# Synthesis and Study of Temperature Dependent Magnetic properties of $Ni^{2+}$ Substituted Mg-Cu-Zn Ferrites Synthesized by Molten Salt Method

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*Abstract:*

Mixed Ferrites with general formula$Mg0.25-_xNi_xCu_{0.25}Zn_{0.5}Fe_2O_4$ (x= 0, 0.05, 0.1, 0.15, 0.2 and 0.25). are investigated. X-ray diffraction study of these compositions revealed the formation of single phase spinels.Study of initial permeability with temperature revealed that the long range ferromagnetic ordering in the compounds having x=0.4 . The sample with x<0.4 and y=0.05 and 0.10 show peaking behavior near curie temperature .temperature dependence normalized AC susceptibility and curie temperature of spinel ferrites study reveals that $MgFe_2O_4$ exhibits multi domain (MD) occurs.

Keywords : Mg-Cu-Zn ferrite, curie temperature etc.

## I. Introduction

Magnetic properties like permeability and susceptibility depends on grain size, grain boundary and domain structure [1]. Domain wall motion and spin rotation contributes to permeability.. The MD particles have domain walls [2] and magnetic changes takes place due to domain wall (DW) motion. SP particle nature reduces magnetic character of the material.$Ni^{2+}$ substitution is interesting substitution in the spinels Addition of $Ni^{2+}$ in$Mg0.25-_xNi_xCu_{0.25}Zn_{0.5}Fe_2O_4$The domain structure changes from MD to SD [3].

In the present investigation the efforts are made to understand the effect of $Ni^{2+}$Substitution on permeability and susceptibility of Mg-Cu-Zn ferrite.

## II. Experimental

A molten salt synthesis method was used for preparing ferrite powder with composition $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ (x= 0, 0.05, 0.1, 0.15, 0.2 and 0.25). A molten-salt synthesis (MSS) method is one of the simplest, most versatile, and cost-effective method used for obtaining crystalline, chemically purified, single phase powders at lower temperature and obtained in a short reaction time with little residual impurities as compared to conventional solid-state reactions [4].

Analytical reagent grade magnesium sulphate, nickel sulphate, copper sulphate, zinc sulphate and ferric nitrate were used as starting precursors. The divalent metal precusors (sulphates), ferric nitrate, NaOH and NaCl were mixed in the molar ratio (1:2:8:10) and then grounded together in an agate mortar for about 30 min. The reaction was exothermic and started spontaneously. As the reaction proceeded, the mixture became mushy and underwent gradual changes in color. The as prepared samples were collected and washed several times with distilled water to remove unwanted reaction by-products. The as prepared powders were calcinated at 700 °C for 2 h.

Further the calcinated powder was used to form the pellets. Initially the calcinated powder was mixed with PVA (organic binder) and grounded in agate mortar. Pellets were formed using a die having 1.5 cm diameter and a hydraulic press machine by applying a pressure of 1.5 ton/cm$^2$. The toroids of samples were fabricated using a die having outer diameter 2.5 cm, inner diameter 1.5 cm with average height of 0.3 cm with hydraulic press machine by applying a pressure of 1.5 ton/cm$^2$. All the samples in present study were sintered at 950 °C for 4 h in air atmosphere.Powder samples were characterized by X-ray diffraction (XRD)on BRUKER D8 advanced X-ray diffractometer using Cu-Kα (λ=1.54056 Å) radiation at 2θ values between 20°and 80°.Low field AC susceptibility measurements were carried out in the temperature range 300k-700k by using double coil set up operating at 323 Hz and 100 Oeof applied field.

The Curie temperature values ($T_c$) were determined from the temperature variations of susceptibility and initial permeability curves.

## III. RESULTS AND DISCUSSION

The presence of the highest intensity peak corresponding to (3 1 1) plane and peaks corresponding to other planes like (2 2 0), (2 2 2), (4 0 0), (4 2 2), (5 1 1) (4 4 0) and (5 3 1) confirmed the formation of cubic spinel structure. No second phase was detected in the XRD patterns of the samples studied in the present work. The broad and well resolved peaks in the XRD patterns, clearly demonstrated fine particles of polycrystalline Ni doped Mg-Cu-Zn ferrite. Fig 1 shows the 0.25 composite of XRD spectrum.

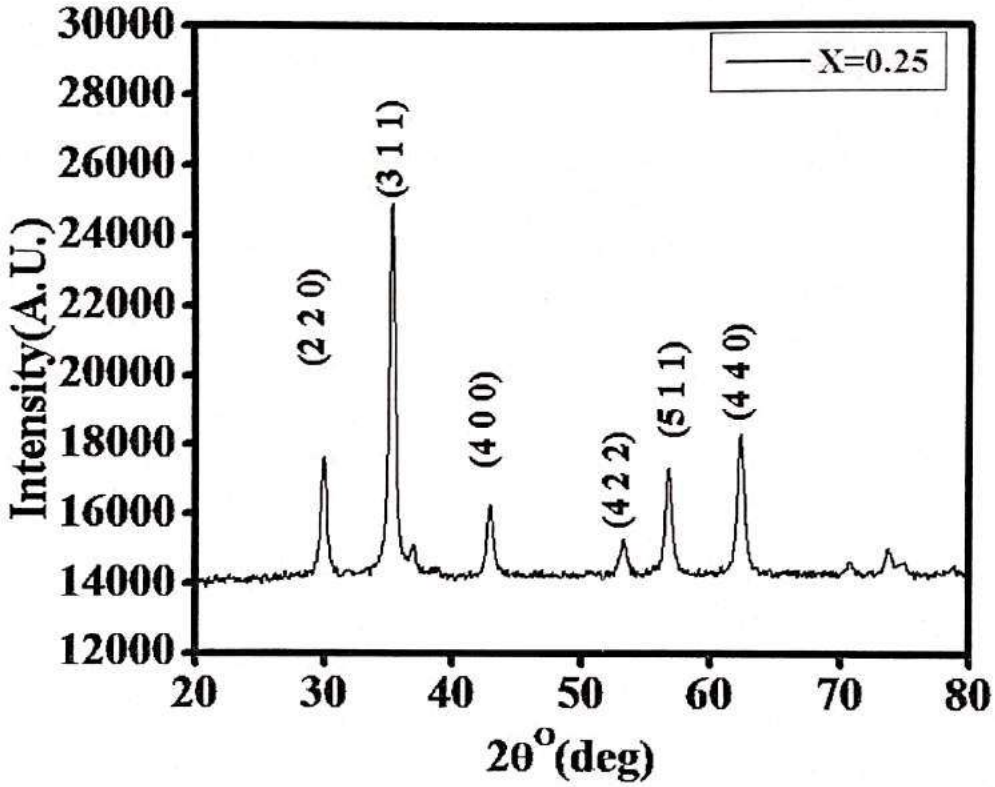


**Figure 1:- XRD spectra of 0.25 composite.**

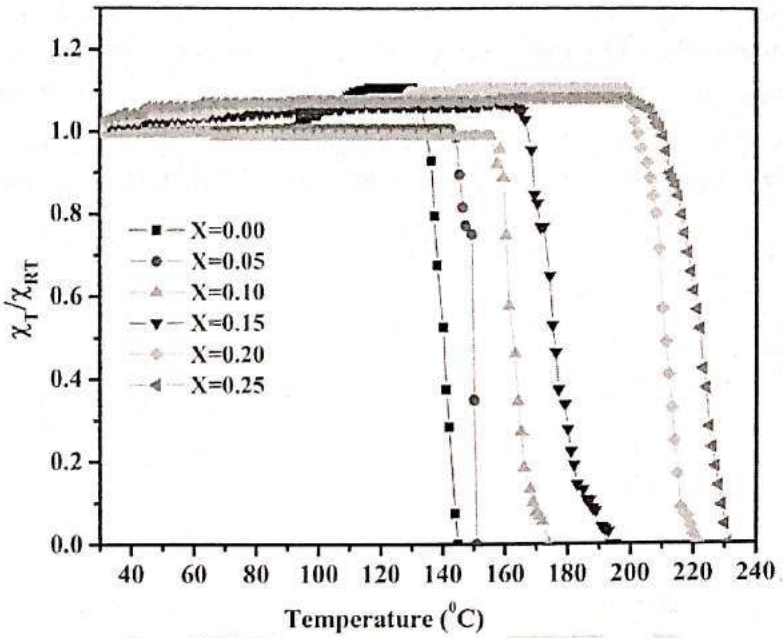


**Figure 2: Variation of normalized AC susceptibility ($\chi_T/\chi_{RT}$) as a function of temperature for sintered $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ ferrites.**

The temperature variation of normalized AC susceptibility of the sintered $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ ferrites samples is shown in Fig.2. The normalized susceptibility for all the samples remains invariant and exhibited sudden drop near the Curie temperature. The decreasing trend in susceptibility with temperature confirmed the ferrimagnetic nature of the material. Upto Curie temperature the available thermal energy was not quite sufficient to disturb the aligned moments of the spins. However, near the transition temperature, the thermal energy is high enough to disturb all the aligned spins where the susceptibility decreases drastically, reaching the paramagnetic region after the transition temperature i.e. Curie temperature.

| X | $M_s$ (emu) | $M_r/M_s$ | $n_B$Expt. | $\rho_R$ (%) | $H_c$ G | D μm | $T_C$(°C) $\chi_{a.c.}$ |
|---|---|---|---|---|---|---|---|
| 0.00 | 220 | 0.0076 | 1.76 | 88 | 6.97 | 0.91 | 145 |
| 0.05 | 240 | 0.0090 | 1.91 | 96 | 8.92 | 1.48 | 151 |
| 0.10 | 248 | 0.0100 | 1.97 | 95 | 10.37 | 0.98 | 174 |
| 0.15 | 270 | 0.0170 | 2.15 | 94 | 17.13 | 2.68 | 194 |
| 0.20 | 288 | 0.0180 | 2.28 | 90 | 18.89 | 1.11 | 220 |
| 0.25 | 325 | 0.0330 | 2.58 | 89 | 36.86 | 1.03 | 231 |

**Table 1: Data on magnetic parameters like $M_s$, Mr/Ms, $n_B$, Hc and relative density ($\rho_R$) and Curie temperature ($T_C$) for different compositions of sintered $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ ferrites.**

Also the Curie temperature is the measure of relative weighted magnetic interactions per formula unit. The Curie temperature for all samples is given in Table 1. From the table it is clear that, the Curie temperature increased with increase in nickel content. This increase in Curie temperature is attributed to the enhancement in ferrimagnetic region (ordered state) at the expense of the paramagnetic region which increases the exchange interaction. In general for ferrites the magnetic properties are

decided by AB interaction. In the mixed compositions, this exchange interaction increases due to the replacement of $Mg^{2+}$ ions by $Ni^{2+}$ ions in B sites. The occupation of $Ni^{2+}$ ions in the B sites will increase the number of $Fe^{3+}$ A –$Fe^{3+}$ B exchange which results in increase in the Curie temperature [5]. The room temperature susceptibility was found to increase with addition of Ni content which may be due to the replacement of $Mg^{2+}$ by $Ni^{2+}$ having high magnetic moment.The magnetic moment in ferrites is mainly from the parallel-uncompensated electron spin of the individual ions, and the spin alignments in the two sublattices are arranged antiparallel. Also, the A–B exchange interaction is predominant over the A–A and B–B interactions. Hence, the net magnetic moment of the lattice is given by the sum of the magnetic moments of A and B sublattices, i.e. $M = M_B - M_A$ [6].

As mentioned above, the variation of initial permeability of Mg-Cu-Zn ferrite was influenced not only by microstructures, but also by magnetostriction, inner stress and other factors. Within a limited Ni substitution, the improvement of magnetic properties of Mg-Cu-Zn ferrites is mainly attributed to the low magnetocrystalline constant, comparatively high saturation magnetization and grain size. The $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ferrite with low Ni content (x=0.05) exhibited high initial permeability as compared to $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ with high Ni content and similar type of observations were reported earlier [7].

| X | $\mu_i$ | $\mu_{rk}$ | $\mu_w$ | D (μm) | $K_1 \times 10^4$ (erg/cc) | Curie temperature Tc(°C) μi |
|---|---|---|---|---|---|---|
| 0.00 | 154 | 12 | 143 | 0.91 | -2.50 | 150 |
| 0.05 | 592 | 14 | 579 | 1.48 | -2.63 | 160 |
| 0.10 | 565 | 14 | 552 | 0.98 | -2.77 | 180 |
| 0.15 | 197 | 16 | 182 | 2.68 | -2.90 | 210 |
| 0.20 | 181 | 17 | 165 | 1.11 | -3.04 | 220 |
| 0.25 | 177 | 21 | 157 | 1.03 | -3.17 | 230 |

**Table 2: Data on initial permeability ($\mu_i$), rotational permeability ($\mu_{rot}$), wall permeability ($\mu_w$) grain size (D) and magneto crystalline anisotropy constant ($K_1$).**

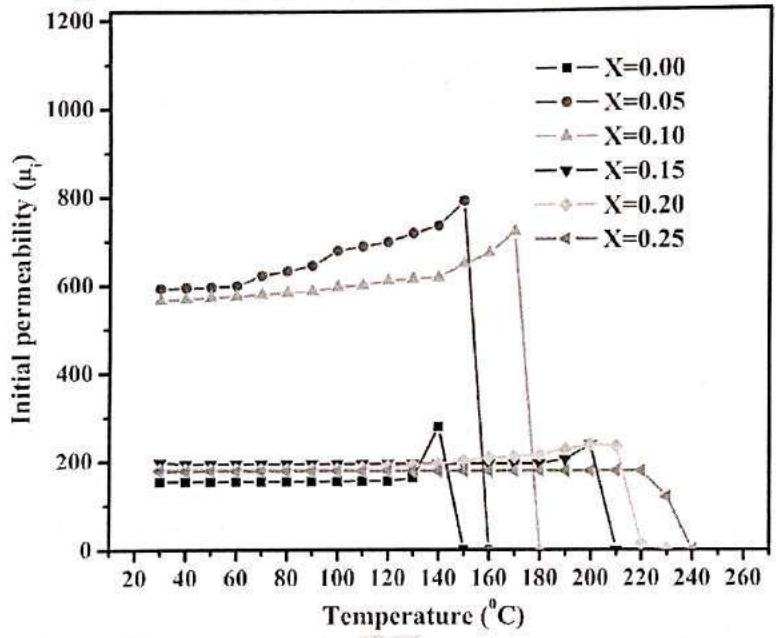


**Figure 3: Temperature dependency of permeability for different compositions of sintered $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ ferrites.**

Fig. 3. shows the temperature dependence of initial permeability of sintered $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ ferrites. It can be noted from the figure that as the temperature increases the initial permeability remains constant up to a certain temperature and increases to a peak value and then falls abruptly to a minimum value. The temperature at which this abrupt fall takes place is the magnetic Curie transition temperature ($T_c$). The Curie transition temperature is usually ascribed as the magnetic transition from ferrimagnetic state to the paramagnetic state. As mentioned earlier, $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ ferrite with x=0.05 exhibited maximum initial permeability and with further increase in Ni content the permeability decreased. The Curie transition temperature for all samples is given in Table 2.

It is observed that the magnitude of Curie transition temperature increased with increase in Ni content and is attributed to the replacement of non-magnetic $Mg^{2+}$ ion by $Ni^{2+}$ ions [8]. The values of rotational permeability and wall permeability have been calculated using formula [9]

$$\mu_{rk} = 1 + 2\pi M^2 s / lk_1$$

$$\mu_w = \mu_i - (\mu_{rk} - 1)$$

Where, Ms – Saturation magnetization, $K_1$ – anisotropy constant, $\mu_{rk}$ – rotational permeability and $\mu_w$- wall permeability.

| X | $\mu_{ih}$ | $\mu_{ic}$ | $\Delta\mu_i$ |
|---|---|---|---|
| 0.00 | 278 | 289 | 11 |
| 0.05 | 790 | 803 | 13 |
| 0.10 | 720 | 734 | 14 |
| 0.15 | 237 | 259 | 22 |
| 0.20 | 317 | 342 | 25 |
| 0.25 | 167 | 198 | 31 |

**Table 3.: Thermal hysteresis of initial permeability for $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ ferrites**

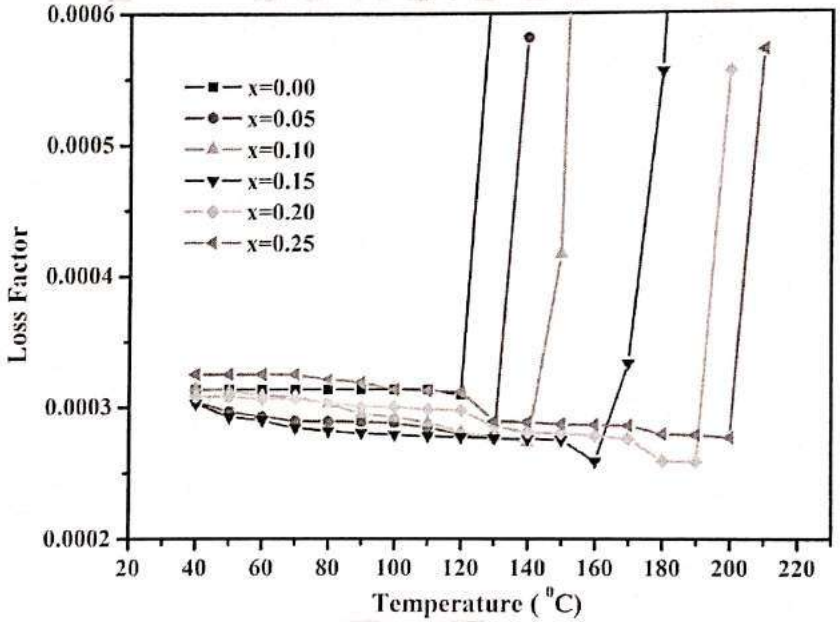


**Figure4.: Temperature dependency of loss factor for different compositions of sintered $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ ferrites.**

Form table 3.it is clearly seen that the magnitude of wall permeability $\mu_w$ is very large in comparison with rotational permeability $\mu_{rk}$ for all compositions of $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ ferrite. Thus, it is concluded that the main contribution to the initial permeability is due to domain wall motion. Hence Globus model is applicable and according to Globus model [10]

$$\mu_i \approx \frac{M_s^2 D}{K_1}$$

where $\mu_i$ is the initial permeability, Ms is the saturation magnetization, D is the average grain size and $K_1$ is magnetocrystalline anisotropy constant. Even though the initial permeability is directly proportional to saturation magnetization, the higher value of initial permeability for x=0.05 is mainly due to the higher sintered density and lower magneto-crystalline anisotropy constant of that sample. The initial permeability values during heating and cooling cycles and $\Delta\mu_i$ are given in Table 3. All the samples have exhibited the thermal hysteresis of initial permeability and $\Delta\mu_i$ increases with increase in Ni content.

Fig. 3.shows the variation of loss factor as function of temperature. The loss factor for all samples almost stayed constant up to a certain temperature and then it increases. It is well known that higher the value of initial permeability lower is the value of

loss factor. The higher value of initial permeability values observed in case of our samples is attributed to the contribution of domain wall motion, which becomes more significant as the sintered density and grain size of the ferrite increases. Generally, a higher initial permeability is achieved through the control of both the density and microstructure, which depends on the sintering conditions [11].

## CONCLUSION

The $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ with X= 0, 0.05, 0.10, 0.15, 0.20, 0.25 have been successfully synthesized by molten salt method. The method has proved its usefulness for the synthesis of fine ferrite particles. The structural study revealed the formation of spinel type material From the dielectric and magnetic studies it is concluded that the $Mg_{0.25-x}Ni_xCu_{0.25}Zn_{0.5}Fe_2O_4$ ferrite with X=0.05 can be the potential candidate for MLCI application with low sintering temperature in view of high density, high initial permeability and saturation magnetization possessed by that composition.

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