Dependence of the Ni$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ spinel ferrite on frequency and temperature using impedance spectroscopy tool.

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Abstract
Ni$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ ferrites were prepared by the conventional solid-state method. The X-ray study–allowed the identification of a single orthorhombic phase. The conductivity of the prepared sample was measured at different frequencies in the temperature range 340 K to 600 K. The obtained results are discussed in terms of charge carriers hopping. We showed that the dielectric permittivity is high at different temperatures and various frequencies. The impedance plane findings present semicircle arcs at various temperatures and an electrical equivalent circuit was determined. In addition, the obtained two relaxation times were established as a function of temperature.

Keywords
High permittivity, impedance spectroscopy, relaxation time, thermally activated, Maxwell-Wagner

I. Introduction
Recently, spinel ferrites are amongst the most versatile materials due to both their rich properties and their potential use in a wide range of advanced applications [1-5]. In particular, the diverse spinel ferrites Zn-Ni, are interesting for their large diversity physics properties, like high electric resistivity, low dielectric loss and large permeability [6-8]. As a matter of
fact, the performance of spinel ferrites depends on the different preparation methods, like hydrothermal, co-precipitation, ball milling method and sol-gel combustion [9,10]. The physical properties of these materials depend on the morphological parameters such as density, grain size and lattice constant [11].

Several researchers were interested to the Ni-Zn systems [12-16]. The insertion of Ni metal ion in ZnFe$_2$O$_4$ ferrite system improves its practical applications. However, Ni-Zn ferrites have been used widely as high frequency ferrite because of high permeability in radio frequency region and high electrical resistivity [17]. For these reasons, the Complex Impedance Spectroscopy (CIS) measurements are right tool to study the electrical properties of ferrite materials which allow the description of the conduction processes and the determination of several dielectric greatness, appropriate to samples [18].

This paper aims to study the X-ray diffraction, impedance spectroscopy, dielectric and electric properties of Ni$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ ferrites with temperatures varying from 340 K to 600 K and in a large frequency range going from 40 Hz to 10 MHz. From the DC and AC conductivity measurements, we explain the charge carriers transport mechanism in the sample. We examine the dependence of the high permittivity as a function of temperature and frequency. Both relaxation time and activation energy values of our samples are determined as a function of temperature and frequency in the materials.

II. Experimental section

Ni$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ Nickel-zinc ferrites were synthesized using solid state reaction technique. The precursor having a purity greater than 99% are mixed and then deposited in uni-axial pressure of $10^5$ N/cm$^2$ for obtain pellets with 10 mm of diameter and 1.5 mm of thickness. We heat the compound for 24h at 1200°C to obtain a better homogeneity of the samples. The dielectric characterization of the sample is done after an aluminum layer deposit of 200 nm thickness.
using thermal heat evaporation under vacuum, on each side of the pellet. To achieve good
conduction of charge carriers, a gold wire is attached to the aluminum layer.

Agilent 4294A impedance analyzer, whose frequency scale is of various from 40 Hz to 100
MHz over large temperature range 340 at 600 recorded the dielectric data. All measurements
are made in dark to avoid the effects of light. We use X-Ray Diffraction (XRD), with
monochromatized Cu–Kα radiation λ_{Cu}=1.5406Å at room temperature for structure
characterization. Phase purity, cell parameters of the ferrites nanoparticles and refinements of
the XRD data that were carried out with the Rietveld refinement using the Fullprof program.

III. Results and discussion

1. X-ray diffraction study

Elemental composition, phase purity homogeneity, lattice structure and cell parameters of
the Ni_{0.4}Zn_{0.6}Fe_{2}O_{4} samples were studied by XRD technique. The powder X-Ray Diffraction
XRD pattern of our compound is presented in Fig. 1-a. We deduce that the sample is in single
phase without any impurity. The refinement of the structure, using Fullprof software,
validates orthorhombic structure with a Pbnm space group in which a = 8.41 Å and V =
595.97 Å³. We determine the average grain size by the Scherrer’s formula [19]

$$D_{sc} = \frac{(0.9 \times \lambda)}{\beta \cos \theta}$$  \hspace{1cm} (1)

with θ is the Bragg angle, λ is the wavelength (λ_{CuKα}=1.5406 Å) and β the Bragg angle the
full width at half hauteur for the most intense diffraction peak.

$$\beta = \left(\beta_{m}^{2} - \beta_{i}^{2}\right)^{1/2}$$  \hspace{1cm} (2)

where β_{i}^{2} is the Full Width at Half Maximum FWHM of a standard silicon sample and β_{m}^{2}
is the experimental FWHM. The value of the average grain size of our samples is 72µm.

2. Electrical conductivity

This physical entity is expressed by the following equation:
\[
\sigma = \frac{hG}{A}
\]  \hspace{1cm} (3)

where \(A\) is the pellet area of the sample, \(h\) is its length and \(G\) is the conductance.

Fig. 2 shows the conductivity evolution as a function of frequency at various temperatures ranging from 340 K to 600 K. We note that the conductivity is almost independent on the frequency until \(10^3\) Hz regardless of the temperature, which coincides with the dc-conductivity. From the \(10^3\) Hz we observe an increase of the conductivity as a function of the frequency for all temperatures. Thus, we can model the behavior observed by this equation:

\[
\sigma(f) = \sigma_d + Af^s
\]  \hspace{1cm} (4)

where \(\sigma_d\) is the electrical conductivity, \(A\) is a constant, \(f\) is the frequency and \(s\) is the slope in region of evolution as a function of frequency \(0 \leq s \leq 1\) [20]. This conductivity evolution is explained by the heterogeneous medium model of Maxwell-Wenger [21] composed of conductive grains and grain boundaries. It forms a potential barrier blocking the transfer of charge. The low conductivity is resulting from the weak electron hopping in the region. So the ac conductivity corresponding to a jump of localized charge carriers in defect sites is separated by potential barriers and therefore it is hopping type conduction.

Fig. 3 exhibits the conductivity variation with the temperature at different frequencies. The curve shows the increase in conductivity with increasing temperature for six frequency values ranging from \(10^2\) to \(10^6\) Hz. This variation is explained by the thermally active charge jump model [22]. Thus, the charge carriers transfer between the sites is improved with the temperature increasing. The evolution of the temperature can be given by the following equation [23]:

\[
\sigma = \sigma_0e^{-\frac{E_a}{k_BT}}
\]  \hspace{1cm} (5)

where \(E_a\) is the activation energy, \(T\) is the absolute temperature, and \(K_B\) is Boltzmann constant. The conductivity evolution according to temperature proves a semiconductor
behavior of this ferrite [24]. The variation of the conductivity logarithm as a function of the inverse of temperature is given in Fig4.

In fact, the fit by the equation (5) (Fig.4) gives the activation energy for each frequency. The electrical conductivity takes place by jump phenomena of charge carriers between defect states in the band gap. We notice the decrease of the activation energy when the frequency increases. This behavior is due to the effect of the frequency which increases the area of space charges.

3. Impedance measurement

Fig.5 presents the variation of imaginary impedance $Z''$ part as a function of real impedance $Z'$ part of complex impedance at various frequencies and temperatures from 340 K to 600 K at each 20 K of Ni$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$. The data shows two semicircles that decrease as a function of temperature. Every semicircle could be associated with contribution; at low frequencies the semicircle can be attributed to the grain boundary and at high frequency the second semicircle is attributed to grain contribution. The presence of both semicircles shows that the dielectric process obeys to a two relaxation phenomena. We note that the decrease of the semicircles diameters decreases with increasing temperature, which can be explained by the increase of the mobility of charge carriers. Therefore, the resistance decreases with increasing temperature.

This behavior is due to the heterogeneity of our compounds where the grains are of low resistance, but in the grain boundaries the displacement of the charge carriers is difficult.

The decentralization of semicircular on the axis of real impedance $Z'$ suggests a non-Debye dielectric relaxation. Thus, the complex impedance is defined by the formalism of Cole-Cole by the following relation [25]:

$$Z^*(\omega) = \frac{R}{1+(\frac{\omega}{\omega_0})^{1-n}}$$

(6)
when \( n \to 0 \), we find the classical relation of Debye’s [26]. Fig.6 shows the fit using Zview software, we simulated the curves of imaginary impedance \( Z'' \) vs. \( Z' \) real impedance as a function of frequency at different temperature. We observe a good agreement between the curves measured and the data obtained by the simulations. We determined that the equivalent circuit corresponds to our composite. The circuit is formed by a series of three R-CPE in parallel combination where CPE is the Constant Phase Element and R is the resistance. The combinations are attributed to non homogeneous sample containing several different conductivity zones. The CPE is determined using the following relation [27]:

\[
Z_{\text{CPE}} = [A(j\omega)^\alpha]^{-1}
\]  

(7)

Where \( A \) is a proportional factor, \( \omega \) is the angular frequency and \( \alpha \) is the exponent between 0 and 1. At \( \alpha = 1 \) the CPE is considered as an ideal capacitive element, whereas for \( \alpha = 0 \) it is equivalent to resistance.

Hence, the total impedance of the structure can be written as:

\[
Z^* = \sum_{i=1}^{n=3} \frac{[A_i(j\omega)^{\alpha_i}]^{-1}}{[A_i(j\omega)^{\alpha_i}]^{-1} + R_i} = Z' - jZ''
\]  

(8)

Where \( Z' \) and \( Z'' \) are given by the following equations:

\[
Z' = \sum_{i=1}^{n=3} \frac{R_i(1+R_iA_i\omega^{\alpha_i}\cos(\frac{\pi}{2}\alpha_i))}{(R_iA_i\omega^{\alpha_i}\sin(\frac{\pi}{2}\alpha_i))^2 + (1+R_iA_i\omega^{\alpha_i}\cos(\frac{\pi}{2}\alpha_i))^2}
\]  

(9)

\[
Z'' = \sum_{i=1}^{n=3} \frac{R_i^2A_i\omega^{\alpha_i}\sin(\frac{\pi}{2}\alpha_i)}{(R_iA_i\omega^{\alpha_i}\sin(\frac{\pi}{2}\alpha_i))^2 + (1+R_iA_i\omega^{\alpha_i}\cos(\frac{\pi}{2}\alpha_i))^2}
\]  

(10)

The values of constant phase element \( A_i \) and resistances \( R_i \) for various temperature between 340 K a 600 K bring together in Table 1. Fig.7 shows the resistance variation as a function of temperature. The data presented a sharp decrease of three resistances when temperature increases. This behavior is explained by different contacts on each side of the pellet and different charge conduction zones. As resistance decreases differently, the conductivity is also increasing differently which gives different activation energies as previously seen. In fact, following the Arrhenius relation we have:
\[ R = R_0 e^{E_{ac}/k_B T} \] (11)

where \( E_{ac} \) is the activation energy, \( R_0 \) is a pre-exponential term and \( k_B \) is the Boltzmann constant. The variation of \( \log(R) \) as a function of the inverse of the temperature is linear which proves that the conduction of the carriers of charges in ferrite is thermally activated. The linear fit of these curves is shown in Fig.8 which gives three different activation energies (0.242 eV, 0.265 and 0.283 eV) associated with three zones of different conductivity [28,29].

Fig.9 exhibits the frequency variations of the real part \( Z' \) of complex impedance at various temperatures. We note that the \( Z' \) amplitude is huge and it decreases with temperature increasing, indicating an increase of the charges carriers conduction in the compound. At low frequency, the real impedance is high then it decreases as the frequency and temperature increases. In the high-frequency region, the amplitudes of \( Z' \) are confused and become independent of the temperature. This behavior of impedance in high frequencies is due to the increase of the area of charge space under the effect of temperature that has a large oscillation. The variation of \( Z' \) in Fig.9 is fitted by the equation (9). A good agreement was found between experimental and theoretical curves deduced from the given equivalent circuit.

Fig.10 shows the variation of the imaginary impedance \( Z'' \) as a function of the frequency at different temperatures. The amplitude of \( z'' \) decreases with increasing temperature. The evolution (of what) shows the presence of two maximum corresponds to two relaxation frequency. The peaks are shifted to high frequencies as the temperature increases. We found the first maximum peak between \( 10^3 \) Hz and \( 10^4 \) Hz. The second is located after \( 10^5 \) Hz.

The presence of two maximum relaxations is due to the morphology of the samples which are formed by grains and grain boundaries [30]. When the temperature is greater than \( 10^7 \), the amplitude of \( Z'' \) becomes independent of the frequency and the temperature [31]. The variations of imaginary impedance as a function of temperature confirm the semiconducting
behavior of the Nickel ferrites. The evolution of $Z''$ fitted by the relation (10) are in a good agreement with theoretical curves.

To study the dielectric relaxation, we have represented the curves of $Z'$ and $Z''$ according to the frequency with a fixed temperature on the Fig.11. The frequency $f_{\text{max}}$, at which the impedance $Z''$ takes its maximum value, corresponds to the inflection point abscissa to the curve of variations of $Z'$ as a function of the frequency. Thus, from the same figure at 500 K, we conclude the values of $f_{1\text{max}}=92.6 \text{ Hz}$ and $f_{2\text{max}}=7.82.4.10^5 \text{ Hz}$.

The Fig.12 illustrates the frequency relaxation variation with temperature. We note an increase of frequency relaxation with the increase of temperature. This is due to an increase of hopping, this evolution is described by the equation:

$$f_{\text{max}} = f_0 e^{\frac{E_a}{k_B T}}$$

(12)

Where $f_{\text{max}}$ is the relaxation frequency, $f_0$ is the pre-exponential term and $k_B$ is the Boltzmann constant. The Fig.13 shows the linear variation of the relaxation frequency as a function of $1 / T$. The linear adjustment of these curves gives us the activation energy values of 0.250 eV and 0.228 eV [32,33].

In order to determine the relaxation time, we use the following relation [34, 35]:

$$\tau = \frac{1}{2\pi f_{\text{max}}}$$

(13)

where $f_{\text{max}}$ is the relaxation frequency.

The variation of relaxation time with temperature is shown in Fig.14. We note a decrease of relaxation time with the increase of temperature. This dependence as a function of heat proves that the charge conduction is thermally activated and that the relaxation frequency is given by the following equation [36]:

$$\tau = \tau_0 e^{E_a / k_B T}$$

(14)

Where $\tau$ is the relaxation time, $\tau_0$ is the pre-exponential term and $k_B$ is the Boltzmann constant.
4. Dielectric study

The evolution of real permittivity as a function of the frequency at temperature range from 340 K to 600 K is shown in Fig. 14. In the curves, we can distinguish three regions. For low frequencies (50 Hz to $10^3$ Hz) the evolution of permittivity as a function of frequency is slow, contrarily in the second region we observe a strong dependence behavior seen from the same spectra. For the third zone, from $f=10^5$ Hz, the permittivity becomes constant. We note giant values of the real permittivity, in particular, at low frequencies, then decreases until it reaches a constant value. The experimental data (the inset in Fig. 15) illustrate that the permittivity increases with temperature at frequency $f<10^3$ Hz, then becomes almost constant in higher frequency. The observation indicated the thermal activation of the charge carriers. This dielectric permittivity evolution is explained by the two-layer model of Maxwell-Wenger [37]. So, by hopping process the electrons reach grain boundary. In this area and due to higher resistance of the grain boundaries, electrons accumulate and produce charge zone. The increase of the frequency decreases the probabilities of sensing the grain boundaries by the electrons, so the polarization decreases [38].

The variation of imaginary permittivity as a function of frequency at different temperatures is shown in Fig. 16. We note for frequency $f<10^3$ Hz) the imaginary permittivity is rapidly decreased. At higher frequency, the imaginary dielectric permittivity becomes constant. At low frequency, we observe a giant value of $\varepsilon''$ that is explained by various parameters: the existence of Fe$^{2+}$ ions, oxygen vacancies, grain boundary defects, etc [39]. These observations are interpreted in the context of the Wagner and Koop's model [40], where the samples contain the minority Fe$^{2+}$ ions and the majority Fe$^{3+}$ ions that formed dipoles. Increases in frequency and temperature accelerate the jump of the electrons between Fe$^{3+}$ and Fe$^{2+}$ Ions. The jump of the electron between Fe$^{2+}$ and Fe$^{3+}$ ions depends on the applied field value, which decreases the dipoles polarization at high frequency in ferrites [41].
The temperature dependence of imaginary permittivity $\varepsilon''$ at various frequencies ranging from $10^2$ Hz to $10^6$ Hz is studied and the results are displayed in the inset in Fig.15. The experimental data illustrate the permittivity increases with temperature increasing at frequency $f<10^3$ Hz, then the curves become in higher frequency almost constant. The evolution of the dielectric permittivity indicated the thermal activation of the charge carriers at low frequency. This shows the semiconducting nature of the compound.

The dielectric loss $\tan\delta$ is the ratio of the imaginary part of the dielectric constant of the real part and is given by the following expression [42]:

$$\tan\delta = \frac{\varepsilon''}{\varepsilon'}$$  \hspace{1cm} (15)

Fig.17 exhibits the frequency dependency of dielectric loss at various temperatures. At low frequency, a decrease in energy loss is observed for different temperatures to reach frequency $f=10^3$ Hz. From $10^4$ Hz, the variation is reversed, there is a rise of dielectric loss. In the last region, we observe a loss decreasing as function of frequency and becomes equal to zero irrespective to temperature. This behavior is explained by Maxwell–Wagner and Koop’s phenomenological models [40], where the electrons jump between Fe$^{2+}$ and Fe$^{3+}$ ions depend on the applied field value, which decreases the dipoles polarization at high frequency in ferrites. We note also a light increase in dielectric loss.

**IV. Conclusion**

Ni$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ spinel ferrite ceramics were synthesized and their structural and dielectric properties were studied. The evolution of ac conductivity as a function of frequency shows two regions and discuss by the Maxwell–Wagner interfacial polarization model. The impedance spectroscopy is investigated as function of temperature and frequency. From the combined spectra of $Z'$ and $Z''$, we show two relaxation frequencies associated with the grain and grain boundaries. Furthermore, We showed that relaxation frequency and time are
temperature depended which proves that the electric charge transport is thermally activated. For better understanding the electrical behavior of the studied compound, We determined the equivalent electrical composed by three series sets of resistance and constant phase in parallel as the appropriate. In the summary, we have shown in the present work that the dielectric constants are giant at low frequency and then decrease as a function of frequency and temperature, and in the end become constant for higher frequency.

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Figures captions

Fig.1 X-ray diffraction data for material Ni$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$

Fig.2 Conductivity as a function of frequency at different temperatures.

Fig.3 Variation of conductivity as function of temperature at different frequency.

Fig.4. Variation of log$\sigma$ Vs 1/T.

Fig.5 Complex impedance spectrum for different temperatures.

Fig.6 Nyquist diagram of Ni$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ as a function of temperature. The inset showing the equivalent circuit.

Fig.7 Variation of resistances with temperature.

Fig.8 Variation of ln(R) with 1/T.

Fig.9 The impedance Z’ with frequency at different temperatures.

Fig.10 Impedance Z” with frequency at different temperatures.

Fig.11 Frequency dependence of impedance Z’ and Z” at 500 K.

Fig.12 Variation of relaxation frequency as function of temperature.

Fig.13. Variation of log$f_{1\text{max}}$ Vs 1/T.

Fig.14 Variation of relaxation time as function of temperature.

Fig.15 $\varepsilon'$ as a function of frequency at different temperatures.

Fig.16 $\varepsilon''$ as a function of frequency at different temperatures.

Fig.17 The loss factor as a function of frequency and temperatures.

List of Table:

Table 1 Variation of impedance parameters at various temperatures.
Fig. 1

Fig. 2

Fig. 3
Fig. 10

Fig. 11

Fig. 12
Table 1

|   | R1(Ω) | CP1T(F)  | CP1P  | R2(Ω)  | CP2T(F) | CP2P  | R3(Ω)  | CP3T(F) | CP3P  |
|---|-------|----------|-------|--------|---------|-------|--------|---------|-------|
| 340| 55081 | 9.174E-10| 0.87446 | 345520 | 1.83E-9 | 0.849 | 16668  | 1.393E-10| 0.9917 |
| 360| 48081 | 2.723E-9 | 0.83684 | 204300 | 3.210E-9| 0.8190 | 14209  | 3.386E-10| 0.93263 |
| 380| 46081 | 1.525E-9 | 0.91799 | 91966  | 1.758E-8| 0.6950 | 11874  | 5.175E-10| 0.8982 |
| 400| 25305 | 5.8359E-8| 1.044  | 79103  | 2.778E-9| 0.8099 | 8446   | 9.652E-10| 0.85894 |
| 420| 19693 | 1.0332E-7| 0.95483 | 46053  | 2.692E-9| 0.8226 | 6547   | 1.475E-9 | 0.82349 |
| 440| 14007 | 1.0542E-7| 0.95168 | 30725  | 2.591E-9| 0.8349 | 5084   | 2.117E-9 | 0.80643 |
| 460| 9634  | 1.3169E-7| 0.92005 | 18163  | 2.460E-9| 0.8497 | 3845   | 2.853E-9 | 0.78799 |
| 480| 6857  | 1.3222E-7| 0.92121 | 11982  | 2.356E-9| 0.8621 | 3147   | 4.270E-9 | 0.76334 |
| 500| 4670  | 1.3749E-7| 0.91868 | 7415   | 2.331E-9| 0.8709 | 2419   | 5.483E-9 | 0.7488 |
| 520| 3460  | 1.3872E-7| 0.92351 | 5356   | 2.437E-9| 0.8719 | 2252   | 6.406E-9 | 0.7412 |
| 540| 1714  | 1.1939E-8| 0.69859 | 3057   | 2.314E-9| 0.8888 | 2049   | 1.4573E-7| 0.92549 |
| 560| 1487  | 1.6974E-8| 0.68226 | 1980   | 2.117E-9| 0.9060 | 1632   | 1.5267E-7| 0.9257 |
| 580| 1207  | 2.0787E-8| 0.67089 | 1156   | 1.930E-9| 0.9240 | 1115   | 1.5492E-7| 0.93033 |
| 600| 994.1 | 2.2023E-8| 0.66792 | 845.6  | 1.809E-9| 0.9336 | 873.6  | 1.64E-7  | 0.92819 |