Investigations of microstructural and impedance spectroscopic properties of $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite prepared using sol-gel method

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Investigations of microstructural and impedance spectroscopic properties of $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite prepared using sol-gel method

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Abstract

Polycrystalline $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite was prepared with sol-gel method. This sample was characterized by powder X-ray diffraction (XRD), Scanning electron microscopy, and impedance spectroscopy. X-ray diffraction analysis combined with the Rietveld refinement confirmed that the sample crystallizes in a single-phase cubic spinel structure with (SG: $Fd\bar{3}m$). Electrical conductivity obeying the Jonscher power law indicates that the prepared material exhibits semiconductor behavior, and the conduction process follows the “non-overlapping small polaron tunnelling” model between neighbors’ sites. The behavior of dielectric constants such as permittivity and loss coefficient has been interpreted based on the Maxwell-Wagner's theory of interfacial polarization. The curves of imaginary parts of impedance (Z'') and modulus (M'') show a dielectric-relaxation phenomenon in the sample with activation energy near to that determined from the *dc* conductivity study. Nyquist plots (Z'' vs. Z') show a monotonic decrease in both grain resistance (R_g) and grain boundary resistance (R_{gb}) with increasing temperature such as $R_{gb} \gg R_g$. This result confirms that the transport mechanism for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ compound is governed by the grain boundaries effect.

Keywords: Spinel ferrites; Sol–gel method; Microstructural Properties; NSPT model; Relaxation phenomenon; grain boundaries effect.

I- Introduction

Among the most important classes of materials possessing a diversity in their magnetic, electrical, electronic, and optical properties are the spinel ferrites [1]. Presently, magnetic oxides made a great attention on scientists in the sake of their wide-ranging applications from industrial use to fundamental research [2, 3]. Spinel ferrites with these properties can be profitable in ferrofluids, gas sensors, transformer cores, biomedical and magnetic refrigeration [4-8]. The spinel ferrites have general formula of MFe_2O_4 , where M is a divalent metal cation [9]. They represent a face-centered cubic structure with merging crystallographic sublattices between tetrahedral (A) and octahedral [B] sites, the spins in these sites are oriented antiparallel to each other. The nature of spinel structure like normal, inverse, or mixed structure relies on the lattice occupancy of the divalent metallic ions in the crystallographic sites [10]. The ionic occupancy as well as preparation method, annealing temperature, stoichiometry, valency etc., present the critical aspect in determining structural, magnetic, and electrical properties in spinel ferrites [11-13].

Among the most interesting spinel systems, $Mg_{1-x}Co_xFe_2O_4$ ferrites are widely used for electronic devices due to their high frequency-permeability, high electrical resistivity, chemical resistance, and cost effectiveness. These interesting properties make these materials good candidates for high frequency applications and microwave devices [14]. In addition, $Mg_{1-x}Co_xFe_2O_4$ materials are also potential candidates for size-antennas miniaturization. [15]. The bibliographic study shown that less studies were presented in the literature on this system with other substitutions. Particularly, no investigation has been cited on the physical properties of Mg–Co ferrites substituted with trivalent metal cation such as Al^{3+} . Although many metal cations have been studied for their effects on electrical and magnetic properties Al^{3+} is a less explored ion. In this report, we presented an investigation on structural and impedance spectroscopic properties of the spinel ferrite $Mg_{0.5}Co_{0.5}Fe_{1.6}Al_{0.4}O_4$ prepared by

sol-gel technique. The process of sol-gel synthesis of this compound will be firstly discussed. Then, the results of the morphological characterization by scanning electron microscope (SEM) are presented, to then discuss and interpret the structural characterization by X-ray diffraction technique and Rietveld method. Electrical measurements by impedance spectroscopy as a function of frequency and temperature were also presented. Variations of electrical conductivity, dielectric constants, electrical modulus, and electrical impedance will be discussed.

II- Experiment

The $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ spinel ferrite was synthesized through the sol-gel process as displayed in [Fig. 1](#). The stoichiometric constituents of $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (purity 99.9%), $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (purity 99.9%), $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (purity 99.9%), and $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (purity 99.9%) were dissolved in distilled water and citric acid ($\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}$). Then, a few amounts of ammonia were added to adjust the pH solution to around 7. Ethylene glycol ($\text{HO}(\text{CH}_2)_3\text{OH}$, 99.9%) was added to the obtained homogeneous solution. The mixtures were maintained under magnetic agitation for 4 h at 90°C transform to become a gel. To eliminate any residual solvent, the collected gel was dried in a furnace at 250°C for 12 h. The obtained precursors were grained in air for 3 h, and then calcined at 600°C for 24 h, and it was grounded again and calcined at 800°C for 24 h. We can notice that the cooling was very slow. In the end, we ground the final calcined precursor, and the resulting powder was undergone a cycle of (grinding→pelleting→sintering) at 1000°C for 24 h.

The final powder was characterized by X-ray diffractometer (Panalytical X'Pert Pro System) using a monochromatic $\text{CuK}\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$) in a range of $15 \leq 2\theta \leq 80^\circ$ with a step of 0.02° . The XRD diffractograms were refined adopting the FULLPROF program [\[16\]](#) founded on the Rietveld analysis [\[17\]](#). Scanning electron microscopy [SEM, Philips XL30 microscope)] was used to probe into the surface morphology of the sample at

20 kV in secondary electron mode. For the electrical characterizations, silver electrodes were sputtered on both sides of the sample (in disk form). The electrical measurements were obtained over a wide temperature range using an Agilent 4294 analyzer under vacuum, in dark and in 100 Hz – 5 MHz frequency range.

III- Results and discussion

A. Microstructure analysis

Fig. 2a shows the secondary electron SEM image for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite. This morphological analysis shows the existence of homogeneous distribution of the spherical grains. We can also observe on this image minor voids associated with the porosity of the sample. Generally, sintering temperature affects the size distribution and the shape of grains along with agglomeration in the synthesized materials. In our case, the sample was prepared at sufficiently higher sintering-temperature (1000 °C). This indicates that this sintering temperature is favorable for good crystallinity and microstructural homogenization for the $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ sample. The statistical particle size distribution using *ImageJ* software is shown in **Fig. 2b**. The histogram fitting using the Gaussian function gives an average particle size around 0.66 μm .

The $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ sample phase purity and structure were approved by analysis of the X-ray diffraction pattern as shown in **Fig. 3a**. All peaks can be indexed to the face-centered cubic phase with $Fd\bar{3}m$ space group. We also note that the sample was described as a pure single phase without the presence of secondary crystallographic phases of impurities. The peaks are indexed as follows: (110), (220), (311), (222), (400), (422), (511), (440), (620), (533), and (622) characteristics of the structure of the single-phase cubic spinel. The average grain size (crystallite) of the studied systems is determined from the most intense (311) diffraction peak, in conformity with the Debye-Scherrer formula [18]:

$$D = \frac{K \lambda}{\beta_{1/2} \cos(\theta)} \quad (1)$$

where, k is a constant such that $k = 0.9$, λ is the X-ray wavelength (1.54059 Å), $\beta_{1/2}$ is the full width at half maximum (FWHM) and θ is the Bragg angle of the peak. The measured average grain size is approximately 37 nm. Hence, the change of growth grain size behavior according to morphological analysis in comparison with that given by the structural analysis can be associated with the difference in particle nature. This latter is secondary particle for morphological analysis whereas for structural one, it is primary particle. To investigate the structural transformation in more details, we realized a simulation on the measured XRD pattern based on the Rietveld refinement technique. The data analysis using the Rietveld method is presented in [Fig. 3b](#). Generally, for AB_2O_4 spinel materials, certain experimental techniques such as Mössbauer spectroscopy have been carried out to establish the cations distribution between both tetrahedral (A) and octahedral (B) sites. The characterization by the Mössbauer technique of $Mg_{1-x}Co_xFe_2O_4$ ferrites [\[19, 20\]](#) and certain Al-substituted ferrites [\[21\]](#) showed that Co^{2+} , Mg^{2+} and Al^{3+} ions are preferred to occupy the octahedral (B) sites, while Fe^{3+} ions are distributed between both (A) and (B) sites. The distribution of all these cations agrees with that presented in previous studies [\[22, 23\]](#). Accordingly, the ions distribution in $Mg_{0.5}Co_{0.5}Fe_{1.6}Al_{0.4}O_4$ compound can be written as: $(Fe_1^{3+})_A[Mg_{0.5}^{2+}Co_{0.5}^{2+}Fe_{0.6}^{3+}Al_{0.4}^{3+}]_BO_4^{2-}$. The refined crystallographic parameters are assembled in [Table 1](#). In particular, the obtained lattice constants are lower than those presented in the literature for the parent compound $Mg_{0.5}Co_{0.5}Fe_2O_4$ [\[22, 24\]](#). This is due to the smaller ionic radius of Al^{3+} ($r_{Al}^{3+} = 0,535$ Å) by comparing with that of Fe^{3+} ($r_{Fe}^{3+} = 0,645$ Å) [\[25\]](#).

B. Electrical conductivity

[Fig.4a](#) presents the conductivity spectra according to the frequency and temperature for $Mg_{0.5}Co_{0.5}Fe_{1.6}Al_{0.4}O_4$ ferrite. According to this figure, the conductivity spectra can be categorically divided into two regions. Almost constant conductivity values were observed at low frequency region extending up to a frequency, known as the hopping frequency (ν_H). The

frequency independence of conductivity in low frequency range is related to the higher activity of grain boundaries which weaken the electrons hopping. Since the conductivity in this range is virtually constant, although the frequency varies, it is often noted, σ_{dc} , as “direct current” conductivity. According to [Fig.4a](#), σ_{dc} grows with temperature rise indicating that the conduction process is thermally activated and that the prepared material exhibits semiconductor behavior over the entire temperature range. Above ν_H , the conductivity increases exponentially with frequency due to the activity of the conductive grains. This promotes the conduction process in the sample due to the increase of electrons hopping. The conductivity at frequencies higher than ν_H corresponds to the conductivity in alternating current (σ_{ac}). The phenomenon of the conductivity dispersion observed for the sample is analyzed by using the universal law of Jonscher [\[26\]](#) given by the following equation :

$$\sigma(T) = \sigma_{dc}(T) + A(T) \omega^s \quad (2)$$

where, $A(T)$ and “ s ” are the pre-exponential and the frequency-exponent parameters, respectively. The electrical conductivity results are well fitted as depicted by continuous red lines in [Fig.4a](#). The fitting curves shows small values for the σ_{dc} conductivity which reflects that the compound exhibits high electrical resistivity. This interesting property make the prepared material good candidate for microwave devices. On the other hand, according to Funke [\[27\]](#), for the ac component of the conductivity ($\sigma_{ac}=A(T)\omega^s$), $s \leq 1$ means that electrons hopping takes place between two sufficiently distant sites. Whereas for $s > 1$, the movement involves hopping between neighboring sites. The $s(T)$ values are plotted in [Fig.4b](#) and they are all superior than 1. So, we can conclude from this figure that the electrons hopping involves translational movement between neighboring sites. Such result was found in other works [\[28, 29\]](#). In addition, an increasing tendency in s values was observed with temperature rise. This is a criterion for a typical thermally activated conduction process. This indicates also that the “non-overlapping small polaron tunnelling” (NSPT model) [\[30\]](#) is valid in the

case of $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ sample. According to this model, the s exponent can be formulated as [30]:

$$s = 1 + \frac{4k_B T}{W_m} \quad (3)$$

where k_B is the Boltzmann constant, and W_m represents the polaron binding energy (i.e. the binding energy of the carrier in its localized site). The obtained W_m value is around 0.134 eV.

The obtained fitting parameters using Jonscher power law analysis were used to describe the hopping frequency (ν_H). This frequency, defined as the frequency at which the conductivity relaxation begins to appear, can be formulated as follows:

$$2\pi\nu_H = \left(\frac{\sigma_{dc}}{A(T)}\right)^{\frac{1}{s}} \quad (4)$$

Basically, dc and ac conductivities were correlated by comparing both $\sigma_{dc}(T)$ and $\nu_H(T)$ curves which are presented in **Fig. 5a**. The correlation between dc conductivity (σ_{dc}) and hopping frequency (ν_H) was examined by representing the logarithmic plot $\log(\sigma_{dc})$ vs. $\log(\nu_H)$ as shown in the inset of **Fig. 5a**. The $\log(\sigma_{dc})$ vs. $\log(\nu_H)$ curve presents a linear variation with slope close to unity. This implies that dc and ac conductivities are well correlated [31]. In the other hand, **Fig. 5a** shows the same temperature dependence of both $\sigma_{dc}(T)$ and $\nu_H(T)$ curves. In fact, the dc conductivity, and the hopping frequency increase with increasing temperature. This confirms the utility of the Arrhenius' law to explain the temperature variation of σ_{dc} and ν_H . Then, the Arrhenius law was used to analyze the $\sigma_{dc}(T)$ and $\nu_H(T)$ behaviors according the two following relations [32]:

$$\sigma_{dc} = \frac{\sigma_0}{T} \exp\left(-\frac{E_a}{k_B T}\right) \quad (5)$$

$$\nu_H = \frac{\nu_0}{T} \exp\left(-\frac{E_a}{k_B T}\right) \quad (6)$$

where σ_0 and ν_0 are pre-exponential factors, and E_a is the activation energy required in the sample. Eq. (5) and (6) are used to calculate the E_a values by plotting $\log(\sigma_{dc} \times T)$ and $\log(\nu_H \times T)$ vs. $(1000/T)$ in **Fig.5b**. The activation energies values derived from the linear fit of

both curves are shown in the same figure. It can be clearly seen that the E_a value deduced from dc conductivity is very close to that estimated from frequency hopping. This shows that charge carriers must overcome the same energy barriers while conducting and relaxing [33]. On the other hand, the obtained E_a value for $Mg_{0.5}Co_{0.5}Fe_{1.6}Al_{0.4}O_4$ sample ($E_a = 0.218$ eV) is smaller than the reported one for the undoped $Mg_{0.5}Co_{0.5}Fe_2O_4$ compound ($E_a = 0.76$ eV) [34]. This reflects the effect of Al on improving the electrical conductivity and the conduction process in $Mg_{0.5}Co_{0.5}Fe_2O_4$ system. In Fig. 5c we present the Arrhenius plots for σ_{ac} at two different frequencies (1 MHz and 5 MHz). As shown in the figure, the σ_{ac} values increase with frequency. The deduced activation energies (E_{ac}) values which are presented in Fig. 5c are lower than the one determined from the σ_{dc} curve. In fact, due to the higher activity of grain boundaries in the dc conductivity regime the electrons hopping requires more energy; however the conductive grains are more active in the ac conductivity regime and consequently low activation energy is required for the electrons hopping [35, 36].

C. Dielectric study

The imaginary part of permittivity (ϵ'') is presented in Fig. 6a versus temperature and frequency for $Mg_{0.5}Co_{0.5}Fe_{1.6}Al_{0.4}O_4$ ferrite. The ϵ'' values decrease with increasing frequency. This behavior can be interpreted considering the Maxwell-Wagner interfacial polarization [37, 38] and Koop's phenomenological theory [39] of dielectrics. Indeed, the dielectric structure of ferrites is assumed to be composed of good conductive grains, separated by grain boundaries slowing the movement of charge carriers, which drops the inter-grains conductivity. During this exchange mechanism, electrons must pass through the grains and grain boundaries of the sample. Due to the high resistance grain boundaries, electrons accumulate at the grain boundaries and produce space charge polarization. Fig. 6a also shows that ϵ'' values decrease rapidly at low frequencies and becomes frequency independent at higher frequencies. This behavior is since the effect of grain boundaries is

greater in the low frequency region while that of grains is more preponderant in the high frequency region [39]. The non-frequency dependence of ε'' for higher frequencies, can be attributed to the fact that beyond a certain frequency, the electrons in the grain can no longer follow the alternating field and consequently, the medium ceases to polarize, hence the constancy of ε'' . In addition, Fig.6a shows an increase of ε'' values with increasing temperature because the hopping of charge carriers is thermally activated. On the other hand, the frequency and temperature dependence of ε'' can be analyzed according to *Giuntini* model [40] which is given as:

$$\varepsilon''(\omega) = (\varepsilon_0 - \varepsilon_\infty) 2 \pi^2 N (ne/\varepsilon_0)^3 K T \tau_0^m W_M^{-4} \omega^m \quad (7)$$

where ε_0 and ε_∞ are the static and optical dielectric constants, respectively; n is the number of electrons that hop, N is the concentration of localized sites, and W_M is the maximum barrier height (the energy required to move the electron from a site to the infinity).

Eq.(7) can be simplified as:

$$\varepsilon''(\omega) = B\omega^m \quad (8)$$

The exponent m was calculated from the negative slopes of the obtained lines in Fig.6a and it was presented in Fig.6b versus temperature. It emerges that m values increase linearly with temperature. According to *Giuntini* model, the behaviors of $m(T)$ (dielectric properties) and the exponents $s(T)$ (measurements of alternating current) show the same tendency by varying temperature [41]. In addition, the exponent $m(T)$ can be expressed according to the following equation:

$$m = \frac{-4k_B T}{W_M} \quad (9)$$

The calculated value of W_M is of the order of 0.138 eV. This value agrees well with that obtained using equation Eq. (3). From Fig.7, one can notice that the loss factor ($\tan\delta$) decreases rapidly at low frequencies and it becomes almost frequency independent in the higher frequencies' values. As mentioned above, the grain boundaries are active in the low

frequency region which requires more energy for the electrons exchange and therefore the loss is higher. However, in higher frequencies region which corresponds to the conductive grains, low energy is required for the electrons transfer. In addition, the $\tan\delta$ values increase with temperature due to the thermally activated conduction process.

D. Electrical modulus

Fig.8a shows the behavior of the real part of the modulus (M') versus frequency and temperature for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite. In the low frequency region, the M' values are very negligible. This confirms that the electrode polarization makes a negligible contribution in the material [42]. With increasing frequency, the M' values increase reaching a saturation at high frequencies especially at low temperatures. This behavior may be due to the short-range mobility of charge carriers [43]. **Fig.8b** shows the variation of the imaginary component of modulus (M''). The $M''(f)$ spectra are characterized by peaks at characteristic frequencies ($f_{M''}^{\max}$) for all temperatures, and the positions of these peaks shift to higher frequencies with increasing temperature. This means that the relaxation time for this process increases with temperature. The $M''(f)$ curves were modeled using the Kohlrausch, Williams and Watts (KWW) function [44]:

$$M'' = M''_{\max} / \left[1 - \beta + \left(\frac{\beta}{1+\beta} \right) \left(\beta \left(\frac{f_{\max}}{f} \right) + \left(\frac{f}{f_{\max}} \right)^{\beta} \right) \right] \quad (10)$$

where $M''_{\max}$ is the peak maxima, and $f_{\max}$ its corresponding maximum frequency. β is the stretching factor ($0 < \beta < 1$) which describes the dielectric-relaxation nature (Debye or non-Debye) [45]. The fitting parameters of $M''(f)$, given in **Table 2**, shows that β values are less than unity, showing so a non-Debye nature of the relaxation phenomenon for the sample.

E. Complex impedance analysis

To distinguish between the grain and the grain boundary contribution to electrical properties for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite, Nyquist plots have been analyzed as depicted in **Fig.9**. The Nyquist plots are represented as semicircular arcs not centered on the real axis.

This decentralization indicates a relaxation process of the non-Debye type. The diameters of the semicircles, limited by the total resistance of the material, decrease with temperature, in relation to the pronounced increase in σ_{dc} . The appearance of a single semicircle for each temperature means that the electrical process obeys a single dipole relaxation [46]. Indeed, the analysis of the dielectric relaxation linked to the grain boundaries in the temperature range 300 K - 500 K has clearly shown that the conduction is controlled by the grain boundaries which explains the appearance of only one semicircle in the Nyquist diagram. These semicircles inform us about the electrical processes occurring in the sample and its correlation with the microstructure of the sample, when we modeled the sample by an equivalent electrical circuit [47, 48]. The experimental data of these semicircles were well fitted by the *ZView* software [49] using an equivalent circuit formed by a grain resistance (R_g) in series with a parallel combination of a grain boundary resistance (R_{gb}) and constant phase element impedance (Z_{CPE}). Such equivalent configuration can be represented as $(R_g + R_{gb} // CPE_{gb})$ [50], as indicated in the insert of Fig.9. All adjusted parameters are summarized in Table 3. We can notice in this table that R_g values are very low, therefore the total resistance R_T of the sample which is the sum of those of R_g and R_{gb} [50] is approximately equal to that of the grains-boundaries. Also, the R_{gb} values decrease with increasing temperature, which indicates the semiconductor behavior for the prepared sample. Such behavior has been observed in other materials [32, 51]. It appears that with the increase in temperature, the grain boundary effect facilitates the passage of charge carriers by lowering the barrier, thus paving the way for increased electrical transport. The normalized representation of the imaginary part of impedance (Z''/Z''_{max} vs. frequency) is shown in Fig.10a. The curves are characterized by well resolved peaks moving to the higher frequencies with temperature rise. Such behavior indicates the existence of a relaxation phenomenon in our material. The peaks maximum allows us the calculation of the f_{max}

relaxation frequency and consequently the estimation of the relaxation time (τ) by using the following equation:

$$\tau = 1/2\pi f_{max}, \quad (11)$$

The Arrhenius variation of $\log(\tau)$ versus $(1000/T)$ is given in **Fig. 10b** according to the following relation [29]:

$$\tau = \tau_0 \exp \left(\frac{E_{Z''}}{k_B T} \right) \quad (12)$$

where, τ_0 is the pre-exponential factor, and $E_{Z''}$ is the activation energy. The linear fit of the curve gives a value of $E_{Z''}$ of the order of 0.206 eV which is close to that found from the *dc* conductivity analysis ($E_a = 0.218$ eV). The slight difference between the two activation energies may be since the relaxation process involves only the hopping energy of charge carriers between localized states, however the conduction mechanism involves the hopping energy, the disorder as well as the binding energy of polarons [52]. This convergence in values confirms that both relaxation and conduction processes may be attributed to the same type of charge carriers. **Fig. 11** shows the plot Z''/Z''_{max} vs. $\log(f/f_{max})$. As shown in this figure, all normalized curves collapse into one curve which indicates that the distribution of the relaxation time is temperature independent [53]. For comparison, the variations of $Z''(f)$ and $M''(f)$ are depicted in **Fig. 12** for two different temperatures (300 K and 400 K). At room temperature ($T = 300$ K), the curves show single peak which appears approximately at the same frequency value. With increasing temperature ($T = 400$ K), the gap between both Z'' and M'' peaks increases confirming the non-Debye-type relaxation behavior for the prepared sample [54].

IV- Conclusion

In the present paper, $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ spinel ferrite was synthesized using sol-gel method, and its microstructural and electrical properties were successively investigated. SEM analysis reveals that sample presents homogeneous surface with grains of regular shape. Rietveld fitted of XRD pattern reveals the cubic structure of the sample having a space group of $Fd\bar{3}m$. According to the Jonscher-law analysis of electrical conductivity, the conduction process for sample having semiconductor behavior occurred between neighboring sites and follows the NSPT model. Dielectric constants decrease with frequency and increase with temperature. Such behavior was interpreted according to the Maxwell-Wagner's theory of interfacial polarization. Results of modeling of Nyquist plots show that the conduction process in the prepared sample is governed by the grain boundaries effect. In conclusion, the synthesized material presents several advantages such as its high resistivity, good electrical properties, low cost and above all its easy synthesis. These features make the $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite good candidate in electronic devices such as for high frequency applications and microwave devices.

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Figures and tables:

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Fig. 2: SEM image (a) and grains size distributions (b) for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite.

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Fig.7: The $\tan \delta$ behavior versus frequency and temperature for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite.

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Fig.9: Nyquist spectra for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite fitting using *Zview* software. The inset shows the used electrical circuit.

Fig. 10: (a) $Z''/Z''_{\max}$ vs. frequency and temperature for $Mg_{0.5}Co_{0.5}Fe_{1.6}Al_{0.4}O_4$ ferrite. (b) Variation of $\log(\tau)$ vs. $(1000/T)$.

Fig.11: $Z''/Z''_{\max}$ vs. $f/f_{\max}$ for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite.

Fig. 12: Combined $Z''(f)$ and $M''(f)$ curves for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite at 300 K (a) and 400 K (b).

Table 1: Structural parameters for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite obtained following the structural refinement by Rietveld method. a : cell parameter; V : cell volume; B_{iso} : isotropic thermal agitation parameter; x , y and z : atomic positions; R_A : distance (Fe – O); R_B : distance (Mg, Co, Fe, Al – O); θ_{A-O-B} : angle (Fe – O – Fe); θ_{B-O-B} : angle (Mg, Co, Fe, Al – O – Mg, Co, Fe, Al); G_s : average grains size. Agreement factors of profile R_p , weighted profile R_{wp} , and structure R_F . χ^2 : the goodness of fit. The numbers in parentheses are estimated standard deviations to the last significant digit.

Sample			Mg _{0.5} Co _{0.5} Fe _{1.6} Al _{0.4} O ₄
Space group			<i>Fd-3m</i>
Cell parameters	<i>a</i> (Å)		8.3549 (3)
	<i>V</i> (Å ³)		583.21 (3)
Atoms	Fe	Wyckoff positions	4c
		Site summetry	-43m
		Atomic positions <i>x</i> = <i>y</i> = <i>z</i>	1/8
		Occupancy factors	1
		<i>B</i> _{iso} (Å ²)	2.87 (2)
	Mg/Co/Fe/Al	Wyckoff positions	16d
		Site summetry	-3m
		Atomic positions <i>x</i> = <i>y</i> = <i>z</i>	1/2
		Occupancy factors	0.5/0.5/0.6/0.4
		<i>B</i> _{iso} (Å ²)	2.13 (2)
	O	Wyckoff positions	32e
		Site summetry	3m
		Atomic positions <i>x</i> = <i>y</i> = <i>z</i>	0.2546 (8)
		Occupancy factors	4
		<i>B</i> _{iso} (Å ²)	2.13 (2)
Structural parameters	<i>D</i> (nm)	37	
	<i>R</i> _A (Å)	1.875 (7)	
	<i>R</i> _B (Å)	2.051 (7)	
	<i>θ</i> _{A-O-B} (°)	123.7 (3)	
	<i>θ</i> _{B-O-B} (°)	92.1 (3)	
Agreement factors	<i>R</i> _p (%)	5.83	
	<i>R</i> _{wp} (%)	7.51	
	<i>R</i> _F (%)	3.74	
	<i>χ</i> ² (%)	1.04	

Table 2: Obtained parameters from the fitting $M''(f)$ curves using KWW function for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite.

$T(\text{K})$	$M''_{\max} \times 10^{-4}$	β	$f_{\max}(\text{Hz})$	χ^2
300	3.110	0.700	2360	0.91
320	3.060	0.720	7134	0.95
340	3.031	0.745	19899	0.97
360	2.851	0.800	49355	0.98
380	2.502	0.840	103992	0.98
400	2.310	0.870	204486	0.99
420	0.950	0.920	341030	0.99
440	0.020	0.935	491279	0.98
460	0.019	0.950	733142	0.97
480	0.090	0.970	979609	0.99
500	0.017	0.980	1076460	0.98

Table 3: Electrical parameters obtained from the fit of Nyquist plots using *Zview* software for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite.

$T\text{ (K)}$	$R_g\text{ }(\Omega)$	$R_{gb}\text{ }(\Omega)$	$Q \times 10^{-11}\text{ (F)}$	α
300	64	514050	17.35	0.971
320	50	157910	16.57	0.978
340	36	55551	15.66	0.984
360	32	22142	13.81	0.993
380	23	9915	13.02	1.002
400	22	4911	12.11	1.008
420	21	2562	2.77	1.114
440	17	1403	8.07	1.204
460	15	865	6.18	1.179
480	10	548	0.91	1.203
500	6	334	0.40	1.276

Figures

Figure 1

Sol-gel synthesis for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite.

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SEM image (a) and grains size distributions (b) for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite.

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Figure 7

The $\tan\delta$ behavior versus frequency and temperature for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite.

Figure 8

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Figure 9

Nyquist spectra for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite fitting using Zview software. The inset shows the used electrical circuit.

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Figure 11

$Z''/Z''_{\max}$ vs. $f/f_{\max}$ for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite.

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Combined $Z''(f)$ and $M''(f)$ curves for $\text{Mg}_{0.5}\text{Co}_{0.5}\text{Fe}_{1.6}\text{Al}_{0.4}\text{O}_4$ ferrite at 300 K (a) and 400 K (b).