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Cation Distribution, Magnetic and Hyperfine Interaction Studies of Ni-Zn Spinel Ferrites: Role of Jahn Teller ion (Cu$^{2+}$) Substitution

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Abstract: The work reports cation distribution, magnetic and hyperfine interaction studies of Cu$^{2+}$ substituted mixed Ni-Zn nanospinel ferrites prepared by combustion technique. X-ray diffraction and electron microscopy were used to study the structural and morphological aspects of all the samples. The Rietveld refined diffraction patterns exhibited cubic-spinel lattice structure with Fd-3m space-group for all the samples. The morphology investigations revealed the spherical morphology of particles with some agglomeration. Magnetic properties performed at 300K and 5K showed the soft ferromagnetic character of all the samples. Magnetization at 5K was progressively enhanced due to the surface effects. Field cooled and zero field cooled measurements were indicated net irreversibility for all the samples. The hyperfine interaction studies revealed the ferrimagnetic nature of Cu$^{2+}$ substituted mixed Ni-Zn spinel nano-ferrites. All the obtained results show that prepared nanoparticles are useful for magnetic fluid hyperthermia and other bio-applications.

Keywords: Ni-Zn nano-ferrites; magnetization; FC-ZFC; hyperfine interaction
1. Introduction

The new progression of the material science area in light of the nano-level has now promoted from synthesis to its real-world application. The ferrite materials in nano-scale are one of the competing contenders which attracted applications in many technological areas. The spinel ferrites in nano-form are prime members of ferrite materials which are the prime interest of the scientist and technologists. In the chemical compositional manner, spinel ferrites can be generalized as ‘M$^{II}$Fe$_2$O$_4$’ where M$^{II}$ symbolizes the two valence metal ions like Zn$^{2+}$, Mg$^{2+}$, Co$^{2+}$, Ni$^{2+}$ etc. The spinel ferrites lattice structure divides into two sub-lattices or intrinsic sites. It has many vital features like magnetic and electrical duality, high stability, super-paramagnetic appearance, and so on. These critical features of spinel ferrite made them employable in different areas like sensing units, microwave absorbing devices, communication antennas, as catalyst, a memory storing chips, medical diagnostic and imaging, photo-catalysis, magnetic switching devices, as magneto-rheological fluids, biotreatments$^{1-5}$, etc. The unusual magnetic and electrical parameters of spinel nano-ferrites depend on the method of preparation, optimization of synthesis parameters, composition, annealing temperature$^{6-9}$ etc. In literature, the number of reports is present on the physical as well as chemical synthesis techniques$^{10-15}$. Recently, the mixed type Ni-Cu-Zn nano-ferrites are of prime interest of the scientist for technological as well as electronic applications. In Ni-Cu-Zn nano-ferrites, a reasonable amount of substitution claimed the magnetic enhancement, wherein the scenery of dopant-substituted determines the association among magnetic interface. Furthermore, emphasis on magnetic properties with the variation in particle size and surface spin is highlighted$^{16}$. It is renowned information that the nano-size particles give rise to canting of spin. Thus, net magnetic exchange interactions give rise to Neel’s and Y-K types of magnetic parameters$^{17, 18}$. Meanwhile, the substitution of diamagnetic ions in a controlled manner is known for the enhanced magnetic properties of ferrites. The presence of diamagnetic cation above the specified limit has resulted in non-collinearity of spins. Besides this, magnetic anisotropy affects the super-paramagnetic behavior in nano-domains. Hence, it is essential to understand the magnetism with diamagnetic ions and its correlation with various properties in nano-domains. In these nano-ferrites, investigation on Cu$^{2+}$ doping provides new insights to illuminate its impact on structural, morphological, magnetic and Mössbauer features. The literature survey of copper substituted Ni-Zn mixed ferrites revealed enhancement in the premium properties$^{19-22}$. Using the VSM and Mössbauer spectroscopy, saturation...
magnetization, cation distribution, superparamagnetism as well as the Curie temperature and local environment of Fe ions can be well understood.

The present work reports the citrate-nitrate synthesis of Cu$^{2+}$ incorporated mixed Ni-Zn nano-ferrites. It focuses on the structural properties investigated by Rietveld analysis, magnetic studies recorded at 300 K and 5 K, as well as FC-ZFC analysis, and Mössbauer spectroscopy examined hyperfine parameters. The study also reports the in-detail impact of Cu$^{2+}$ ions concentration on these properties.

2. Experimental details
The nitrate-citrate route was employed to prepare Cu$^{2+}$ incorporated Ni-Zn mixed nano ferrite samples having the generic formula as Ni$_{0.70-x}$Cu$_x$Zn$_{0.30}$Fe$_2$O$_4$ ($x = 0.00, 0.05, 0.15,$ and 0.25). A weighed amount of nitrate for each composition and citric acid was dissolved separately in 50ml DI H$_2$O. The metal nitrate to citric acid ratio calculated using propellant chemistry was 1:3. The pH of a mixed solution of nitrates and citric acid adjusted at seven by inserting the ammonium solution. Then the combined solution was continuously stirred on a hot plate at 90°C until it transforms to gel. After the gelation of a sol, the thermal heat was raised to 115°C. The viscous gel evaporated all the water molecules and burnt with glowing flints to form a loose powder. The as-prepared powder was ground using agate pestle mortar and annealed at 600°C for six h to achieve good crystalline nature. Fig. 1 and 2 display the schematic flowchart of sol-gel auto combustion technique and the mechanism of the chemical reaction of NiCuZn ferrite, respectively. In the mechanism the gases liberated are not shown. The given mechanism is compared with that of for nanocrystalline spinel ferrites prepared using sol-gel auto combustion technique$^{23}$.

X-ray diffractometer (Panalytical Xpert Pro) with Cu-kα X-ray radiation ($\lambda=1.5409$ Å) was employed to gather structural information of prepared samples. The high-resolution transmission electron microscopy (Philips-Tecnai G2, F30) operated at accelerated voltage 300 kV was used to visualize the morphology. It has a magnification of 58x to 1 million x with resolution 2 Å (point) and 1 Å (line). Vibrating sample magnetometer was employed to record the M-H plots at 300 K and 5 K as a function of an applied magnetic field $\pm$10 kOe. Furthermore, the field cooled (FC), and Zero Field cooled (ZFC) measurement of each sample was performed in the temperature range 5-350 K at 500 Oe. Moreover, Mössbauer spectrometer (FAST Com-Tec 070906) was employed for Mössbauer analysis of Ni-Cu-Zn nano-ferrite samples. The Mössbauer spectrum of each sample was analyzed using the MossWinn 4.0 program.
3. Results and discussion

3.1 Rietveld analysis

XRD patterns of Ni-Cu-Zn spinel ferrite nanoparticles fitted with Rietveld refinement scheme using Fullprof have appeared in Fig. 3. The experimentally obtained XRD patterns are almost coinciding with that of calculated XRD patterns. Thus, the difference is remarkably small. The solid circle line (black color) denotes the experimental data while the solid lines (red color) indicate the calculated intensities. The vertical lines mean allowed Bragg’s position and the baseline (blue color) provides the dissimilarity among experimental and calculated intensities. The observed structure favours the single-phase structure since there is no detectable impurity found. The profile parameters obtained from fitted patterns for all the samples are presented in Table 1 and indicate that the refinement is reliable. There are nine peaks indexed by Miller indices as (220), (311), (222), (400), (422), (511), (440), (620) and (533) which are evident from powder XRD patterns. The average crystallite size, as appeared in Table 1, varies between 22 – 34 nm. The lattice parameter shows the increasing trend with a copper substitution (Table 1). This phenomenon attributes to the lattice expansion raised after the incorporation of large ionic radius Cu$^{2+}$ (0.074 nm) ion instead of comparatively small Ni$^{2+}$ (0.069 nm) ion.

3.3 HR-TEM analysis

HR-TEM analysis was employed to study the morphological and spinel phase aspects of typical Ni-Cu-Zn nano-ferrite samples (x = 0.00, 0.05 and 0.15) as shown in Fig. 4-6. The presence of separated as well as agglomerated particles with some level appears in TEM micrographs. The inter-magnetic interface between the particles is the main reason for this agglomeration. A similar result of spinel ferrite materials is reported. The values of the average particle size deduced from the TEM images was in the scope of 24-45nm. The SAED patterns showed diffraction rings which apply to the spinel-cubic lattice. Moreover, the absence of other diffraction rings or dots belonging to any different phase which indicates the high crystallinity of the prepared Ni-Cu-Zn spinel ferrites. HR-TEM images (Figure 4, 5 and 6) feature visible fringes with d-spacing for each sample as 0.18nm, 0.18nm and 0.24nm corresponding to (422), (422) and (222), as well as (220) and (422) planes of a cubic phase for x = 0.00, 0.05 and 0.15 respectively. Further, this shows that the nanoparticles are well crystallized. The lattice image of a single grain contains equally spaced lattice rows which signify that the region is well crystalline and free from any lattice defects. It can be treated as a coherent diffraction domain, as revealed by the X-ray diffraction analysis. Our results on
HR-TEM are analogous to NiCuZn spinel ferrite nanoparticles prepared using the co-precipitation synthesis technique.

### 3.4 VSM analysis

Fig. 7 and 8 show the M-H loops of all the samples recorded at 300 K and 5 K respectively. The soft magnetic nature in M-H plots showed moderate saturation with insignificant coercivity. The values of all the magnetic parameters signify the ferrimagnetic behavior of all the samples. Fig. 9 displays, the variation of magnetic parameters with Cu$^{2+}$ incorporation. The change in magnetic parameters is due to the influence of cationic stoichiometry and occupancy to specific sites. In this work, saturation magnetization decreases with Cu$^{2+}$ incorporation except for composition $x = 0.05$. The corresponding magnetic moment of Ni$^{2+}$, Cu$^{2+}$, Zn$^{2+}$, and Fe$^{3+}$ is two $\mu_B$, one $\mu_B$, zero $\mu_B$, and five $\mu_B$ respectively. Neel’s Model can explain the observed behavior of magnetization with Cu$^{2+}$ substitution. The following relation displays the formula for the theoretical magneton number for spinel ferrites.

\[ \eta_B^V = M_B - M_A \]  

where, $M_B$ and $M_A$ are magnetic moments of [B] and (A) sites respectively. As magnetization of sample $x = 0.05$ increased, it indicates that Zn$^{2+}$ ions and Ni$^{2+}$, as well as Cu$^{2+}$ ions, must have occupied [B] and (A)-sites respectively. Zn$^{2+}$ ions have occupied octahedral B-sites as per cation distribution performed using magnetic data. The cation distribution given in table 2 matches well with that of obtained by XRD data. The present work notices the deviation in regular preferences of cations. The partial occupation of Ni$^{2+}$ and Zn$^{2+}$ ions at tetrahedral A-sites must have forced Fe$^{3+}$ ions to transfer to octahedral [B]-site, in that way rising the magnetic moment of [B] sub-lattice. Similar results on zinc substituted Cu-Ni spinel ferrite synthesized using co-precipitation technique and copper doped Ni-Zn spinel ferrite were reported. Both experimental and calculated magneton number increases for sample $x = 0.05$ and found to match well as presented in table 2. M-H loops recorded at 5 K (Fig 8) shows progressive enhancement as compared to that of recorded at 300 K can be explained by the surface effects. As the thermal energy decreases the magnetic moment get aligned in the direction of applied field compared to 300 K where magnetic moment experiences many disordered states. At low temperature the surface spins freezing to a certain direction causes the increment in magnetization. Projection of magnetic moments towards the way of the exterior field may get enhanced. Hence, magnetization at 5 K shows progressive enhancement as compared to that of at 300 K.
Figure 9 represented coercivity as a function of Cu$^{2+}$ ion concentration and found to increase. Increase in coercivity attributed to the appearance of anisotropy field which found to increase in this work indicating magnetically disordered state. An extra energy will be required to switch the spins of disordered state to ordered state which will bind the magnetization in a preferred direction. Thus, the coercivity increases with Cu$^{2+}$ ions concentration. As noticed, coercivity values are much lower revealing ferrimagnetic nature of Cu$^{2+}$ substituted Ni-Zn ferrites. Remanence ratio (Fig. 9) increases with copper substitution, as $M_S$ reduces with Cu incorporation. The observed values of the remanence ratio at 300 K and 5 K are in the scope of 0.035-0.094 and 0.306-0.328. It is renowned information that the lesser value (<0.5) of the remanence ratio is the prerequisite for the application of spinel ferrites in high-frequency devices.

The magnetic spin orients in the easy axis direction, which can be determined by magneto-crystalline anisotropy. The relation for the anisotropy field ($H_K$) is

$$H_K = \frac{2K_1}{M_s}$$  \hspace{1cm} (2)

ZFC-FC experiments studied the effect of magnetic anisotropy on the super-paramagnetic behavior of ferrite. Fig. 10 depicts the resulting ZFC-FC curves. In FC measurement, the total magnetization was noted at the lowermost thermal range (5K), as the magnetic moment orient in the way of the exterior magnetic field. As temperature increases, the FC curve of each sample shows a continuous decrease in magnetization. It attributes to the thermal agitation which randomizes magnetic moments. In FC curves, magnetization as a function of Cu$^{2+}$ substitution was decreased except $x = 0.05$ and followed the same trend as that of at 300 K and 5 K with copper. The variation is due to the particle size or magneto-crystalline anisotropy and cation distribution. The blocking temperature ($T_B$) is the temperature at which a maximum magnetization reached. Above $T_B$, both ZFC and FC curves coincide to a specific temperature. The blocking temperature increases with the Cu incorporation $^{33}$, and it shifted from 27 - 112 k. It is observed that the copper substitution risen the blocking temperature which can be explained by Neel’s theory who explains the dependence of blocking temperature on magneto-crystalline anisotropy. As a function of copper substitution the magnetic anisotropy increased (fig. 9). Consequently, blocking temperature increased with copper concentration. ZFC curves become broader with Cu incorporation, which can be explained by the increase in particle size. A substantial spin frustration above $T_B$ decreases the magnetization. Above $T_B$, the super-paramagnetism appears due to higher thermal energy than the magnetic energy barrier. Neel’s theory $^{34}$ elucidate the reliance of $T_B$ on effective
magneto-crystalline anisotropy ($K_{\text{eff}}$) and volume of particle size ($V_p$). The following relation gives its dependence,

$$T_B = \frac{K_{\text{eff}} V_p}{25 K_B} \quad (3)$$

At low temperature, the difference in ZFC and FC curves relates to the variation in coercivity. It is related to the anisotropy constant of Cu$^{2+}$ substituted Ni-Zn spinel ferrite$^{35}$. At a certain temperature, both ZFC and FC curves bifurcate that temperature is known as bifurcation temperature ($T_B$). With copper substitution, $T_B$ changes from 133-311 K. In ZFC curves, domain structure differs at low temperature than at high temperature whereas, in FC curve, it retains its state in the high-temperature configuration. It is noticeable that $T_B$ is greater than $T_B$, indicating the existence of substantial dipole-dipole interaction among spins$^{36}$. It is a well known fact that the Curie temperature of the sample plays a vital role in controlling the temperature of the material under AC magnetic field for hyperthermia application. In our earlier report$^{37}$ we have reported the Curie studies by DC electrical properties. It is found that the increase in Copper content lowered the Curie temperature which was attributed to the A-B exchange interaction due to distribution of Fe$^{3+}$ ions over the sub-lattices.

### 3.5 Mössbauer spectroscopy

Mössbauer spectroscopy deals with the valence states and is a powerful method to know the location of the Fe matrix and to investigate the super-paramagnetic behaviour of nano ferrites. In ferromagnetically aligned magnetic moments, the superparamagnetism can be accounted for by Neel-Arrhenius relation, while for single domain particles it is given by Neel’s relaxation theory$^{34}$. Fig. 11 displays the Mössbauer spectra of all the samples. A six lines asymmetry advocates the occurrence of at least two hyperfine magnetic sextets corresponding to Fe$^{3+}$ ions at (A) and [B] sites. The presence of sextets corresponding to hyperfine interaction indicates the presence of different-sized nanoparticles, and associated superparamagnetism characterizes the presence of strong Quadrupole doublet. A mixed spinel ferrite possesses two exchange interactions where Fe$^{3+}$ ions exist over (A) and [B] sites while Cu$^{2+}$, Ni$^{2+}$ ions located at [B] site and Zn$^{2+}$ ions accommodate at (A) and [B] sites. The spectral parameters such as line width, isomer shift, quadrupole splitting and hyperfine field obtained from Mössbauer spectra for (A) and [B] sites as well as for doublet are tabulated in Table 3. The line width values for (A) sites are slighter than [B] site. The gained values of $I_S$
matches well with the literature \(^{35}\). The observed \(I_S\) values are lesser than 0.5 mm/s, which signifies the subsistence of \(\text{Fe}^{3+}\) valence state and rules out the possibility of \(\text{Fe}^{2+}\) ion presence. The hyperfine field occurring from the magnetic moment varies between 34.40 - 47.89 T and decreases with copper substitution except for \(x = 0.05\). The reduction in \(H_f\) with copper is predictable because \(\text{Cu}^{2+}\) (1\(\mu\)B) of small magnetic moment replace elevated magnetic moment \(\text{Ni}^{2+}\) (2\(\mu\)B) ion. It is a well-renowned fact that AB exchange interaction is stronger than AA and BB interactions. Moreover, the relative area at (A) and [B] sites determines the fraction of \(\text{Fe}^{3+}\) ions at respective sites. As observed from Table 3, it indicates that the larger fraction of \(\text{Fe}^{3+}\) ions occupy tetrahedral sites favoring spin canting and leads to Y-K type magnetism. It is well known and accepted that the doublet initiates due to the paramagnetic or super-paramagnetic behaviour of nanoparticles. Sarveena et al. have reported the same kind of behavior \(^{38}, 39\). Hence, in the present case Mössbauer spectra indicate the ferrimagnetic-ordered state and the presence of doublet, which interpret a paramagnetic phase. Overall, \(\text{Cu}^{2+}\) substitution weakens the A-B super-exchange interaction as witnessed by M-H behavior, M-T results and magnetic hyperfine interactions. Thus, magnetization with copper substitution exhibits decreasing the trend.

4. Conclusions

Nanocrystalline \(\text{Cu}^{2+}\) incorporated Ni-Zn spinel nano-ferrite samples were synthesized efficiently by the nitrate-citrate route. The single phasic cubic-spinel structure of prepared nano-ferrite samples was maintained throughout the copper substitution. TEM images witnessed the spherical morphology with fine particle size distribution. M-H plots reveal the ferrimagnetic behavior of \(\text{Cu}^{2+}\) incorporated Ni-Zn nano-ferrite samples which further supported by Mössbauer spectroscopy. Magnetization at 5K was progressively enhanced due to the surface effects. Field cooled and zero field cooled measurements were indicated net irreversibility for all the samples. The hyperfine interaction studies revealed the ferrimagnetic nature of \(\text{Cu}^{2+}\) substituted mixed Ni-Zn spinel nano-ferrites.

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Conflict of Interest
None

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Fig. 1: Schematic flowchart of nitrate-citrate route employed to synthesize Ni$_{0.70-x}$Cu$_x$Zn$_{0.30}$Fe$_2$O$_4$ ($x = 0.00, 0.05, 0.15$ and $0.25$)

Fig. 2: Schematic of reaction mechanism of Ni$_{0.70-x}$Cu$_x$Zn$_{0.30}$Fe$_2$O$_4$ ($x = 0.00, 0.05, 0.15$ and $0.25$) synthesis
Fig. 3: Rietveld refined XRD patterns of Ni$_{0.70-x}$Cu$_x$Zn$_{0.30}$Fe$_2$O$_4$ (x = 0.00, 0.05, 0.15 and 0.25)
Fig. 4: TEM image, particle size distribution, SAED and HR-TEM patterns of Ni$_{0.70}$-\(x\)Cu$_x$Zn$_{0.30}$Fe$_2$O$_4$ (\(x = 0.00\))
Fig. 5: TEM image, particle size distribution, SAED and HR-TEM patterns of Ni$_{0.70-\chi}$Cu$_{\chi}$Zn$_{0.30}$Fe$_2$O$_4$ (\(\chi = 0.05\))
Fig. 6: TEM image, particle size distribution, SAED and HR-TEM patterns of Ni$_{0.70-x}$Cu$_x$Zn$_{0.30}$Fe$_2$O$_4$ (x = 0.15)
Fig. 7: M-H plots for Ni$_{0.70-x}$Cu$_x$Zn$_{0.30}$Fe$_2$O$_4$ (x = 0.00, 0.05, 0.15 and 0.25) recorded at 300 K.
Fig. 8: M-H plots for Ni$_{0.70-x}$Cu$_x$Zn$_{0.30}$Fe$_2$O$_4$ ($x = 0.00$, 0.05, 0.15 and 0.25) at 5 K.
Fig. 9: Magnetic parameters for Ni$_{0.70-x}$Cu$_x$Zn$_{0.30}$Fe$_2$O$_4$ ($x = 0.00, 0.05, 0.15$ and $0.25$) at $300$ K and $5$ K
Fig. 10: FC-ZFC curves for Ni$_{0.70-x}$Cu$_x$Zn$_{0.30}$Fe$_2$O$_4$ (x = 0.00, 0.05, 0.15 and 0.25)
Fig. 11: Mossbauer spectra of Ni$_{0.70-x}$Cu$_x$Zn$_{0.30}$Fe$_2$O$_4$ ($x = 0.00$, 0.05, 0.15 and 0.25)
Table 1: Goodness of fit ($\chi^2$), profile reliability index ($R_p$), weighted profile reliability index, ($R_{wp}$), lattice parameter (a), average crystallite size (D), average particle size (t) for Ni$_{0.70-x}$Zn$_{0.30}$Fe$_2$O$_4$ nanoparticles

| Comp | $\chi^2$ | $R_p$ | $R_{wp}$ | $R_{exp}$ | a (Å) | D$_{XRD}$ (nm) | t$_{TEM}$ (nm) |
|------|----------|-------|----------|----------|-------|---------------|----------------|
| 0.00 | 1.02     | 11.20 | 10.98    | 12.0     | 8.359 | 29            | 24             |
| 0.05 | 1.01     | 12.32 | 12.21    | 9.33     | 8.368 | 30            | 25             |
| 0.15 | 1.04     | 12.27 | 11.80    | 8.04     | 8.376 | 34            | 45             |
| 0.25 | 2.18     | 2.63  | 1.78     | 1.93     | 8.386 | 28            | -              |

Table 2: Cation distribution, magneton number ($\eta_B$), and blocking temperature ($T_B$) for Ni$_{0.70-x}$Zn$_{0.30}$Fe$_2$O$_4$ nanoparticles at temperatures 300K and 5K

| Comp | Cation distribution (from Magnetization) | Cation distribution (from XRD) | $\eta_B$ | $T_B$ (K) |
|------|----------------------------------------|--------------------------------|----------|------------|
|      | (Ni$_{0.02}$Zn$_{0.10}$Fe$_{0.88}$)    | (Ni$_{0.02}$Zn$_{0.10}$Fe$_{0.88}$) | 2.51     | 27         |
| 0.00 | [Ni$_{0.68}$Zn$_{0.20}$Fe$_{1.12}$]    | [Ni$_{0.68}$Zn$_{0.20}$Fe$_{1.12}$] |          |            |
|      | (Ni$_{0.015}$Cu$_{0.01}$Zn$_{0.12}$Fe$_{0.85}$) | (Ni$_{0.015}$Cu$_{0.02}$Zn$_{0.12}$Fe$_{0.85}$) | 2.76     | 53         |
| 0.05 | [Ni$_{0.635}$Cu$_{0.04}$Zn$_{0.175}$Fe$_{1.15}$] | [Ni$_{0.635}$Cu$_{0.04}$Zn$_{0.175}$Fe$_{1.15}$] |          |            |
|      | (Ni$_{0.01}$Cu$_{0.02}$Zn$_{0.12}$Fe$_{0.86}$) | (Ni$_{0.01}$Cu$_{0.02}$Zn$_{0.12}$Fe$_{0.86}$) | 2.55     | 89         |
| 0.15 | [Ni$_{0.54}$Cu$_{0.14}$Zn$_{0.18}$Fe$_{1.14}$] | [Ni$_{0.54}$Cu$_{0.14}$Zn$_{0.18}$Fe$_{1.14}$] |          |            |
|      | (Ni$_{0.005}$Cu$_{0.02}$Zn$_{0.12}$Fe$_{0.86}$) | (Ni$_{0.005}$Cu$_{0.02}$Zn$_{0.12}$Fe$_{0.86}$) | 2.46     | 112        |
| 0.25 | [Ni$_{0.445}$Cu$_{0.24}$Zn$_{0.178}$Fe$_{1.137}$] | [Ni$_{0.445}$Cu$_{0.24}$Zn$_{0.178}$Fe$_{1.137}$] |          |            |

Table 3: Line width ($\Gamma$), Isomer shift ($\delta$), Quadrupole Splitting ($\Delta$), Hyperfine Field ($H_f$) and relative area ($R_A$) of Ni$_{0.70-x}$Zn$_{0.30}$Fe$_2$O$_4$ (x = 0.00, 0.05, 0.15 and 0.20)

| Comp | Sub Spectrum | $\Gamma$ (mm/s) | $\delta$ (mm/s) | $\Delta$ (mm/s) | $H_f$ (T) | $R_A$ (%) |
|------|--------------|-----------------|-----------------|-----------------|-----------|-----------|
| 0.00 | S1           | 0.50            | 0.26            | 0.00            | 34.40     | 65.55     |
|      | S2           | 0.71            | 0.27            | 0.00            | 31.22     | 30.24     |
|      | D1           | 0.69            | 0.20            | 0.58            | ----      | 04.21     |
| 0.05 | S1           | 0.41            | 0.15            | 0.02            | 47.89     | 71.19     |
|      | S2           | 0.42            | 0.18            | -0.06           | 42.95     | 21.96     |
|      | D1           | 0.35            | 0.20            | 0.49            | ----      | 06.86     |
| 0.15 | S1           | 0.41            | 0.14            | 0.01            | 47.66     | 66.49     |
|      | S2           | 0.41            | 0.17            | -0.12           | 42.33     | 27.01     |
|      | D1           | 0.35            | 0.15            | 0.68            | ----      | 06.49     |
| 0.25 | S1           | 0.41            | 0.17            | 0.06            | 47.32     | 72.30     |
|      | S2           | 0.42            | 0.13            | 0.05            | 42.02     | 23.20     |
|      | D1           | 0.36            | 0.12            | 0.58            | ----      | 04.50     |
Effect of Cu$^{2+}$ substitution on structural, morphological, cation distribution, magnetic and Mossbauer properties of mixed Ni-Zn Spinel Ferrites is studied.