STRUCTURAL AND ELECTRICAL PROPERTIES OF NICKEL – ZINC FERRITES

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ABSTRACT

Ferrites with the formula $Ni_{1-x}Zn_xFe_2O_4$ (where x=07, 0.9 and 1) were synthesized via solid state double firing ceramic method. The final firing of the specimens was carried out at 1300 ^{o}C for 2 hours. The spinel phase crystallization was investigated by x-ray diffraction technique. The crystallite size (D), lattice constant (a) and tetrahedral and octahedral sites radii of the ferrite were calculated. The microstructure and surface topology was studied by using atomic force microscopy. The AC conductivity behavior as s function of frequency was discussed in view of Koop's theorem. Investigation of the dielectric constant, loss factor and loss tangent (δ) have been performed in the range from 100 Hz to 10 MHz.

KEYWORDS: ferrites, dielectrics, ferro materials.

INTRODUCTION

For the applications of the magnetic substances in circuits working at high and ultra high frequency, the best material is still ferrites. It is unlikely to be substitute ferrites with another magnetic substance in these applications. Ferrites are known as low priced materials, which made them employed in analog devices, microwaves, transformers, top-efficiency filters and variety of frequency circuiting. The ferrites electrical characteristics like dc conductivity are strongly dependent upon the material composition and synthesis conditions *e.g.* forming pressure, firing temperature and soaking time, grain size, pore size and pores distribution[1]. One of the most widely famous spinel magnetic materials is Nickel-Zinc ferrite. The ferrite $NiFe_2O_4$ possesses inverse spinel structure, which consists of tetrahedral and octahedral sites taken up by Fe^{2+} ions and by Fe^{2+} and Ni^{2+} ions, respectively. While the substituted ferrite of the structure $Ni_{1-x}Zn_xFe_2O_4$, are substances having a remarkable technological importance in many applications such as communications and entertainment devices. The ferrites of nickel-zinc are one of the most broadly utilized soft magnetic substances in high frequency apparatus motivated by its higher resistivity leading to reduced energy dissipation due to circulation currents[2]. The dielectric properties of ferrites are strongly dependant on its microstructure, composition and synthesis method[3]. Nickel-zinc ferri-materials are suitable for use in the high frequency band because of its good electrical properties with adequate low losses for microwave applications and are widely used in many other electronic instruments because of its environmental stability and remarkable performance over the radio frequency part[4]. This work aims to investigate the influence of frequency and Zn^{2+} ions addition on the Ac conductivity and dielectric properties of Nickel ferrites.

EXPERIMENTAL

Ferrite specimens having the composition of $Ni_{1-x}Zn_x$ Fe_2O_4 (x=07, 0.9, 1) were synthesized. The double firing method of ceramic powders was employed in the specimen preparation. Analytical grade oxides supplied by Aldrich company having purity of 99.9 % of Fe_2O_3, NiO, and ZnO were utilized. The oxides have been mixed in accordance with the exact molar ratios. The oxide mixtures were grinded. Then fine powder mixtures were calcined at temperature of 1000°C for 3 hours. The calcined mixtures were grinded and mixed again very well to get perfect homogeneity. After then, the specimens have been pressed uniaxialy. A pressure of 4 tons used to make samples into pellet shape. The pellet diameter is about 1.5 cm. The specimens were fired at 1300 ^{o}C for 2 hours and left to cool naturally to ambient temperature. The specimen's structure was examined by using XRD. The XRD radiation is Cu K with (= 1.540598). The XRD was performed on D2Phaser Bruker. The crystallite size (D) for the cubic cell structure was obtained

Microstructure was investigated using AFM performed on Angestrom Advanced Inc., 2008, USA. The pellets side faces were polished and then thermally coated with aluminum thin layer. These Ohmic contact aluminum electrodes have been prepared for electrical investigations. Measurements of the dielectric constant, loss and loss tangent have been performed over the frequency range 100Hz - 10MHz. The test performed on HP4284A LCR meter.

RESULTS AND DISCUSSION

Figures 1, 2, and 3 illustrate the XRD patterns of the ferrite $Ni_{1-x}Zn_x Fe_2O_4$, where x=07, 0.9, 1, respectively. The characteristic peaks of the diffraction patterns belonging to the Fd3m cubic spinel group. The patterns indicate the crystallization of pure cubic phase spinel structure with the main peak at (311). According to these data the existence of excellently polycrystalline ferrite structure could be confirmed. However, there were no secondary phases could be noticed[5]. Table 1 shows the results of crystallite size (D), lattice constant (a) and A- and B-sites radii. Fig. 4 reveals that the lattice constant

varies approximately linearly with increase in Zn content. However, this behavior indicates that the $Ni_{1-x}Zn_x$ Fe_2O_4 ferrite follows Vegard's law. Basically, the change in lattice constant with addition of Zn^{2+} may be ascribed to the greater ionic radii of Zn^{2+} (0.82 Å) than that of Ni^{2+}

(0.78 Å)[6]. The tetrahedral and octahedral sites were found to increase upon the increase in Zn addition. The increase in A- and B- site indicates that the Zn^{+2} ions were distributed over the tetrahedral and octahedral site.
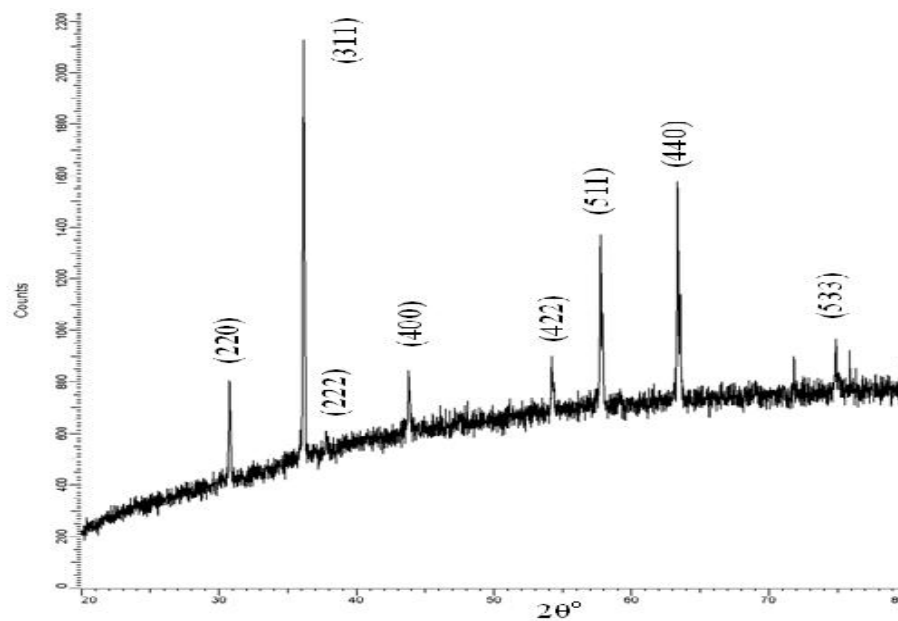
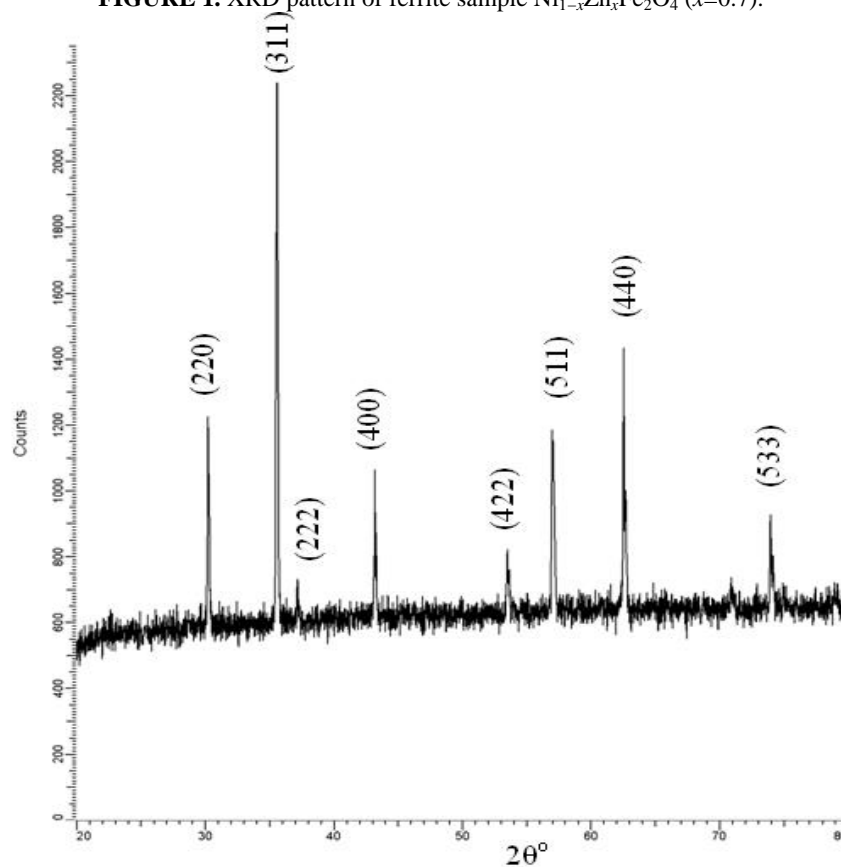**FIGURE 1.** XRD pattern of ferrite sample $Ni_{1-x}Zn_xFe_2O_4$ (x=0.7).

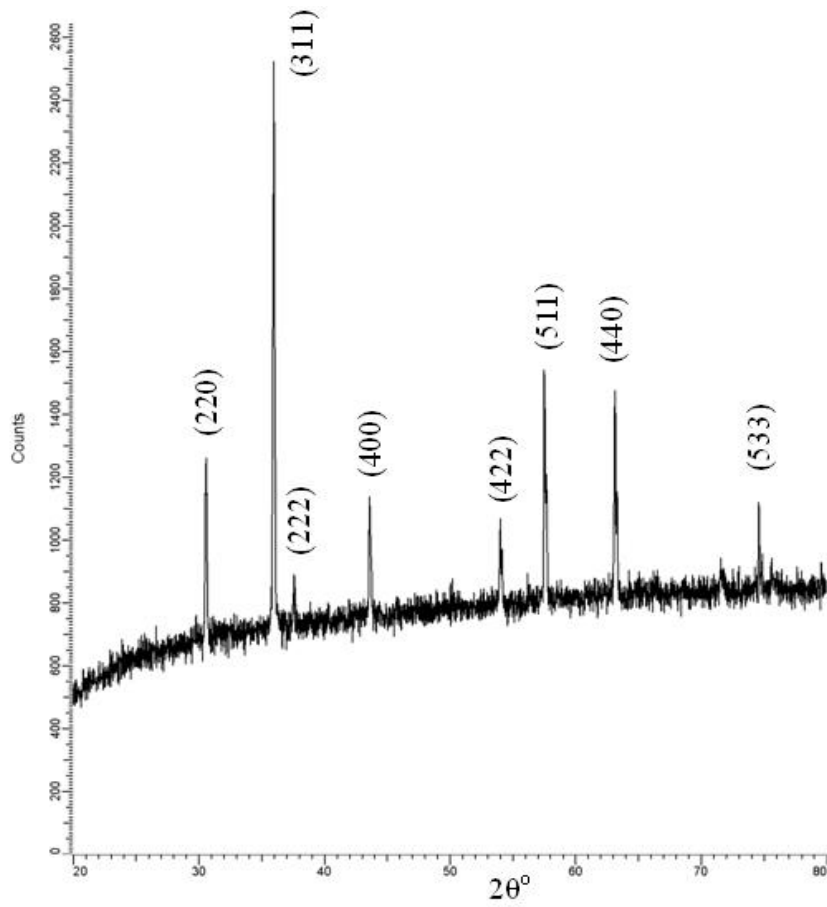**FIGURE 2.** XRD pattern of ferrite sample $Ni_{1-x}Zn_xFe_2O_4$ (x=0.9).

FIGURE 3. XRD pattern of ferrite sample $Ni_{1-x}Zn_xFe_2O_4$ ($x=1$).

TABLE 1. The crystallite size (D), lattice constant (a), A- and B-sites radii of $Ni_{1-x}Zn_xFe_2O_4$

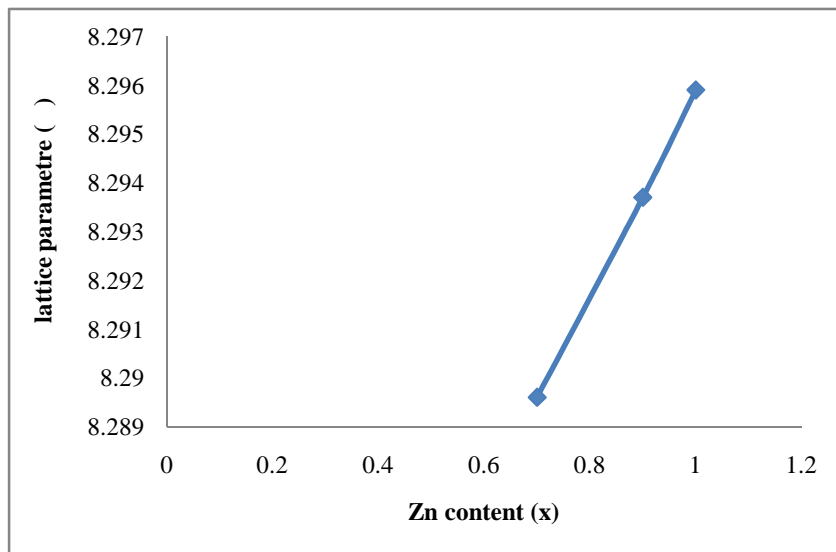| x	a	r_A	r_B	D nm
0.7	8.2896	0.4314	0.7024	101.62
0.9	8.2937	0.4748	0.7525	103.51
1	8.2959	0.4761	0.7539	112.36

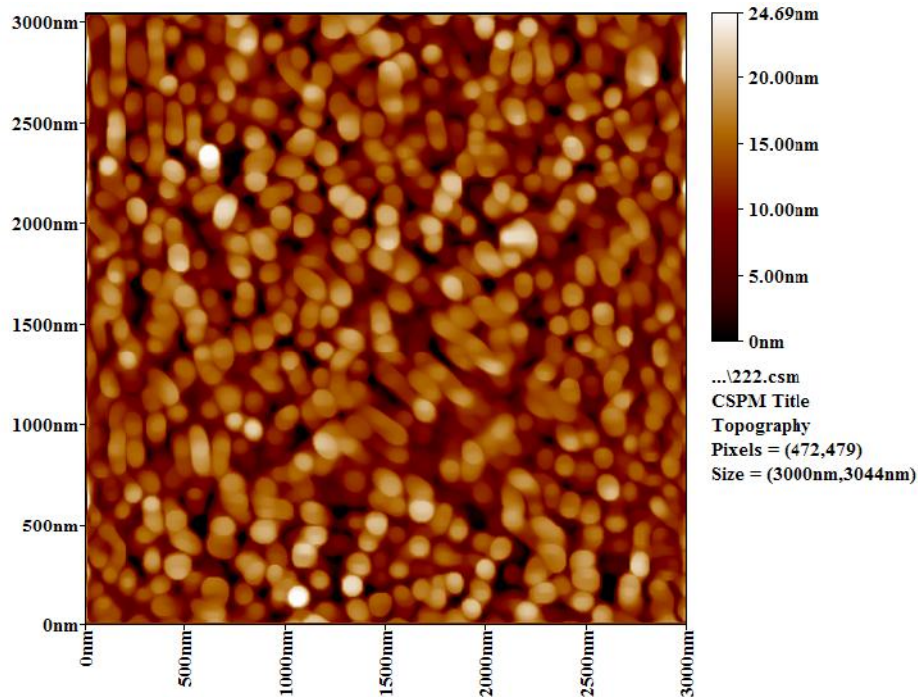**FIGURE 4**. Lattice constant versus Zn content (x)

FIGURE 5. AFM image of ferrite sample $Ni_{1-x}Zn_xFe_2O_4$ ($x=0.7$).

Fig.5 illustrates the atomic force microscopy photo of the prepared specimen with x=0.7 showing a homogeneous topography with little amount of porosity exist. The high densification of the sample may be attributed to the high sintering temperature and long sintering time of the prepared sample.
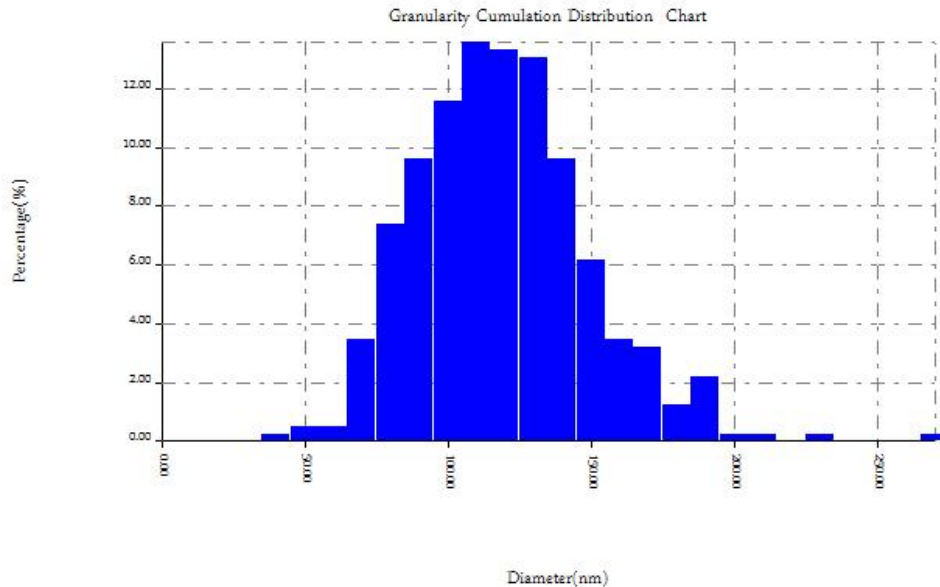

FIGURE 6. Grain size distribution of the ferrite sample $Ni_{1-x}Zn_xFe_2O_4$ ($x=0.7$).

Fig. 6 illustrates the granularity cumulating distribution chart of the specimen with x=0.7. The average grain size diameter of the specimen was found to be 114 nm. This is anticipated to reflect positive effects on the electrical properties of the material.

The AC conductivity is shown in Fig. 7. The increment in conductivity was noticed as a well behaved function of frequency. However, this behavior agrees perfectly with the relationship:

$$\sigma_{AC} = \omega \varepsilon_o \varepsilon'' = \omega \varepsilon_o \varepsilon' \tan \delta$$

where $_o$ is the vacuum permittivity, $'$ is the real part of dielectric parameter, $''$ is the imaginary part and tan is the tangent of loss angle. The relationship between dielectric properties of the material and frequency was debated according to Koop's theorem. It assumes that the ferrimagnetic substance demonstrates a multilayer capacitor behavior. Koop's theorem, considers the ferrimagnetic grain and its boundaries possess dissimilar properties. The influence of the multilayer capacitor rises as the frequency increase. This may cause the electrical conductivity of the ferrite material to increase [7].
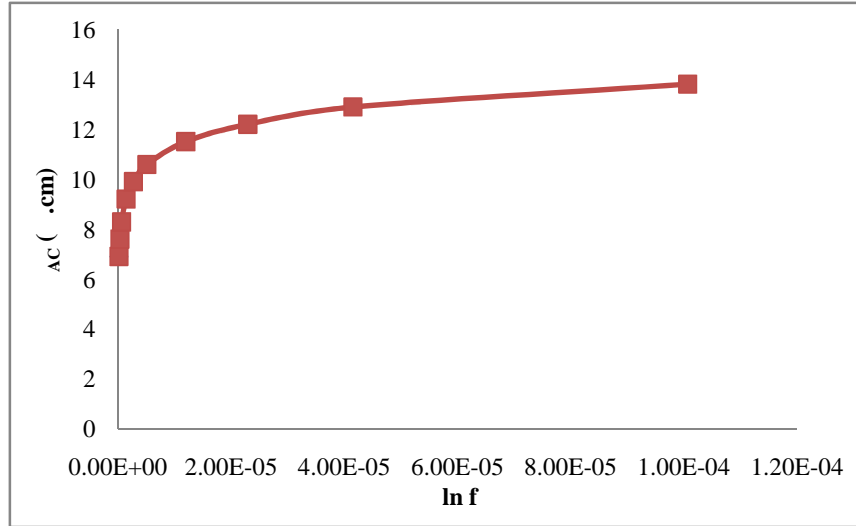

FIGURE 7. AC conductivity versus frequency of ferrite sample with the chemical formula $Ni_{1-x}Zn_xFe_2O_4$ (x=0.7).
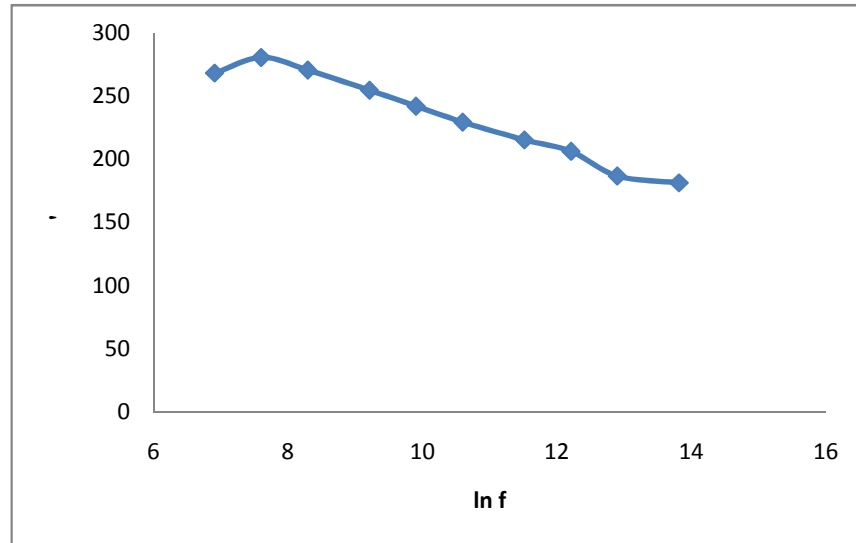

FIGURE 8. dielectric constant versus frequency of ferrite sample with the chemical formula $Ni_{1-x}Zn_xFe_2O_4$ (x=0.7).

Fig. 8 illustrates the change of dielectric constant of the samples with frequency. It shows a decrease in the dielectric constant as the frequency increase. The components of substance polarizability are known as $_e+ _i+ _o+ _s$. These components represent the susceptibilities accompanied with, electronic, ionic, dipolar, and space charge polarizability, respectively. The $_e$ occurs as a result of displacement of electron clouds in the dielectric field far-off from the equilibrium position. The material then shows net dipolar polarization. This happens in whole solid materials all the way to the optical frequencies ~10^{16} Hz. The $_i$ component occurs due to analogous ionic shift in the dielectric field which appears near the IR region of 10^{10}–10^{13} Hz. dissimilarly, the dipolar part of polarizability depends on frequency and temperature, because it symbolize the dipole orientation as well as the ionic hopping polarization. Space charge polarization which is the last part is caused by the mobility of charge carriers. Basically, the carriers display varying frequency responses which may be assorted. The lower frequencies region, the whole components of polarization exist and should be considered. However, within the optical frequencies region, *i.e.*, frequencies higher than the optical phonon one, only the $_i$ component dominates. When the dipolar component of polarization does not exist, then $*$ must be non-dispersive in the microwave

frequencies range. Therefore, dispersion below microwave frequencies is caused by dipolar component influences and sometimes by space charge component. Consequently, they exhibit a leading effect on the dielectric properties [8-10].
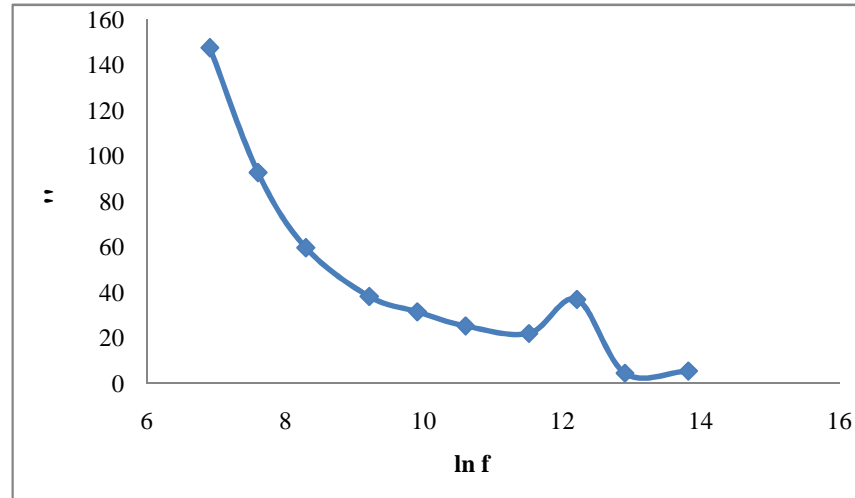

FIGURE 9. Dielectric loss versus frequency of ferrite sample with the chemical formula $Ni_{1-x}Zn_xFe_2O_4$ (x=0.7).

The change in dielectric loss as a function of frequency is illustrated in figure 9. It indicates that the dielectric loss decreases persistently with rising of frequency with small peaking of loss. The peaking behavior occurs as the jumping frequency of charge carriers (*i.e.* electrons) through the Fe^{2+} and Fe^{3+} becomes equivalent to the applied field frequency. If the material resistivity is too high, as is the case for the majority of microwave ferrites, as given by the equation

$$\tan = 1/ _o$$

where $_o$ represents the angular frequency of the maximum value of tan . Therefore, the loss tangent for such ferrite material is anticipated to fall down nearly inversely as a function of frequency [11].

CONCLUSION
The X-ray diffraction patterns of ferrites revealed pure cubic spinel phase, without any alter in the crystalline form of the primary phases. The substitution of Zn in the ferrite increased the lattice constant. The dielectric dispersion relation was discussed in the sense of electron interchange of the ions Fe^{2+} and Fe^{3+}. It was suggested that the polarization process in such ferrites was analogous to the mechanism of electrical conduction.

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