

# Effect of Cu Incorporation on Structure, Densification and Magnetic Properties of Polycrystalline Li<sub>x</sub>Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>2+x</sub>O<sub>4</sub> Ceramics

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# Abstract

The polycrystalline  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{x+2} \text{O}_4$  and  $\text{Li}_x \text{Ni}_{0.1} \text{Cu}_{0.1} \text{Zn}_{0.8-2x} \text{Fe}_{x+2} \text{O}_4$  (x = 0.0, 0.1, 0.2, and 0.3) ferrites were synthesized by the standard solid-state reaction method. The compound was sintered at 1150°C for 5 hours. The effect of Cu substitution and its impact on the crystal structure, microstructure, complex initial permeability and magnetization of the Ni-Zn ferrites were studied. The effect of Li<sup>+</sup> incorporation on the properties mentioned above was also investigated. X-ray diffraction patterns of the samples indicated a single cubic spinel structure for both the compound. No effect of Cu addition on crystal structure was observed. The density of the ferrites was found to be enhanced because of adding Li whereas the porosity of the samples decreased with the content of Li ions. The average value of grain size increased with the addition of Li content. The samples having Cu ions formed bigger size grains. Frequency-dependent complex initial permeability, loss tangent, and relative quality factor were studied at room temperature using an Impedance analyzer in the range of 100 Hz - 120 MHz regions. In the low-frequency region, the prepared samples exhibited a high value of permeability and after a certain frequency, the permeability falls. The value of permeability enhanced with the increase in Li whereas loss tangent was found to be reduced. The relative quality factor graphs described that the compound has excellent frequency stability up to a certain frequency which is suitable to be used in inductors, resistors, capacitors, etc. Initial permeability for Li<sub>x</sub>Ni<sub>0.1</sub>Cu<sub>0.1</sub>Zn<sub>0.8-2x</sub>Fe<sub>x+2</sub>O<sub>4</sub> ferrites was found high than Li<sub>x</sub>Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>x+2</sub>O<sub>4</sub> which might be attributed to having bigger size grains of Cu containing samples because of easy movement of domain wall in bigger size grains. The values of saturation magnetization  $(M_s)$  were calculated for both compounds from M-H hysteresis loops and it enhanced with the increase in Li content which might be related to the modification of predominant exchange interactions between the cations. The Cu-containing compound exhibited higher values of saturation magnetization. The cation distribution reflects this increment because ferromagnetic  $Ni^{2+}$  and paramagnetic  $Cu^{2+}$  ions occupied in the B-sites and the diamagnetic Zn and paramagnetic Li occupied in the A-sites; therefore, net magnetic moments increased gradually. The studied materials might be used as an alternative to Pb-based compounds and would be environment friendly.

#### **Keywords**

Ceramics, Ferrites, Complex Initial Permeability, Saturation Magnetization

# 1. Introduction

The demand for magnetic ceramics in the modern technological world is increasing day by day because of their versatile contribution to the miniaturization of many electrical devices. Ferrites are an important class of magnetic ceramics that are composed of iron oxides and other metallic ions. They are non-conducting magnetic compounds. The general chemical formulae of ferrites are MeFe<sub>2</sub>O<sub>3</sub>, where M stands for suitable divalent metal ions such as  $Co^{2+}$ ,  $Mn^{2+}$ ,  $Ni^{2+}$ ,  $Fe^{2+}$ ,  $Zn^{2+}$ ,  $Cd^{2+}$ ,  $Mg^{2+}$ , etc. and Fe is the trivalent ferric ion. Based on crystal structure, ferrites are of four types: spinel, garnet, ortho, and hexagonal. Depending upon the magnetic properties they might be soft ferrites and hard ferrites. The spinel soft ferrites have high demand because of their highest possible number of symmetries [1] [2] [3]. In research, it gained a lot of interest because of its low price and easy magnetizing and demagnetizing properties. X-ray crystallography described that the spinel ferrites have a face-centered cubic structure where oxygen ions are packed closely together with the much smaller divalent and trivalent metal ions in the interstitial sites. There are two types of interstitial sites. If the interstitial sites are denoted by A- and B- respectively, then there are 64Aand 32B-sites in the unit cell of the structure. Considering the substitution atoms in A-site and B-site the spinel ferrites can exhibit ferrimagnetic, antiferromagnetic, and paramagnetic behavior [4].

The cubic structured soft ferrite Ni-Zn possesses good magnetic and electric properties and there is much research is going on to improve their properties. The Ni-Zn ferrites have potential applications in antenna rods, loading coils and core materials for power transformations in electronics and telecommunications. Nowadays, a large number of researchers are working on Ni-Cu-Zn ferrites because Ni-Zn ferrites create cation vacancies, unsaturated oxygen ions and excessive electrons due to the loss of Zn during the calcination and sintering process [5]. These unwanted problems are reduced by the substitution of nonmagnetic metal ions (such as Cu) in Ni-Zn ferrites. The substitution of magnetic ion Ni by the Cu modifies the cation distribution process in the system and plays an important role to enhance various properties of Ni-Cu-Zn ferrites [6]-[11]. The

Ni-Cu-Zn ferrites have various excellent properties such as high resistivity, low cost and low sintering temperature. They are used for multilayer chip inductors and electromagnetic interference (EMI) filters. Li substitution in place of Zn in Ni-Zn ferrites is also emerging research because of their high demand in industrial and technological applications. Research on Li substitution in Ni-Cu-Zn ferrites requires more study because a small number of foreign ions in the ferrite can dramatically change its properties. Li and Li substituted ferrites are performed as an excellent material in high-density recording media and absorbers and microwave devices because of their low cost, high saturation magnetization, high Curie temperature, etc. [12]-[17]. They exhibit comparatively better performances over other spinel ferrites. Detailed studies of the effect of Cu<sup>2+</sup> incorporation on structure, density, microstructure, complex initial permeability, and magnetization of Li doped Ni-Zn ferrites have been reported here.

# 2. Experimental Details

# 2.1. Synthesis

The polycrystalline  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{x+2} O_4$  and  $\text{Li}_x \text{Ni}_{0.1} \text{Cu}_{0.1} \text{Zn}_{0.8-2x} \text{Fe}_{x+2} O_4$  (x = 0.0, 0.1, 0.2, and 0.3) ferrites were synthesized following the conventional solid-state reaction method from the raw materials of  $\text{Li}_2O_3$  (99.9%),  $\text{Ni}_2O_3$  (99.9%), CuO (99.99%), ZnO (99.9%) and  $\text{Fe}_2O_3$  (99.9%). The raw powders were mixed with acetone by a mortar and pestle. To obtain a homogeneous mixture the mixed powders were blended for 6 h and then calcined at 850°C for 5 h. The powders were blended again for 2 h. One drop of polyvinyl alcohol was added with the mixed powders as a binder for sample preparation. Then toroid- and disc-shaped samples were prepared using a hydraulic press. Finally, the prepared samples were sintered at 1150°C for 5 h.

#### 2.2. Characterization

X-ray diffractometer (Philips PANalytical X'PERT-PRO) with  $CuK_a$  radiation ( $\lambda = 1.541$  Å) was used to study the crystal structure of the sintered samples. The lattice parameter (a) was calculated using the following relation:

$$d^2 = \frac{a^2}{h^2 + k^2 + l^2} \tag{1}$$

where h, k, and l are the Miller indices and d is the interplanar distance. The exact value of a was calculated using the Nelson-Riley function:

$$F(\theta) = \frac{1}{2} \left( \frac{\cos^2 \theta}{\sin \theta} + \frac{\cos^2 \theta}{\theta} \right)$$
(2)

where  $\theta$  is the Bragg angle. A straight-line fit was achieved and the exact value of *a* was obtained from the extrapolation of these lines to  $F(\theta) = 0$ .

Theoretical density (  $\rho_{th}$  ) was calculated using the following formula:

$$\rho_{th} = \frac{ZM}{N_A a^3} \tag{3}$$

where Z is the number of atoms per unit cell, M is the molecular mass of the sample, and  $N_A$  is Avogadro's number. The bulk density ( $\rho_B$ ) of the sintered samples was calculated using the following equation:

$$\rho_B = \frac{m}{\pi r^2 t} \tag{4}$$

where m is the mass, r is the radius and t is the thickness of the sample. The porosity (P) of the samples was determined using the following relation:

$$P = \frac{\rho_{th} - \rho_B}{\rho_{th}} \times 100\%$$
(5)

To study the microstructural analysis Scanning Electron Microscopy (SEM) was used. The average grain diameter ( $\overline{D}$ ) was determined using the Image J software. Wayne Kerr Impedance Analyzer (model: Wayne Kerr 6500B) was used to analyze the  $\mu'_i$ . The following equation was used to calculate the  $\mu'_i$ .

$$\mu_i' = \frac{L_s}{L_0} \tag{6}$$

where  $L_s$  is the inductance of the sample and  $L_0 = \frac{\mu_0 N^2 S}{\pi d}$  is the inductance of the winding coil without the sample, where *N* represents the number of turns of coil (*N* = 4), *S* is the area of cross-sectional area and  $d\left(=\frac{d_i + d_0}{2}\right)$  is the mean diameter of the sample. The relative quality factor (RQF) was calculated using the following equation:

$$RQF = \frac{\mu'_i}{\tan \delta_M}$$
(7)

where  $\tan \delta$  is the magnetic loss.

A Vibrating Sample Magnetometer (VSM) was used to measure the *M*-*H* hysteresis loop and magnetization, coercivity, etc. of the materials were determined from the hysteresis loop.

#### 3. Results and Discussion

#### 3.1. Crystal Structure

**Figure 1(a)** and **Figure 1(b)** show the XRD pattern of various  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{x+2} \text{O}_4$  (x = 0.0, 0.1, 0.2 and 0.3) and  $\text{Li}_x \text{Ni}_{0.1} \text{Cu}_{0.1} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} \text{O}_4$ , (x = 0.0, 0.1, 0.2, 0.3) ferrites. Narrow and clear peak of different planes are observed in the XRD pattern which suggest good crystallinity of the prepared samples. The peaks of the XRD pattern are indexed following previously published articles [18].

It is seen that the high index peaks are shifting to the right-hand side with the substitution of Li content and this tendency is clearly noticed in both XRD patterns. The shifting of the peaks to the right-hand side suggests shrinkage of the unit cell volume of the compound. It is found that the XRD pattern matched with the XRD pattern of Ni-Zn type ferrites which indicates that the prepared



**Figure 1.** XRD pattern of various (a)  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{x+2} \text{O}_4$  and (b)  $\text{Li}_x \text{Ni}_{0.1} \text{Cu}_{0.1} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} \text{O}_4$ , (*x* = 0.0, 0.1, 0.2, 0.3) ferrites sintered at 1150°C temperature.

samples have cubic spinal structure. The lattice constant for each sample is determined using the formula of cubic crystal structure. The values of lattice constants corresponding to all peaks for an individual sample are plotted against the Nelson-Riley function  $F(\theta)$  [19]. The Nelson-Riley function can be written as,

$$F(\theta) = \frac{1}{2} \frac{\cos^2 \theta}{\sin \theta} + \frac{\cos^2 \theta}{\theta}$$

where  $\theta$  is Bragg's angle. The relation between  $F(\theta)$  and lattice constant of  $\text{Li}_{x}$ . Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>x+2</sub>O<sub>4</sub> ferrites has been shown in **Figure 2**. The accurate value of lattice constant for a single sample is estimated from the extrapolation of the fitted lines to the y-axis. Values of lattice constant for different amount of Li content compound of Li<sub>x</sub>Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>x+2</sub>O<sub>4</sub> (x = 0.0, 0.1, 0.2, 0.3) samples have been listed in **Table 1**.

There is a decreasing trend of lattice constant with the variation of Li content is noticed for the prepared samples. The variation of lattice constant for  $\text{Li}_{x-}$  $\text{Ni}_{0.2}\text{Zn}_{0.8-2x}\text{Fe}_{x+2}\text{O}_4$  is found to be from 8.41 to 8.35 Å. The reduction of the lattice constant is might be due to the variation of ionic radius of the samples with the addition of Li in the compound. A few differences in ionic radius of Li (0.59 Å) and other elements [Fe<sup>3+</sup> (0.645 Å), Ni<sup>2+</sup> (0.69 Å) and Zn<sup>2+</sup> (0.60 Å)] [20] is observed. The ionic radius of Li<sup>+</sup> (0.59 Å) is smaller than that of Zn<sup>2+</sup> (0.60 Å) ions which results in a decrease of lattice constant with Li addition.

**Figure 1(b)** shows the XRD pattern of Cu incorporated  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{x+2} O_4$  ferrites. The crystal structure of  $\text{Li}_x \text{Ni}_{0.1} \text{Cu}_{0.1} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} O_4$  is found to be a cubic spinal structure which implies that addition of Cu does not affect the crystal structure of  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{x+2} O_4$  ferrites. The lattice constant for the Cu incorporated Li-Ni-Zn ferrites is also reducing with the Li substitution. Some impurity peaks are noticed in the XRD pattern of  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{x+2} O_4$  (*x* = 0.2) ferrites. The unwanted peaks are found to be disappeared when Cu is incorporated which might be attributed to the good crystallinity of the ferrites because of Cu

Li content (x)	Ni <sub>0.2</sub> Zn <sub>0.8-2x</sub> Li <sub>x</sub> Fe <sub>2+x</sub> O <sub>4</sub>				$Li_xNi_{0.1}Cu_{0.1}Zn_{0.8-2x}Fe_{2+x}O_4$			
	$a = b = c (\text{\AA})$	$ ho_B{ m gm/cm^{-3}}$	P(%)	<i>D</i> μm	$a=b=c(\mathrm{\AA})$	$ ho_B{ m gm/cm^{-3}}$	P(%)	$D\mu{ m m}$
0	8.41	4.00	24.99	7.192	8.42	3.8909	27.15	9.629
0.1	8.40	3.73	28.25	9.665	8.31	3.7988	29.70	12.394
0.2	8.37	3.89	23.82	13.188	8.39	3.9605	22.18	15.006
0.3	8.35	4.06	18.50	13.192	8.36	3.8909	22.07	16.440

Table 1. The values of lattice constant, bulk density ( $\rho_B$ ), porosity (P) and grain size (D) for Li<sub>x</sub>Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>x+2</sub>O<sub>4</sub> and Li<sub>x</sub>-

 $Ni_{0.1}Cu_{0.1}Zn_{0.8-2x}Fe_{2+x}O_4$ , (x = 0.0, 0.1, 0.2, 0.3) samples sintered at 1150°C for 5 hours.





**Figure 2.** Lattice constant as a function of  $F(\theta)$  of  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{x+2} O_4$  for (a) x = 0.0, (b) x = 0.1, (c) x = 0.1= 0.2 and (d) x = 0.3 ferrites.

addition. The phase formation kinetics of the compound speeded up because of Cu addition.

#### **3.2. Density Measurement**

Figure 3(a) illustrates the variation of X-ray density (theoretical density) and bulk density with Li content of Li<sub>x</sub>Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>x+2</sub>O<sub>4</sub> ferrites. Values of density have an important effect on various properties such as electric as well as magnetic properties of ferrite compounds. Materials having high density contain lower



Figure 3. Variation of (a) theoretical density and bulk density and (b) porosity with Li content.

porosity and exhibit good electric and magnetic properties. It is seen in **Figure 3(a)**; the X-ray density decreases with the addition of Li content whereas bulk density is found to be increased. X-ray density is calculated using molecular weight, lattice parameter, etc. Molecular weight of Li (6.941 gm/mol) is lower than that of Zn (65.4 gm/mol) and thus the X-ray density is reducing gradually with the addition of a low molecular weight element by substituting heavy element.

The variation of porosity with Li content is described in **Figure 3(b)**. It is seen that porosity is reducing with Li content. Voids, pores, etc. remove because of adding Li and thus the compound becomes denser. The sintering environment may also be an important factor to be denser than the prepared samples. Measured values of X-ray density, bulk density, porosity, etc. are presented in **Table 1**. It is also noticeable from the graph (**Figure 3(a)**), the X-ray density is higher than that of bulk density which means when the samples are sintered, they might contain cracks and pores on the microscopic scale and vacancies in the lattice on the atomic scale [21]. The bulk density may have these defects but the X-ray density is precisely measured from the lattice constant, the volume of the unit cell is free from these defects.

Variation of X-ray density and bulk density for  $\text{Li}_x \text{Ni}_{0.1} \text{Cu}_{0.1} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} \text{O}_4$ , (*x* = 0.0, 0.1, 0.2, 0.3) ferrites is illustrated in **Figure 4**. It shows the same characteristics like the compound  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} \text{O}_4$  but the values of bulk density are found to be enhanced because of Cu incorporation.

#### 3.3. Microstructural Study

**Figure 5(a)** and **Figure 5(b)** shows the SEM images of  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} O_4$  and  $\text{Li}_x \text{Ni}_{0.1} \text{Cu}_{0.1} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} O_4$  ferrites respectively sintered at 1150°C for 5 hours. Microstructure has an influence on the magnetic properties of ferrite materials especially on the measurement of permeability. Microstructures of both ferrites consist of non-uniform, non-repetitive and randomly oriented grains in shape and size. Some intergranular pores are also noticed in the SEM images. The



**Figure 4.** Variation of theoretical density and bulk density of  $\text{Li}_x \text{Ni}_{0.1} \text{Cu}_{0.1} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} \text{O}_4$ , (*x* = 0.0, 0.1, 0.2, 0.3) ferrites with Li content.



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**Figure 5.** Scanning Electron Microscope (SEM) images of (a)  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} \text{O}_4$  and (b)  $\text{Li}_x \text{Ni}_{0.1} \text{Cu}_{0.1} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} \text{O}_4$  for x = 0.0 to 0.3 respectively.

non-uniform grains are obtained may be due to the inhomogeneous driving force of grain boundary movement whereas some uniform grains are existed because of the homogeneous driving force of grain boundary in the grain growth mechanism. The average value of grain size is determined by the linear intercept technique. The magnitude of grain size for both ferrites is presented in **Table 1**. The average values of grain size are found to be increased with the addition of Li content. The gradual improvement of grain size is attributed might be due to the enhancement of magnetic dipole interaction between the ions because of replacing  $Zn^{2+}$  and Fe<sup>3+</sup> ions with Li<sup>+</sup>.

It is seen in **Figure 5(b)** and in **Table 1**, the size of grains for  $\text{Li}_{x}$ . Ni<sub>0.1</sub>Cu<sub>0.1</sub>Zn<sub>0.8-2x</sub>Fe<sub>2+x</sub>O<sub>4</sub> ferrites is bigger than the grains obtained for Li<sub>x</sub>. Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>2+x</sub>O<sub>4</sub> which suggest that the addition of Cu ions modify microstructure. The increase of grain size with the addition of Cu might be due to the fact that Cu ions help to complete the sintering process properly and thus increase the molecular concentration at the crystal surface and accelerate the grain growth mechanism.

# 3.4. Study of Magnetic Properties

#### 3.4.1. Complex Initial Permeability

Information about the factors that control the magnetic properties of materials and also the adjustability of magnetic materials in high-frequency use can be obtained from complex initial permeability. The complex initial permeability is given by,  $\mu_i^* = \mu_i' - \mu_i''$  where  $\mu_i'$  is the real part of complex initial permeability and  $\mu_i''$  is the imaginary part of complex initial permeability. The  $\mu_i'$  represents the energy stored in the system and  $\mu_i''$  describes the dissipation of energy in the system. The variation of the real part of complex initial permeability for the various components of Li<sub>x</sub>Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>2+x</sub>O<sub>4</sub> ferrites with frequency is shown in Figure 6(a-i).

The values of  $\mu_i^{\prime}$  for all compositions are found to be almost constant up to a certain frequency. After a certain frequency, they show a slightly rising trend and then fall. The value of  $\mu_i^{\prime}$  is improved than the parent sample and the best



**Figure 6.** Variation of real part of permeability, loss tangent and relative quality factor of (a)  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} \text{O}_4$ and (b)  $\text{Li}_x \text{Ni}_{0.1} \text{Cu}_{0.1} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} \text{O}_4$  ferrites with frequency.

result is obtained for x = 0.1. There are two mechanisms to originate initial permeability in magnetic materials such as spin rotation in the domain and domain wall displacement. Hence, the complex initial permeability is written as [22] [23]:

$$\mu_i' = 1 + \chi_{spin} + \chi_{dw}$$

where  $\chi_{spin}$  and  $\chi_{dw}$  denote the magnetic susceptibility of spin and domain wall motion respectively.  $\chi_{spin}$  and  $\chi_{dw}$  can be written as [24]:  $\chi_{spin} = \frac{2\pi M_s^2}{K}$ and  $\chi_{dw} = \frac{3\pi M_s^2 D}{4\pi}$ , where *D* is the average grain size and  $\gamma$  is the domain wall

energy.

The influence of spin rotation on initial permeability was found very small than domain wall motion because the motion of the domain wall can continue still in a weak magnetic field [25]. The displacement of the domain wall is varied because of changing sintering temperature and grain size. The motion of the domain wall reduces with the decrease of grain size because the smaller grain contains a lesser number of domain walls and the easy reversal of the domain walls gives initial permeability in the direction of the applied magnetic field. On the other hand, previously published article of Ross [26] and Perduijin et al. [27] reported, when the grain size increases in the ferrite, the permeability increases, and vice versa. However, the Li<sub>x</sub>Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>2+x</sub>O<sub>4</sub> compound exhibited enhanced complex initial permeability with the addition of Li content which might be attributed to an increase in the size of grains (Figure 5) with Li content. The sample having 10% Li content (x = 0.1) shows the best result but all the substituted samples have an improved value of  $\mu_i^{\prime}$  than that of unsubstituted samples which might be due to porosity. The substituted compound has a lower amount of porosity and exhibited better results as pores act as a barrier to the domain wall motion and induce local demagnetizing fields which results in the reduction of  $\mu_i^{\prime}$  [9] [28].

#### 3.4.2. Loss Factor (tan $\delta$ )

The frequency dependence of magnetic loss factor  $(\tan \delta)$  at room temperature of  $\operatorname{Li}_x \operatorname{Ni}_{0.2} \operatorname{Zn}_{0.8-2x} \operatorname{Fe}_{2+x} O_4$  compound sintered at 1150°C for 5 hours is shown in **Figure 6(a-ii)**. The loss tangent  $(\tan \delta)$  describes the energy dissipated in the system. All the samples have a high value of loss factor at low frequency. The values of tan $\delta$  reduce with the increase in frequency and then remain constant. At low frequency, the motion of the domain wall follows the variation of frequency with the change in an external magnetic field but at high frequency, the domain wall becomes unable to follow the variation of the external magnetic field and this might be attributed to exhibiting constant value. It is seen in **Figure 6**, the Li content samples have a lower value of tan $\delta$  than unsubstituted ones which indicates that the addition of Li affects the reduction of loss tangent.

#### 3.4.3. Relative Quality Factor (RQF)

**Figure 6(a-iii)** illustrates the variation of relative quality factor (RQF) with the change in frequency. The RQF determines a range of frequencies where the samples exhibit better performance and it is one of the criteria of the materials to

be used in microwave devices [29]. RQF has the maximum value at that frequency level where the magnetic loss tangent has the minimum value. The value of RQF first increases slowly with frequency and then rises quite abruptly making a peak at a certain frequency named resonance frequency. All the samples have a broad peak except the unsubstituted sample. The parent sample may have a peak at a high frequency beyond our measurement range.

**Figures 6(b(i-iii))** shows the variation of the real part of permeability, loss tangent, and relative quality factor respectively with the frequency of Cu included  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} O_4$  ferrites where Ni was substituted by Cu. A noticeable change is observed in the magnetic properties of  $\text{Li}_x \text{Ni}_{0.1} \text{Cu}_{0.1} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} O_4$  ferrites although the magnetic material Ni was replaced by nonmagnetic Cu. The overall permeability was found to be enhanced and followed Snoek's law [30]. The addition of Cu in  $\text{Li}_x \text{Ni}_{0.2} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} O_4$  ferrites influences density measurement, porosity, and grain size. Moreover, the addition of Cu helps to complete the sintering process properly. The properties of Cu mentioned above contribute to improving the permeability, loss tangent, and relative quality factor.

#### 3.4.4. M-H Hysteresis Loop

Magnetization vs magnetic field (M-H) hysteresis loops for each sample of Li added Ni-Zn (Li<sub>x</sub>Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>2+x</sub>O<sub>4</sub>) ferrites have been described in Figure 7. Like a normal behavior of ferromagnetic materials, magnetization of the prepared ferrite samples is found to increase with applied field and attains a maximum value at higher field except pure Ni-Zn samples. Hysteresis loops of ferrite materials having very low saturation magnetization, approximately zero remanence magnetization indicate paramagnetic behavior at room temperature [31] [32]. The values of saturation magnetization  $(M_s)$ , remanent magnetization  $(M_r)$ and coercivity  $(H_c)$  for the prepared samples have been determined from the M-H hysteresis loops (Figure 7). The magnetization increased with a sluggish nature until it gets the maximum value due to the spin rotation up to x = 0.2. This variation of room temperature saturation magnetization with the increase in Li and Fe content is related to 1) the modification of predominant exchange interactions between the cations; 2) substitution of paramagnetic (Li) and ferromagnetic (Fe) ions for the diamagnetic  $Zn^{2+}$ ; 3) cation redistribution in the spinel lattice due to addition of Li and Fe ions [33].

The mechanism has also been explained in terms of spin-disorder, spin canting, and spin-glass-like state in the surface layers of the nanoparticles due to the local chemical disorder, broken exchange interaction and a dissimilar local symmetry for those atoms near the surface. As the divalent  $Cu^{2+}$  and  $Zn^{2+}$  cations have a tendency to occupy both sites (octahedral and tetrahedral sites), so they can form the mixed spinel structure. Mixed spinel configurations are characterized by the degree of inversion, which depends strongly on the preparation procedures [34]. The increase in saturation magnetization with increasing Li and Fe concentration from x = 0 to 0.20 suggests Neel's two-sub-lattice model which stated that the resultant magnetization of the samples varies with the difference



Figure 7. M-H loop ofLi<sub>x</sub>Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>x+2</sub>O<sub>4</sub> spinel ferrites at room temperature.

of the B-site and A-site magnetization  $(M = M_B - M_A)$  [35] [36]. The possible cation distribution for Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Li<sub>x</sub>Fe<sub>2+x</sub>O<sub>4</sub> ferrites for  $0 \le x \le 0.20$  can be expressed as the following:

$$\left(Zn_{0.8-2x}^{2+}Li_{x}^{+}Fe_{0.85}^{3+}\right)_{A}\left[Ni_{0.2}^{2+}Fe_{1.15+x}^{3+}\right]_{B}O_{4}$$

Here, the bracket () and [] specify the tetrahedral A-site and octahedral B-site, respectively. The cation distribution is attained from a few particulars [37]. They are as follows: 1) Diamagnetic  $Zn^{2+}$  ion prefers to occupy both the tetrahedral A-site and octahedral B-sites [38] [39]; 2) Ni<sup>2+</sup> prefers to go to the octahedral B-site [40]; 3) it is evident that the Li<sup>+</sup> ion has tetrahedral site preference in spinel structure and Cu<sup>2+</sup> has octahedral site preference [41].

According to the above cation distribution of the prepared samples and using the values of the magnetic moments of  $Zn^{2+}$  (0  $\mu_B$ ), Ni<sup>2+</sup> (2  $\mu_B$ ), Li<sup>+</sup> (0  $\mu_B$ ), and Fe<sup>3+</sup> (5  $\mu_B$ ) [41] [42], the total magnetization for x = 0.00 to 0.20 is expected to be high, where  $\mu_B$  is the Bohor magneton number (9.27 × 10<sup>-24</sup> joule per tesla). Due to the high amount of diamagnetic Zn content on the tetrahedral sites (≥0.4), a deficiency of Fe<sup>3+</sup> ions on the tetrahedral sites creates and the moments of both sites are not ordered anymore. Then the B lattice divides itself into sub-lattices; the magnetization of the B sub-lattices makes Y-K angle with each other differing from 0 or 180° regarding the strength of B-B to A-B interactions [43] [44]. In that case, the total magnetization can be rewritten as (**Table 2**),

$$M = (5x + 1.9\theta YK) \mu_B$$

So, the cation distribution and the exchange interactions between A-sites and B-sites may justify the observed variation in  $M_{s}$ . For the x = 0.30 sample the saturation magnetization is decreased due to the possible influx of the paramagnetic Zn<sup>2+</sup> ions to the octahedral sites in replace of ferromagnetic Fe<sup>3+</sup> ions to the tetrahedral sites. This result not only decreases the magnetic moment difference between the A & B-sites but also modifies largely the prevailing exchange interactions. The rearrangement of cations distribution and magnetization for x = 0.30 sample are,

$$\left(\mathrm{Li}_{x}^{+}\mathrm{Fe}_{0.80+x}^{3+}\right)_{\mathrm{A}}\left[\mathrm{Zn}_{0.8-2x}^{2+}\mathrm{Ni}_{0.2}^{2+}\mathrm{Fe}_{1.20}^{3+}\right]_{\mathrm{B}}\mathrm{O}_{4} \text{ and } M = (2.4-5x)\mu_{B}$$

**Figure 8** depicts the room temperature M-H loops of Ni-Li-Cu-Zn ferrites that have been sintered at 1150°C for 5 h. Paramagnetic divalent Cu ions can occupy both sites of the spinel ferrite [33] [41]. It is observed that when 10% ferromagnetic Ni ions are replaced by 10% paramagnetic Cu ions, the net magnetic moments of (difference between B-sites and A-sites) the spinel ferrites have been decreased. Since Li-ions occupied in the tetrahedral sites and Fe ions occupied in both sites the possible cations distribution can be written as,

$$\left(Zn_{0.8-2x}^{2+}Li_{x}^{+}Fe_{0.85}^{3+}\right)_{A}\left[Ni_{0.1}^{2+}Cu_{0.1}^{2+}Fe_{1.15+x}^{3+}\right]_{B}O_{4}$$

From **Figure 8**, it is also noticeable that saturation magnetization is gradually increased with Li and Fe content. The cation distribution reflects this increment because ferromagnetic Ni<sup>2+</sup> (2  $\mu_B$ ) and paramagnetic Cu<sup>2+</sup> (1  $\mu_B$ ) ions occupied in the B sites and the diamagnetic Zn and paramagnetic Li occupied in the A-sites; therefore, net magnetic moments have been increased gradually. Expected increased magnetization can be written as,

$$M = (1.8 + 5x) \mu_B$$

where  $\mu_B$  is the Bohor magneton number. The experimental number of Bohr magnetons has been determined from the formula  $\frac{M_s M}{5585}$  [45], where  $M_s$  is the

**Table 2.** Molecular weight (*M*), saturation magnetization ( $M_s$ ), experimental ( $\mu_{exp}$ ) and theoretical ( $\mu_{th}$ ) Bohr magneton number for Li<sub>x</sub>Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>2+x</sub>O<sub>4</sub> and Li<sub>x</sub>Ni<sub>0.1</sub>Zn<sub>0.8-2x</sub>Fe<sub>2+x</sub>O<sub>4</sub> spinel ferrite.

<b>X</b> -	$\text{Li}_{x}\text{Ni}_{0.2}\text{Zn}_{0.8-2x}\text{Fe}_{2+x}\text{O}_{4}$				Li <sub>x</sub> Ni <sub>0.1</sub> Cu <sub>0.1</sub> Zn <sub>0.8-2x</sub> Fe <sub>2+x</sub> O <sub>4</sub>			
	M(amu)	$M_s$ (emu/gm)	$\mu_{exp}(\mu_B)$	$\mu_{tb}(\mu_B)$	M (amu)	$M_s$ (emu/gm)	$\mu_{exp}(\mu_{B})$	$\mu_{th}(\mu_B)$
0.00	239.759	25	1.07	1.9	240.244	24.5	1.05	1.8
0.10	227.479	54	2.20	2.4	233.449	46.00	1.92	2.3
0.20	215.199	68	2.62	2.9	226.65	48.00	1.95	2.8
0.30	202.919	11	0.40	0.9	219.859	60.00	2.38	3.3



**Figure 8.** Room temperature M-H loop of  $\text{Li}_x \text{Ni}_{0.1} \text{Cu}_{0.1} \text{Zn}_{0.8-2x} \text{Fe}_{2+x} \text{O}_4$  ferrites for x = 0.00 to 0.30 respectively sintered at 1150°C temperature.

experimental saturation magnetization, M is the molecular weight of the Ni-Li-Zn, and Ni-Li-Cu Zn ferrite samples. The theoretical number of Bohr magnetons ( $\mu_{tb}$ ) is obtained from the cation distribution and is strictly valid at 0 K by considering the spin moments on the A and B-sublattices aligned exactly antiparallel and are slightly higher compared to the experimental value. This variation can be explained based on the fact that the experimental value has been determined from the saturation magnetization measured at room temperature in which the magnetic spins on the A and B-sites may not be truly antiparallel as an effect of temperature. The variation of remanent magnetization and coercivity can be explained based on the magnetocrystalline anisotropy, particle morphology, dimension of the crystals, residual stress, and crystal imperfections [46].

# 4. Conclusion

Various compositions of non-magnetic Cu ion added and Li doped Ni-Zn ferrites were synthesized by the conventional solid-state reaction technique. The effect of Cu, as well as Li on different properties of the prepared samples such as crystal structure, density and porosity, microstructure, complex initial permeability, loss factor and magnetization, were studied. Polycrystalline Li,  $Ni_{0.2}Zn_{0.8-2x}Fe_{x+2}O_4$  and  $Li_xNi_{0.1}Cu_{0.1}Zn_{0.8-2x}Fe_{x+2}O_4$  (x = 0.0, 0.1, 0.2 and 0.3) ferrites had spinal cubic structure. All the compositions exhibited good crystallinity. There was no effect of Cu addition on the crystal structure. The studied samples had high density. The values of density increased with the addition of Li content whereas porosity reduced gradually. The Cu included samples showed better results. Microstructure of  $\text{Li}_{x}\text{Ni}_{0.2}\text{Zn}_{0.8-2x}\text{Fe}_{x+2}\text{O}_{4}$  and  $\text{Li}_{x}\text{Ni}_{0.1}\text{Cu}_{0.1}\text{Zn}_{0.8-2x}\text{Fe}_{x+2}\text{O}_{4}$  was measured using Scanning Electron Microscope (SEM) and average grain size was determined from the collected images following linear intercept technique. The values of grain size were found to be enhanced with the addition of Li. Nonuniform and nonrepetitive grains were seen. Li<sub>x</sub>Ni<sub>0.1</sub>Cu<sub>0.1</sub>Zn<sub>0.8-2x</sub>Fe<sub>x+2</sub>O<sub>4</sub> compound had bigger size grains. The overall complex initial permeability properties such as real part of complex initial permeability, loss tangent and quality factor were improved. The real part of complex initial permeability increased whereas loss tangent reduced with the rising of Li content. The compound containing Cu showed better results because of the bigger size grains of these compositions. The M-H hysteresis loops were determined using Vibrating Sample Magnetometer (VSM) and the values of magnetization were measured for both Lix-Ni<sub>0.2</sub>Zn<sub>0.8-2x</sub>Fe<sub>x+2</sub>O<sub>4</sub> and Li<sub>x</sub>Ni<sub>0.1</sub>Cu<sub>0.1</sub>Zn<sub>0.8-2x</sub>Fe<sub>x+2</sub>O<sub>4</sub> ferrites. Saturation magnetization was increased with the rising of Li content and the Cu added samples exhibited higher values and this might be attributed to change in cation distribution.

# **Conflicts of Interest**

The authors declare no conflicts of interest regarding the publication of this paper.

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