

THE ELECTRICAL PROPERTIES OF Mg-Cu-Zn FERRITES IRRADIATED BY γ -RAYS OF ^{60}Co SOURCE

^{1,2}Mohamed Eltabey, ^{3,4}H. Ebrahim Hassan and ³Ismail Abd Elrahim Ali

¹Department of Basic Engineering Science, Faculty of Engineering, Menoufiya University, Shebin El-Kom, Egypt

²Department of Science Physics, Preparatory Year Deanship, Faculty of Medicine, Jazan University, Saudi Arabia

³Department of Physics, Faculty of Science, Taif University, Taif, 888 Hawiya, Saudi Arabia

⁴Cyclotron Facility, Nuclear Research Center, Atomic Energy Authority, Cairo 13759, Egypt

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ABSTRACT

Ferrite samples with chemical formula $\text{Mg}_x\text{Cu}_{0.5-x}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ ($x = 0.0, 0.2$ and 0.4) was prepared by the conventional ceramic method. The enhanced changes in the dc/ac resistivity and the dielectric constant due to γ -rays irradiation by ^{60}Co source were measured using the two probes technique. The samples showed decreasing of their resistivity at room temperature after irradiation for both dc and ac applied electric fields. Temperature dependence curves of the dc resistivity, before and after irradiation, showed three regions with different activation energies. It was also observed that the activation energy decreased after irradiation at every region. The frequency dependence of ac resistivity and dielectric constant parameters were investigated before and after irradiation process. Both real part (ϵ') and imaginary part (ϵ'') of the dielectric constant were decreased due to irradiation. These results were discussed in the view of gamma rays interaction with ferrite lattice.

Keywords: Gamma Irradiation, Mg-Cu-Zn Ferrites, Resistivity, Activation Energy, Dielectric Constant

1. INTRODUCTION

Ferrites have many important applications in industry, modern telecommunication and electronic devices and they are still of interest as promising materials for miniature electro-optic modulators, pyroelectric detectors, piezoelectric sensors, high quality filters, transformer cores, ferrite isolators, memory core industry, multilayer chip inductor, rod antennas, radiofrequency circuits, wave guides and electronic memory elements as well as a recording media (Muralidharan *et al.*, 2010; Wu *et al.*, 2011; Kramer *et al.*, 2006; Ahmed *et al.*, 2007). The Mg-Cu-Zn ferrites have become important to industry because of their applications in intermediate frequency transformer and antenna cores. Also, these ferrites have magnetic properties similar to those of Ni-Cu-Zn ferrites with the

advantage that they are economical and easy to synthesize (Zhou *et al.*, 2012). Therefore, Mg-Cu-Zn ferrites are considered a promising material for Multilayer Current Inductors (MLCI) with high-performance and low cost. In addition, they are useful in the fabrication of cores of Intermediate Frequency Transformers (IFT) for amplitude modulation (Sujatha *et al.*, 2011).

Radiation interaction with materials is under consideration in the past few years due to broad development of electronic industry in nuclear facilities, accelerators, spacecrafts and satellites. Gamma ray can generate defects of various types such as point, cluster, beside excitation and ionization of the atoms. The effect of γ irradiation on microstructure, diffusion coefficient of oxygen ions, dielectric properties, thermoelectric power and thermal conductivity of Co-Zn ferrites was investigated (Ateia, 2006; Tashtoush,

Corresponding Author: Mohamed Eltabey, Department of Basic Engineering Science, Faculty of Engineering, Menoufiya University, Shebin El-Kom, Egypt

2005; Hemeda and El-Saadawy, 2003). Okasha (2010) found that, the γ -irradiation improved the magnetic properties of Mg-Mn and Mn-Ni ferrites nano-particles (Okasha, 2010; Hassan *et al.*, 2013). On the other hand, in previous work (Eltabey *et al.*, 2011), the authors reported the influence of γ -irradiation on structure and magnetic properties of Mg-Cu-Zn ferrites samples. It was found that, there was disimprovement in the magnetic properties. Where, the values of magnetization, initial permeability and Curie temperature in addition to the homogeneity were decreased due to irradiation. The present work reported the effect of irradiation on the electrical parameters as dc/ac resistivity, conduction activation energy and dielectric constant for the same samples.

2. MATERIALS AND METHODS

Samples of $\text{Mg}_x\text{Cu}_{0.5-x}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ ferrite system with $x = 0.0, 0.2$ and 0.4 were synthesized using conventional ceramic method which previously described elsewhere (Eltabey *et al.*, 2011). The XRD was performed using a diffractometer of type X'Pert Graphics and identified with Cu K_α radiation and morphological structure of the samples are discussed (Eltabey *et al.*, 2011). For measuring the electrical resistivity, the samples were inserted between two silver electrodes where the silver paste was used as a contact material. The resistivity of the sample (ρ) is calculated using the relation $\rho = RA/d$ where, A and d are the cross-section area and the thickness of the sample, respectively. The current passing through the sample is obtained by measuring the voltage drop across a standard resistance connected to the circuit. The temperature of the sample was measured using thermocouple type K coupled with digital thermometer type BK-710. Parallel plate Capacitance (C_p) and dielectric parameters were measured for the tablet samples using LCR bridge meter model HIOKI 3532-50- LCR HiTESTER. The real part of the dielectric constant (ϵ') was calculated using the formula $\epsilon' = C_p d / \epsilon_0 A$ (Lipare *et al.*, 2004) where C_p is the capacitance of the parallel plate, d is the thickness of the tablet, A is the cross-sectional area of the flat surfaces of the sample and ϵ_0 is the permeability of free space ($\epsilon_0 = 8.85 \times 10^{-12}$ F/m). The ac resistivity (ρ_{ac}) was obtained from the values of (ϵ') and the loss tangent by using the relation $\rho_{ac} = 1 / (\epsilon' \epsilon_0 \omega \tan \delta)$ (Lipare *et al.*, 2004) where ω is the angular frequency. The parameters ϵ' , $\tan \delta$ and ρ_{ac} were measured as a function of frequency within the range 45 Hz-5 MHz at room temperature. The samples were exposed to ^{60}Co

radioactive source in the gamma irradiation cell at Cyclotron Facility, Nuclear Research Center, Atomic Energy Authority, Cairo, Egypt. They were irradiated with 1.9 MGy dose at dose rate of 5kGy/h.

3. RESULTS

3.1. X-Ray Analysis

The x-ray spectra of the investigated samples were presented in our previous paper (Eltabey *et al.*, 2011). It was shown that all the investigated ferrite samples have single spinel phase of the structure before and after irradiation. It was also observed that some peaks intensities changed after irradiation.

3.2. Dc Resistivity At Room Temperature

Figure 1 shows the variation of the dc resistivity (ρ_{dc}) at room temperature with Mg-concentration (x) for $\text{Mg}_x\text{Cu}_{0.5-x}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ ($x = 0.0, 0.2$ and 0.4) before and after irradiation.

3.3. Temperature Dependence of the DC Resistivity

Figure 2-4 show the temperature dependence of the DC resistivity, before and after irradiation, expressed as $\log(\rho_{dc})$ versus $1000/T$ for $\text{Mg}_x\text{Cu}_{0.5-x}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ with $x = 0.0, 0.2$ and 0.4 respectively. **Table 1** gives the determined values of the activation energy in the paramagnetic region, E_p (region III) and of ferrimagnetic one, E_f (region II). For all samples, the first region ranged from room temperature up to nearly 335 K.

3.4. Frequency Dependence of ac Resistivity

The variation of the ac resistivity (ρ_{ac}) as a function of frequency at room temperature, before and after γ -irradiation for the sample with $x = 0$, as an example, is shown in **Fig. 5**. It is observed that, ρ_{ac} decreases with increasing frequency and its value for irradiated samples are lower than that of unirradiated ones.

3.5. Frequency Dependence of Real Part of Dielectric Constant (ϵ')

Figure 6 shows the plotting of ϵ' as a function of frequency at room temperature for the sample with $x = 0.2$, as an example, in $\text{Mg}_x\text{Cu}_{0.5-x}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ ferrite system before and after γ -irradiation. It could be seen that, ϵ' initially decreases rapidly with increasing the frequency then it becomes almost frequency independent at high frequencies i.e., the dispersion is high in the low frequency region.

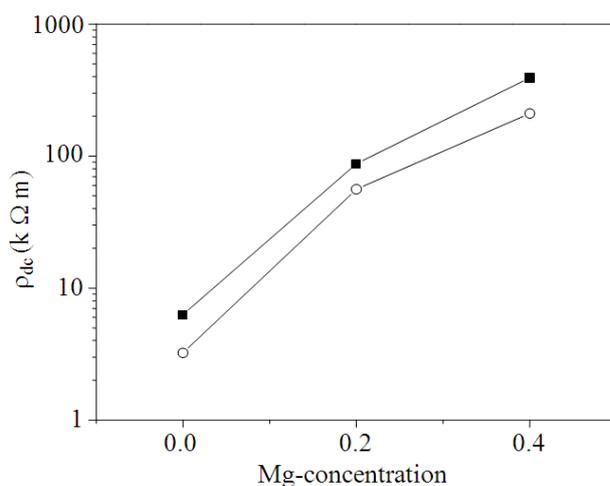


Fig. 1. Dc resistivity at room temperature, after and before irradiation, as a function of Mg-concentration

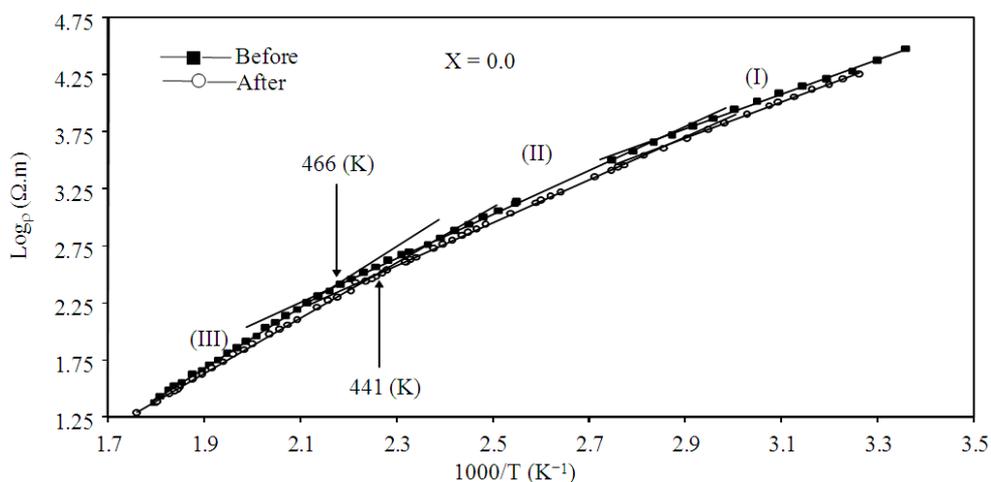


Fig. 2. Temperature dependence of ρ_{dc} for sample with $x = 0.0$ before and after irradiation

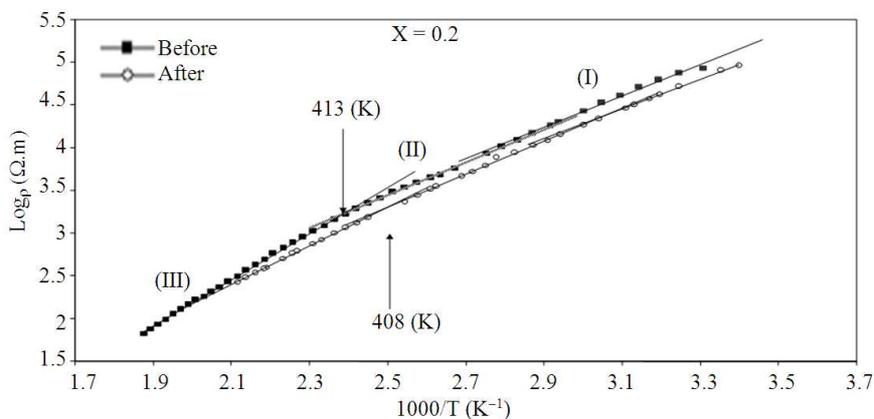


Fig. 3. Temperature dependence of ρ_{dc} for sample with $x = 0.2$ before and after irradiation

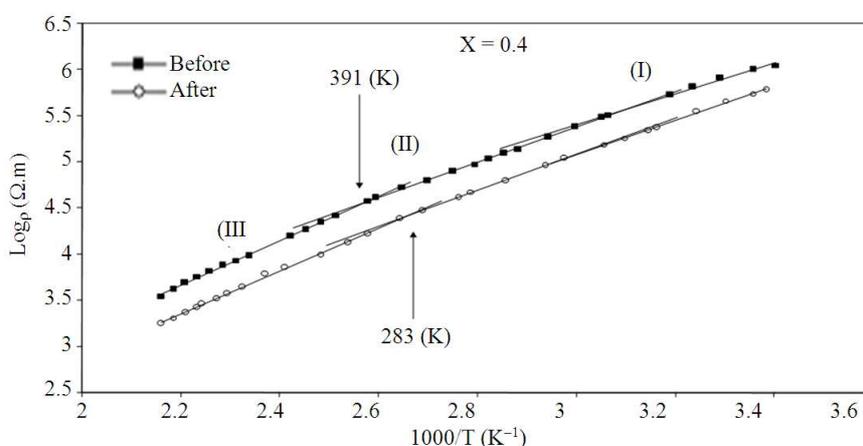


Fig. 4. Temperature dependence of ρ_{dc} for sample with $x = 0.4$ before and after irradiation

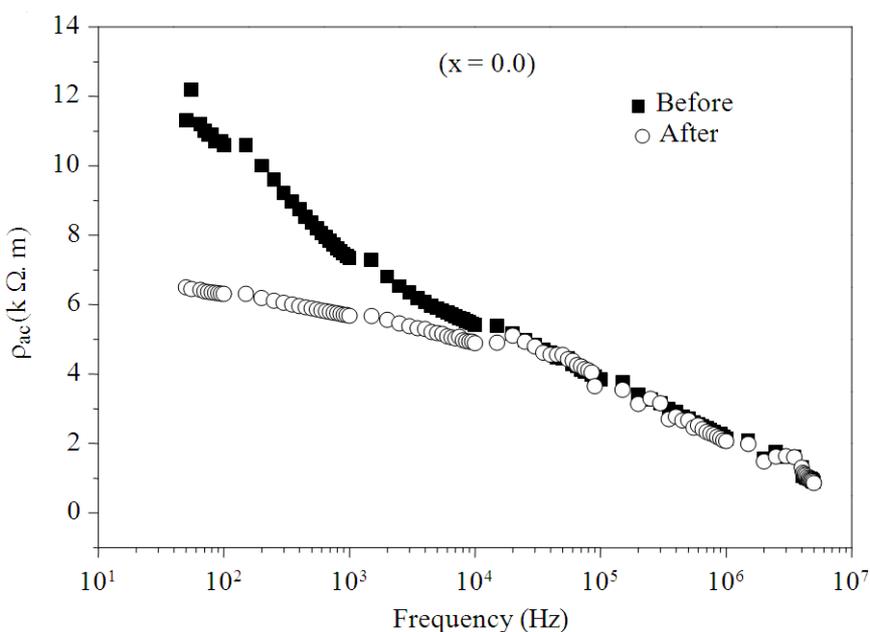


Fig. 5. Variation of the AC resistivity (ρ_{AC}) with frequency for $Mg_xCu_{0.5-x}Zn_{0.5}Fe_2O_4$ sample with $x = 0.0$ of Mg content before and after γ -irradiation

3.6. Frequency Dependence of Imaginary Part of Dielectric Constant (ϵ'')

The frequency dependence for the imaginary part of the dielectric constant ϵ'' for the investigated sample with $x = 0.4$ as an example, before and after γ -irradiation, at room temperature is shown in **Fig. 7**. It is observed that, ϵ'' decreases continuously with increasing the frequency up to a value of 10 kHz and then becomes frequency independent. The value of ϵ'' increased after γ -irradiation for all the investigated samples.

3.7. Composition Dependence of the Dielectric Properties

Figure 8 represents the composition dependence of ρ_{ac} , ϵ' and ϵ'' (at frequency = 300 Hz) for the samples of $Mg_xCu_{0.5-x}Zn_{0.5}Fe_2O_4$ ferrite system, with $x = 0.0, 0.2$ and 0.4 . This figure shows that ρ_{ac} increase with increasing Mg content for all investigated samples. This behavior is similar to that for ρ_{dc} (sec. 3.2). On the other hand, ϵ' and ϵ'' , before irradiation, have almost a reverse trend to ρ_{ac} .

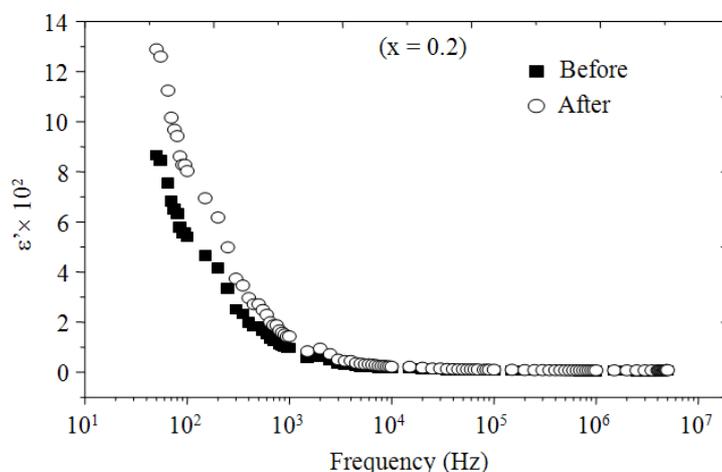


Fig. 6. Frequency dependence of the ϵ' for $Mg_xCu_{0.5-x}Zn_{0.5}Fe_2O_4$ sample with $x = 0.2$ of Mg content before and after γ -irradiation

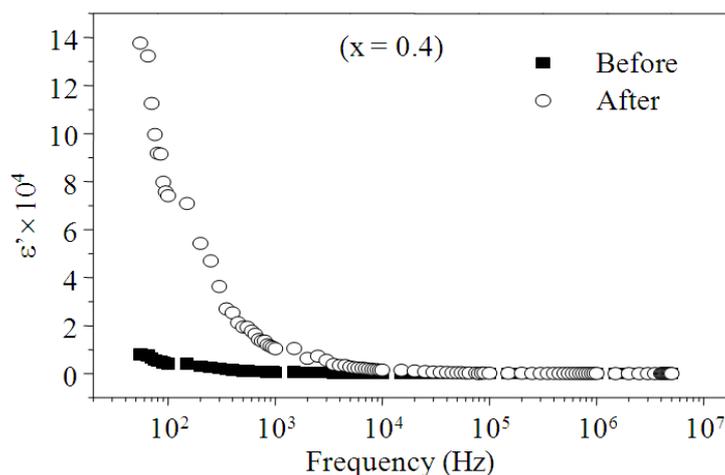


Fig. 7. Frequency dependence of the ϵ'' for $Mg_xCu_{0.5-x}Zn_{0.5}Fe_2O_4$ sample with $x = 0.4$ of Mg content before and after γ -irradiation.

4. DISCUSSION

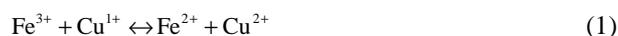
4.1. Morphological structure

The morphological structure and changes in the values of lattice parameter and porosity due to irradiation of the studied samples were discussed elsewhere (Eltabey *et al.*, 2011).

4.2. DC Resistivity

From Fig. 1 it is obvious that, the resistivity increases as Mg-concentration increases and their values for irradiated samples are lower than that of unirradiated ones. According to previous studies, it was reported that

the main factors affecting the ferrites resistivity (ρ_{dc}), are the Fe^{2+} ions concentration and porosity of the samples (Sattar *et al.*, 2007). The decrease in the Fe^{2+} ions concentration limits the hopping probability of the conduction electrons between Fe^{2+} and Fe^{3+} ions in B-sites. On the other hand, the increasing in the sample porosity hinders the motion of the charge carriers. According to (Yue *et al.*, 2001; Murthy, 2001; Maria *et al.*, 2013), in the case of Mg-Zn-Cu ferrite, the B-sites are occupied by both stable Mg^{2+} ions and Fe^{3+} and Cu^{2+} ions. The following equilibrium may exist during the sintering process Equation (1):



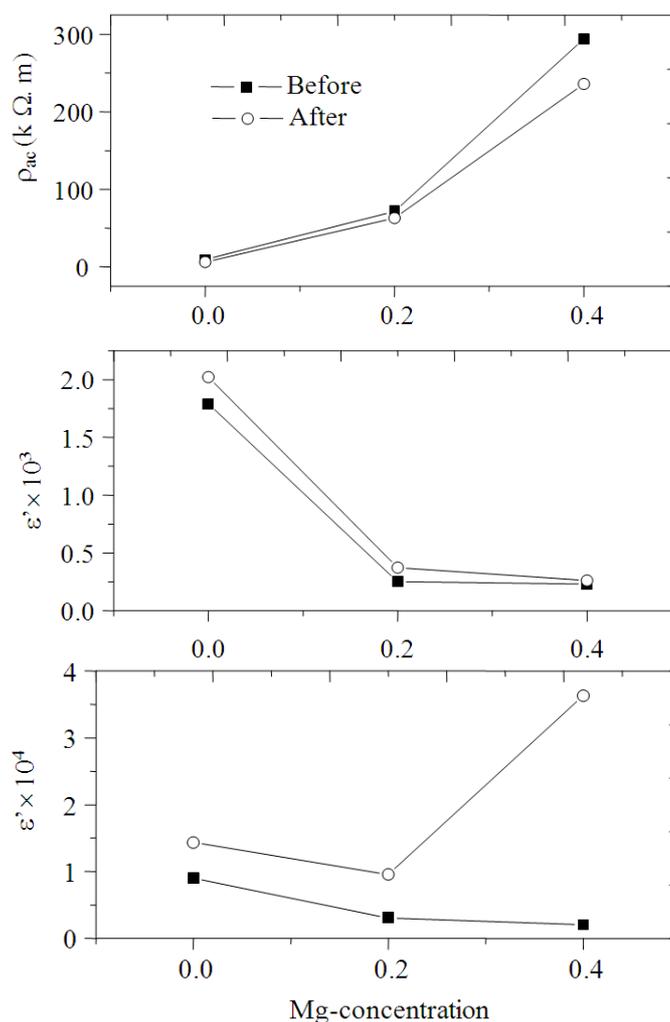


Fig. 8. Composition dependence of the ρ_{ac} , ϵ' and ϵ'' for $Mg_xCu_{0.5-x}Zn_{0.5}Fe_2O_4$ ferrite system $x = 0.0, 0.2$ and 0.4 Mg content respectively, before and after γ -irradiation

Table 1. Numerical values of ferri and para magnetic activation energies (E_f and E_p) before and after irradiation

Mg content	E_f (eV) region (II)		E_p (eV) region (III)	
	Before irradiation	After irradiation	Before irradiation	After irradiation
0.0	0.165	0.163	0.230	0.212
0.2	0.165	0.122	0.232	0.195
0.4	0.169	0.168	0.208	0.192

Under oxidizing conditions, the tendency is towards the right side of the process. Thus, some of the Fe^{2+} ions can be formed in ferrite which increase the probability of electron hopping. Therefore, in $Mg_xCu_{0.5-x}Zn_{0.5}Fe_2O_4$ system, with increasing Mg content, i.e., decreasing the Cu ions content, the probability of Fe^{2+} ion formation

decreases. Consequently, the probability of electron hopping decreases leading to increase the resistivity. The decreasing of ρ_{dc} values by irradiation could be attributed to the increase in the ratio of Fe^{2+}/Fe^{3+} on B-sites (Goldman, 2006). Thus, the decrease of ρ_{dc} after irradiation indicates that the effect of increasing the

Fe²⁺/Fe³⁺ ratio on B-sites dominate that of porosity. Similar results were reported by several authors for different types of ferrites (Hemeda and El-Saadawy, 2003; Hemeda, 2005; Hamada, 2004).

It is obvious from the graphs of temperature dependance of resistivity (Fig. 2-4) that, the electrical resistivity of all samples decreases with increasing temperature, i.e., the resistivity exhibits a normal semiconductor behavior. This behavior could be described by Arrhenius relation (Sattar *et al.*, 2007) Equation (2):

$$\rho = \rho_0 \exp(E_p / kT) \quad (2)$$

where, E_p is the activation energy, k is Boltzmann's constant and ρ₀ is a temperature independent constant. The activation energy is determined by the slope of the fitting lines shown in Fig. (2-4). Moreover, one can notice that there are three linear regions of different activation energies (E_p).

The values of the determined activation energy given in Table 1 refer to three conduction mechanisms of the ferrites samples. The electrical conduction in region (I) is attributed to the presence of impurities i.e., extrinsic conduction mechanism (Ravinder, 2000; Sattar *et al.*, 2005). The formation of such impurities is due to the oxygen loss during the sintering process. The loss of oxygen leads to the formation of Fe²⁺ ions on the account of Fe³⁺ ions for charge compensation. These Fe²⁺ ions act as donor centers (Bhise *et al.*, 1996; Patil *et al.*, 1994; 1996).

It was observed that, for all Mg concentrations, the transition temperature values before and after irradiation (T_p) are close to our previously reported data of the Curie temperature (T_c) that were determined from the magnetic permeability curves (Eltabey *et al.*, 2011). Thus, T_p could be considered the transition temperature between the ferri and para magnetic regions indicating that ρ_{ac} is sensitive to the magnetic transition.

From Table (1) it is clear that, the activation energy in the paramagnetic region E_p (region (III)) are greater than that of ferrimagnetic one E_f (region (II)) for all the investigated samples. This increase in activation energy due to magnetic transition is explained as follows: According to (Goodgenough, 1973), the magnetic transition can be considered as second order one, which is characterized by a large temperature range. This second order transition may be accompanied by volume expansively (Zemansky, 1952), i.e., an increase in the jumping length between the ions and hence an increase in activation energy.

It is also noticed that the values of activation energy decreased after irradiation in every region. This result

could be attributed to generation of some vacancies at different depths, which acts as trapping centers cause a depressing of the jumping length of electrons leading to decrease the activation energy (Ahmed *et al.*, 2003; 2007).

4.3. Ac Resistivity

The results of frequency dependance of ac resistivity shown in Fig. 5 could be explained as mentioned above for the dc resistivity; the conduction mechanism in this system is due to the electron hopping between Fe²⁺ and Fe³⁺ ions which have exchangeable ionization states. Increasing of frequency enhances the electron hopping rate and hence increases the conductivity i.e., decreases the resistivity. At high frequencies, the decreasing rate of ac resistivity with frequency is lower than that at low frequency. This is due to the fact that the electron hopping frequency cannot follow the external electric field and thus lags behind it (Mousa *et al.*, 1989; Bellad and Chougule, 2000; Lipare *et al.*, 2004). The frequency dependence of the resistivity could be explained theoretically using Koops's model (Koops, 1951). According to this model, the polycrystallite ferrite is considered to be composed of two layers: Grains and grains boundaries. The grains are wide and of low resistivity (ρ₁), while the grain boundaries are thin and of high resistivity (ρ₂). According to Koops's assumption that ρ₂ >> ρ₁, y << 1 (where y is the ratio of the grain boundaries thickness to that of the grain), one can write the total impedance as Equation (3):

$$\rho \cong \rho_1 + \left[\frac{\gamma \rho_2}{1 + \frac{b \rho_1 \rho_2 \omega^2}{\gamma}} \right] \quad (3)$$

where, b is constant (Rahman *et al.*, 2012). Thus at very high frequency, the second term can be neglected and the impedance ρ[∞] is given by Equation (4):

$$\rho^\infty = \rho_1 \quad (4)$$

i.e., the resistivity at high frequency originates mainly from grain, which have low resistivity. On the other hand, at very low frequency the impedance ρ⁰ is given by Equation (5):

$$\rho^0 = \rho_1 + \gamma \rho_2 \quad (5)$$

According to the assumption that γρ₂ > ρ₁, the impedance at low frequency results mainly from the

resistivity of the grain boundaries, which have high resistivity. The decrease of ρ_{ac} for the investigated samples after γ -irradiation could be attributed, as mentioned above also with dc resistivity, to the increase of Fe^{2+}/Fe^{3+} ratio (3.2 sec). The decrease of ferrite ac resistivity due to irradiation by γ -rays was reported by different authors (Ahmed *et al.*, 2003; 2007).

4.4. ϵ' and ϵ''

The behavior of ϵ' values as a function of frequency shown in **Fig. 6** is similar to that reported by (Rezlescu and Rezlescu, 1974; Reddy *et al.*, 1999). This behavior of ϵ' could be explained on the basis of the dielectric properties and space charge polarization, which are mainly governed by the conduction mechanism in ferrites (Lipare *et al.*, 2004), wherein the electron hopping takes place. The electron hopping is favorable at low applied field frequency. Therefore, at low frequencies the dielectric constant is higher. The electron exchange between Fe^{3+} and Fe^{2+} gives local displacement of electron in the direction of the applied electric field and consequently induces electric polarization. Beyond a certain frequency, the electron exchange does not follow the alternating field and so the dielectric constant reaches a constant and small value (Wagner, 1913; Murthy and Sobhanadri, 1976; Kharabe *et al.*, 2006; Ajmal and Maqsood, 2008). In addition, from **Fig. 6**, it is noticed that there is a slightly increase in the ϵ' after γ -irradiation. This could be explained in a view of interaction of γ -rays with the matter, which is summarized as follows, gamma irradiation interacts with ferric ions as Equation (6):



This interaction creates ferrous ions at the octahedral sites and increases the ratio Fe^{2+}/Fe^{3+} at these sites (Ahmed *et al.*, 2007; Hemeda and El-Saadawy, 2003; Mousa *et al.*, 1989). The jumping electrons are oriented in the field direction and consequently contribute the rising of ϵ' .

The decrease in ϵ'' could be discussed as follows. The parameter ϵ'' represents the dielectric loss in the ferrites core which is attributed to three main factors; these are eddy current, electric dipole loss and hysteresis loss. The eddy current loss is inversely proportional to the resistivity of ferrite. The resistivity of the ferrite samples is inversely proportional to the frequency which consequently increases the eddy current loss. On the other hand, the dipole loss decreases as the frequency increases, especially at high frequencies, as the dipole orientation cannot follow up the applied field.

Furthermore, it was reported that the hysteresis loss is directly proportional to the frequency (Otsuki *et al.*, 1999). It can be concluded that the decrease in the dielectric parameter ϵ'' is the resultant loss due to these factors. The increase of ϵ'' after γ -irradiation could be attributed to the decrease of the resistivity due to irradiation process. This increases the eddy current loss and then ϵ'' increases which is in a good agreement with the following relation (Lipare *et al.*, 2004):

$$\rho_{ac} = \frac{1}{\epsilon'' \epsilon_0 \omega} \quad (7)$$

4.5. Composition Dependence of ρ_{ac} , ϵ' and ϵ''

The inverse relation between ρ_{ac} and ϵ' has been reported by (Reddy *et al.*, 1999; Bellad *et al.*, 1999). The inverse proportionality with Mg concentration of ρ_{ac} with ϵ' could be explained on the basis of the relation between the mobility of the electron hopping and resistivity Equation (8):

$$\sigma = \frac{1}{\rho} = ne\mu \quad (8)$$

where, n and μ are concentration and the mobility of charge carriers, respectively and e is the electron charge. If the electron exchange between Fe^{2+} and Fe^{3+} ions is easy, i.e., the mobility is large and then it leads the resistivity to decrease. Meanwhile, such exchange causes the polarization to increase i.e., ϵ' increases. Moreover, the reverse behavior of the ρ_{ac} and ϵ'' is expected as the increase of resistivity decreases the loss ϵ'' and vice versa which is in a good agreement with Equation (7).

5. CONCLUSION

- The dc resistivity of the $Mg_xCu_{0.5-x}Zn_{0.5}Fe_2O_4$ system increased as the Mg-concentration increases. After γ -irradiation, the all investigated samples showed lower value of which is attributed to the increase in the ratio of Fe^{2+}/Fe^{3+} on B-sites as consequence of hopping reaction
- The temperature dependence of ρ_{dc} showed three conduction regions with different activation energies. Also, it is noticed that the values of activation energy decreased after γ -irradiation in every region
- Dispersion curves of the dielectric constant ϵ' , dielectric loss ϵ'' and ρ_{ac} with frequency were found to be decreased with frequency for each composition. The values of ϵ' and ϵ'' decreased also

after γ -irradiation and that results were explained in the light of two layers model (Koops's model)

- The increases of the conductivity, dielectric constant and dielectric loss after γ -irradiation is related to the increase of the ratio $\text{Fe}^{2+}/\text{Fe}^{3+}$
- These results reveal that the structure, the magnetic and the electrical properties of Mg-Cu-Zn ferrites are highly affected by gamma rays irradiation

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