at 1200°C is shown in **Figure 10(a)**.

It is observed that there is a sudden drop in μ'_i at Néel temperature, T_N , where the magnetic state of the ferrite changes from ferrimagnetic to paramagnetic state. This is because at T_N , the thermal agitation is so high that it reduces the alignment of the magnetic moment along a given axis to zero [29]. **Figure 10(b)** shows the variation of T_N with Cu²⁺ content. It is observed that T_N increases

835



Figure 10. (a) The temperature dependent initial permeability and (b) Néel temperature of $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ with Cu^{2+} content sintered at 1200°C.

With increase in Cu^{2+} substitution for Mn^{2+} in $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ due to strengthening of A-B interaction between the two sublattices. This could be attributed to the decrease in distance (hooping length) between the magnetic ions of A- and B-sites and is confirmed by the decrease in the lattice parameter with increase in Cu^{2+} content as shown in **Figure 2**. The shorter distance between magnetic cations leads to the increase in A-B interaction and consequently T_N increases.

3.3.3. B-H Loops of Mn_{0.50-x}Zn_{0.50}Cu_xFe₂O₄

Figure 11(a) shows the B-H loops of

 $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ sintered at 1200°C at constant frequency 1000 Hz and **Figure 11(b)** is the B-H loops for low field.

The magnetic induction, B of all samples increases linearly with increase in applied magnetic field up to 120 A/m (depending on compositions). Beyond 120 A/m, B increases slowly and then reaches to the saturation induction, B_s . Increasing B with applied low field H indicates that all compositions are in ferromagnetic state at room temperature. From the loops B_s has been calculated. It is observed that B_s increases with increasing Cu^{2+} content which is shown in **Figure 12(a)**.

The variation in saturation magnetization with Cu^{2+} content could be explained by cation distribution and exchange interaction. It is well known that Zn^{2+} ion has strong A-site occupancy, Mn^{2+} ions prefer to go both sites (80% A- and 20% B-sites) [16]. Also Cu^{2+} and Fe^{3+}

ions can occupy both A- and B-sites. Substitution of Cu^{2+} in place of Mn^{2+} results into migration of some of Fe^{3+} from A- to B-site. The change in B_r (residual induction) and B_s with Cu^{2+} content are shown in **Figure 12(b**). The coercive field (H_c) for each sample has been measured from the B-H loops of $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ which is shown in **Figure 12(b**). It is observed that H_c decreases



Figure 11. (a) B-H loops of $Mn_{0.50-x}Zn_{0.50}Cu_xFe_2O_4$ (b) B-H loops at low field sintered at 1200 °C.



Figure 12. (a) The B_s and B_r and (b) Coercive field of $Mn_{0.50\mathchar`s}Zn_{0.50}Cu_xFe_2O_4$ with Cu^{2+} content sintered at 1200°C.

with increase in Cu^{2+} content, whereas μ'_i increases. Mazen and Abu-Elsaad reported that H_c is directed to gradually decreasing with D [29]. Our experimental result is in agreement with the reported value. Therefore, samples having larger grains are expected to have lower H_c .

4. Conclusion

Substitution of Cu²⁺ causes appreciable change in the structural and magnetic properties of the Mn-Cu-Zn ferrites. The XRD patterns confirm single phase cubic spinel structure of Mn_{0.50-x}Zn_{0.50}Cu_xFe₂O₄. The *a*₀ decreases with the increase in Cu²⁺ content while ρ_B , D, μ'_i and T_N increase with the increase in Cu²⁺ content. μ'_i is greatly enhanced from 258 to 1061 (~400%). The highest RQF (5420) has been found for the sample

 $Mn_{0.20}Zn_{0.50}Cu_{0.30}Fe_2O_4$ sintered at 1250°C. These ferrites are important and suitable for technological applications because of their high permeability.

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