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**RESEARCH ARTICLE**

# Studying the Structure, the Electric and Magnetic Properties of Ni-Zn Ferrites.

**Jafer Fahdel Odah**

Department of Physics, College of Science, Al-Muthanna University , Samawa, Iraq.

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***Corresponding Author**

**Jafer Fahdel Odah**

## *Abstract*

The two types of ferrites have been prepared using co-precipitation technique ($Ni_{0.6}\,Zn_{0.4}\,Fe_2O_4$ and $Ni_{0.4}\,Zn_{0.6}\,Fe_2O_4$). The prepared ferrites were sintered at 800C˚. Four samples halve of these as pressed pellets (12.3mm diameter, 5-4mm thickness) and later halve as triodes ($R_{in}$=9.85mm, $R_{out}$=19.4mm). All of them were sintered at (1100 °C) for (2 hrs.). (X-ray diffraction (XRD) technique was used to study the structure and the scanning electron microscope also was used to determine the grain size. The electrical characteristics for these samples including measure the real and imaginary parts of dielectric constant ($\varepsilon_r$ *and* $\varepsilon_i$ which used to determine the loss factor ($tan\delta$) over the frequency range (20Hz - 3MHz). From the values of loss factor, the quality factor could be obtained. Also the electrical resistivity was calculated over the same range of the frequencies. The magnetic properties were measured using the same equipment, such as the initial permeability ($\mu_i$) after measuring the inductors (L) and the inductor factor ($A_L$) also was calculated.



## Introduction:-

Ni-Zn spinal type ferrites are interesting materials due to their magnetic properties. These materials are widely used from the end of the last century. These ferrites have been commercially used in recording heads, antennas rods, loading coils, microwave devices, and so on [1] and [2]. Spinal ferrites are good dielectric materials and they have wide applications ranging from microwave frequency to radio frequency the dielectric properties of these ferrite are very sensitive to the method of preparation and sintering condition [3].

As Ni-Zn ferrites are transition metal oxide semiconductors, their charge carrier concentration and electrical conductivity is mainly influenced by the quantity of metal ion pairs with different valence or the concentration of oxygen or cation vacancies. The microstructure of the polycrystalline ferrites plays also a great role on their transport properties. The hopping type charge transport in spinel ferrites takes place between ions in octahedral crystallographic site. Iron ions provide electron hopping $Fe^{3+} + e^- \rightarrow Fe^{2+}$ (n-type), but nickel ions provide hole hopping $Ni^{2+} + h^+ \rightarrow Ni^{3+}$ (p-type) [4].

Ni-Zn ferrites, technologically important materials, have spinel configuration based on a face-centered cubic lattice of the oxygen ions, with the unit cell consisting of 8 formula units of the type $(Zn_yFe_{1-y})[Ni_{1-y}Fe_{1+y}]O_4$, were the metallic cation in ( ) occupy the tetrahedral sites (A) and the metallic cations in [ ] occupy the octahedral sites [B][5]. Both dielectric and magnetic properties of these oxides depend on the type of cations and their distribution among the two interstitial positions, which in turn depends on the method of synthesis and calcination conditions [6,7]. The grain size increases with increasing calcination temperature of Ni-Zn ferrites [8,9] and the dielectric constant increases when the grain grows [10]. Many researchers have studied Li–Cd [11], Cu–Zn [12], Li–Mg [13], Cu–Cd [14], Ni–Zn [15], Co–Cd [16.], Mg–Zn [17] ferrite systems.

Nano ferrites form an important class of very good electrical materials because of their high resistivity and low energy loses (eddy currents) and hence has very vast technological application over a wide range of frequencies [18]. The order of magnitude of the conductivity greatly influences the dielectric and magnetic properties of ferrites. A lot of work is being carried out on the electrical properties of ferrites both in bulk and in nano form. Azadmanjiri [19], studied the electrical properties of Zn substituted $NiFe_2O_4$. He found that substitution of Zn has significantly decreased the electromagnetic properties such as dielectric constant and loss tangent. Thakur et al. [20] studied the dielectric properties of Mn–Zn nano ferrites. They reported that the resistivity of the studied system has increased 100 times than those samples prepared by the conventional ceramic methods. Yue et al. [21] studied the Ni–Cu–Zn nano ferrite system. They reported that the doping of Mn in formulation has largely affected the grain size and electromagnetic properties of the system. They also found that dielectric constant and dissipation factor are affected by the $MnO_2$ doping. In the present work, two types of ferrites have been prepared by co-precipitation technique ($Ni_{0.6}Zn_{0.4}$ $Fe_2O_4$ and $Ni_{0.4}Zn_{0.6}$ $Fe_2O_4$) and then the structure of these ferrites and some magnetic and dielectric parameters were studied.

## Methods:-

The powders of Ni- ferrites and Zn- ferrites were prepared using Co-precipitation in (400) ml distil water, Ferrites have been synthesized using aqueous ammonia to avoid incorporation of sodium. Control of synthesis parameters has resulted in the production of ferrite particles , having a meta stable cation distribution as revealed from micro structural and magnetic studies. The pH of the solution was adjusted to 7.5-8.5 using ammonia solution. The solution was uniformly heated at 100˚C with constant stirring to transform it into a gel and then filtered gel was obtained by dehydration process. The dried gel was combusted with the evolution of large amount of gases and it resulted in the formation of loose powder. The prepared powder was then calcined at 800˚C.Two types of ferrite were synthesized which are of Ni- ferrites, Zn- ferrites to formed final production with composition $Ni_{0.6}$ Zn $_{0.4}Fe_2O_4$ and $Ni_{0.4}$ Zn $_{0.6}Fe_2O_4$,then it was pressed ($5tons/Cm^2$) as rings ($R_n$ ) and ($3tons/Cm^2$) as pelts ($D_n$). the temperature of final sintering in this case was kept at 1100˚C. The pellets samples were well polished to remove any roughness and the two surfaces of each pellet were coated with silver paste as contact material for electrical and dielectric measurements using ***L***.C.R type (micro test 6379), the dielectric measurements were done as a function of frequency in the range ( 20 Hz – 3MHz) at room temperature. The real part of dielectric constant was calculated using equation

$$\varepsilon_r = \frac{\mathrm{C\,d}}{\varepsilon^\circ\,\mathrm{A}} \qquad (1)$$

Where C is the capacitance measured of the pellet in pF, d the thickness of the pellet, A the cross-sectional area of the flat surface of the pellet and ε˚ the constant of permittivity for free space.

Dielectric loss tangent (tanδ) was calculated using the relation

$$tan\delta = \frac{\varepsilon_i}{\varepsilon_r} \qquad (2)$$

The quality factor ($Q$) also calculated using the relation

$$Q = \frac{1}{tan\delta} \qquad (3)$$

The electric resistivity (ρ) was calculated from the equation

$$\rho = \frac{\mathrm{RA}}{\mathrm{d}} \qquad (4)$$

Where R is the resistance of sample which measured by LCR meter under same range of frequencies.

Magnetic properties were measured using the same equipment. Initial permeability (μi) was calculated after measuring the inductor (L), and then using the formula

$$\mu_i = \frac{\mathrm{L}}{\mathrm{L}_\circ} \qquad (5)$$

Where $L_o=4.6\ N^2d\ \log(r_{out}/r_{in})\times10^{-9}$ , rin is the inner radius and rout is the outer radius, and N is the numbers of turns.

The inductor factor ($A_L$) was calculated using the following formula

$$A_L = \frac{L}{N^2} \qquad (6)$$

## Results and discussion:-

### XRD discussion:-

The phase identification and lattice constant determination were performed by x-ray diffraction pattern (XRD). Typical XRD pattern of ferrites sample is shown in Fig.(1) and (2).

Both samples show good crystallization, with well-defined diffraction lines. The structure can be indexed as a single-phase cubic spinal structure. It is obvious that the characteristic peaks for spinel ferrites appear in the samples as the main crystalline phase. The peaks (2 2 0), (3 1 1), (2 2 2), (4 0 0), (4 2 2), (5 1 1) and (4 4 0) correspond to spinal structure.

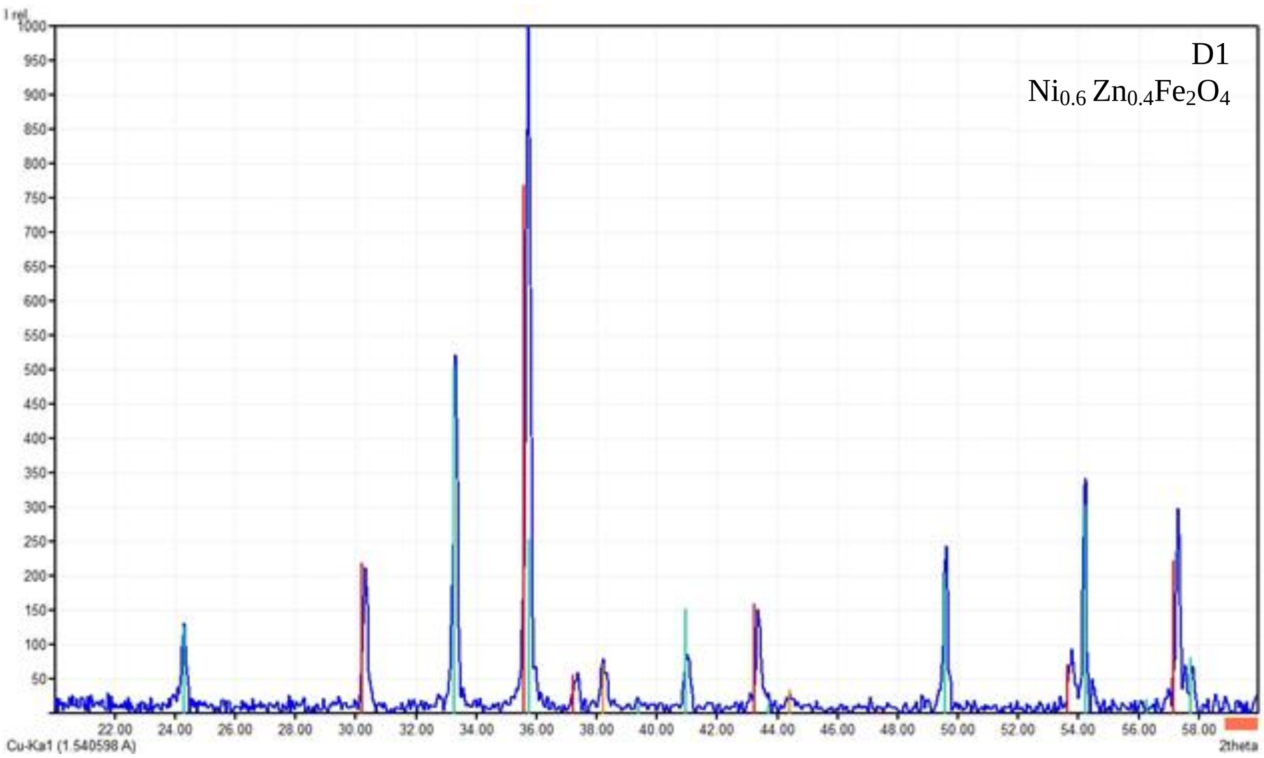


Figure (1) X-ray diffraction patterns of the sample with composition of $Ni_{0.6} Zn_{0.4}Fe_2O_4$

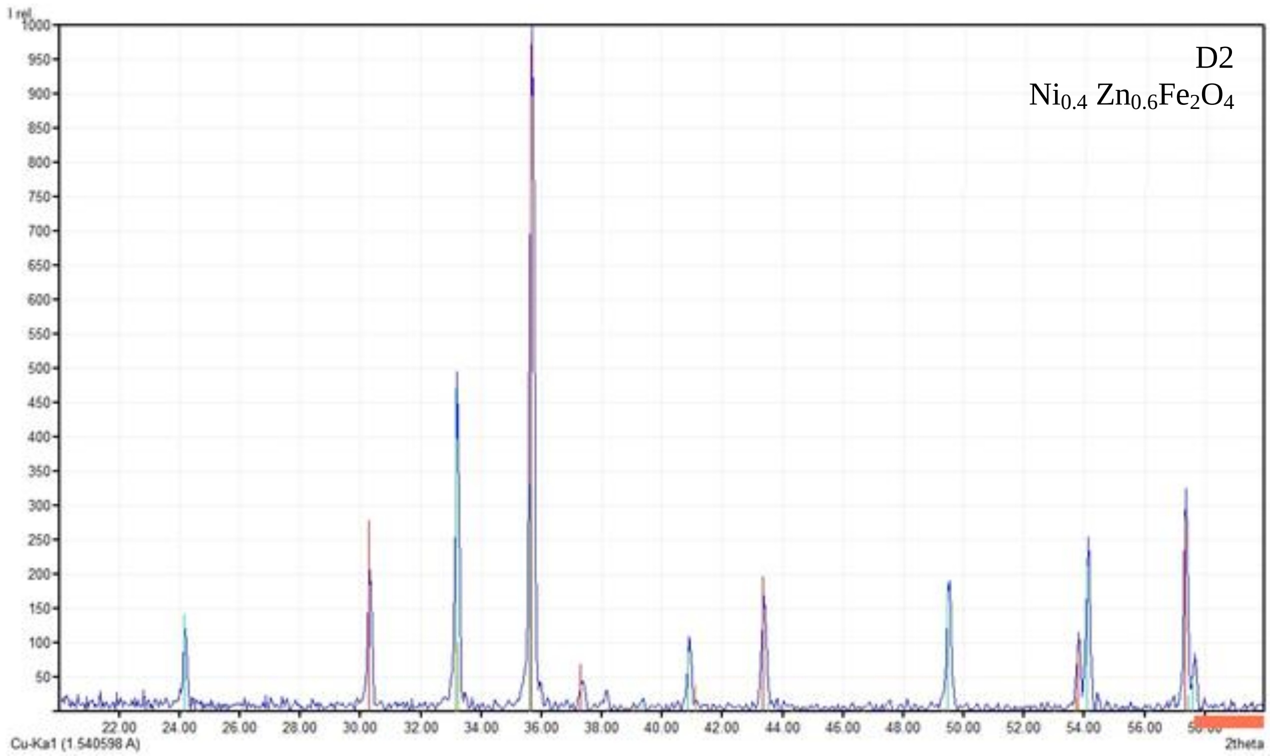


Figure (2) X-ray diffraction patterns of the sample with composition of $Ni_{0.4}$ $Zn_{0.6}Fe_2O_4$

**Scanning Electron Microscope (SEM):-**

Figures (3) and (4) show SEM of fractured surfaces for the Ni-Zn ferrites sintered at 800 $C^o$. The grain size increases with the zinc content because a zinc addition can enhance the diffusion of elements and cause the particles to grow large.

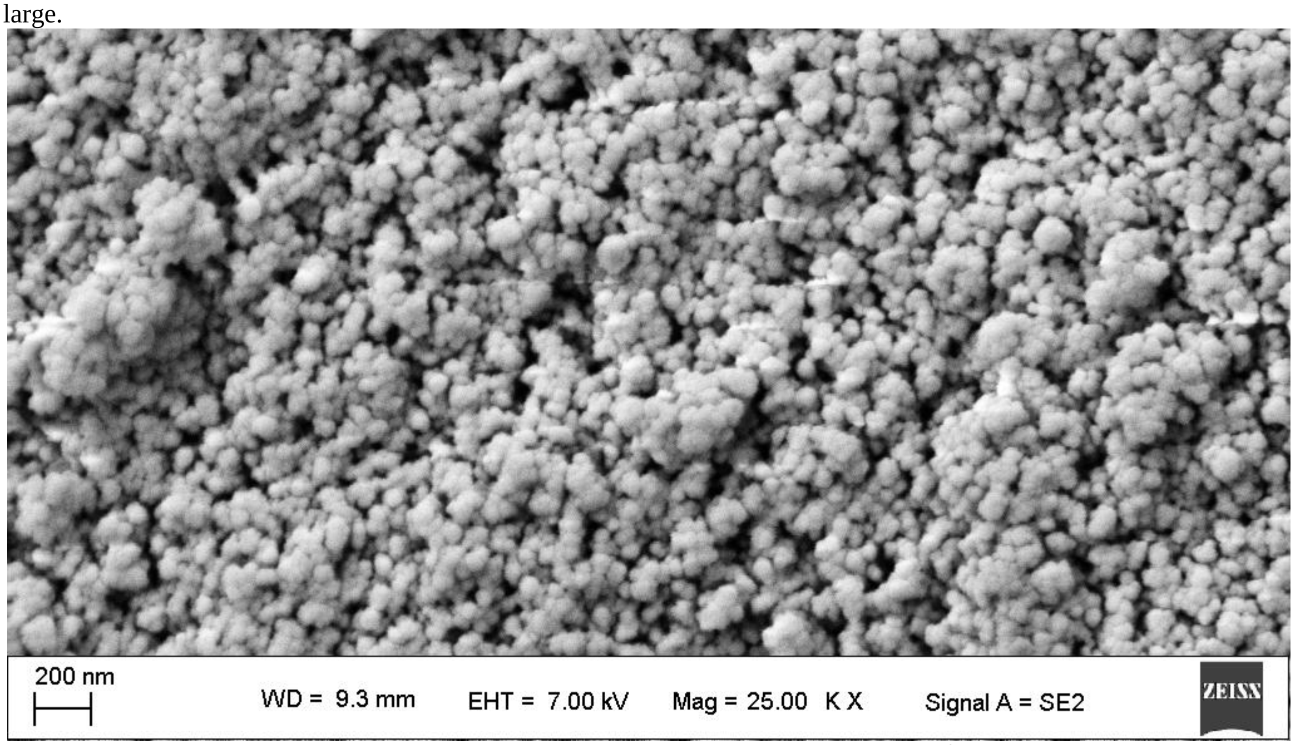


Figure (3) SEM micrograph of $Ni_{0.6}Zn_{0.4}Fe_2O_4$ sintered at 800 $^oC$

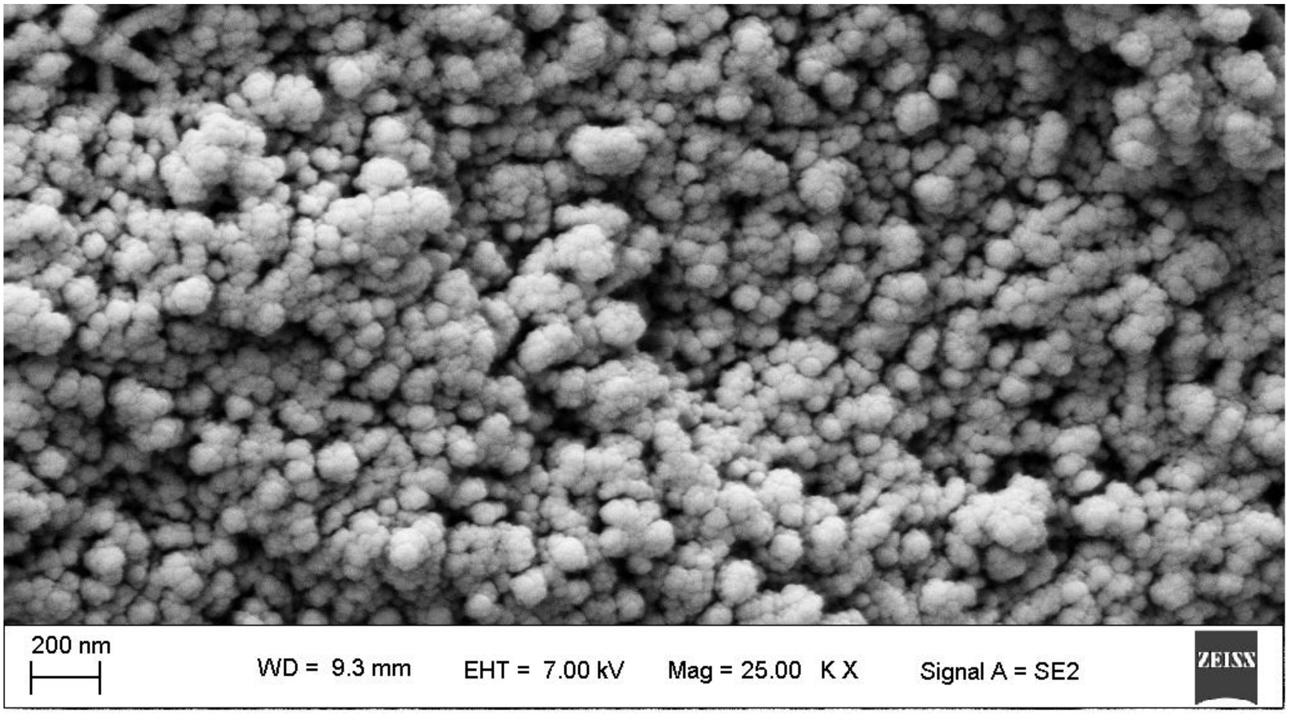


Figure (4) SEM micrograph of $Ni_{0.4}Zn_{0.6}Fe_2O_4$ sintered at 800 $^oC$

**Electric and Magnetic Properties Calculations:-**

The dielectric behavior in ferrite can be explained based on the assumption that the mechanism of dielectric polarization is similar to that of conduction. Many scientists have established a strong correlation between the conduction mechanism and dielectric constant of ferrite [22,10]. In the studies, they have explained the dielectric behavior based on a number of available $Fe^{2+}$ ions. The electronic change such as $Fe^{2+} \leftrightarrow Fe^{3+}$ results in a local displacement of electron, which determines the polarization and thus the dielectric constant of ferrite .we observed that from figure (5) which shows the variation of the real part of dielectric constant ($\varepsilon_r$) behavior with increasing frequency of two samples. A assembly of space charge carriers in the inhomogeneous dielectric structure, is discussed by Maxwell and Wagner [22] and [23]. Since an assembly of space charge carrier in the inhomogeneous dielectric structure described requires finite time to line up their axis parallel to an alternating electron field, the dielectric constant naturally decrease.

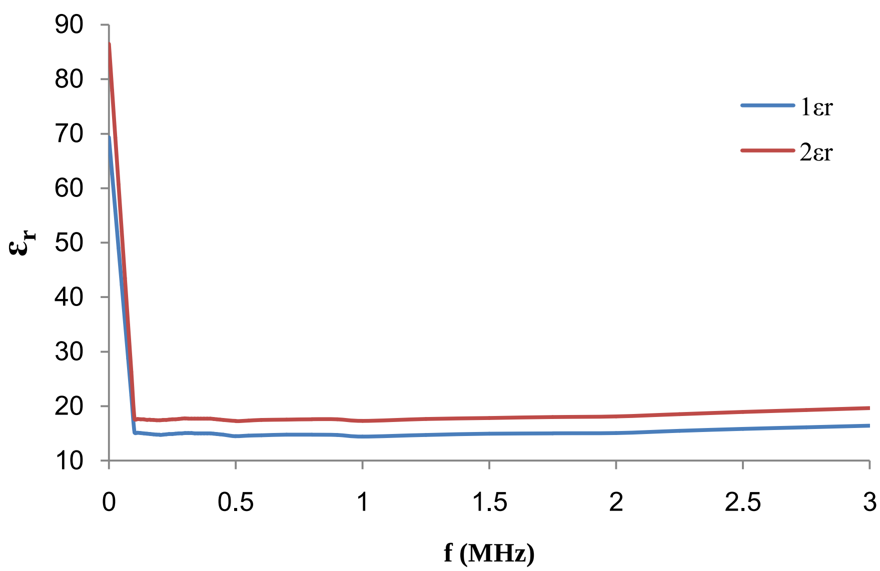


Figure (5) Variation of real part of dielectric constant with frequency of D1 and D2.

It is clear that from figure (5) that the dielectric constant for both the two samples decreases with increasing frequency, this decreasing is a normal dielectric behavior of spinal ferrite and is due to the reason that, as the frequency of the externally applied field increases gradually, though the number of ferrous ions is present in the ferrite material, the dielectric constant decreases. The reduction occurs because beyond a certain frequency of the externally applied electric field, the electronic exchange between ferrous and ferric ions, i.e. $Fe^{2+} \leftrightarrow Fe^{3+}$ cannot follow the alternating field.

The decrease of dielectric constant with increase of frequency as observed in the case of mixed nickel–zinc ferrites is a normal dielectric behavior of spinel ferrites. The normal dielectric behavior was also observed by several investigators in the case of Li–Ti [24], Ni–Cu–Cn [25], Mg–Ti–Zn [26] and Co–Zn [27] ferrites.

Figure (6) shows the variation of the imaginary part of dielectric constant ($\varepsilon_i$ ) behavior with increasing frequency of two samples. can be explained on the basis of Koop's two layer model and Maxwell- Wagner polarization theory. To interpret the frequency response of dielectric constant in ferrite and dielectric materials , Koops [28] suggests a theory in which relatively good conduction grains and isolating grain boundary layers of ferrite material can be represent with the behavior of an inhomogeneous dielectric structure.

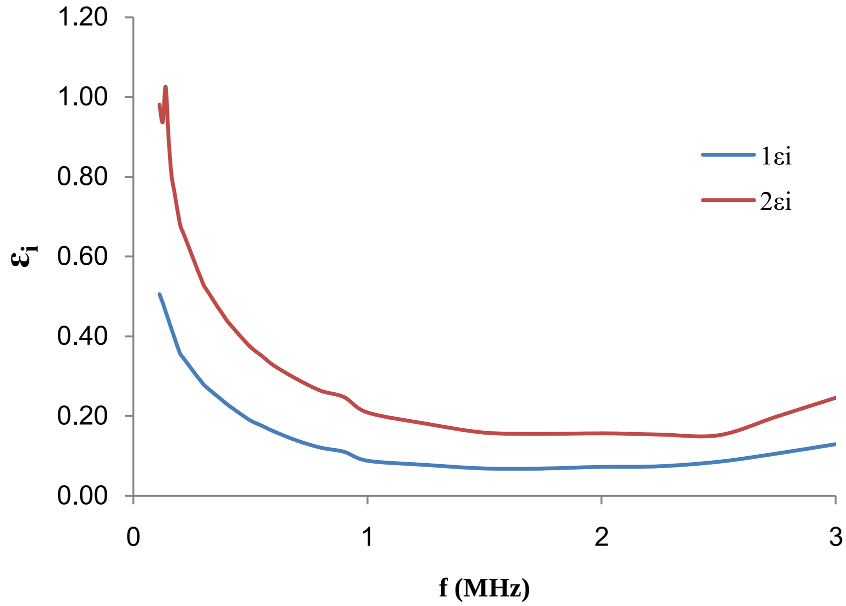


Figure (6) Variation of the imaginary part of dielectric constants with frequency of D1 and D2

The decrease in imaginary part of dielectric constant is pronounced more in comparison to real dielectric constant. The low dielectric values make these ferrites useful in higher frequency applications.

Figure (7) shows the variation of dielectric loss (tanδ) factor with frequency, at room temperature. The loss tangent decreases as the frequency of the alternating field increases. The values of (tanδ) depend upon a number of factors such as stoichiometry, $Fe^{2+}$ content and structural homogeneity, which in turn depend upon the composition and sintering temperature of the samples. The decrease of (tanδ) with an increase of frequency can be explained on the basis of Koop’s phenomenological theory.

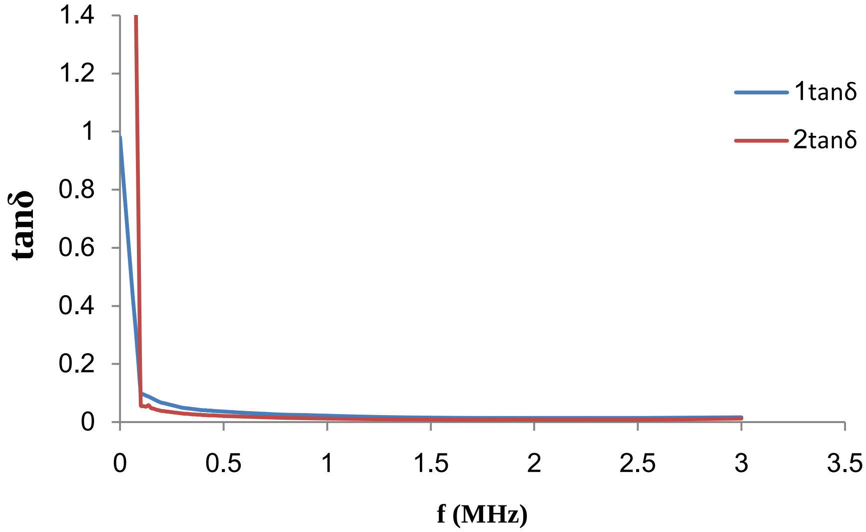


Figure (7) Variation of loss tangent (tanδ) with frequency

Frequency dependent relative quality factor of the samples sintered at 1200 $^oC$ is shown in Fig. (8). The high value of Q-factor is expected for high frequency magnetic applications .This factor is increases with an increasing of frequency showing a peak and decreases with further increase of frequency. It is seen that the Q-factor deteriorates beyond 2 MHz. The loss is due to the lag of domain wall motion with respect to the applied alternating magnetic field and is attributed to the various domain effects [29] such as non-uniform and non-repetitive domain wall motion, domain wall bowing, localized variation of flux density, nucleation and annihilation of domain walls. This happens at the frequency where the permeability begins to drop with frequency. This phenomenon is associated with the ferrimagnetic resonance within the domains and at the resonance maximum energy is transferred from the applied magnetic field to the lattice resulting in the rapid decrease in Q-factor.

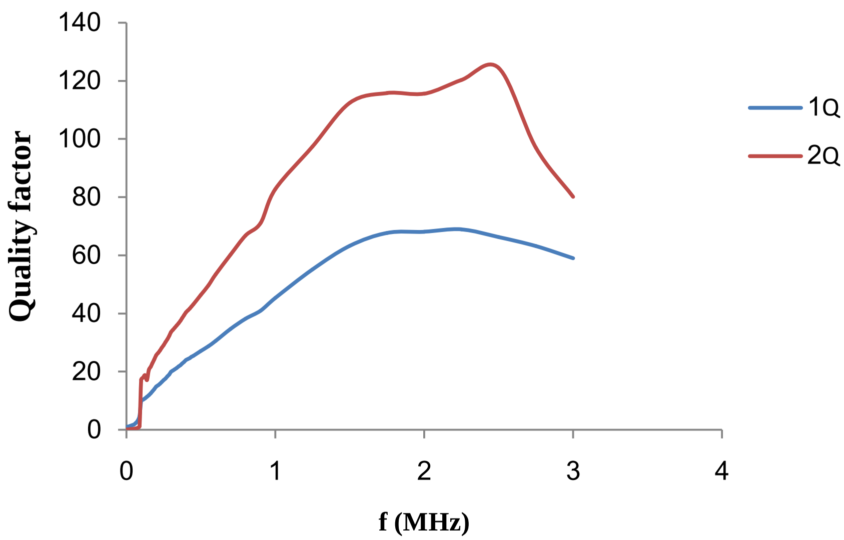


Figure (8) Variation of quality facto (*Q)* with frequency

The variation of DC resistivity versus frequency for both two samples is represented in Figure (9).

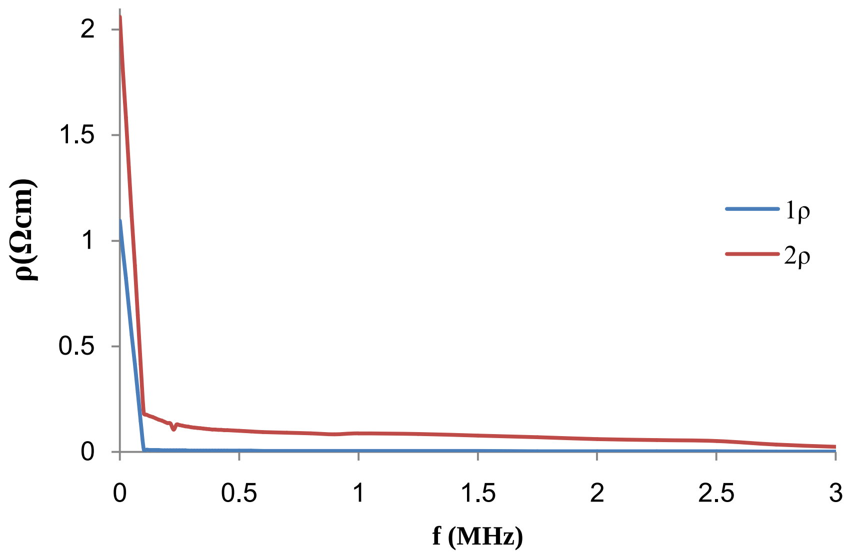


Figure (9) Variation of the resistivity of the two samples D1 and D2 with the frequency

It is clear from figure (9) that the increasing in frequency causing increase in the localized vibration of electrons this leads to increasing the electron transitions between $Fe^{2+} \leftrightarrow Fe^{3+}$, and the result of these transitions leads to decreasing in resistivity. The resistivity of the samples is also found to increase with Ni content decreasing. The increasing trend of resistivity (ρ) with Ni content can be attributed to the microstructural aspects.

Figure (10) show the initial permeability variation against the frequencies for the two prepared Ni-Zn ferrites with different Ni and Zn content.

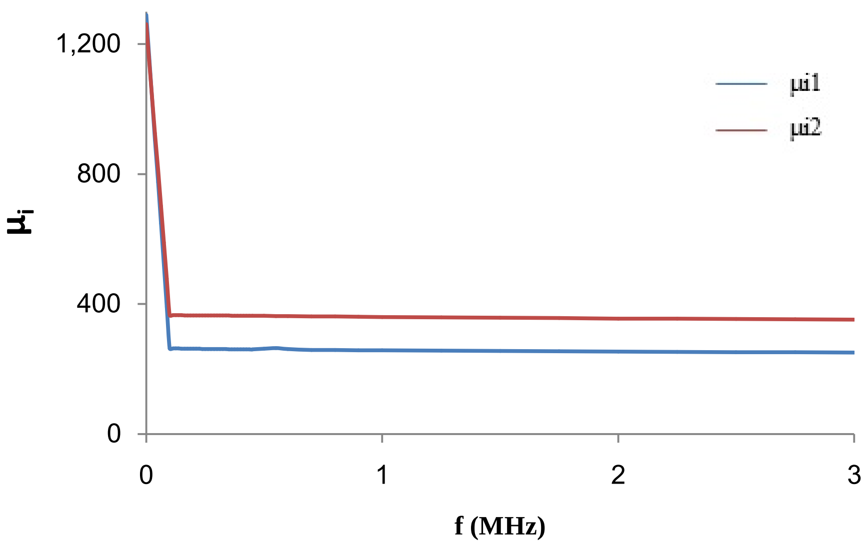


Figure (10) Variation of the initial permeability of the two samples D1 and D2 with the frequency

The general characteristic of the spectra is that initial permeability, at low frequency it is found that the initial permeability falls sharply and it is remains constant in a certain frequency range, i.e. at higher frequencies for both two samples. It is also found from Fig. (10) that the initial permeability increases with increasing Zn content and with decreasing Ni content, this increment may be related to the replacement of $Ni^{2+}$ ion by $Zn^{2+}$ ion [30].

The inductor factors versus the frequencies of R1 and R2 are shown in figure (11).

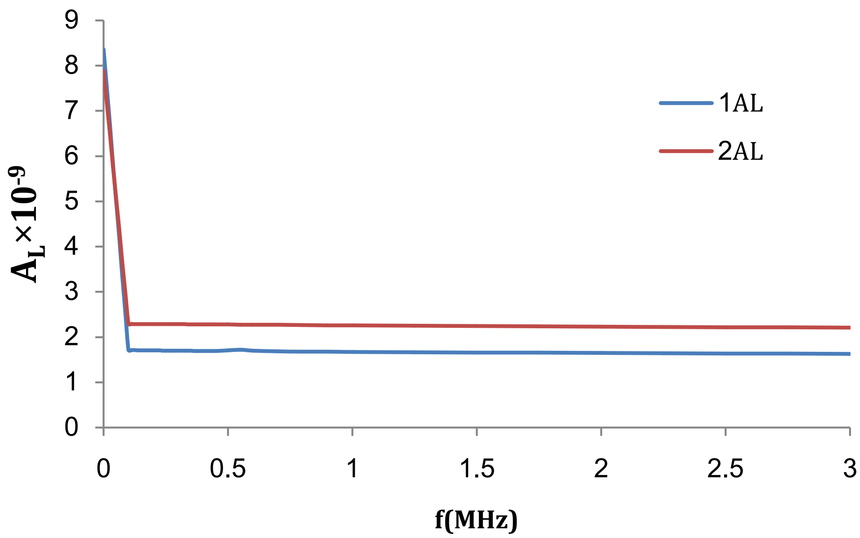


Figure (11) Variation of the inductor factors of the two samples R1 and R2 with the frequency

At low frequencies the inductor factor decreases rapidly up to 100KHz due to the low conductivity of ferrites which caused eddy currents strongly weaken the magnetic field in the core end that leads to decrease the inductance. At high frequencies one can observe that the inductor factor independent of frequency more than 100KHz.

## Conclusions:-

The spinal ferrite, especially Ni-Zn ferrite, it is possible to influence magnetic and electric properties of ferrite by changing ratio between nickel ion and zinc ion or by making non-stoichiometric compounds. Composites synthesized by co-precipitation technique were characterized using X-ray diffraction and SEM, the XRD patterns show the formation of a pure Ni-Zn spinel structure. SEM images show that the grain size gets larger with the increasing of zinc concentration. The composition $Ni_{0.4}Zn_{0.6}Fe_2O_4$ exhibits highest of values of quality factor, dielectric constant, dielectric loss tangent, also the initial permeability has been increased with the increase of zinc content and attributed to the grain growth. The resistivity of the samples was studied. It is observed that the resistivity of Ni–Zn ferrites prepared by co-precipitation technique is increases with Ni content increasing.

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