

Investigation of the Structural, Dielectric and Electrical properties of Zn-substituted Li-Ni ferrite

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Abstract: Substitution of Zn content on the structural, dielectric and electrical properties of $\text{Li}_{0.15}\text{Ni}_x\text{Zn}_{0.85-2x}\text{Fe}_{2+x}\text{O}_4$ ($x = 0.00, 0.10, 0.20, 0.30$ and 0.40) ferrites prepared by standard double sintering ceramic technique, sintered at 1150°C for 5 hours have been studied. It has been observed that Zn-substitution on Li-Ni ferrites plays a remarkable role in improving its structural, electrical and dielectric properties. The bulk density, x-ray density and lattice constant of the samples decreases with increasing the Ni content. The porosity of the prepared samples didn't show any trend with Ni content. The X-ray diffraction (XRD) pattern confirmed the single-phase cubic spinel structure of the samples without having any other intermediate phases. The real part of permeability, Loss factor and AC resistivity have been found to be decreased while the Quality factor increased with the increase in Ni-content. Frequency dependent dielectric constant decreased with increasing the frequency as well as Ni-content.

Keywords: Standard double sintering ceramic technique, Lattice constant, Spinel ferrites, Permeability and Dielectric constant,

I. Introduction

Ferrites constitute a special class of magnetic oxide materials with general formula $\text{MO.Fe}_2\text{O}_3$, where M is a divalent metal ion such as Mn^{2+} , Zn^{2+} , Ni^{2+} , Cu^{2+} , Mg^{2+} , Co^{2+} , Cd^{2+} . These materials show semiconducting characteristics which are of great technological applications since they have high electrical resistivity as well as better magnetic quantities [1]. Among the soft magnetic materials for high frequency power electronics Li-Ni ferrites is one of the most popular and smart candidate of highest rank [2]. These types of polycrystalline spinel ferrites have wide variety of applications such as power electronics, ferro-fluid technology, magnetic data storage, in transformers, choke coils, noise filters and as contrast agent in magnetic resonance imaging [3-5]. It is well established that in Li-Ni ferrites most of the Ni^{2+} ions occupy in tetrahedral A sites [6] which results in a lowering of Fe^{3+} in octahedral A-sites. Moreover, Structural and other properties of ferrites are dependent on the quality of new materials, milling technique, preparation temperature, concentration of the dopants and impurities [7]. For these reasons, it is possible to introduce various metallic ions to these ferrites to improve their electronic and magnetic properties significantly [8]. In the last couple of decades a very large number of researches observed the effect of various additives on different properties of ferrites [9-32]. The effect of magnetic properties of Ni concentration in Li-Zn ferrite was studied by Soiban et al. [12]. From their investigation of saturation magnetization, Curie temperature and Mossbauer studies, it has been observed that Ni substitution greatly affects the magnetic properties of the Li-Zn ferrites system. Magnetic studies of Co substituted Li-Zn-ferrites were also studied by Soibam. Magnetisation measurements indicated that cobalt shows anomalous behavior when substituted in Li-ferrites in the presence of Zn [13]. The structure and magnetic properties of Mg-doped $\text{Li}_{0.41}\text{Fe}_{2.41}\text{Mg}_{0.17}\text{O}_4$ ferrites were investigated by Widatallah et al. [14]. The saturation magnetization and Curie temperature were found to decrease and the material increasingly turned super-paramagnetic as milling proceeded at higher milling times. Tasakin et al. [15] studied the effect of sintering atmosphere without O_2 at high sintering temperature where high density was expected. This causes to decrease in permeability which might be attributed to the variation of chemical composition caused by volatilization of Zn. Rosses [17] reported that very high permeability is restricted to certain temperature ranges and the shapes of permeability versus temperature curves are strongly affected by any inhomogeneity in the ferrite structure.

Rezlescu [19] reported that the sintering behavior and microstructure of the ferrites largely affected by

PbO addition. El-Shabasy [22] studied the DC electrical resistivity of $Zn_xNi_{1-x}Fe_2O_4$ ferrites. He shows that the ferrite samples have semiconductor behavior where DC electrical resistivity decreases on increasing the temperature.

The present work is aimed at finding the effect of Ni-addition on the structural, dielectric and electrical behaviors of Li-Ni ferrites.

II. Experimental Details

Ferrite samples with chemical formula $Li_{0.15}Ni_xZn_{0.85-2x}Fe_{2+x}O_4$ ($x = 0.00, 0.10, 0.20, 0.30$ and 0.40) were prepared by the standard double sintering ceramic technique. The raw materials Fe_2O_3 , LiO, ZnO and NiO were used as starting materials. The analytical research grade oxides as supplied by the manufacture E. mark of Germany were used for the sample preparation process. Appropriate amount of raw metal oxide were mixed, crushed, grinded and milled. Milling was carried out in a wet medium to increase the degree of mixing. To avoid iron contamination, stainless steel balls were used in a steel ball milling machine and fluid such as acetone is used to convert the mixture into slurry. The grinded powders were presintered in air or oxygen at $900^{\circ}C$.

Later on, the presintered powders were again crushed into the fine powders. Using die-punch assembly different shapes of the dies were used to produce the finished products. Two types of samples: pallet and ring were prepared by using Universal Testing Machine (UTM). The finished products were sintered at constant of $1150^{\circ}C$ for five hours. A heating and cooling rate $5^{\circ}C/min$ was maintained throughout the sintering process.

A PHILIPS X' PERT PRO X-ray diffraction (XRD) system was used to get X-ray data of the samples at Pilot Plant & Process Development Center, Bangladesh Council of Scientific & Industrial Research (BCSIR), Dhaka 1205, Bangladesh. The powder diffraction technique was used with a primary beam power of 40 kV and 30 mA for Cu radiation. All the data of the samples were stored in the computer memory and analyzed by computer software "X PERT HIGHSCORE". The two surfaces of each pellet shaped samples were coated with silver paste as contact material for electrical and dielectric measurements. Dielectric properties of ferrites were carried out by using Keithley Electrometer and Hewlett Packart Impedance Analyzer (WAYNE KERR 6500B). Dielectric measurements have been done as a function of frequency in the range of 3 kHz to 10^7 Hz at room temperature. The ac electrical resistivity measurements carried out by the 6500B Precision Magnetics Analyzer over the frequency range 0 Hz to 500 Hz.

III. Results and Discussions

The phase identification and lattice parameters are determined by an X-ray diffractometer. The X-ray diffraction pattern of the prepared samples sintered at $1150^{\circ}C$ for 5 hours are shown in Fig. 1.

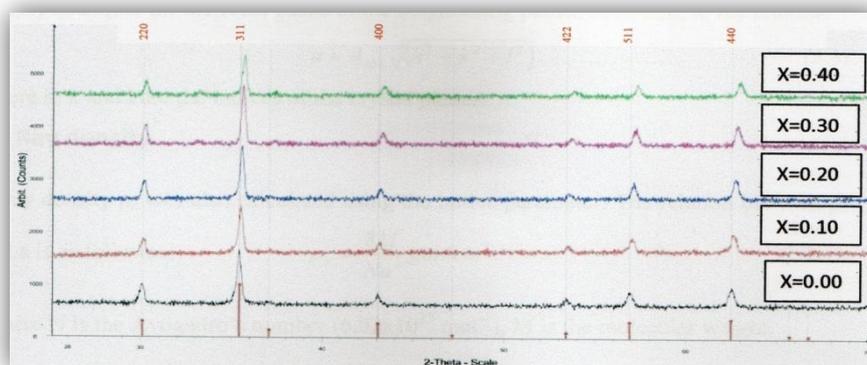


Fig. 1: X-ray diffraction (XRD) pattern of $Li_{0.15}Ni_xZn_{0.85-2x}Fe_{2+x}O_4$ (Where $x=0.00, 0.10, 0.20, 0.30$ and 0.40) sintered at temperature $1150^{\circ}C$.

The XRD patterns confirmed the single phase cubic spinel structure of the samples without having any other intermediate phases. All the samples have been found to show crystallization, with well defined diffraction lines. According to the XRD data various diffraction peaks appeared at different planes (220), (311), (400), (422), (511) and (440). It is observed that all the diffraction peaks either all odd or all even which indicate the samples are spinel in phase. Generally, for the spinel ferrites the peak intensity depends on the concentration of magnetic ions in the lattice. The intensity of the samples has been found to be increased as the amount of Ni-content is increased.

The lattice parameter a of cubic sample has been calculated from d-spacing corresponding to the different peaks of the neighboring planes according to the relation

$$a = d_{hkl} \sqrt{(h^2 + k^2 + l^2)} \dots \dots \dots (1)$$

where h, k and l are the Millar indices of the crystal planes.

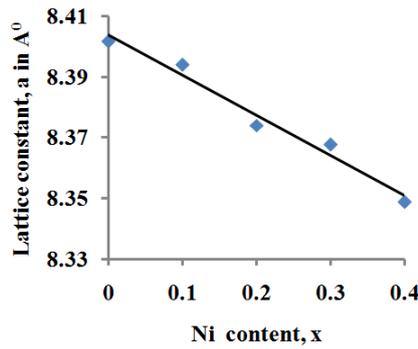


Fig. 2. Variation of lattice constant with Ni content.

In Fig. 2, lattice constants of all the samples were plotted against Ni-content. From Fig. 2 it is observed that lattice constant decreases with the increasing Ni concentration.

X-ray density ρ_x was also calculated using the lattice parameter. The relation between ρ_x and a is as follows:

$$\rho_x = \frac{8M}{Na^3} \text{ gm/cm}^3 \quad \dots\dots\dots (2)$$

Where N is the Avogadro's number ($6.02 \times 10^{23} \text{ mol}^{-1}$), M is the molecular weight.

The bulk density ρ_b of pure Li-Ni ferrites was calculated by using the following relation:

$$\rho_b = \frac{m}{V}, \dots\dots\dots (3)$$

where ρ is the density, m is the molar mass and V is the volume of the samples.

The difference between the theoretical density (measured by usual mass and dimensional consideration) and ρ_x gave us the measure of porosity. Porosity (P) in percentage will be calculated using the following equation

$$P = \left(1 - \frac{\rho_b}{\rho_x}\right) \times 100\% \quad \dots\dots\dots (4)$$

The calculated lattice constant, X-ray density, bulk density and porosity for different samples are given in Table 1.

Table-1: Lattice constant, X-ray density, bulk density and porosity for the samples with the composition $\text{Li}_{0.15}\text{Ni}_x\text{Zn}_{0.85-2x}\text{Fe}_{2+x}\text{O}_4$.

Ni content, x	Lattice constant, a in Å	X-ray density, ρ_x (g/c.c.)	Bulk density, ρ_b (g/c.c.)	Porosity, P (%)
0.0	8.4017	5.2055	4.9786	4.36
0.1	8.3940	5.1833	4.8877	5.70
0.2	8.3741	5.1100	4.7300	7.40
0.3	8.3678	4.8700	4.6600	4.30
0.4	8.3489	4.8001	4.4900	6.46

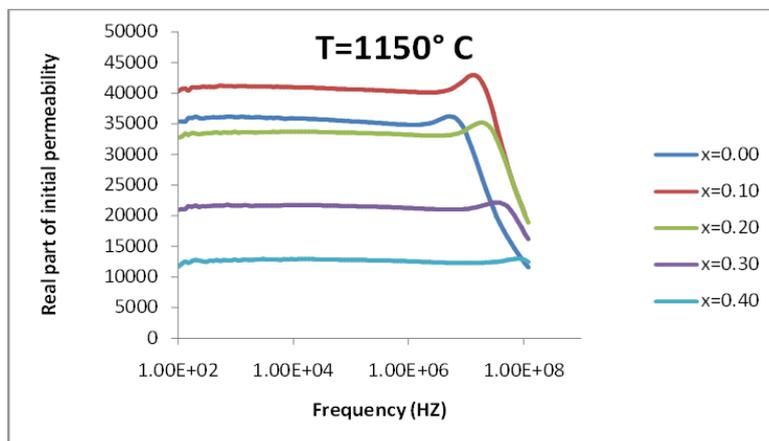


Fig. 3 : The variation of Real part of Permeability Spectra for $\text{Li}_{0.15}\text{Ni}_x\text{Zn}_{0.85-2x}\text{Fe}_{2+x}\text{O}_4$ (Where $x=0.00, 0.10, 0.20, 0.30$ and 0.40) sintered at temperature 1150°C .

Permeability measures the degree of penetration of magnetic field through a magnetic substance. According to the permeability data presented in the Fig. 3, it is observed that the real part of the initial permeability of Li-Ni ferrites decreases with the enhance of Ni content. It may be stated that with a small addition of Ni on the Li-Zn ferrites the density might have been decreased which lead to decrease in permeability. Our result has been found to be in agreement with the previous investigations: ferrites with higher density and large average grain size posses a high initial permeability and vice versa [37].

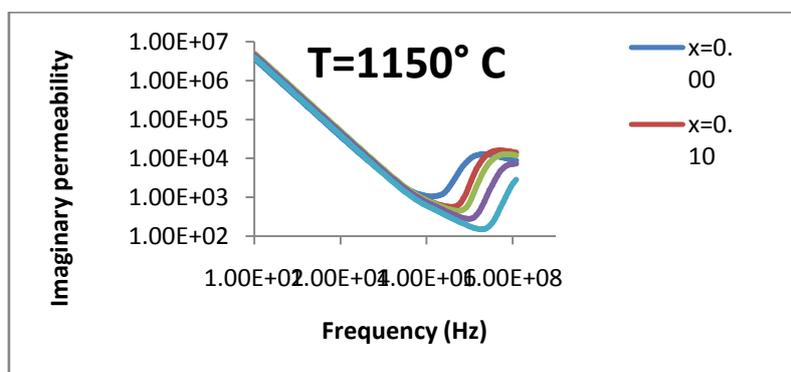


Fig. 4: The variation of Imaginary part of Permeability Spectra for $\text{Li}_{0.15}\text{Ni}_x\text{Zn}_{0.85-2x}\text{Fe}_{2+x}\text{O}_4$ (Where $x=0.00, 0.10, 0.20, 0.30$ and 0.40) sintered at temperature 1150°C .

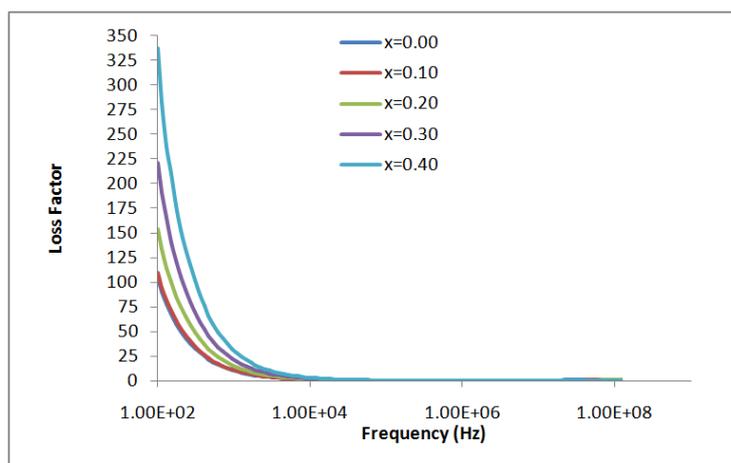


Fig. 5: The variation of loss factor with frequency for $\text{Li}_{0.15}\text{Ni}_x\text{Zn}_{0.85-2x}\text{Fe}_{2+x}\text{O}_4$, where $x=0.00, 0.10, 0.20, 0.30$ and 0.40 sintered at 1150°C .

Permeability of polycrystalline ferrites is corrected with two types of mechanisms [38-39]. The mechanisms are: domain wall motion and the spin rotation magnetization inside the domains. As the Ni is added to Li-Zn ferrites Nickel iron oxide generates and segregate at the grain boundaries pinning at the domain wall. For this reason, domain wall energy is increased and the permeability is decreased [39]. The intragranular pores might also be responsible for the pinning of the domain walls which results to deterioration of magnetic properties of Li-Ni ferrites [25]. In our investigation, it was also observed that over the enter frequency range the permeability remains almost constant. This might reflect the fact no structural relaxations or resonances are taking place in the observed frequency spectra.

Fig. 5 shows the variation of loss factor with frequency for $\text{Li}_{0.15}\text{Ni}_x\text{Zn}_{0.85-2x}\text{Fe}_{2+x}\text{O}_4$, where $x=0.00, 0.10, 0.20, 0.30$ and 0.40 sintered at 1150°C . According to the data, it is found that the loss factor of Li-Ni ferrites decreases gradually with the increase in Ni content. At lower frequency side around 100 Hz, the loss factor is high but it starts decreasing with increasing the frequency and high frequency it becomes almost unaffected with the frequency. In ferrites loss tangent, generally, arises due to the non-uniform domain wall motion, various domain defects, and variations of flux density and annihilation of domain walls [41]. It is known that at high frequencies the losses are to be lower if domain wall motion is inhibited and the magnetization is forced to change by rotation [42]. The decrease of loss factor with the increase in frequency

might attribute the relaxation phenomena of magnetization of domain walls [43]. Higher the loss factor at low frequency side may be due to the interfacial type of polarization [44].

Variations of Q-factor with frequency for the Li-Zn ferrites without and with different amount of Ni-contents are shown in Fig-06. From these diagrams it is observed that the Q-factor of Li-Zn ferrites increases with the addition of Ni-content to the pure Li-Zn ferrites which is consistent with your loss factor measurements data. It is also observed that at low frequency side, the Q-factor is almost constant with frequencies. Above 1 KHz, Q-factor increases with the increase of frequency. From the frequency characteristics of Q-factor a perfect frequency band can be identified at which the samples work as a good soft magnetic material with low loss. At low frequency region materials with low Q-values work as better filters. These types of filters can be used to minimize undesirable oscillations and high frequency content rapidly passing signals. These materials are successfully used to deduce ringing at high frequency applications.

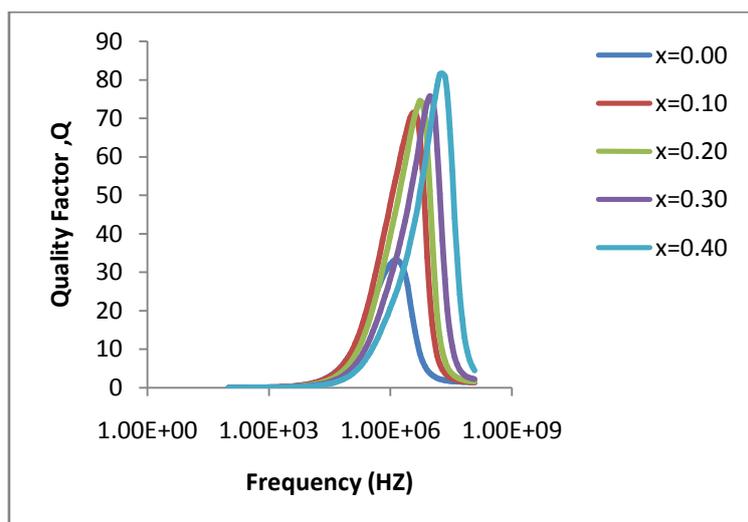


Fig. 6: The variation of relative Quality Factor with Frequency for $\text{Li}_{0.15}\text{Ni}_x\text{Zn}_{0.85-2x}\text{Fe}_{2+x}\text{O}_4$ ferrite (Where $x=0.00, 0.10, 0.20, 0.30, 0.40$) sintered at 1150°C .

Dielectric constant of Li-Ni ferrites has been calculated by measuring the capacitance using the formula:

$$\epsilon' = \frac{cd}{\epsilon_0 A}$$

where ϵ' is the dielectric constant, C is the capacitance of the sample, d is the thickness of the sample, ϵ_0 is a constant and A is the area of the circular sample $= \pi r^2$, where r is the radius of the sample. Fig. 7 presents the variation of dielectric constant with frequency at room temperature. The dielectric constant, ϵ' decreases with increasing frequency, which is rapid at lower frequencies and slower at higher frequencies. At much higher frequency it is very small and becomes independent of frequency. This is a normal dielectric behavior observed in most of the ferromagnetic materials, which may be due to the interfacial polarization as predicted by Maxwell and Wagner [43]. Similar types of dielectric dispersions are also reported in earlier [26, 47-48].

A strong correlation between the dielectric constant and conduction mechanism are available in the literature [40]. Dielectric constant of ferrites depends upon the number of available Fe^{2+} ions on the B-sites. The hopping of electrons between Fe^{2+} and Fe^{3+} , and Ni^{3+} and Ni^{2+} causes the local movements of electrons. Dielectric polarizations arise due to these local displacements of electrons which in turn gives rise the dielectric constant of ferrites [49]. Addition of Ni forms a non-magnetic Nickel-iron-oxide which segregates at grain boundaries results in an increase in dielectric constant. It is also known that dielectric constant of ferrites are corrected to the different types of polarization mechanisms [50]. Space charge polarization becomes dominant at low frequencies [50]. Increase of frequency weakens the space charge polarization which in turn causes to decrease of dielectric constant. However, various types of defects and dislocations in the crystal lattice invite the interfacial polarization to be active in the low frequency region [51-53]. For this reason, dielectric constant is higher at low frequency side.

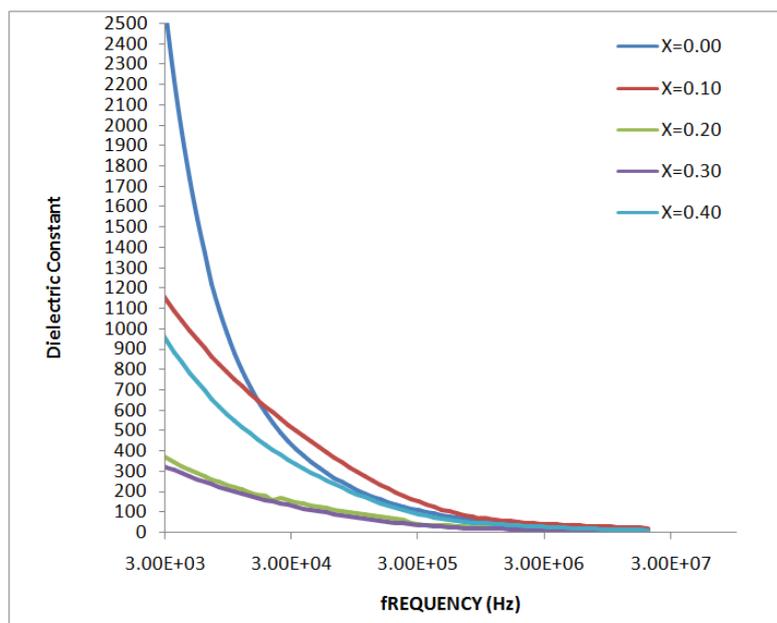


Fig. 7: The variation of Dielectric Constant with Frequency for $\text{Li}_{0.15}\text{Ni}_x\text{Zn}_{0.85-2x}\text{Fe}_{2+x}\text{O}_4$ ferrite (Where $x=0.00, 0.10, 0.20, 0.30, 0.40$) sintered at 1150°C .

Resistivity of ferrites depends upon the method of preparation, chemical composition, grain size and sintering conditions. Fig. 8 represents the variation of AC resistivity of Ni-Zn ferrites without and with Ni addition as a function of frequency at room temperature. According to the resistivity data, it has been noticed that with the addition of Ni content AC resistivity of Li-Zn ferrites decreases. It has also been found that at lower frequency region the resistivity decreases faster comparing to the higher frequency region and at much higher frequency the resistivity of the Li-Zn becomes almost independent of frequency. This means that all the samples show typical ferromagnetic behavior with frequency [54]. The conduction mechanism in ferrites is similar to the dielectric dispersion which is already discussed in the dielectric properties of ferrites in this paper. With the addition of Ni to the Li-Zn ferrites the mobility of charge carriers is increased. The increased mobility results in an increase of conductivity and thereby resistivity is decreased.

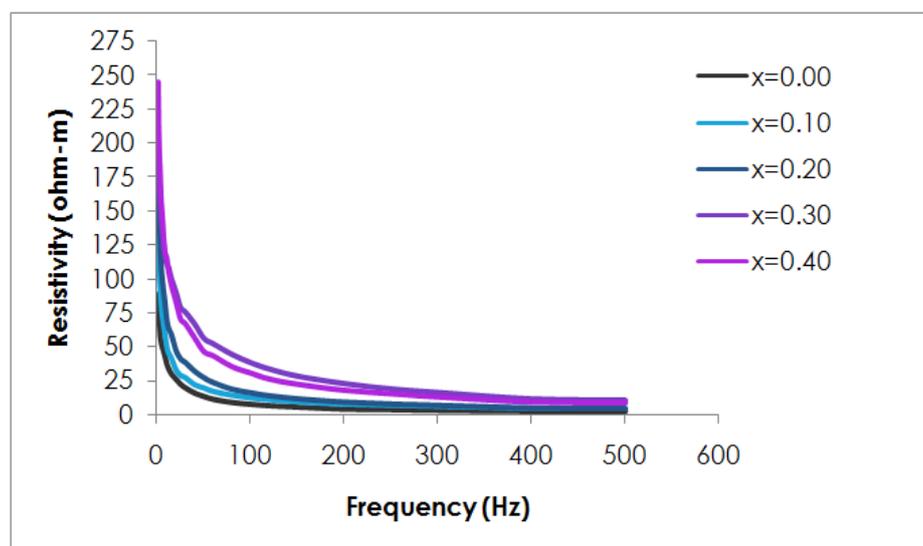


Fig. 8: The variation of AC resistivity with Frequency for $\text{Li}_{0.15}\text{Ni}_x\text{Zn}_{0.85-2x}\text{Fe}_{2+x}\text{O}_4$ ferrite (Where $x=0.00, 0.10, 0.20, 0.30, 0.40$) sintered at temperature 1150°C .

At high frequency region, the hopping frequency can no longer follow the frequency of the applied field and hence the resistivity becomes almost invariant [53]. During the moment when the jumping frequency of hopping charge carriers are equal to that of the applied field; resistivity becomes minimum and resonance takes place.

IV. Conclusions

Substitution of Zn in pure Li-Ni ferrites has been studied in this work. In our investigations, it has been observed that Ni-addition on Li-Zn ferrites plays a remarkable role in improving its magnetic, electrical and dielectric properties. The bulk density and x-ray density of the samples decrease with the increase in Ni-content. In XRD studies, the XRD pattern confirmed the single-phase cubic spinel structure of the samples without having any other intermediate phases. Magnetic properties such as complex permeability, loss factor and Q-factor have been measured as a function of frequency. Frequency stability of the real part of permeability increases with increasing Ni-content. The decrease in the permeability with increasing Ni-content might be connected with increased density with addition of Ni. Throughout the study, has been investigated that the real part of complex initial permeability remains unaffected at high frequency with the addition of Ni-content. The loss Factor decreases with increases frequency and Ni-content as well. Q-factor increases with increasing frequency and Ni-content. The dielectric constant decreases with the increase in frequency, which is faster at lower frequencies and slower at higher frequencies. The observed decrease in dielectric constant with increase in frequency might be due to the fact above certain frequencies the electronic exchange between Fe^{2+} and Fe^{3+} ions does not follow the frequency of the applied AC field. AC resistivity also observed to be decreased with the increase in frequency and increase with the increase in Ni-content. The minimum resistivity occurred when the frequency of the hopping charge carriers is equal to the applied field frequency termed as resonance frequency.

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